ELECTRON IMPACT IONIZATION OF 
NITROGEN AND METHANE

A thesis submitted to The University of Manchester
for the degree of Doctor of Philosophy
in the Faculty of Science and Engineering

2019

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Abstract

This thesis presents the results for experimental (e,2e) studies on both nitrogen ($N_2$) and methane ($CH_4$) molecules. The data from the experiments were compared to theoretical calculations using various models and approximation methods: the molecular three-body distorted wave (M3DW) approximation, the distorted wave Born approximation (DWBA), the Ward–Macek (WM) approximation, the orientation-averaged molecular orbital (OAMO) approximation, the proper averaging (PA) approach, and the generalized Sturmian functions (GSF) approach. The Manchester (e,2e) spectrometer is described in detail along with new experimental hardware and software used for experimental control, optimization, data acquisition, extraction, visualization, and analysis.

Two in-depth (e,2e) studies were carried out for the nitrogen ($N_2$) molecule. The first was carried out in both coplanar and non-coplanar geometries, for incident electron energies ~10 and ~20 eV above the ionization potential of the $3\sigma_g$, $1\pi_u$ and $2\sigma_u$ states of $N_2$. The results used outgoing electron energies with $E_1 = E_2 = 4.6\text{ eV} \pm 0.5\text{ eV}$, $E_1 = E_2 = 9.7\text{ eV} \pm 0.5\text{ eV}$, $E_1 = 4.6\text{ eV} \pm 0.5\text{ eV}$ and $E_2 = 14.5\text{ eV} \pm 0.5\text{ eV}$. The experimental results show that the cross-sections are very sensitive to the states from which the ionization occurs, as well as the incident energy and post collisional interactions. These measurements were compared to calculations based on three models, all of which confirmed the importance of post collision interactions at these energies. The second (e,2e) study was a continuation of the first. It was carried out with incident electron energies ~20 and ~40 eV above the ionization potential for the $3\sigma_g$ and $1\pi_u$ states of $N_2$, using equal energies for the outgoing electrons. The data were obtained with the incident electron beam in the scattering plane. This experiment produced six sets of measurements of the ionization triple differential cross-sections, with fixed angles of 45°, 90° and 125° with respect to the incident electron beam for one of the outgoing electrons. A disagreement was found between theory and experiment which is thought to be due to limitations of the OAMO technique used to average over all molecular orientations.

The final (e,2e) study detailed in the thesis was carried out for ionization of the highest occupied molecular orbital (HOMO) $1\epsilon_2$ state and the next highest occupied molecular
orbital (NHOMO) 2a_1 state of CH_4 at an incident electron beam energy of 250 eV, for
ejected electron energies of 50 eV and 30 eV. Five fixed scattering angles (20°, 22.5°, 25°,
27.5°, and 30°) were used in these coplanar asymmetric experiments. The measured triple
differential cross-sections were compared to experimental results of the Afyon group in
Turkey. Both experimental results were also compared to theoretical calculations using
the M3DW and GSF models. The results showed very good agreement between both
experiment and theory. The structure and peak locations in the results for 50 eV ejected
electrons are in good agreement with the theoretical calculations. However, the agreement
at 30 eV was not as satisfactory. The M3DW calculations were slightly better in their
predictions of the location and magnitude of the experimental peaks than the GSF model.

A significant portion of this thesis details the modernization of the Manchester (e,2e)
spectrometer that was carried out by the author, and which was then used to perform
the experiments that are detailed here. New computer-controlled variable voltage supplies
were designed, built, and integrated with the (e,2e) spectrometer, alongside the new (e,2e)
LabVIEW computer application used for experimental control and data acquisition. The
inner-workings of the (e,2e) spectrometer, the vacuum system, and the experimental
hardware and software are all detailed in the thesis.
Declaration

I hereby declare that no portion of the work referred to in the thesis has been submitted in support of an application for another degree or qualification of this or any other university or other institute of learning.

1 November 2019, Manchester, UK

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To my parents Abdulghani and Fairouz.
For their love, selfless efforts, sacrifices, and guidance.
First and foremost, I would like to thank my supervisor, Professor Andrew James Murray, for giving me the opportunity to join his group and to work on the Manchester (e,2e) coincidence experiment. I am grateful for his guidance, for giving me the opportunity to attend international conferences, for inspiring me to pick up the guitar, and for all the coffee breaks and barbecues. He has shown me what it means to be an experimental physicist. Professor Murray is and will continue to be an inspiration to me.

I would also like to thank Professor George King for his continued support, the many helpful discussions, and his interest in my well-being. A special thanks to Dr. Matthew Alexander Harvey for being the best colleague one could ever have. He is a brilliant problem solver who I learned a lot from. I am grateful for all the ways he has helped, supported, and guided me throughout my PhD. I am very fortunate to have him as a friend. Many thanks go to his wife Alison, and their two amazing children Adam and Matilda, for all the warmth and kindness they have shown me. I am thankful for all the useful discussions and fun times I had with others in the Atomic & Molecular Physics group, in particular James Pursehouse and Michael Jones, as well as Parinya Udommai and Manish Patel. I would like to thank the University of Manchester for this wonderful opportunity, the many resources, and for the overseas scholar award that helped make this academic journey possible. Many thanks go to the theoreticians involved in the work carried out during my PhD.

I am extremely grateful for my parents Abdulghani and Fairouz, for their love, support, sacrifices, patience, and for giving me the opportunities that led me here. I would not be here if it wasn’t for them. I am thankful for my sisters and best friends Marwah and Zenah for believing in me and cheering me on throughout my journey. I would also like to thank my former supervisor at CSU Fullerton, Professor Murtadha Khakoo, for exposing me to fundamental research, involving me in the entire scientific process, and for giving me the research opportunity that led me to pursue my PhD at the University of Manchester. I am grateful for his continued guidance and support.

Last but not least, I would like to acknowledge Reality itself, for its sphere of knowledge and frontiers that inspire the mind to seek the unknown.
CHAPTER 1

Introduction

1.1 Introduction

The greater part of scientific endeavor, research, and discovery in physics is essentially the study of matter; the substance that makes up 4.6% of the universe. Most of what physicists know about the universe comes from the fundamental studies of matter-matter and light-matter interactions. The Manchester (e,2e) coincidence experiment is one such example; a method used to extract information on the structure of an atomic or molecular target through electron impact with and ionization of the target itself. This experiment gives physicists an in-depth examination of the angular correlations that arise between the post-collision scattered electron and the ejected valence shell (or inner-shell) electron. A wealth of information and data can be collected due to the experiment’s accessibility to all possible geometries.

1.2 Kinematics of (e,2e) Reactions

The analysis of the single event processes that take place in the ionization of atoms and molecules by electron impact are very subtle and quite challenging to understand. The difficulty lies in dealing with the physics of a many-body problem as well as the infinite range of the Coulomb interactions that arise from the particles involved in the collision. The following section will shed light on the physics of (e,2e) reactions and how information is obtained on the structure of atoms and molecules in (e,2e) coincidence experiments.

Understanding the kinematics of (e,2e) reactions begins by concentrating on the most basic process of ionization by electron impact. Let us first consider the (e,2e) reaction seen in Figure 1.1. In the initial phase of the interaction, a beam of electrons of energy $E_0$ and momentum $k_0$ is incident on an atomic or molecular target. Following the collision at the interaction region is the second phase, in which two emerging electrons with momenta $k_1$ and $k_2$ and corresponding energies $E_1$ and $E_2$ are detected in coincidence by energy analyzers at angles $\xi_1$ and $\xi_2$ [1, 2]. Therefore, electron impact ionization of an atomic or
molecular target $A$ can be described by the following reaction:

$$e_0(E_0, k_0) + A \rightarrow e_1(E_1, k_1) + e_2(E_2, k_2) + A^+(Q)$$  \hspace{1cm} (1.1)

where $e_0$ is the incident electron, $e_1$ and $e_2$ are the outgoing detected electrons, and $Q$ is the recoil momentum of the target $A$. Through ionization, the target shifts from the ground state to an ionic state by a separation energy $\varepsilon$ which meets the energy conservation condition:

$$\varepsilon = E_0 - E_1 - E_2 = E_0 - E$$  \hspace{1cm} (1.2)

where $E = E_1 + E_2$, and the energy of the incident electron $E_0$ must be high enough to ionize the target. The ionization potential is simply:

$$E_0 = E_1 + E_2 + IP$$  \hspace{1cm} (1.3)

The recoil momentum $Q$ of the ion can be found by conservation of momentum in the rest frame of the initial target:

$$k_0 = k_1 + k_2 + Q$$  \hspace{1cm} (1.4)

It is important to note that the recoil momentum $Q$ is separate from the momenta ($k_1$ and $k_2$) transferred by the incident electron $e_0$ to the atomic or molecular target $A$ [3]. The $(e,2e)$ coincidence experiment is kinematically complete since the momenta $k_0$, $k_1$, and $k_2$, and the recoil momentum $Q$ are all determined and known respectively. The detection plane shown in fig. 1.1 is the plane formed by the trajectories of the scattered and ejected electrons, where $\Psi$ measures the deviation from the coplanar geometry.

The kind of information one can extract from such ionization processes is done through the measurement of the probability that an incident electron of energy $E_0$ and momentum $k_0$, will produce a scattered electron and an ejected electron of respective energies $E_1$ and $E_2$, and momenta $k_1$ and $k_2$; outgoing electrons that are emitted into their respective solid angles and detected in coincidence with each other. Such a measurement produces triple differential cross sections (TDCSs), from which one can obtain single and double differential cross sections, as well as total ionization cross sections that depend only on the energy of the incident electron, $E_0$ [2]. It is clear by now that the energy and momentum conservation are the imposed constraints to the kinematics of $(e,2e)$ reactions, mainly because the $(e,2e)$ coincidence experiments described in this thesis do not account for (or
1.2 Kinematics of (e,2e) Reactions

Figure 1.1: A drawing of the (e,2e) coincidence detection plane. The incident electron beam has an energy $E_0$ and momentum $k_0$, and is directed at an angle $\Psi$ with respect to the detection plane. The scattered (or ejected) electron has an energy $E_1$ and momentum $k_1$, and is detected at an angle $\xi_1$ from the $z$-axis. The ejected (or scattered) electron has an energy $E_2$ and momentum $k_2$, and is detected at an angle $\xi_2$ from the $z$-axis. The coplanar and perpendicular geometries are at angles where $\Psi = 0^\circ$ and $\Psi = 90^\circ$ respectively. Another non-coplanar geometry used in the experiments described in this thesis is when the electron gun is moved to an angle $\Psi = 45^\circ$. The gas jet delivers the atomic or molecular target to the interaction region.

measure) the spin of any of the particles involved in the interactions. These constraints allow the 'scattered' and 'ejected' electrons to carry a range of different energies and momenta. Therefore, for an incident electron with energy $E_0$, the ionization of the atomic or molecular target is usually described by a triple differential cross-section (TDCS):

$$\text{TDCS} = \frac{d^3\sigma}{dE_1d\Omega_1d\Omega_2}$$  \hspace{1cm} (1.5)

where the solid angles are given by $d\Omega_1$ and $d\Omega_2$ which are essentially defined by the momenta of the outgoing 'scattered' and 'ejected' electrons and the acceptance solid angle of the electron energy analyzers. The energy of one of the outgoing (scattered or ejected) electrons is represented by $E_1$.

The experimental measurement of cross-sections is carried out by placing constraints on the number of independent variables, to yield a number of arrangements or kinematic geometries in which the (e,2e) collisions occur. For example, one such collision could be in the coplanar geometry; an occurrence on the same plane between the momenta $k_0, k_1,$
and $k_2$. Another important arrangement is when the collision occurs in a perpendicular geometry, where the gun is placed at $90^\circ$ with respect to the plane at which the outgoing electrons are being detected. Finally there are symmetric and asymmetric geometries; asymmetric is where a fast scattered electron $E_1$ is detected in coincidence with a slow ejected electron $E_2$, and symmetric geometries are where the angles $\xi_1$ and $\xi_2$ of the outgoing electrons $E_1$ and $E_2$ are equal as well as their respective energies $E_1$ and $E_2$ [4]. The other possible kinematic conditions is where the outgoing electrons $E_1$ and $E_2$ are equal in energy but are detected asymmetrically where $\xi_1 \neq \xi_2$, or the opposite where $E_1 \neq E_2$ and $\xi_1 = \xi_2$.

1.3 A Brief History of (e,2e) Spectroscopy

Electron impact ionization studies using the (e,2e) coincidence techniques have been successfully carried out for the last fifty years [5]. The purpose of these experiments was twofold: to obtain information on the fundamental interactions and correlations that arise from single ionization of an atomic or molecular target, and to provide direct measurements of the energy-momentum density of electrons using the methods of electron momentum spectroscopy (EMS) [6, 7]. In addition to these experiments, various methods and techniques were developed to tackle few and many-body systems such as the Frankfurt Cold Target Recoil Ion Momentum Spectroscopy (COLTRIMS) technique [8, 9]. The results of many of the experiments carried out in this time period were compared to theory and used to improve and develop new quantum mechanical models.

The first (e,2e) coincidence experiments were carried out and published in 1969 by Ehrhardt et al. (Germany) [5] and Amaldi et al. (Italy) [10]. The work of Ehrhardt was an investigation of the ionization process by measuring the angular correlation of the scattered and ejected electrons by electron impact ionization of Helium. These (e,2e) coincidence measurements were carried out using ionizing incident energies of 50 eV and 114 eV, which also determined the energies of the "scattered" and "ejected" electrons. These results were compared to the solutions of the Lippmann-Schwinger equation, which found that the angular correlation distributions were more sensitive than the energy loss from electron impact ionization [5]. Several attempts were made to explain the obtained experimental results for helium with little to no success. The Born approximation [11–13], along with the inclusion of Coulomb effects in various ways [14–16] all failed in predicting the experimental results. The models usually failed to predict the ratio of the magnitude of the peaks (recoil vs. binary encounter peak), and/or the directions of these peaks [3]. These challenges are due to the distortion of electron waves from the assumed plane waves or Coulomb waves; challenges that become the motivation for the development of more sophisticated
Theoretical models.

The experiments carried out by Amaldi and co-workers focused on studying the electronic structure of the target, in contrast to the ionization dynamics studied by Ehrhardt. The methods used by Amaldi were carried out to determine the one-electron momentum density of the target in question. These experiments used incident energies in the 0.1–10 keV regime, two hemispherical electron energy analyzers with a high energy resolution and a short resolving time of 4.3 ns, and two channel electron multipliers for electron detection. The energy spectra in the results contained a well resolved peak due to the K shell electrons in carbon and oxygen [10].

Following the results of Ehrhardt and Amaldi in 1969, Weigold and McCarthy carried out experiments that laid down the foundations of electron momentum spectroscopy (EMS). These experiments exploited the ideal conditions of (e,2e) coincidence reactions which enabled direct measurements of electron energy-momentum densities for specific electronic states of the target ion. The experiments measured the difference between the electrons’ incident and total final energies and momenta. This measured difference gave the separation or binding energy for the orbitals from which the electrons are ejected, as well as the correlation between the incident and final states of the system. The results of these experiments showed the sensitivity of the ionization differential cross-section measurements to that of the electron energy-momentum densities [7].

As mentioned above, various experimental methods and techniques were developed to tackle few and many-body systems such as the Cold Target Recoil Ion Momentum Spectroscopy (COLTRIMS) technique. COLTRIMS is essentially a momentum microscope for scattering experiments; an imaging technique that allows for the measurement of the momenta of the recoiling target ions and electrons that result from atomic or molecular reactions. These measurements are obtained in a three dimensional momentum vector space using analyzers with a high resolution and 4\(\pi\) solid angle. The studies carried out using the COLTRIMS technique yield results that provide complete and detailed pictures of the dynamics of atomic and molecular processes [8]. The success of COLTRIMS led to the development of another technique known as the reaction microscope. This technique allows for an investigation of the dynamics of many particle systems under external electromagnetic fields [9].

1.4 The Manchester (e,2e) Coincidence Experiment

The Manchester (e,2e) apparatus is comprised of an (e,2e) spectrometer and vacuum system, experimental hardware, and experimental software. The (e,2e) spectrometer is composed of an electron gun, two electron energy analyzers, a Faraday cup, and an atomic
or molecular target beam source. The electron gun consists of an electron source, and a two-stage electrostatic lens system that can deliver a collimated electron beam to the interaction region [17]. The electron gun's beam energy can vary between 20 eV to 300 eV, with a beam current up to 5 μA. Each of the two electron energy analyzers use a triple cylindrical lens system to detect the scattered and ejected electrons (that are detected in coincidence), by imaging them onto the entrance aperture of a hemispherical energy selector, which guide the energy selected electrons onto the entrance slit of a channel electron multiplier (for detection) [18]. The Faraday cup, which is positioned opposite to the electron gun, measures the current of the electron beam and reduces the number of scattered electrons from the interaction region. The measurements of electron-impact ionization coincidence events are carried out in a coplanar or a non-coplanar geometry. These measurements are possible because the electron gun can be rotated out of the detection plane spanned by the electron energy analyzers. The experimental hardware is composed of a set of power and voltage supply units, control and data acquisition modules, and coincidence detection electronics. The experimental software is a series of applications and routines designed for control and optimization of the (e,2e) spectrometer, and data acquisition and analysis.

1.5 An Overview of Manchester (e,2e) Coincidence Experiments

The motivation behind developing the Manchester (e,2e) coincidence experiment, which began in 1982, was to study the angular correlations that arise between the post-collision scattered electron and the ejected valence shell (or inner-shell) electron. A number of experiments (including the ones discussed in this thesis) have been carried out over the past 3+ decades including several modifications and upgrades.

1.5.1 The Manchester (e,2e) Coincidence Experiment (1982-1990)

The experiment was first commissioned and designed by Hawley-Jones (1984) [18]. The first series of experiments were carried out using a hemispherical energy selected electron gun, which studied the Wannier effect by measuring close to threshold ionization cross-sections in the perpendicular plane geometry [19].

These experiments were followed by the work of M.B.J. Woolf (1989) [17] who designed and used an unselected electron gun to study helium by measuring the angular correlated differential cross-sections. The experiments were performed in the perpendicular plane with incident energies that ranged from 10 to 80 eV above the ionization potential, and equal energies for the outgoing "scattered" and "ejected" electrons.

Following these experiments, experimental computer-controlled hardware and software
were designed and installed by B.C.H. Turton (1990), mainly to perform automated optimization routines during (e,2e) coincidence scans. The experimental hardware and software were tested by performing scans to optimize the signal from the resonance states found in helium [20].

1.5.2 The Manchester (e,2e) Coincidence Experiment (1990-2002)

Following the experiments mentioned above, the (e,2e) apparatus’ hardware and software underwent considerable modifications, that would allow the apparatus to be fully computer-controlled and optimized to perform (e,2e) coincidence experiments 24 hours a day. In addition, significant improvements were made to increase the timing resolution and efficiency of the electron energy analyzers. Full details on the computer-control experimental hardware can be found in [21]. These modifications and improvements, including the experiments detailed below were carried out by Andrew James Murray.

Several experiments in the perpendicular plane were performed using the computer-controlled apparatus to:

1. Test the optimization routine and verify the experimental results obtained by Woolf.
2. Provide a comparison of experimental results obtained using the computer-controlled optimization routine with those from manually optimized experiments.

The results verified the efficiency of the fully computer-controlled system and showed the significant improvements in the obtained data when compared to the results obtained from manual operation.

Following the verifications of the computer control systems mentioned above, data were obtained for equal and non-equal energies in the perpendicular geometry with incident energies ranging from $34.6 \, eV$ to $104.6 \, eV$ [22]. These results were then coupled with symmetric differential cross-section measurements obtained in the coplanar geometry with incident energies ranging between $44.6 \, eV$ and $74.6 \, eV$ [23, 24]. The results from these studies were then parameterized in terms of a set of orthogonal angular functions that define the correlation between the momenta vectors of the incident, scattered, and ejected electrons [25–27]. Additional experiments were carried out to measure doubly-symmetric helium ionization cross-sections with excitation of the residual ion in a coplanar geometry; studies that confirmed the reliability, stability, and advantages of the fully computer-controlled (e,2e) apparatus [28].

Further work on helium was carried out in collaboration with Dr. Nick Bowring (1997-1999). The first of those studies were for near-threshold doubly symmetric (e,2e) measurements on helium [29]. Following these measurements, the dips and backward peaks
in the helium (e,2e) differential cross-section at low energies were detailed [30]. Lastly, the unexpected sharp dip in the helium differential cross-sections at 64.6 eV was explored using asymmetric angles for the outgoing 'scattered' and 'ejected' electrons. This study proposed a two-electron interference model to explain the existence of the dip with a constant angle between the outgoing electrons over a wide range of asymmetric angles [31].

Following these experiments, (e,2e) differential cross-section measurements were obtained for neon, in the low energy regime in both coplanar and non-coplanar geometries [32]. A comparison study was then performed between the differential cross-section measurements of argon and helium at 64.6 eV using equal energies and symmetric angles for the outgoing electrons [33]. Lastly, a symmetric non-coplanar (e,2e) study was carried out to explore the deep narrow interference minima for helium and neon [34].

1.5.3 The Manchester (e,2e) Coincidence Experiment (2002-2007)

Experimental studies in this period of time were carried out by Murray, Cvejanovic, and Hussey. The first of those studies was on (e,2e) differential cross-section measurements of the 3σ_g and 1π_u molecular orbitals of N_2 at incident electron energies between 25.6 and 76.7 eV [35]. An atomic beam oven for combined laser and electron impact ionization experiments was also designed at this time [36], as well as a non-magnetic translator for use in high vacuum systems [37], an automatic controller for filling and maintaining liquid nitrogen levels in dewars [38], and a non-magnetic high accuracy linear translator for use in high vacuum were also designed and built as part of an ongoing development program to study ionization from laser excited atoms [39]. Several studies were then carried out for calcium: single ionization by electron impact [40], low energy (e,2e) super-elastic scattering from calcium’s 4^1P_1 state [41], and low energy symmetric (e,2e) cross-section measurements in the coplanar geometry at incident energies that varied between 10.1 to 64.6 eV [42]. Additional (e, 2e) studies were also conducted on H_2 from the X^1Σ_g^-  H_2^+ ground state at incident energies ranging from 10 eV through to 40 eV [43]. Ionization differential cross-section measurements were then carried out for the 1π_g and 4σ_g orbitals of the CO_2 molecule at low incident energies ranging from 10 eV to 80 eV above the ionization potential [44]. An extensive (e,2e) survey was then conducted for Na, Mg, K and Ca from near-threshold to intermediate energies [45]. This was followed by coplanar asymmetric experimental and theoretical studies on hydrogen at an incident energy of 75.3 eV [46]. Lastly, (e,2e) ionization cross-section measurements were carried out for the 3σ_g and 2σ_u^* states of the N_2 molecule to compare the experimental results with the theoretical predictions of the effects of two-centre interference effects from a diatomic target [47].
1.5.4 The Manchester (e,2e) Coincidence Experiment (2007-2015)

The studies in this period of time were mainly an investigation of the $H_2$ and $H_2O$ molecules. The first of these studies was an investigation of Young’s double slit type interferences via electron impact ionization of various molecules at low to intermediate energies [48]. This was followed by electron impact ionization studies in the low to intermediate energy regime from the $1b_1$ state of $H_2O$, in a coplanar geometry using symmetric and asymmetric kinematic conditions [49]. A publication was submitted to Nature Physics in which it highlighted the significance of electron impact ionization experiments and quantum mechanical calculations, and their significance in constructing accurate pictures of such processes [50]. This was followed by 35.4 eV electron impact ionization studies that reported differential cross-section measurements of oriented $H_2$ molecules using a non-perturbative close-coupling technique [51]. An experimental and theoretical electron impact ionization study was then carried out which explored the deep interference minima by measuring triple differential cross-sections in non-coplanar geometries for small atoms and molecules [52]. An additional study on $H_2$ molecules was carried out in which the triple differential cross-sections are measured for equal and unequal outgoing electron energies [53]. Low energy symmetric electron impact ionization studies from the $3a_1$ state of the $H_2O$ molecule were carried out in coplanar and non-coplanar geometries [54]. Lastly, electron impact ionization cross-section measurements were reported for the $H_2$ molecule for outgoing electron with energies of 1 to 10 eV [55].

1.5.5 The Manchester (e,2e) Coincidence Experiment (2015-2019)

New (e,2e) studies and hardware and software developments carried out during this time period are detailed in this Ph.D. thesis. The new experimental hardware, upgrades, and modification are found in chapter 5. The new experimental computer applications, scripts, and routines are detailed in chapter 6. New (e,2e) studies on nitrogen and methane molecules are then detailed in chapter 8 and chapter 9, respectively.

1.6 The New Manchester (e,2e) Coincidence Experiment

The purpose of the work carried out during the course of this Ph.D. was twofold: modernization of the (e,2e) coincidence experimental hardware and software, and conducting experimental electron-impact ionization coincidence studies on the nitrogen ($N_2$) and methane ($CH_4$) molecules to test fundamental quantum theories used to describe atomic and molecular interactions.

The modernization of the Manchester (e,2e) coincidence experiment required the development of new computer-controlled voltage supply units for the electron gun and electron
energy analyzers, a computer-controlled stepper motor and data acquisition unit, and a set of computer applications for voltage supply control, optimization, measurement, data acquisition, and data analysis. The development phase included the design of the power and voltage supply circuitry, the making of printed electronic circuit boards, the design and implementation of algorithms for the development of experimental control computer applications in LabVIEW [56], code for Arduino-controlled voltage supplies [57], and scripts for data extraction, analysis, and visualization [58].

The (e,2e) studies on the \( N_2 \) and \( CH_4 \) molecules were carried out in both the low and intermediate energy regimes, using both equal and non-equal outgoing electron energies, symmetric and asymmetric conditions, and a range of geometries from the coplanar to the perpendicular plane. The work was carried out in collaboration with several theoreticians, whose models were tested against the obtained experimental results. The studies resulted in four publications (with one pending as of September 2019) [59–61].

1.7 Layout of Thesis

This thesis is divided into four main sections. The first section is composed of two chapters which give an introduction to (e,2e) coincidence experiments and the theoretical frameworks used to describe (e,2e) reactions. The second section consists of five chapters which detail all components of the (e,2e) apparatus, including a comparison between the previous and modernized setup, as well as the experimental procedures for operating the modernized (e,2e) experimental hardware and software. The third section is comprised of two chapters which detail the experimental (e,2e) studies on the nitrogen (\( N_2 \)) and methane (\( CH_4 \)) molecules. The final section contains the conclusion and a brief overview of current and prospective projects, as well as potential upgrades that could be made to the modernized (e,2e) apparatus.

Following this chapter, chapter 2 gives an overview of electron collisions including the types of scattering found in atomic and molecular interactions, and brief introductions to the theoretical models that were tested against the experimental results discussed in chapter 8 and chapter 9. These models include:

- The distorted wave Born approximation (DWBA).
- The molecular three-body distorted wave (M3DW) approximation.
- The orientation averaged molecular orbital (OAMO) approximation.
- The Ward–Macek (WM) approximation.
- The Generalized Sturmian Functions (GSF) approach.
Chapter 3 is an overview of the fundamental components making up the (e,2e) apparatus, and also introduces chapters 4 to 7. This chapter gives a comparison between the previous and modernized (e,2e) apparatus setup, highlights the modernized components of the (e,2e) apparatus, and the reasons for such modifications, replacements, and/or upgrades.

Chapter 4 is an overview of the entire (e,2e) vacuum system, with details on the components and inner workings of the (e,2e) spectrometer’s electron gun, electron energy analyzers, Faraday cup, and gas jet.

Chapter 5 details all components, modules, and units making up the modernized experimental hardware. The chapter details how the printed circuit boards (PCBs) were made, and covers the circuit designs and schematics used to build the (e,2e) spectrometer power and voltage supply units, the Arduino-based control boards, and the stepper motor control and data acquisition unit. It gives an overview of the hardware carried over from the previous control systems to the new setup; the constant current filament supply, the detection electronics, and the channel electron multiplier EHT supplies.

Chapter 6 details the design of the new computer applications and programs used for experimental control, voltage supply control, communication, and data exchange, extraction, analysis, and visualization. The three main sections in this chapter cover the (e,2e) LabVIEW computer application [56], the programmable Arduino micro-controllers [57], and the use of Python and Jupyter notebooks for data analysis [58, 62].

Chapter 7 details the experimental procedures used to prepare the vacuum system, operate and optimize the (e,2e) spectrometer, perform (e,2e) coincidence and rate scans, and analyze the data generated by the (e,2e) coincidence experiment.

Chapter 8 gives an introduction to the nitrogen molecule, and then details the (e,2e) coincidence studies that were carried out on molecular nitrogen which resulted in two publications in the Journal of Physics B: Atomic, Molecular and Optical Physics [59, 60].

Chapter 9 discuss the electron-impact ionization studies on the methane (CH$_4$) molecule at intermediate incident energies, with comparisons to experimental results from another experimental group in Afyon, Turkey, and with theoretical calculations based on the M3DW and GSF models. The work discussed in this chapter was part of an international collaboration that resulted in publication in the Journal of Chemical Physics [61].

Chapter 10 concludes and summarizes the contents of the thesis, and gives a brief review of current and prospective projects, and potential upgrades and/or modifications that could be carried out on the modernized (e,2e) apparatus.
CHAPTER 2

Theory

2.1 Introduction

Speculations on the ambiguous nature and properties of the electron stretch back to the time of the ancient Greeks. The earliest interactions with this particle were through one of its properties: its electric charge. It was not until 1838, when Richard Laming proposed the first hypothesis on the concept of a unit electric charge, an atomic core, and the significance of this charge to the chemical properties of the atom it occupies [63, 64]. This electric charge was then named an \textit{electron} in 1891 by George Johnstone Stoney [65]. The first discovery of the electron was in 1897 by J. J. Thomson with his colleagues John S. Townsend and Harold A. Wilson; a result of an exploration of the properties of cathode rays [66]. Thomson’s work suggested the relative size of the electron to that of the hydrogen atom, as well as its unchanging mass regardless of the type of atom it occupies. This discovery was monumental as it led to the development of the first model of the atom by Ernest Rutherford in Manchester (1911) [67].

An electron \( (e^-) \) is thought of as a point like (dimensionless) elementary particle belonging to the first generation of the lepton family of particles. The mass of an electron is \( \frac{1}{1836} \) of the proton and is approximately \( 9.10938356(11) \times 10^{-31} \) kg [68]. It has an intrinsic angular momentum (spin) of \( \frac{1}{2} \), and carries a negative electric charge of \( -1.6021766208(98) \times 10^{-19} \) C which generates a surrounding electric field. The properties that an electron exhibits allow it to participate in a number of interactions, one of which is electromagnetic. This kind of interaction is the most relevant to the theoretical models and experiments detailed in this thesis.

This chapter will detail the interactions of electrons with other electrons, atoms and molecules, the forces and processes that contribute to these interactions, and the collision and scattering types by which experiments such as the Manchester \((e,2e)\) coincidence experiment are carried out. This is followed by highlighting the complexities that arise when dealing with n-body problems, and the motivation for the development of sophisticated theoretical models that can predict the result of atomic and molecular interactions and
reactions. This is then followed by covering the theoretical models used in conjunction
with the (e,2e) coincidence experiments detailed here.

2.2 Electron Collisions

Electron collision studies are fundamental to the understanding of nature, and to many
processes in science and industry. These studies provide information on the structure of
atoms and molecules, due to the many interactions that occur between them. A complete
understanding of these interactions are gained via experiments that allow the development
and fine-tuning of theoretical models that predict the behavior of these processes. Such
models can then be applied to an array of different uses in science and industry.

Electron collisions with atoms and molecules must be considered carefully due to the
many interactions that occur between the incident electron and the atomic or molecular
target. These interactions occur prior to, during, and after the collision with the target.
The first of these interactions to consider is the long range Coulomb interaction between
a single incident electron and an atomic or molecular target as shown in fig. 2.1. This
interaction behaves according to Coulomb’s inverse-square law which is a measure of the
electrostatic force between charged particles [69]. The next thing to consider is the time it
takes for an electron to interact with an atomic or molecular target. This amount of time
is dependent on the energy of the electron. If the incident electron has an energy of 250 eV,
then the incident electron is traveling with a non-relativistic velocity of:

\[ v_{250 \text{ eV}} = \sqrt{\frac{2E_{\text{inc}}}{m_e}} = \sqrt{\frac{2 \times 250 \times 1.6 \times 10^{-19}}{9.1 \times 10^{-31}}} \approx 9.4 \times 10^6 \text{ m/s} \] (2.1)

If the electron is traveling an interaction distance of 100 Å, then the time for the interaction
2.2 Electron Collisions

to occur is:

\[
t_{250 \text{eV}} = \frac{100 \times 1 \times 10^{-10}}{9.4 \times 10^6} \approx 1.1 \times 10^{-14} \text{sec}
\]

(2.2)

which is approximately 11 fs. The short interaction times such as the one calculated above, lead to coherent excitation of target states. The energy (and interaction time) an electron travels with can limit the effects of the interactions it encounters prior to, during, and after the collision with an atomic or molecular target. However, the simplest of targets are an n-body problem, with each body exerting its own electrostatic force on other bodies in the system. For this reason the interaction of an electron with an atom or molecule cannot be solved exactly. An example of such an interaction is shown in fig. 2.2, where the incident electron is interacting with the atomic nucleus and its orbiting electrons.

![Diagram of electron interactions](image)

**Figure 2.2:** An n-body interaction problem where the incident electron \(e_1\) experiences repulsive electrostatic forces \(F_{12}\) and \(F_{13}\) with electrons \(e_2\) and \(e_3\) respectively. The incident electron \(e_1\) experiences an attractive electrostatic force \(F_{1N}\) with the nuclei of the atomic or molecular target. Repulsive electrostatic forces between the bound electrons \(e_2\) and \(e_3\), as well as the attractive forces between the bound electrons and the nucleus are indicated by arrows.

For complex atoms and molecules such as molecular nitrogen \((N_2)\) and methane \((CH_4)\) where the number of bound (core and valence) electrons are much higher than those shown in fig. 2.2, it becomes more difficult to theoretically solve such n-body problems. For an accurate solution, the theoretical models would have to account for all electron-electron and electron-nuclear interactions. Such a solution is physically impossible to obtain at the present time due to computational and processing limitations. For this reason, many models use approximation methods such as freezing and averaging the core (nucleus and core electrons) and only considering the valence electrons involved in the interactions. In addition to the challenges n-body problems pose, the theoretical models have to take into account processes such as quantum distortions of the electronic wavefunctions, post-collisional Coulomb interactions, multiple collisions, and polarization of the target (as shown in fig. 2.3).
The quantum wave distortions shown in subfigure (a) of fig. 2.3 are the result of the distortion of the initial and final electron Coulomb wavefunctions. The post collisional interactions (PCIs) in subfigure (b) of fig. 2.3 are due to the outgoing electron-electron electrostatic repulsion following collision with and ionization of the atomic or molecular target. In an experiment the outgoing electrons usually cannot be detected at $\xi_1 = 0^\circ$ and $\xi_2 = 180^\circ$, because the electron energy analyzers would run into each other at these angles. However, due to post collisional interactions (PCIs), and for electrons having equal energies the electron energy analyzers are not required to reach those angles because the triple differential cross-sections $TDCS(\xi_1 = 0^\circ) = TDCS(\xi_1 = 180^\circ) = 0$. The multiple scattering shown in subfigure (c) of fig. 2.3 shows a multiple collision of the incident electron with the nucleus then with a valence electron to yield two outgoing electrons. The process shown in subfigure (d) of fig. 2.3 is the polarization of the target due to the Coulomb interaction between the incident electron, the nucleus of the target and its bound electrons [50]. At high energies the effects of some of these interactions and processes are minimized and

**Figure 2.3**: Processes of electron collisions. See text for details.
so can be neglected, thereby making the calculations easier. However, since many of the atomic and molecular spectroscopy experiments (including those detailed in this thesis) are carried out at low to intermediate energies, the contributions by all interactions and quantum processes shown in fig. 2.3 must be equally considered and accounted for in the theoretical models.

The electron-atom and electron-molecule interactions in electron impact experiments can produce a variety of scattering types. Electron scattering is the process by which the incident electrons are deviated from their original trajectory due to impact with an atomic or molecular target. Some of the scattering phenomena observed in these experiments are:

- **Elastic scattering**: these are electron collisions where there is no loss of incident energy, but a possible change in momentum of the incident electron upon impact.

- **Inelastic scattering**: these are electron collisions where the incident electron leaves an atomic or molecular target in an excited state.

- **Ionization**: these are electron collisions where the incident electron ionizes the atomic or molecular target, and ejects a valence electron from the target. This process is dependent on the incident energy of the electrons being high enough to ionize the target. Additional ionization processes are ionization with excitation, as well as multiple ionization of the target.

Scattering processes have been fundamental in the discovery and understanding of the structure of atoms. Following the discovery of the electron by J. J. Thomson in 1897 [66], Ernest Rutherford and his colleagues Hans Geiger and Ernest Marsden performed the famous gold foil experiment at the University of Manchester (between 1908 and 1913) [67]. In this experiment a beam of alpha particles were fired at foils of gold. The results of this experiment disproved Thomson’s plum-pudding model of the atom [70], and showed the true structure of the atom; that all of the atom’s positive charge and most of its mass are concentrated in a core called the nucleus.

The gold foil experiment shown in fig. 2.4 was carried out by pointing a beam of alpha particles (helium nuclei) and firing them at a foil of gold that is only a few atoms thick. A fluorescent screen (not shown in fig. 2.4) surrounded the apparatus upon which a scattering pattern emerged following the firing of these alpha particles at the gold atoms. The pattern on the fluorescent screen revealed that the alpha particles scattered in all directions, including the direction back at the alpha particle source. A tiny fraction (around 1 in 8000) of the fired alpha particles were deflected (elastically scattered) by more than 90°. This very scattering pattern was what disproved Thomson’s plum-pudding model of the atom.
that hypothesized that all alpha particles would have gone straight through and past the gold atoms. These measurements suggested that in order for the alpha particles to scatter in all directions, a strong electrostatic force must have caused some of these deflections to occur, which in turn implied that the atom’s positive charge was densely concentrated in the core of the atom. These conclusions explained why the positively charged alpha particles were being strongly repelled and deflected the closer they were in their approach towards the nucleus of the gold atom. These conclusions explained why the strength of the repulsive electrostatic force and deflection angle increased with closer distances between an approaching positively charged alpha particle and a nucleus of a gold atom. The small number of alpha particle deflections were explained by the small size of the nucleus. These monumental results led to Rutherford proposing his model for the atom which consisted of mostly empty space, and a cloud of electrons surrounding a dense nucleus where all of the atom’s positive charge was concentrated. The gold foil experiment is a matter-matter interaction and an example of elastic scattering; a process that also occurs in electron collisions with atoms and molecules over all incident electron energies.

2.2.1 Elastic Scattering

Elastic scattering due to electron impact with atoms or molecules (as shown in fig. 2.5) is a process where the energy of an incident electron is not changed, however the momentum may change. An incident electron $e$ with energy $E_{\text{inc}}$ and momentum $k_{\text{in}}$ collides with an atomic or molecular target, and scatters at an angle $\theta$ with respect to the direction of the incident electron with a final momentum $k_{\text{out}}$. The momentum of an elastically scattered electron following impact with an atomic or molecular target is dependent on
2.2 Electron Collisions

Figure 2.5: Elastic scattering with varying impact parameters. The incident and outgoing energies $E_{inc1}$ and $E_{inc2}$ of electrons $e_1$ and $e_2$, respectively, are unchanged. The momenta $k_{inc1}$ and $k_{inc2}$ of the incident electrons change upon impact with the target and become $k_{out1}$ and $k_{out2}$ respectively. The impact parameters $p_1$ and $p_2$ are proportional to the change in momentum and scattering angle of the incident electron upon impact. The angles $\theta_1$ and $\theta_2$ are the scattering angles for electrons $e_1$ and $e_2$ respectively.

the interaction of the incident electron with the target’s bound and valence electrons. The angle by which the electron scatters away from the target depends on the impact parameter $p$ which is defined by the distance between the trajectory of the electron and the parallel axis running through the atomic or molecular target. The incident electrons that follow a trajectory far from the nucleus encounter a small Coulomb force and are deflected only by a small angle. On the other hand, the electrons that follow a trajectory close to the nucleus encounter a strong attractive Coulomb force and are deflected by angles of up to $180^\circ$. The elastic cross-section $\sigma_{elastic}$ for such scattering experiments can be measured and expressed in the following manner:

$$\sigma_{elastic}(\theta_e, E_{inc}) = \frac{d\sigma}{d\Omega}$$

(2.3)

where $d\Omega$ is the solid angle of detection of an electron energy analyzer. The cross-section $\sigma_{elastic}$ does not depend on the angle $\psi$ with respect to the detection plane (see fig. 1.1).

Elastic scattering processes can lead to quantum mechanical interferences of the incident electron wave and the scattered electron following impact with the target. These quantum mechanical interferences are seen as deep and sharp dips in the cross-section measurements. If the incident electron energy is low, resonances can occur and appear in the elastic
cross-section measurements. An elastic resonance occurs when an incident electron with low energy $E$ collides with a target and is captured for a time $t_{res}$ around the atom until it is ejected (as shown in fig. 2.6). During the interaction time $t_{int}$ of the electron with the target, a temporary negative ion is formed for a time $t_{res}$. The two different interaction trajectories are when the incident electron is captured then ejected creating a temporary negative ion in the process:

$$|E_{inc}, k_{in}\rangle + |\psi_{ground}\rangle \xrightarrow{\text{indirect}} E \approx 0; |\psi_{ion}^{\text{exc}}\rangle \xrightarrow{\text{emission}} |E_{inc}, k_{out}\rangle + |\psi_{ground}\rangle \quad (2.4)$$

or when the incident electron is deflected upon impact with the target:

$$|E_{inc}, k_{in}\rangle + |\psi_{ground}\rangle \xrightarrow{\text{direct}} |E_{inc}, k_{out}\rangle + |\psi_{ground}\rangle \quad (2.5)$$

where $|\psi_{ground}\rangle$ is the target ground state wavefunction. These two different interaction trajectories lead to an interference in the elastic cross-section measurements which appears as a sharp dip with a width given by Heisenberg’s uncertainty principle where $\Delta E = \frac{\hbar}{2t_{res}}$.

An example of this is the 19.34 eV elastic resonance observed from electron collisions with Helium [71].

![Figure 2.6: Elastic resonance occurs by capture of the incident electron with incident energy $E_{inc}$ and momentum $k_{in}$ for a time $t_{res}$. The electron is then ejected with the same energy as $E_{inc}$ and momentum $k_{out}$. A temporary negative ion is formed for the time $t_{res}$ during which elastic resonance occurs.](image)

2.2.2 Inelastic Scattering

Inelastic scattering processes occur when the incident electron of energy $E_{inc}$ and a well defined momentum $k_{in}$, excites the target upon impact to an excited state $|\psi_{exc}\rangle$ of a
discrete energy $E_{\text{exc}}$ and so scatters inelastically. This process is useful as varying incident electron energies can allow for the excitation of all states of a target; states that are not accessible via photon excitation. The inelastic scattering reactions by electron impact are simply:

$$E_{\text{inc}}, k_{\text{in}}, |\psi_{\text{ground}}\rangle \xrightarrow{\text{interaction}} E_{\text{inc}} - E_{\text{exc}}, k_{\text{out}}, |\psi_{\text{exc}}\rangle$$ (2.6)

with the experimental inelastic cross-section measurements expressed as follows:

$$\sigma_{\text{inelastic}}(\theta_e, E_{\text{inc}}, E_{\text{exc}}) = \frac{d\sigma}{d\Omega}$$ (2.7)

where $\theta_e$ is the inelastic scattering angle with respect to the direction of the incident electron. The solid angle of the electron energy analyzer is given by $d\Omega$, which is tuned to detect electrons of energies $E_{\text{inc}} - E_{\text{exc}}$. It is important to note that the different substate contributions cannot be identified in inelastic differential cross-section measurements $\sigma_{\text{inelastic}}$ because the states are degenerate in energy. These measurements are usually carried out with electron energy analyzers with high energy resolution. The measurements produce electron energy loss spectra in which it shows the probability of excitation (or vibration) of a state as a function of the incident electron energy $E_{\text{inc}}$ and scattering angle $\theta_e$. An example of this are studies such as the one carried out by Khakoo et al. [72], in which cross-section measurements were obtained for both elastic and vibrationally inelastic scattering of low energy electrons from the ethylene ($C_2H_4$) molecule. These measurements were obtained at incident energies ranging from 0.5 to 100 eV and scattering angles ranging from 5° to 130°.

Another example of an inelastic scattering process is seen in Compton’s experiment which was carried out by Arthur Holly Compton in 1923 [73]. This experiment involved the scattering of photons (X-rays or gamma rays) from electrons in a graphite target, which found an increase in the wavelength (Compton effect) of the scattered electron relative to that of the incident photon, and a transfer of energy from the incident photon to the recoiling electron.

2.2.3 Ionization

Ionization by electron impact is a process by which the incident electron travels an interaction distance with an energy sufficient to ionize the target. The result of this interaction are two electrons: the scattering of the incident electron, and the ejection of a valence electron from the atomic or molecular target. Electron impact ionization studies of atoms and molecules are carried out using experiments such as the Manchester (e,2e)
coincidence spectrometer. The kinematics of the ionization process by electron impact is shown in fig. 2.7, where the incident electron $e_0$ travels with an energy $E_0$ and momentum $k_0$. Upon impact, the atomic or molecular target $A$ is ionized and becomes $A^+$ resulting in a scattered (or ejected) electron $e_1$ with energy $E_1$ and momentum $k_1$, and an ejected (or scattered) electron with energy $E_2$ and momentum $k_2$. The scattering angles of the outgoing electrons are denoted by $\xi_1$ and $\xi_2$, to distinguish between them and the angles $\theta_1$ and $\theta_2$ used to describe the kinematics of the elastic and inelastic scattering processes in the previous sections. From eq. (1.1), the ionization reaction with target $A$ is

$$e_0(E_0, k_0) + A \rightarrow e_1(E_1, k_1) + e_2(E_2, k_2) + A^+(Q)$$

where

$$E_0 = E_1 + E_2 + IP$$

is known from eq. (1.3), and the same is known for the initial and final momenta of the system from eq. (1.4)

$$k_0 = k_1 + k_2 + Q$$
The energies of the initial and final state of the system are

\[ E_0 = \frac{|k_0|^2}{2m_e}; \quad E_1 = \frac{|k_1|^2}{2m_e}; \quad E_2 = \frac{|k_2|^2}{2m_e}; \quad T = \frac{|Q|^2}{2M_{A^+}} \]  

where \( Q \) is the recoil momentum of the target ion and \( M_{A^+} \) is the mass of the ionized target. The energy \( T \) of the recoiled ionized atomic or molecular target can be ignored since it is small in comparison to the energy of the incident and outgoing electrons. Electron impact ionization evolves a system from an initial state with one charged particle to a final state with three charged particles, making the entire interaction an \( n \)-body problem. In addition, the contributions of the Coulomb interactions and the processes shown in fig. 2.3 play a critical role in determining the exact and final state of the system. The challenges that arise from these complex \( n \)-body scattering problems become motivations for the development of sophisticated theoretical models for predicting the behavior of these interactions. The models that were tested against the experimental results detailed in this thesis are covered in the next section.

### 2.3 Theoretical Electron-Impact Ionization Models

The previous section shed light on the interactions and processes of \( n \)-body problems and the theoretical challenges they pose. The \( n \)-body (also known as as the few-body) problem is an unsolved fundamental problem in atomic and molecular physics. The reason for the difficulty in finding a solution to this problem, is due to the fact that the Schrödinger equation has no analytical solution for more than two mutually interacting particles. In a system containing three (or more) particles such as the one found in an \((e,2e)\) coincidence experiment, the theoretical models required to describe these interactions would have to use various methods, techniques, and approximations, and enough computational resources to perform numerous calculations. The accuracy and verification of these models are determined by a comparison of their calculations with experimental results.

Several different theoretical methods and techniques were developed over the last fifty years [74]. However, only the models used for the experiments detailed in this thesis will be explained here. These electron impact ionization experiments were carried out for the nitrogen (\( N_2 \)) and methane (\( CH_4 \)) molecules and tested against the calculations of the following theoretical models:

- The distorted wave Born approximation (DWBA).
- The molecular three-body distorted wave (M3DW) approximation.
- The orientation averaged molecular orbital (OAMO) approximation.
• The Ward–Macek (WM) approximation.
• The proper averaging (PA) approach.
• The Generalized Sturmian Functions (GSF) approach.

These models provide approximations for the electron impact single ionization of molecules such as nitrogen ($N_2$) and methane ($CH_4$). The models have been developed to deal with different atomic and molecular targets, incident electron energies, and scattering geometries. Each model has a unique set of approximation methods, which are tested and improved when compared to experimental results.

2.3.1 The Distorted Wave Born Approximation (DWBA)

The distorted-wave Born approximation (DWBA) is one of the most successful approaches for calculating the differential cross-sections for atomic and molecular targets [75, 76]. However, the model has limitations due to the poor treatment of post collision interactions [77], which in turn motivated the development of the molecular three-body distorted wave (M3DW) approximation. The inclusion of molecular orientation averaging was later introduced, which then posed computational challenges that were dealt with using orientation averaged molecular orbital (OAMO) approximation [74]. This section will give an overview of the approximation methods mentioned above, highlight the similarities and differences between them, and motivation behind each model.

Obtaining ($e, 2e$) cross-sections using the distorted wave Born approximation (DWBA) begins with the exact transition amplitude for the ionization process. This is given by the following $T$-matrix:

$$T = \langle \Psi_f | H - H_0 | \Phi_i \rangle$$

(2.9)

where the initial and final state wavefunctions $\Psi_f$ and $\Phi_i$ are eigenfunctions of the two Hamiltonians $H$ and $H_0$, where $H$ is the full Hamiltonian for the system and $H_0$ is an approximate initial-state Hamiltonian, where:

$$H |\Psi_f\rangle = E |\Psi_f\rangle$$

(2.10)

$$H_0 |\Phi_i\rangle = E |\Phi_i\rangle$$

(2.11)

The interactions included in the initial and final state wavefunctions $\Psi_f$ and $\Phi_i$ are contained to all orders of perturbation theory, while the operator $(H - H_0)$ interactions are contained to only the first order of perturbation. The $T$-matrix in eq. (2.9) is evaluated by approximating $\Psi_f$ and choosing $H_0$. 
Choosing the initial-state Hamiltonian $H_0$ leads to the first-order distorted-wave Born approximation (DWBA), where $H_0$ is chosen to be:

$$H_0 = H_{\text{target}} + T_p + U_i$$  \hspace{1cm} (2.12)

where $H_{\text{target}}$ is the Hamiltonian for the neutral target with eigenfunction $\psi_{\text{target}}$, $T_p$ is the kinetic energy operator for the projectile (incident electron), and $U_i$ is an initial-state spherically symmetric distorting potential. The initial-state distorting potential, as it is known, approximates the interaction between the target and the projectile. This is achieved by considering the target’s nuclear contribution and the interaction between the projectile (incident) electron and the target electrons. The charge density of the target electrons (obtained using the Hartree-Fock method) [78], and the projectile (incident) electron, are used to evaluate a spherically symmetric approximation for the interaction between the two.

The next step is to consider the initial and final state of the system. As mentioned previously, the electron impact ionization process evolves a system from an initial state with one charged particle to a final state with three charged particles. One of the challenges that arise from such a system is the quantum mechanical wave distortions (shown in subfigure (a) of fig. 2.3) of the initial and final electron Coulomb wavefunctions. To obtain the initial state distorted wave eigenfunction $\chi_i$ of the projectile (incident) electron, the initial state distorting potential $U_i$ is used:

$$(T_p + U_i)\chi_i = \varepsilon_i \chi_i$$  \hspace{1cm} (2.13)

where $\varepsilon_i$ is the energy of the incident projectile electron. Hence, the initial state wavefunction $\Phi_i$ of the system can be written as:

$$\Phi_i = \chi_i\psi_{\text{target}}$$  \hspace{1cm} (2.14)

The exact final state of the system is approximated using the product of the ionized target and the two outgoing electron wavefunctions:

$$\Psi_f \approx \chi_{\text{proj}}\chi_{\text{ejec}}\psi_{\text{ion}}$$  \hspace{1cm} (2.15)

where $\psi_{\text{ion}}$ is the final state wave function for the ionized target. The final state distorted wave eigenfunctions $\chi_{\text{proj}}$ and $\chi_{\text{ejec}}$ are obtained in a similar manner to $\chi_i$ in eq. (2.13),
by using the final state distorting potential $U_{ion}$:

\begin{align}
(T_p + U_{ion}) \chi_{proj} &= \varepsilon_{proj} \chi_{proj} \\
(T_p + U_{ion}) \chi_{eject} &= \varepsilon_{eject} \chi_{eject}
\end{align}

(2.16) \hspace{1cm} (2.17)

where $\varepsilon_{proj}$ and $\varepsilon_{eject}$ are the energies of the projectile and ejected electrons respectively. It is worth noting that the 'projectile' in the final state of the system refers to the scattered electron following impact with the target. The final state distorting potential $U_{ion}$ is evaluated in a similar manner to the initial state distorting potential $U_i$; it uses the continuum electron and the bound electrons of the ionized target, instead of the projectile electron and bound electrons of the neutral target, respectively.

The full Hamiltonian $H$ of the system is given by:

$$H = H_{target} + T_p + V_i$$

(2.18)

where $V_i$ is the initial state interaction between the projectile (incident) electron and the target. Using eq. (2.12), the operator $(H - H_0)$ in the $T$-matrix of eq. (2.9) can be expressed as:

$$H - H_0 = (H_{target} + T_p + V_i) - (H_{target} + T_p + U_i)$$

$$= V_i - U_i$$

(2.19)

Using eq. (2.14), eq. (2.15), and eq. (2.19), the exact transition amplitude for the ionization process of eq. (2.9) becomes the direct scattering distorted-wave Born approximation (DWBA) $T$-matrix for molecules:

$$T_{dir}^{DWBA} = \langle \Psi_f | H - H_0 | \Phi_i \rangle$$

$$= \langle \chi_{proj} \chi_{eject} \psi_{ion} | V_i - U_i | \chi_i \psi_{target} \rangle$$

(2.20)

The physical effects due to interactions and quantum mechanical processes contained in the wavefunctions of the $T$-matrix $T_{dir}^{DWBA}$ of eq. (2.20) are as follows:

- The initial state of the system: with the distorted wave $\chi_i$ as an eigenfunction of the initial state distorting potential $U_i$, the Coulomb (elastic scattering) interactions of the projectile (incident) electron with the neutral target’s nuclei and bound electrons are contained to all orders of perturbation theory.

- The final state of the system: with the distorted waves $\chi_{proj}$ and $\chi_{eject}$ as eigenfunctions of the final state distorting potential $U_{ion}$, the Coulomb (elastic scattering)
interactions of the projectile (scattered) electron with the ionized target’s nuclei and bound electrons are contained to all orders of perturbation theory.

- The \((V_i - U_i)\) interaction: this is the first order non-spherical component of the initial state interactions between the projectile (incident) electron and the bound electrons of the target only.

### 2.3.2 The Molecular Three-Body Distorted Wave (M3DW) Approximation

The distorted-wave Born approximation (DWBA) is one of the most successful theoretical methods used to calculate the fully differential cross-sections (FDCSs) for high energy electron impact ionization of complex atomic or molecular targets. The first of these DWBA calculations were carried out in 1977 by Madison et al. \cite{76} for the ionization of helium. Despite the success over the last 40 years, the DWBA model had limitations that skewed the calculations when the incident electron energy fell below \(\sim 100\) eV. One known cause behind this limitation was the poor treatment of the final state of the system, in particular, the post collision interaction shown in subfigure (b) of fig. 2.3 between the outgoing projectile (scattered) and ejected electrons. This was demonstrated in a 1989 low energy electron-hydrogen scattering study by Brauner et al. \cite{77}, where good agreement with experiment was found when including the interaction between the outgoing electrons in the final state wavefunction \(\Psi_f\). In the approach used by Brauner et al., the final state wavefunction \(\Psi_f\) is approximated using the so-called 3\(C\) wavefunction \cite{77}:

\[
\Psi_f \approx CW_{\text{proj}}CW_{\text{ejec}}C_{\text{proj-ej}} (2.21)
\]

where \(CW_{\text{proj}}\) and \(CW_{\text{ejec}}\) are Coulomb waves for electrons exposed to a nuclear charge, and \(C_{\text{proj-ej}}\) is the Coulomb distortion factor containing the post collisional interactions (PCIs) between the outgoing projectile (scattered) and ejected electrons in the final state of the system. The 3\(C\) wavefunction is used for lighter atomic and molecular targets by treating them as point charges, and for this reason, it must be generalized to the distorted-wave method to accommodate heavier targets. The distorted-wave Born approximation (DWBA) equivalent for the final state wavefunction \(\Psi_f\) would then be:

\[
\Psi_f \approx \chi_{\text{proj}}\chi_{\text{ejec}}C_{\text{scat-ej}}\psi_{\text{ion}} (2.22)
\]

which is also known as the 3-body distorted-wave (3DW) wavefunction. Including this function yields the direct-scattering molecular 3-body distorted-wave \(T\)-matrix:

\[
T_{\text{dir}}^M3DW = \langle \chi_{\text{proj}}\chi_{\text{ejec}}C_{\text{scat-ej}}\psi_{\text{ion}} | V_i - U_i | \chi_i\psi_{\text{target}} \rangle (2.23)
\]
The initial state interaction $V_i$ for the M3DW approach is approximated as:

$$V_i = \frac{1}{r_{ab}} + U_{ion}$$  \hspace{1cm} (2.24)

where $\frac{1}{r_{ab}}$ is the interaction of the projectile (incident) electron with the active electron in the molecular target, and $U_{ion}$ is the interaction of the nuclei and bound electrons of the target with the projectile electron. As for the initial state distorting potential, it is written as:

$$U_i = U_a + U_{ion}$$  \hspace{1cm} (2.25)

where the spherically symmetric potential $U_a$ is the interaction of the projectile and the active electron in the molecular target. Using eq. (2.24) and eq. (2.25) yields the following perturbation:

$$V_i - U_i = \frac{1}{r_{ab}} - U_a$$  \hspace{1cm} (2.26)

The reason why this perturbation is considered non-spherical, is because both terms represent the difference between interaction of the projectile with the active electron in the target and the spherically symmetric approximation for the same interaction.

The physical effects due to interactions and quantum mechanical processes contained in the wavefunctions of the $T$-matrix $T^{M3DW}_{dir}$ of eq. (2.23) are as follows:

- The initial state of the system: this is similar to the DWBA approach. The Coulomb (elastic scattering) interactions of the projectile (incident) electron with the neutral target’s nuclei and bound electrons are contained to all orders of perturbation theory.

- The final state of the system contains the following Coulomb interactions to all orders of perturbation theory:
  - the projectile (scattered) electron with the ionized target’s nuclei and bound electrons.
  - the ejected electrons with the ionized target’s nuclei and bound electrons.
  - the outgoing projectile (scattered) and ejected electrons.

- The $(V_i - U_i)$ interaction: this is similar to the DWBA approach. It only contains the first order non-spherical component of the initial state interactions between the projectile (incident) electron and the bound electrons of the target. The $(V_i - U_i)$ operator is modified for the M3DW approach (see above).
2.3.3 Methods for Molecular Orientation Averaging

The DWBA and M3DW approximations can be modified to include the effects of the orientations of molecular orbitals. To achieve this, the following must be determined:

- $\psi_{\text{ion}}$: the final state ion wavefunction for the molecule.
- $\psi_{\text{target}}$: the initial state target wavefunction.

and declaring the following:

- $\xi_{\text{coord}}$: this represents the coordinates of all the passive electrons.
- $\textbf{r}_a$: this represents the coordinates (radial distance) of the active electron.
- $\textbf{R}$: this represents the orientation of the molecule.

which yield the following dependencies: $\psi_{\text{ion}}(\xi_{\text{coord}}, \textbf{R})$ and $\psi_{\text{target}}(\xi_{\text{coord}}, \textbf{r}_a, \textbf{R})$. The orientation of the molecule is assumed to be the same for the initial state and the final state of the system due to the short interaction time of electron impact. Integrating over all coordinates of the passive electrons yields the following:

$$\langle \psi_{\text{ion}}(\xi_{\text{coord}}, \textbf{R}) | \psi_{\text{target}}(\xi_{\text{coord}}, \textbf{r}_a, \textbf{R}) \rangle = \psi_{\text{Dyson}}(\textbf{r}_a, \textbf{R})$$

(2.27)

where $\psi_{\text{Dyson}}(\textbf{r}_a, \textbf{R})$ (also written as $\phi_{\text{Dyson}}(\textbf{r}_a, \textbf{R})$) is the so-called Dyson orbital. Lastly, the perturbation discussed above is dependent only on the projectile and active electron coordinates, and for this reason the spherically symmetric potential is written as $U_a(\textbf{r}_a)$ since it is dependent on the radial distance of the projectile electron. Using the perturbation terms of eq. (2.26) and the dependency of the orientation of the molecule from eq. (2.27), the direct-scattering $T$-matrix of eq. (2.23) then becomes:

$$T_{\text{dir}}^{M3DW} = \left\langle \chi_{\text{proj}}(\textbf{r}_a) \chi_{\text{ejec}}(\textbf{r}_b) C_{\text{scat-ejec}}(\textbf{r}_{ab}) \left| \frac{1}{\textbf{r}_{ab}} - U_a(\textbf{r}_a) \right| \psi_{\text{Dyson}}(\textbf{r}_a, \textbf{R}) \chi_i(\textbf{r}_b) \right\rangle$$

(2.28)

The molecular orientation averaging using the Dyson orbital of eq. (2.27) can be applied to the DWBA direct-scattering $T$-matrix of eq. (2.20) to yield:

$$T_{\text{dir}}^{\text{DWBA}} = \langle \chi_{\text{proj}}(\textbf{r}_a) \chi_{\text{ejec}}(\textbf{r}_b) | V_i - U_i | \psi_{\text{Dyson}}(\textbf{r}_a, \textbf{R}) \chi_i(\textbf{r}_b) \rangle$$

(2.29)

The dependency of the direct-scattering $T$-matrix of eq. (2.28) (or eq. (2.29)) on the orientation of the molecule poses a computational challenge, because it requires long
computing time and enough processing power to evaluate eq. (2.28) (or eq. (2.29)) over the large number of molecular orientations to obtain an accurate solution. This problem was considered by using the orientation-averaged molecular orbital (OAMO) approximation by Gao et al. [74], which averages the molecular orbitals instead of the cross-sections. However, Gao et al. [74, 79] showed that the OAMO approximation is valid for molecules with highly symmetric (gerade) orbitals such as \( N_2 \); one of the molecules used in the (e,2e) studies detailed in this thesis.

The M3DW direct-scattering \( T \)-matrix then becomes:

\[
T_{dir}^{M3DW} = \langle \chi_{proj}(r_a) \chi_{ejec}(r_b) C_{scat-ejec}(r_{ab}) \left| \frac{1}{r_{ab}} - U_a(r_a) \right| \psi_{Dyson}^{OAMO}(r_a) \chi_i(r_b) \rangle (2.30)
\]

and the DWBA direct-scattering \( T \)-matrix becomes:

\[
T_{dir}^{DWBA} = \langle \chi_{proj}(r_a) \chi_{ejec}(r_b) \left| V_i - U_i \right| \psi_{Dyson}^{OAMO}(r_a) \chi_i(r_b) \rangle (2.31)
\]

where \( \psi_{Dyson}^{OAMO}(r_a) \) is the Dyson orbital averaged over all molecular orientations.

2.3.4 The Ward–Macek (WM) Approximation

The Ward-Macek (WM) [80] approximation is covered briefly here. This approximation can be used along with either the DWBA or the M3DW approximation, and is used for the post collisional Coulomb interaction term between the outgoing electrons in eq. (2.22):

\[
\Psi_f \approx \chi_{proj} \chi_{ejec} C_{scat-ejec} \psi_{ion}
\]

Since the functions for the distorted electron waves depend on the radial distance, and the Coulomb interaction term depends on the relative distance and momentum between the outgoing electrons, the final state wavefunction can be rewritten as:

\[
\Psi_f \approx \chi_{proj}(r_a) \chi_{ejec}(r_b) C_{scat-ejec}(r_{ab}, k_{ab}) (2.32)
\]

where

\[
C_{scat-ejec}(r_{ab}, k_{ab}) = e^{-\pi \gamma^2 / 2 \Gamma(1 - i \gamma)} _1 F_1(i \gamma, 1, -i[k_{ab} r_{ab} + k_{ab} \cdot r_{ab}]) (2.33)
\]

where the terms in the equation above are:

- \( \Gamma(1 - i \gamma) \): is a gamma factor.
- \( _1 F_1 \): is a confluent hypergeometric function.
• $r_{ab}$: is the relative distance between the two outgoing electrons.
• $k_{ab}$: is the relative momentum between the two outgoing electrons.
• $\gamma$: is the Sommerfeld parameter ($1/v_{ab}$), where $v_{ab}$ is the relative velocity between the two outgoing electrons.

The post collision interactions are treated in the WM approximation by averaging the separation distance between the two outgoing electrons. This is achieved by replacing the $[k_{ab}r_{ab} + k_{ab} \cdot r_{ab}]$ term in eq. (2.33) with $2k_{ab}r_{ab}^{\text{ave}}$ where $r_{ab}^{\text{ave}}$ is the average separation distance between the outgoing scattered and ejected electrons.

$$C_{\text{scat-ejec}}^{WM}(r_{ab}^{\text{ave}}, k_{ab}) = e^{-\pi\gamma/2} \Gamma(1 - i\gamma) \frac{1}{4\pi} F_1(i\gamma, 1, 1 - 2ik_{ab}r_{ab}^{\text{ave}})$$ (2.34)

The $C_{\text{scat-ejec}}^{WM}$ factor can be then be used with the DWBA or M3DW approximation, to include post collision interactions (PCIs) in the final state wavefunction to all orders of perturbation theory, such that:

$$T_{\text{dir}}^{WM} = C_{\text{scat-ejec}}^{WM}(r_{ab}^{\text{ave}}, k_{ab})T_{\text{dir}}^{D\text{WBA}(M3\text{DW})}$$ (2.35)

### 2.3.5 The DWBA and M3DW Fully Differential Cross-Section Calculations

The final step before calculating the fully differential cross-sections, is accounting for the exchange $T$-matrix for the desired model. This is necessary because the projectile and ejected electrons are indistinguishable from each other. Using the DWBA-OAMO direct-scattering $T$-matrix from eq. (2.31), the DWBA exchange $T$-matrix is written as:

$$T_{\text{exc}}^{D\text{WBA}} = \left\langle \chi_{\text{proj}}(r_b)\chi_{\text{ejec}}(r_a) \right| V_i - U_i \left| \psi_{\text{Dyson}}(r_a)\chi_i(r_b) \right\rangle$$ (2.36)

and the M3DW-OAMO direct-scattering $T$-matrix from eq. (2.37), the M3DW exchange $T$-matrix is:

$$T_{\text{exc}}^{M3\text{DW}} = \left\langle \chi_{\text{proj}}(r_b)\chi_{\text{ejec}}(r_a)C_{\text{scat-ejec}}^{WM}(r_{ab}) \right| \frac{1}{r_{ab}} - U_{\text{a}}(r_a) \left| \psi_{\text{Dyson}}(r_a)\chi_i(r_b) \right\rangle$$ (2.37)

The exchange $T$-matrices for the DWBA (or M3DW) models which use the WM approximation can be found in a similar manner as to those in eq. (2.36) and eq. (2.37). Finally, by using the direct-scattering and exchange $T$-matrices, the fully differential cross-sections (FDCSs) for molecular targets can be found by using the following:

$$\frac{d^5\sigma}{d\Omega_a d\Omega_b dE_b} = \frac{1}{(2\pi)^5} \frac{k_a k_b}{k_i} (|T_{\text{dir}}|^2 + |T_{\text{exc}}|^2 + |T_{\text{dir}} - T_{\text{exc}}|^2)$$ (2.38)
Refer to Madison et al. [75] for additional details on the effects and calculations of other quantum mechanical processes such as the spherically symmetric distorting potentials for molecules, the spherically symmetric static distorting potential, the exchange distortion, the correlation-polarization potential, and the full final state Coulomb interaction $C_{\text{proj-ejec}}$.

2.3.6 The Proper Averaging (PA) Approach

The proper averaging (PA) approach developed by Don Madison’s group [81], is a method that considers the random orientations of the molecules in the experiments, by calculating the theoretical triple differential cross-section (TDCS) for each orientation $R$, which are all then averaged over all possible molecular orientations. This proper average (PA) is calculated using:

$$
\sigma^{PA} = \frac{\int \sigma^{TDCS}(R)d\Omega_R}{\int d\Omega_R}
$$

The results of approximations that make use of the proper averaging (PA) approach are considerably more accurate than those that do not include this approach. Poor agreement between experimental results and theoretical calculations that did not include the proper averaging (PA) approach, was seen in the studies on $H_2$ [50, 53, 82], and $CH_4$ [83, 84]. The $(e,2e)$ study on $CH_4$ [61] detailed in this thesis included the proper averaging (PA) approach in its TDCS calculations, the results of which showed good agreement between theory and experiment. The disadvantage of the proper averaging (PA) approach is that it is computationally demanding in time and in resources. The lack of access to these resources prevented the use of this approach in past experiments (mentioned above). The advanced computational resources that were made available in 2019, were used to perform PA-based calculations for the $(e,2e) CH_4$ study detailed in this thesis [61]. The results of this study showed the necessity for the proper averaging (PA) approach in providing accurate predictions of ionization cross-sections of molecular targets.

2.3.7 The Generalized Sturmian Function (GSF) Approach

The Generalized Sturmian Functions approach [85, 86] is another method for dealing with the three-body problem. The motivation behind the development of this approach stems from the many challenges posed by atomic and molecular processes, as well as the limitation of other theoretical models. These problems range from the simultaneous excitation and ionization, multiple ionization of atomic and molecular targets, and the effects of the ejected electrons. The methods for approximating the effects of the bound electrons of the target in an interaction, the post collision interaction of the outgoing
electrons, and the initial state of the system were difficult for other theoretical models; challenges that led to the use of the Generalized Sturmian Functions approach. This approach was used in parallel with the M3DW approximation, and the calculations from both were tested against the \((e,2e)\) studies on the methane \((CH_4)\) molecule \([61]\) detailed in this thesis. This section will provide a very brief overview of the Generalized Sturmian Functions approach. For details on this approach with respect to electron impact ionization of molecules, refer to work provided by Ancarani, Granados, and Gasaneo in \([85, 86]\).

The Generalized Sturmian Functions approach is a treatment of the bound states and the collisional processes. The so-called spectral method uses a basis set of Generalized Sturmian Functions, which are by definition solutions of a Sturm–Liouville problem (explained below) \([87]\). The Generalized Sturmian Functions are efficient in that they can be adapted to include physical properties for a given scattering problem. As an example, in electron impact ionization of a neutral target, an ejected electron is set to have a specific energy and interact with a unit charge over large distances.

The mathematicians Jacques Charles François Sturm and Joseph Liouville developed a Sturm–Liouville theory \([87]\) of a real second-ordered linear differential equation of the form:

\[
\frac{d}{dx} \left[ p(x) \frac{dy}{dx} \right] + q(x)y = -\lambda \omega(x)y \quad (2.40)
\]

where \(p, q,\) and \(w\) are specified such that \(p(x) > 0, q(x) > 0,\) and \(\omega(x) > 0\) for \(x \in (a,b)\), \(\omega(x)\) is a known function called the density or weighing function, and the function \(y\) and constant \(\lambda\) are both unknown. Determining the values of \(\lambda\) that satisfy the boundary conditions defined by eq. \((2.40)\) is the Sturm–Liouville problem. When determined, the values of \(\lambda\) and solutions for each \(\lambda\) become the eigenvalues and eigenfunctions of this problem respectively.

In the Generalized Sturmian Functions approach, the ionization process is described by the driven equation from a first-order perturbative approximation \([88]\):

\[
(E - \hat{H}_0)\psi^1(\mathbf{r}_a,\mathbf{r}_b; \hat{\mathbf{R}}) = \hat{T}\psi^0(\mathbf{r}_a,\mathbf{r}_b; \hat{\mathbf{R}}) \quad (2.41)
\]

where the terms in the driven equation above are:

- \(\hat{H}_0\): is the unperturbed Hamiltonian of the molecular target.
- \(E\): is the total energy of the system.
- \(\hat{T}\): is the transition (first-order perturbation) operator such that \(\hat{T} = V(\mathbf{r}_a,\mathbf{r}_b)\), where
$V$ describes the interaction potential between the projectile and the molecular target. The projectile (incident) electron is represented by a plane wave which is described by the following initial and final state wavefunctions:

\[
\psi^0(r_a, R; \hat{R}) = (2\pi)^{-3/2} e^{ik_a r_a} \psi_0(r_a; \hat{R}) \]

\[
\psi^1(r_a, R; \hat{R}) = (2\pi)^{-3/2} e^{ik_a r_a} \phi^1(r_b; \hat{R}) \]

where $\phi^1(r_b; \hat{R})$ is the wavefunction of the ejected electron. The driven equation in eq. (2.41) is solved by using the initial state one-center expansion wavefunction calculated by Moccia [89], which describes any given molecular orbital $i$ as:

\[
\phi_i(r) = \sum_j A_{ij} R_j(r) S_{\ell m j}\]

where

\[
R_j(r) = N_j r^{\ell_j - 1} e^{-\zeta_j r} \]

The terms in eq. (2.44) and eq. (2.45):

- $A_{ij}$: this contains the expansion coefficients.
- $S_{\ell m j}$: this contains the real spherical harmonics.
- $N_j$: this is the normalization coefficient.
- $\ell_j$: this is the quantum number of the orbital angular momentum
- $m_j$: this is the magnetic quantum number.
- $\zeta_j$: this is the orbital exponent.

The values of $A_{ij}$, $\ell_j$, $m_j$, and $\zeta_j$ are determined by the type of molecule involved in the interaction. For the methane ($CH_4$) molecule, these values are given by Moccia in [89].

The final state wavefunction for the ejected electron $\phi^1(r_b; \hat{R})$ is separated into angular and radial components, the latter of which is expanded in Generalized Sturmian Functions:

\[
\phi^1(r_b; \hat{R}) = \frac{1}{r_b} \sum_{\ell m}\sum_j a^{(\ell,E)}_j(\hat{R}) S^E_j(r_b) Y^m_\ell(\hat{R}_b) \]

this reduces the driven equation (eq. (2.41)) to a linear system of equations for finding the expansion coefficients $a^{(\ell,E)}_j(\hat{R})$ for a given molecular orientation $\hat{R}$. As mentioned above,
Generalized Sturmian Functions are solutions of a Sturm-Liouville problem \[86\] and satisfy the two-body non-homogeneous Schrödinger-type equation \[85, 90, 91\]:

\[
\left[-\frac{1}{2} \frac{d^2}{dr^2} + \frac{\ell (\ell + 1)}{2r^2} \mathcal{U}(r) - E_S \right] S_{J}^{(\ell, E_S)}(r) = -\beta_J^{(\ell, E_S)} \mathcal{V}(r) S_{J}^{(\ell, E_S)}(r)
\]  

(2.47)

where

- \(E_S\): is an externally fixed parameter.
- \(\beta_J^{(\ell, E_S)}\): this term contains the generalized charge eigenvalues for a given angular momentum \(\ell\).
- \(\mathcal{V}(r)\): is a short range generating potential that determines the size of the inner region in which most of the interactions occur.
- \(\mathcal{U}(r)\): is a long range auxiliary potential that dictates the asymptotic behavior of all Generalized Sturmian Functions.

The electron impact ionization processes are studied by imposing two boundary conditions on eq. (2.47), and solving the driven equation (2.41) using the initial and final state expansions of eq. (2.42) and eq. (2.46) which transform eq. (2.41) into a radial equation. This radial equation is solved using the definition of Generalized Sturmian Functions in eq. (2.47), which then transforms the driven equation of eq. (2.41) into a linear system of equations. This is followed by projecting onto an individual element of the basis set \(S_{i}^{(\ell, E)}(r_b)\), where \(E\) is chosen to be the physical energy. The results above are calculated using computational numerical methods and subroutines \[85, 90, 92\].

The theoretical triple differential cross-section (TDCS) calculations are given by \[93, 94\]:

\[
\frac{d^3\sigma}{d\Omega_a d\Omega_b dE} = (2\pi)^4 \frac{k_a k_b}{k_i} |f(k_i, k_a, k_b, \hat{r}_b; \hat{\mathcal{R}})|^2
\]  

(2.48)

An angular average is performed on eq. (2.48) due to the random orientation of the initial molecular target. The calculated triple differential cross-sections are then tested and verified against the experimental results for the same molecular target.

The Generalized Sturmian Function approach is a computationally efficient model, and includes properties such as the correct asymptotic behavior of the ejected electron, and the interaction between the ejected electron and the ionized molecular target. However, improvements to the Generalized Sturmian Function approach can still be made, such as potentials of the initial state of the system, the distorted projectile electron waves, and the outgoing electron exchange and post collisional interaction effects.
2.4 Conclusion

The theoretical models detailed in this chapter provided calculations that were tested against experimental results for the three (e,2e) studies detailed in chapter 8 and chapter 9 of this thesis.

The first experimental study detailed in section 8.3 was performed with incident electron energies \( \sim 10 \) and \( \sim 20 \text{ eV} \) above the ionization potential of the \( 3\sigma_g, 1\pi_u \) and \( 2\sigma_u \) states for \( N_2 \), using both equal and non-equal outgoing electron energies and a range of geometries from the coplanar to the perpendicular plane. The publication [59] of this work was carried out in collaboration with Don Madison’s groups from Missouri University of Science & Technology (USA) using the following models:

- The molecular three-body distorted wave (M3DW) approximation.
- The distorted wave Born approximation (DWBA).
- The DWBA using the Ward–Macek (WM) approximation.

The work resulted in experimental and theoretical ionization triple differential cross-sections.

The second experimental study detailed in section 8.4 was also performed on the \( N_2 \) molecule. The experiment was performed with the incident electron beam in the scattering plane where \( \psi = 0^\circ \), at symmetric energies \( \sim 20 \) and \( \sim 40 \text{ eV} \) above the ionization potential for the \( 3\sigma_g \) and \( 1\pi_u \) states for \( N_2 \), and for a number of fixed angles for one of the outgoing electrons. This experiment tested the following theoretical models from Don Madison’s group:

- The distorted wave Born approximation (DWBA).
- The molecular three-body distorted wave (M3DW) approximation.
- The M3DW using the Ward–Macek (WM) approximation for post collision interactions (PCI).

This experiment was an extension of the first (section 8.3), to further test the theoretical models and see if their predictions improved at higher energies [60].

The final (e,2e) experiments detailed in chapter 9 are on a series of experiments and theoretical investigations carried out on the ionization of the highest occupied molecular orbital (HOMO) \( 1\epsilon_2 \) and the next highest occupied molecular orbital (NHOMO) \( 2\alpha_1 \) states of \( CH_4 \) at an incident electron beam energy of \( 250 \text{ eV} \), for ejected electron energies of \( 50 \text{ eV} \) and \( 30 \text{ eV} \). The fixed scattering angles (\( 20^\circ, 22.5^\circ, 25^\circ, 27.5^\circ, \text{ and } 30^\circ \)) were used to perform these experiments. The measured triple differential cross-sections were compared
to the experimental results of the Afyon group in Turkey. Both experimental results were also compared to theoretical calculations using the following models:

- The molecular three-body distorted wave (M3DW) approximation with Don Madison’s group (USA).
- The Generalized Sturmian Function (GSF) approach with Ugo Ancarani’s group (France).

This successful international collaborative effort resulted in publication in 2019 [61]. Following from these experiments, further measurements have now been carried out for lower incident energies of 20 eV and 40 eV in both coplanar and non-coplanar geometries. These results have now been compared to the M3DW calculations using the proper averaging approach from the Rolla group (Don Madison’s group in Missouri), and have now been submitted for publication.

The next chapter will introduce a series of five chapters on the (e,2e) apparatus used to perform (e,2e) coincidence experiments and test the theoretical models detailed here. The first in this series is an overview of the entire (e,2e) apparatus, including the modernized experimental components, and a comparison of the old and modernized experimental hardware and software. This is then followed by a chapter detailing the (e,2e) spectrometer and vacuum system, two chapters detailing the experimental hardware and software, and a chapter on the experimental procedures for carrying out (e,2e) coincidence experiments, and performing data extraction and analysis routines. This series is then followed by two chapters that detail the direct application of the modernized (e,2e) apparatus, via the experimental studies carried out on $N_2$ and $CH_4$, along with their published results and comparisons to theory.
CHAPTER 3

The (e,2e) Apparatus

3.1 Introduction

The Manchester (e,2e) coincidence experimental apparatus is comprised of an (e,2e) spectrometer and vacuum system, experimental hardware, and experimental software. It is used to perform experiments such as the ones detailed in chapter 8 and chapter 9, on Nitrogen \((N_2)\) and Methane \((CH_4)\), respectively. This chapter is an overview of the fundamental components making up the (e,2e) apparatus (as shown in the block diagram in fig. 3.1), and is an introduction to:

- chapter 4: this chapter covers the (e,2e) spectrometer, and the vacuum system in which it is housed (including the vacuum system’s top flange, the pumping system, and the target gas line).
- chapter 5: this chapter details the modernized and old experimental hardware; power supplies, voltage supplies, stepper motor control and data acquisition unit, detection electronics, and the filament constant current supply.
- chapter 6: this chapter details the new experimental software designed for control, measurement, and analysis.
- chapter 7: this chapter details the experimental procedures needed to perform (e,2e) coincidence experiments using the modernized hardware and software.

Large portions of the (e,2e) coincidence experiment were modernized to improve and increase efficiency, timing, control, and monitoring of the system. This current chapter will also highlight the modernized components of the (e,2e) apparatus, and the reasons for such modifications, replacements, and/or upgrades.
Figure 3.1: A color-coded block diagram of the fundamental components making up the (e,2e) apparatus; the (e,2e) spectrometer and vacuum system (green), the experimental hardware (red), and the experimental software (blue); the dash-outlined blocks represent the modernized components.

3.2 An Overview of the (e,2e) Apparatus

The (e,2e) apparatus is divided into three fundamental parts; the (e,2e) spectrometer and the vacuum system in which it is housed, the experimental hardware, and the experimental software. Each part is made up of various components, each of which performs the functions seen in fig. 3.1. This block diagram is a general overview of the (e,2e) apparatus, and is not specific to the old or the modernized setup. The colors in the block diagram of fig. 3.1 correspond to the three fundamental parts mentioned above, while the dash-outlined blocks correspond to the modernized (modified, replaced, and/or upgraded) components.

The (e,2e) vacuum system is made up of a vacuum chamber, a top flange, and a pumping system. The vacuum chamber is for housing the (e,2e) spectrometer, the top flange is for supporting and providing access to and from the (e,2e) spectrometer via its feedthroughs, and the pumping system is for evacuating the vacuum chamber and maintaining pressures suitable for performing (e,2e) coincidence experiments.

The (e,2e) spectrometer (shown in fig. 3.2 and fig. 4.3) is composed of a two-stage non energy-selected electron gun [17], a pair of electrostatic energy analyzers [18], the gas jet, and a Faraday cup. The (e,2e) spectrometer is supported by four struts attached to a set of rotating gear plates that support the electron energy analyzers, and a rotating arm with
Figure 3.2: 3D model of the top flange and (e,2e) spectrometer. The labeled items are: (A) electron gun, (B) analyzer 1, (C) analyzer 2, (D) Faraday cup, (E) interaction region, (F) analyzer rotating gear plates, (G) rotating arm for moving the electron gun to coplanar and non-coplanar geometries, (H) a cross-section of the outer \( \mu \)-metal cylinder, (I) a cross-section of the vacuum chamber, (J) a cross-section of the inner \( \mu \)-metal cylinder, (K) analyzer 2 voltage feedthrough, (L) analyzer 2 EHT and signal feedthrough, (M) support struts for the (e,2e) spectrometer, (N) analyzer 1 voltage feedthrough, (O) top flange, (P) analyzer 1 EHT and signal feedthrough, (Q) analyzer 1 rotary feedthrough, (R) crane bracket, (S) analyzer stepper motors, (T) electron gun rotary feedthrough, (U) analyzer 2 rotary feedthrough, (V) interlocks’ system feedthrough, (W) gas line feedthrough, and (X) 52 way electron gun voltage feedthrough. The electron gun in this model is drawn in the perpendicular plane (\( \psi = 90^\circ \)). This 3D model is for demonstration purposes and is not drawn to scale.
an x-y translation table for the electron gun.

The (e,2e) experimental hardware is composed of computer-controlled voltage supplies for the (e,2e) spectrometer’s electrostatic elements, a stepper motor control module for positioning the electron energy analyzers, the channel electron multiplier supplies, the filament constant current supply, the fast detection electronics, the data acquisition electronics, and the analog-to-digital measurement electronics.

The (e,2e) experimental software is a set of tools, computer programs, and scripts used for experimental control of the voltage supplies and the stepper motor unit, and is used for data acquisition via the fast detection electronics, and data analysis.

3.3 The Modernized (e,2e) Apparatus

The modernization of the (e,2e) apparatus involved the modification, replacement, and upgrade of many components. This section compares the previous (e,2e) apparatus with the modernized setup, and addresses the motivation(s) behind such changes and upgrades.

Figure 3.3: A photograph of the previous (e,2e) apparatus; (e,2e) spectrometer, vacuum system, experimental hardware, and computer system.
The previous (e,2e) apparatus (shown in fig. 3.3), a block diagram of which can be seen in fig. 3.4, was a fully computer controlled and computer optimized system. It was based on a DOS-based 80286 PC with bus electronics, and a slave 8086 computer interfaced to a Keithley digital voltmeter. The programs and routines used to control the experiment and for analyzing the data were written in TURBO BASIC. The 80286 PC communicated with various digital-to-analog converter (DAC) cards, to supply voltages to the (e,2e)
spectrometer electrostatic elements. The electron energy analyzers were positioned using a custom-built (Compumotor-68000-CPU-based) stepper motor driver unit, addressed via a serial port located on the 80286 bus. The signals produced by the fast detection electronics, specifically the time-to-amplitude converter (TAC) and the two ratemeters, were fed to a multi-channel analyzer (MCA) card, and 32-bit counters, located inside the 80286 PC respectively. A data logger module composed of analog-to-digital converters was used to report measurements such as the position of the electron energy analyzers and the vacuum pressure.

![Figure 3.5: A photograph of the modernized (e,2e) apparatus; (A) the (e,2e) spectrometer and vacuum system, (B) the new experimental hardware, (C) the new computer control system.](image)

The previous setup of the (e,2e) apparatus required an upgrade for several reasons. The electronic components and devices were outdated and discontinued which made it difficult to replace them in the event of a malfunction or a failure. The computer system used DOS-based and TURBO BASIC programs, which made it difficult to upgrade and introduce new and improved features (control, optimization, acquisition, and analysis).

The modernization of the (e,2e) apparatus (seen in fig. 3.6) includes the following upgrades and modifications, a photograph of which can be seen in fig. 3.5:
Figure 3.6: A block diagram of the interfacing of the modernized (e,2e) apparatus; the (e,2e) spectrometer and vacuum system components (green), the experimental hardware components (red), and the experimental software (blue); the dash-outlined blocks represent the modernized components.

- Variable voltage supplies: The previous electron energy analyzer and electron gun variable voltage supplies were computer controlled via digital-to-analog converter cards that were difficult to troubleshoot due to outdated and discontinued technology. Each DAC is coupled with an amplifier which then drives an individual lens or deflector element. The variable voltage supplies were upgraded to opto-isolated printed circuit boards, that use fewer and inexpensive electronic components.
are capable of receiving data signals from micro-controllers, and delivering outputs with higher resolution. The new variable voltage supplies are consolidated into 19-inch rack units, one for the electron energy analyzers, and another for the electron gun and Faraday cup, each with its dedicated power supply unit (for supplying voltage rails). The new variable voltage supplies are controlled by Arduino MEGA 2560 micro-controllers [57] with both manual and computer control, using front panel rotary encoders, and via serial communication from a newly designed (e,2e) LabVIEW computer application. The outputs of the variable voltage supplies are mapped according to the previous feedthrough pin assignments, which (if needed) made switching to the older setup possible.

- Current monitor: A simpler current monitor board was designed for measuring the Faraday cup current. The output voltage of the current monitor board is sent to the (e,2e) LabVIEW computer application via a data acquisition card (explained below).

- Measurement: The previous (e,2e) apparatus used a 70 relay switching board for measuring the voltage or the current on each electrostatic element via a digital voltmeter. The additional measurements such as vacuum pressure and stepper motor potentiometers, were done using an analog-to-digital data logger. Similar to the previous variable voltage supplies, many of the electronic components in the relay switching board and data logger were outdated and discontinued. The new (e,2e) apparatus includes small and robust 12-bit resolution analog-to-digital converter printed circuit boards for measuring the voltages for each electrostatic element. The stepper motor potentiometers are measured by analog-to-digital pins on a dedicated stepper motor Arduino MEGA 2560 micro-controller. The measurements such as vacuum pressure and Faraday cup current, are carried out using the analog-to-digital converter pins on the PCI6221 LabVIEW data acquisition breakout board (CB-68LP) [56]. The new measurement modules are reliable, affordable, and easier to troubleshoot.

- Data Acquisition: The previous (e,2e) apparatus used an outdated multi-channel analyzer (MCA) board and 32-bit counters to measure the time-to-amplitude (TAC) signals and analyzer count rates, respectively. The modules were located on the bus inside the 80286 PC. A cost-effective and efficient replacement was to use the PCI6221 LabVIEW data acquisition card and CB-68LP breakout board for measuring both the TAC output signals and the analyzer count rates. This PCI6221 LabVIEW data acquisition card and its CB-68LP breakout board were designed by National Instruments to be used in conjunction with its own LabVIEW platform [56].
• Stepper Motor Control: As mentioned above, the electron energy analyzers were positioned using a custom-built (Compumotor-68000-CPU-based) stepper motor driver unit. Three voltage references were used for measurement of the three potentiometers located on the electron energy analyzer and electron gun drive shafts. For similar reasons, the unit was replaced with an Arduino MEGA 2560 based stepper motor control unit, capable of measuring the stepper motor potentiometers (with new voltage reference sources) and receiving angular position commands from the (e,2e) LabVIEW computer application. In addition, the actual stepper motors for both electron energy analyzers were replaced due to aging and potentiometer noise.

• Control and Acquisition Software: The previous (e,2e) apparatus used a DOS-based computer for control, optimization, and data acquisition. Loading the settings for an (e,2e) coincidence scan was done by editing text files with the the desired angular positions, runtimes, and other relevant information pertaining to the experiment. The DOS-based PC and its TURBO BASIC programs, were hence replaced with a new Windows-based computer running National Instrument’s LabVIEW platform. LabVIEW was used to design an experimental application for controlling and optimizing the experiment via the Arduino MEGA 2560 based variable voltage supplies and stepper motor control unit, measuring and acquiring analyzer count rates and coincidence signals, collecting and formatting the data in organized files using the JSON format [95], and more. The (e,2e) LabVIEW computer application can be easily upgraded and modified to include newer features.

• Analysis Software: The analysis on the machine was limited and experimental files had to be exported via floppy disk, and imported into a Windows-based machine running a program capable of extracting, analyzing and visualizing the data. The entire process was time-consuming, lacked a robust error handling mechanism, and required a machine that could perform all functions from performing a scan to analyzing the data. The new data extraction, analysis, and visualization routines were achieved by designing and running scripts, using the Python programming language [58] and Jupyter notebooks [96], respectively. The entire process (data extraction, analysis, and visualization) can be done by calling pre-written Python functions in a matter of seconds. Updates on the experiment in real-time can be sent to mobile phones via the Slack messaging application [97].

The (e,2e) spectrometer, vacuum chamber, pumping system, channel electron multiplier EHT supplies, and the filament constant current supply, were carried over to the new setup.
3.4 Conclusion

The purpose of this chapter was to give the reader a general overview of the \((e,2e)\) apparatus ((\(e,2e\) spectrometer and vacuum system, experimental hardware, and experimental software), the differences between the previous and the modernized \((e,2e)\) apparatus, and the reasons for the changes and upgrades. Details on the \((e,2e)\) spectrometer and vacuum system can be found in chapter 4. The details for all hardware and software (including upgrades and modifications) are covered in chapter 5 and chapter 6. The experimental procedures for operating the \((e,2e)\) spectrometer via its experimental hardware and software are detailed in chapter 7.
CHAPTER 4

The (e,2e) Spectrometer and Vacuum System

4.1 Introduction

This chapter is an overview of the entire (e,2e) vacuum system, with details on the components and inner workings of the (e,2e) spectrometer. The (e,2e) vacuum system (shown in fig. 4.1) is made up of a vacuum chamber, a top flange, and a turbo-molecular pump backed by a roughing pump. The initial designs for the entire machine were drawn up and built in 1984 by Jones, Read, and Cvejanovic, who worked closely with Vacuum Generators for the construction and manufacture of the vacuum chamber; more details can be found in Jones’ Ph.D. thesis [18]. The (e,2e) spectrometer is mounted from the top flange via four support struts, and lives inside the vacuum chamber. The (e,2e) spectrometer is composed of a non energy-selected electron gun, two hemispherical electron energy analyzers, a Faraday cup, and a gas jet. The electron energy analyzers are positioned by a pair of stepper motors, and an interlocks’ system prevents them from running into each other or into other components of the (e,2e) spectrometer.

4.2 The (e,2e) Vacuum System

The vacuum chamber is fabricated from non-magnetic 304 type stainless steel. It is manufactured with a protective surface layer with a high resistance to corrosion and oxidation, giving the steel an extremely low outgassing rate. For magnetic screening, 3 mm thick cylindrical inner and outer μ-metal shielding is placed (as shown in fig. 4.2). The chamber was designed with the dimensions of the (e,2e) spectrometer in mind, with a total inner chamber length of one meter, allowing the (e,2e) spectrometer and the interaction region to sit as far as possible from the top flange and the vacuum pump. The vacuum system employs a turbo-molecular pump and a roughing pump to achieve low background working pressures.
Figure 4.1: A drawing of the vacuum system.
Figure 4.2: Top view of the top flange of the (e,2e) vacuum system shown in fig. 4.1. The labeled items are: (A) interlocks' system feedthrough, (B) 52 way electron gun voltage feedthrough, (C) crane bracket, (D) analyzer 2 voltage feedthrough, (E) analyzer 2 EHT and signal feedthrough, (F) analyzer 1 voltage feedthrough, (G) gas line, (H) analyzer 1 EHT and signal feedthrough, (I) electron gun rotary feedthrough, (J) stepper motor, and (K) analyzer rotary feedthroughs. The (e,2e) spectrometer is supported by four support struts attached to the underside of the top flange. A crane bracket is attached to the upper side of the top flange, to lower and remove the (e,2e) spectrometer into and from the vacuum chamber.

4.2.1 The Top Flange

The top flange of the vacuum chamber is a 25 mm thick stainless steel "lid", that supports the (e,2e) spectrometer from its underside, and acts as a two-way system between the inside of the chamber and the experimental hardware on the outside. A crane bracket is attached to the upper side of the top flange, to lower and remove the (e,2e) spectrometer into and from the vacuum chamber. This is achieved by means of an overhead crane which attaches to the (crane) bracket’s latching mechanism. To ensure a good vacuum, a viton 'O' ring on the upper lip of the vacuum chamber is situated for the top flange to sit on. The two-way system of the top flange employs eleven CF70 Conflat flanges, and a larger central flange (as shown in fig. 4.2) which is not used in any of the experiments described.
in this thesis. The CF70 flanges accommodate feedthroughs for electrical, mechanical, and gas connections to the (e,2e) spectrometer. An oxygen-free copper gasket (acting as a vacuum seal) is placed between every feedthrough and CF70 port on the chamber. The feedthroughs used in the experiments described in this thesis are as follows:

- 1x 52-Way electron gun voltage feedthrough.
- 2x 19-Way feedthroughs for the energy analyzer voltages.
- 2x High voltage channeltron/EHT and pulse signal output feedthroughs.
- 3x Rotary motion feedthroughs for electron gun and analyzers.
- 1x Gas line feedthrough.
- 1x Interlocks feedthrough.
- 1x Grid power supply feedthrough.

4.2.2 The Pumping System

The vacuum system (as shown in fig. 4.1) can achieve background vacuum pressures of around $2 \times 10^{-8}$ torr and more, with the aid of a Balzers TPU-510 500/1 turbo-molecular pump, which is backed by an Edwards RV12 roughing pump. An ionization gauge used to measure pressure in the vacuum chamber, is accommodated by a side (CF70) flange which sits at a halfway point between the top flange and the base of the vacuum chamber, and approximately 50 cm from the interaction region. An ionization gauge controller is used to control the ionization gauge and report the pressure back to the computer control system.

The turbo-molecular pump is mounted from the underside of the vacuum chamber, which in turn is attached to the chamber via a DN150CF Conflat flange. The roughing pump is located on the same side as the ionization gauge. It is used to evacuate the vacuum chamber prior to turning on the Balzers turbo-molecular pump, while a second pump is used to evacuate the gas line before introducing the target gas into the vacuum chamber.

4.2.3 The Target Gas Line

As mentioned in section 4.2.1, the target gas line has a dedicated CF70 feedthrough via the top flange of the (e,2e) vacuum chamber. The target gas line is broken up into two parts by this feedthrough; the outer portion responsible for storing and introducing the target gas into the vacuum chamber, and the inner portion which delivers the target gas from the feedthrough to the interaction region.
The Outer Portion of the Gas Line:

The gas line feedthrough has a Negretti needle shut-off valve which is used to do the following:

- To control the flow of the target gas into the vacuum chamber prior to and during the experiment.
- To prevent the target gas from entering the vacuum chamber during a tuning of the (e,2e) spectrometer, or prior to opening up the (e,2e) vacuum chamber.

A short flexible pipe sits in between the gas line feedthrough, and a long ¼ inch grade 316 stainless steel tubing that runs back to the compressed target gas cylinder. These two pipes are joined together by a 3-way Swagelok pipe fitting, one side of which runs to a shut-off valve attached to the roughing pump. This shut-off valve is used when evacuating the gas line prior to running an experiment. The compressed target gas cylinder is equipped with a gas regulator to help maintain the gas pressure in the stainless steel tubing at approximately 1 bar. For flexibility when replacing the compressed target gas cylinder, a short and flexible ¼ inch copper tubing sits in between the gas regulator and the stainless steel tubing.

The Inner Portion of the Gas Line:

The target gas is introduced into the vacuum chamber via the CF70 feedthrough mentioned above. The target gas is delivered to the interaction region through a PTFE hose inside the chamber. The hose is shielded using a copper braid which is grounded, to prevent it from charging up by stray electrons during experimentation. At the exit end of the PTFE hose is a 20 mm long platinum iridium hypodermic needle with an exit aperture of 0.5 mm in diameter. The hypodermic needle, which produces the target gas beam at the interaction region, is attached to the rear of the electron gun at a 90° angle to the incident electron beam, and is set approximately 5 mm from the interaction region.

4.3 The (e,2e) Spectrometer

The (e,2e) spectrometer (shown in fig. 4.3) is composed of a two stage non energy-selected electron gun designed by Woolf (1989) [17], a pair of electrostatic energy analyzers designed by Jones (1984) [18], the gas jet, and a Faraday cup. A total of four struts support the main body of the (e,2e) spectrometer as seen in fig. 4.3. The struts are attached to a set of rotating gear plates that support the electron energy analyzers, and a rotating arm with an x-y translation table for the electron gun. The following sections will cover the inner workings of the (e,2e) spectrometer in more detail.
Figure 4.3: Photograph of the (e,2e) spectrometer with the electron gun in the perpendicular plane ($\Psi = 90^\circ$). The labeled items are: (A) rotating arm for moving the electron gun to coplanar and non-coplanar geometries, (B) electron gun (backside), (C) analyzer 1, (D) analyzer 2, (E) gas line, (F) Faraday cup, and (G) analyzer rotating gear plates. The (e,2e) spectrometer is supported by four support struts attached to the underside of the top flange. A crane bracket is attached to the upper side of the top flange, to lower and remove the (e,2e) spectrometer into and from the vacuum system. The shields around the analyzer lenses have been removed for this photograph.

4.3.1 The Electron Gun

The gun shown in fig. 4.4 consists of an electron source, a pair of triple element aperture lenses, and three pairs of x-y deflectors. All elements are electrically isolated using PTFE annuli, so that each element can sit at a different potential. The voltages provided by the computer-controlled power supplies are delivered to each element via PTFE insulated constantan wire. The electron gun is wired via an internal connector block so that the gun can be easily dismounted without rewiring the feedthrough. This internal connector block is a PTFE 25-way plug and socket between the 52-way electrical feedthrough and the electron gun.
Figure 4.4: A drawing of the components of the non energy-selected electron gun. The focusing elements of the triple aperture lenses are GL1B and GL2B. The x-y deflectors are GD1X/Y, GD2X/Y, and GD3X/Y. The defining 1 mm apertures are GA1 and GA2. The field free voltage for the defining apertures is GL1C.

The Electron Source

The gun’s electron source is composed of a triode arrangement of filament, grid, and anode. The filament is a commercially available tungsten hairpin, preferred for its robust qualities, and ability to withstand high current. The filament is mounted on a ceramic base, and can be easily replaced for a new filament if needed. The emission characteristics of the filament determine the electron energy resolution, which in turn classifies the electron gun as non energy-selected. This setup is ideal since the experiments explained in this thesis require high currents, electron incident beams with high energy, and do not require high energy resolution. The grid is a Pierce electrode [98], which essentially is a 3 mm stainless steel circular conical output aperture that sits after the tungsten filament, the tip of which slightly protrudes (0.5 mm) through the grid’s aperture. The anode is comprised of a 20 mm diameter cylindrical electrode, and a 1 mm diameter aperture, both of which are constructed from molybdenum. The anode is located after the grid, and together with the tungsten filament, they produce a relatively collimated beam of electrons at the aperture of the anode. The tungsten filament is electrically heated with the aid of a constant current supply, and by thermionic emission the filament produces electrons. The electron cloud is then shaped by the grid, which sits on a potential of anywhere between $-0.1 \text{ eV}$ and $-2 \text{ eV}$. The anode, which sits on a potential of anywhere between $60 \text{ eV}$ and $80 \text{ eV}$, extracts the electrons to be focused and defined by the second stage of the electron gun.
The Electron Gun Lens System

The electron beam at this stage of the gun is focused and then corrected using the deflectors explained in this section. Once the beam has been produced by the electron source, it is then focused by the pair of triple element aperture lenses that follow the anode. The twin lens system GL1B and GL2B in this stage is the most crucial, because alone it can focus and produce a narrow well-defined electron beam, with a zero beam angle at the interaction region. The potential of each of the focusing elements GL1B and GL2B can reach a $V_{max}$ of approximately 850 eV. The electron beam is collimated by the defining apertures GA1 and GA2, each of which have a diameter of 1 mm. These defining apertures sit in a field free region in a high potential of anywhere between 50 eV and 120 eV as set by the voltage GL1C. The effects of patch fields on the electron trajectories are reduced due to the potential in this field free region. The last step in this stage of the electron gun handles corrections due to any misalignment(s) caused by electrostatics, the physical placement, or by small imperfections in the electron gun. These corrections are done by the three sets of $x$-$y$ deflectors GD1, GD2, and GD3. Each set of deflectors consists of two pairs of parallel plates fixed at right angles to each other; an arrangement that allows for the electron beam to be steered in the $x$ and $y$ directions and onto the interaction region. The relative 0 V of deflector GD1 sits on the anode’s potential, while the voltages of deflectors GD2 and GD3 both sit on the voltage which sets GL1C. More details on the construction, inner-workings, and performance of the electron gun can be found in Woolf’s PhD thesis [17].

4.3.2 The Electron Energy Analyzers

The electron energy analyzers were first designed and built by Jones [18]. Changes were later made to the system by Turton in 1990 [20], followed by further modifications by Professor Andrew Murray. There are two identical electron energy analyzers, constructed from molybdenum, each of which consists of a hemispherical deflector energy selector, a triple aperture lens system, and a channel electron multiplier (CEM) as seen in fig. 4.5. The analyzer voltages are wired to the appropriate analyzer elements and channel electron multipliers, via two 19-way feedthroughs and two channeltron feedthroughs, respectively. There are two rotating gear plates, an inner and an outer, each of which supports an analyzer. The analyzer angles are adjusted using stepper motors, which move the electron energy analyzers around the x-z plane (as seen in fig. 1.1) within the physically allowed and computer-controlled angles. In addition, there is an interlocks system composed of nine photodiode/phototransistor pairs that prevent the analyzers (and electron gun) from running into each other and other components in the (e,2e) spectrometer such as the electron gun and Faraday cup. Additional modifications were made to the stepper control...
and interlocks systems for the experiments as detailed in this thesis.

Figure 4.5: A drawing of the components and voltage supplies for the electron energy analyzers: a triple cylindrical lens system, a pair of x-y deflectors, a hemispherical energy selector, and a channel electron multiplier (CEM). The interaction region is imaged onto a 1 mm real aperture and a second 1 mm aperture at the output sets the energy resolution. The distance between the interaction region and defining aperture is 130 mm, so that \( P = Q = 4D \), where \( D \) is the lens diameter. The gap between lenses is set to 0.1 \( D = 1.625 \) mm. See text for more details.
The Electron Energy Analyzer Elements

Each electron energy analyzer is composed of a triple cylindrical lens system, a pair of x-y deflectors, a hemispherical energy selector, and a channel electron multiplier (CEM) as shown in fig. 4.5. The electron energy analyzer elements are all constructed from molybdenum. The triple cylindrical lens system is composed of:

- AA1, a conical element referenced to earth that is closest to the interaction region.
- AL1A, a lens element referenced to earth that is physically connected to AA1.
- AL1B, a focusing lens element.
- AL1C, or simply known as the the analyzer mean (AM), which is a field free region.
- AD1X/Y, a set of x-y deflectors whose $V$ is referenced to the potential of AL1C (AM).

The conical element AA1 defines a $5^\circ$ acceptance half angle, which reduces the electron scattering angle that the analyzer accepts. This element acts as the entrance slit, and along with the lens AL1A, they aid in the detection of the scattered and ejected electrons, the energies of which are selected by tuning the residual energy of each analyzer. The electrons are then focused by the lens element AL1B, into the field free region AL1C (or AM). As mentioned above, AL1C is known as the analyzer mean, because its voltage defines the pass energy of the analyzer. Within the cylindrical element AL1C is a set of x-y deflectors AD1X/Y; a set of parallel plates similar in principle and function to those that exist in the electron gun. The analyzer deflectors provide corrections due to any mechanical or electrostatic misalignment(s), or physical imperfections in the lens element AL1C (or AM). The deflector $+/-$ power supplies are referenced to the AL1C field free region’s potential, and can have a range of $+/-15\, V$. Once the deflector corrections have been made, the electrons are guided into the entrance slit of the hemispheres. Each of the hemispherical deflectors are composed of:

- AIH, an inner hemisphere.
- AOH, an outer hemisphere.

Each inner and outer hemisphere is referenced to its own power supply; the inner hemisphere AIH is referenced to a power supply with a $V_{\text{max}}$ of 60 eV, while the power supply of the outer hemisphere can reach $V_{\text{max}}$ of 30 eV. The inner hemisphere has a radius $r_{\text{inner}} = 17.2\, \text{mm}$ and a potential $V_{\text{inner}}$, and the outer hemisphere has a radius $r_{\text{outer}} = 33.6\, \text{mm}$ and a potential $V_{\text{outer}}$. An electric field is generated between the two hemispheres due to the
applied potentials $V_{\text{inner}}$ and $V_{\text{outer}}$. As a result, electrons with different kinetic energies follow different circular paths between the two hemispheres. The electrons that follow the mean circular path at a radius $r_{\text{mean}} = 25.4$ mm halfway between the inner and outer hemispheres, undergo energy selection as they pass through the exit aperture of the hemispheres. These electrons have a kinetic energy $E = eV_{\text{mean}}$ referred to as the pass energy. The potentials $V_{\text{inner}}$ and $V_{\text{outer}}$ are given by:

$$V_{\text{inner}} = V_{\text{mean}} \left[ \frac{2r_{\text{mean}}}{r_{\text{inner}}} - 1 \right] \tag{4.1}$$

and

$$V_{\text{outer}} = V_{\text{mean}} \left[ \frac{2r_{\text{mean}}}{r_{\text{outer}}} - 1 \right] \tag{4.2}$$

The tuning conditions for the inner and outer hemispheres are determined by the desired pass energy for a scan. The pass energy of the electrons arriving at the entrance aperture of the hemispherical energy selector is set by the analyzer mean (AM) voltage. The analyzer mean, and the inner and outer hemisphere voltages are usually tuned and left untouched during experimentation. The pass energy, and the diameter of the entrance and exit apertures of the hemispheres, determine the energy resolution of a hemispherical electron energy analyzer, which is given by:

$$E_{\text{res}} = \frac{Ed_{\text{aperture}}}{2r_{\text{mean}}} \tag{4.3}$$

where $E$ is the pass energy determined by the analyzer mean, and $d_{\text{aperture}}$ is the resolving aperture diameter of the hemispheres. For a pass energy of 15 eV and a fixed exit aperture diameter of 1 mm, the energy resolution $E_{\text{res}}$ of a hemispherical electron energy analyzer is $\approx 300$ meV. The Jost correctors (anuli) shown in fig. 4.5 are referenced to the analyzer mean potential, and are there to correct the fringing fields by terminating the electric field lines at the entrance and exit apertures of the hemispheres. Once the electrons pass around the hemispheres, the energy selected electrons are then guided onto the input of the Mullard X919BL channel electron multipliers.

The Channel Electron Multiplier

The diameter of the input aperture of a Mullard X919BL channel electron multiplier is 10 mm, which makes it easier for the CEM to pick up electrons from the hemisphere’s exit aperture diameter of 1 mm. The front cone of the channel electron multiplier is referenced to
the potential $V_{\text{mean}}$, and its output (anode) is set to a high voltage of approximately 2.3 kV. The high voltages are provided by two Brandenberg 5 kV supplies. The bombardment of the energy selected electrons upon the surface of the front cone of the channel electron multiplier creates secondary emission of electrons with a gain of up to $\sim 10^8$. This outputs a negative going pulse of approximately 25 - 40 mV into a 50 Ω load. To prevent pickup noise from traveling into and out of the system, a low pass filter is placed in between the CEM and its EHT supply, as shown in fig. 4.6. This filter has a slow response time which prevents the pulse detection electronics from seeing any sudden high voltage spikes from the EHT supply. The fast pulses that correspond to the electrons detected by the CEM are channelled through a 1 nF 6 kV capacitor to eliminate the DC voltage, which in turn gets fed to the input of a preamplifier.

The pulse detection electronics are decoupled from the EHT line using two 1 nF 6 kV capacitors. The 1 kΩ is decoupled to earth from any CEM high frequency pulses via the 1 nF 6 kV capacitor (C2). The voltage across the 1 kΩ resistor is connected to the 50 Ω input of the preamplifier via the 1 nF 6 kV capacitor (C3). The 10 MΩ resistor is for safety purposes; it protects the preamplifier when it is connected or disconnected to the pickoff and filter circuit.

4.3.3 The Stepper Motor and Interlocks System

The rotating gear plates that support the electron energy analyzers (shown in fig. 4.3), are controlled by external stepper motors attached to the rotary feedthroughs of the top flange. Since the analyzers are mounted from these gear plates, they are fixed in a plane. The analyzers are able to view the interaction region between the angles of 25° and 130° in symmetric angular scans, while each analyzer can independently move to a minimum of 20°, which can be useful in asymmetric angular scans. The reason for this limitation is mechanical, because the size and shape of the analyzers do not allow them to move to
smaller angles (or larger angles) before running into each other. New stepper motors were installed due to aging and unstable (noisy) holding current of the previous models. The new stepper motors rotate (with an angular resolution of) $1.8^\circ$ per step, and are connected to an 18:1 gearbox which drives a 10:1 gear; a combination ($1.8/18/10$) that requires the stepper motors to move 100 steps per degree. Attached to each stepper motor rotary feedthrough is a potentiometer that reports back a voltage to the Arduino MEGA 2560 based stepper motor control board (detailed in section 5.5.1); a voltage that corresponds to the position of an electron energy analyzer (or electron gun). It is worth noting that the current potentiometers are noisy, and will be replaced in the next iteration with an optical encoder that would eliminate the problems with noise and the effects of potentiometer aging.

The interlocks system is a hardware system that prevents the electron energy analyzers from running into each other and other components in the (e,2e) spectrometer such as the electron gun and Faraday cup. The interlocks system is comprised of nine photodiode/phototransistor pairs inside the vacuum chamber. If the analyzers are moved to a position where a collision could occur, then a flag passes in between the photodiode/phototransistor pair, shutting off the light and sending a signal to the Arduino-based stepper motor control board (through the opto-isolators). This then brings the stepper motors to a stop until the fault is rectified. A dedicated 19 way CF70 feedthrough is used to send signals to the Arduino MEGA 2560 based stepper motor control board, from the nine photodiode/phototransistor pairs inside the vacuum chamber.

The previous hardware used to control the stepper motors and communicate with the internal interlocks’ system was replaced with the new stepper motor control and data acquisition unit. In addition, and as mentioned above, the old stepper motors were also replaced. These replacements and modifications are detailed in section 5.5.

4.4 Conclusion

This chapter detailed the components and inner workings of the (e,2e) spectrometer and the entire vacuum system. The next chapter will cover the new external hardware, which includes the computer-controlled voltage supply units used to operate and control the (e,2e) spectrometer, the (e,2e) coincidence detection electronics, the stepper motor and data acquisition unit, the high voltage EHT supplies, and the filament constant current supply.
CHAPTER 5

Experimental Hardware

5.1 Introduction

The modernized (e,2e) system includes new computer-controlled Arduino MEGA 2560 based power supplies for the electron gun, the electron energy analyzers, and the Faraday cup, a computer-controlled stepper motor control system for positioning the energy analyzers, a LabVIEW-based data acquisition module, and a Windows-based computer containing and running the essential hardware and software, respectively. The (e,2e) spectrometer and the vacuum system, the channel electron multiplier EHT high voltage supplies, and the pulse detection electronics were carried over from the old to the new setup.

The electronics behind the new power supplies, the variable voltage supplies, and the stepper motor control and data acquisition unit were all designed and built in-house. Electronic circuits were designed, then used to make printed circuit boards (PCBs), which were manually chipped with the appropriate electronic components. The new setup delivers voltages to 7 elements in each electron energy analyzer, and to 18 elements making up the electron gun, and the Faraday cup. These units are also equipped with power supplies for the Arduino MEGA 2560 micro-controllers, the stepper motors, the analyzer analog-to-digital converter boards, and the current monitor board.

The experimental hardware is shown in the color coded block diagram in fig. 3.6. The dash-outlined blocks of the block diagram represent the new hardware additions and replacements; two power supply units, two variable voltage supply units, and one stepper motor control and data acquisition unit. The new and old experimental hardware are all housed in a 19-inch rack cabinet.

The power supply units (explained in section 5.3) deliver the voltage rails needed to power the variable voltage supply boards’ electronic components. The interfacing between a power supply unit and a variable voltage supply unit is shown in fig. 5.1. The variable voltage supply units (explained in section 5.4) consist of an Arduino MEGA 2560 based control panel, digital-to-analog electronics, variable voltage supply electronics, and input and output back panel interface boards. The Arduino MEGA 2560 micro-controller allows
for manual and computer control of the variable voltage supply boards, and is equipped
with TFT LCD screens for voltage monitoring. The additional electronics for the Faraday
cup supply and a current monitor are included in the electron gun and Faraday cup power
and variable voltage supply units.

Figure 5.1: Block diagram of the interfacing between a power supply unit with a variable
voltage supply unit. The dash-outlined box for the Faraday cup supply and current monitor
electronics are located in the electron gun power supply and variable voltage supply units. The
remaining components of this block diagram are common to all power supply and variable
voltage supply units.

Figure 5.2: Block diagram of the stepper motor control and data acquisition unit.
The stepper motor control and data acquisition unit (explained in section 5.5) is designed for stepper motor control, and measurement and detection purposes. The stepper motor control side of this unit drives and controls the electron energy analyzer stepper motors, reports their position as well as that of the electron gun, and prevents the (e,2e) spectrometer elements from running into each other in the event of a malfunction. The data acquisition side is responsible for transmitting output signals from the high speed pulsed detection electronics, and sending pressure and current measurements to the LabVIEW-based computer control system. A block diagram of the stepper motor control and data acquisition unit’s internal components is shown in fig. 5.2.

The experimental hardware that remained from the previous setup (covered in section 5.6 and section 5.7) include the constant current filament supply, the two channel electron multiplier high voltage supplies, and the high-speed detection electronics (constant fraction discriminators, ratemeters, delay generator, and time-to-amplitude converters).

5.2 Printed Circuit Boards

Printed circuit boards (PCBs) were the method of choice for developing the new electronics for the (e,2e) spectrometer’s power supply and computer controlled variable voltage supply units, as well as the stepper motor electronics. Printed circuits boards are affordable, easy to make, and reduce the number of faulty or loose connections by replacing wires with copper tracks.

The reasons for using printed circuit boards were several: printed circuit boards are usually made for use with dual in-line (DIP) package electronic components, most of which can be secured by inserting the pins of the electronic components through the PCB’s hand-drilled holes, then soldered directly onto the PCB’s copper pads. The DIP package of electronic components are widely accessible from electronic component vendors, are relatively affordable when compared to other packages, are large enough for the naked eye to see, and are easier to troubleshoot or replace in the event of a malfunction compared to surface mount components.

A few electronic components with surface mount technology were used in developing the electronics. The disadvantage in using such components is the smaller footprint and troubleshooting difficulty. However, with surface mount electronics becoming lower in cost, the next logical step in developing electronics would be to send PCB layouts to manufacturing and assembly services, which are becoming more affordable. For the electronics detailed in this thesis however, all printed circuit boards (PCBs) were made by hand in the lab at Manchester.
5.2.1 Making Printed Circuit Boards

A printed circuit board is essentially a non-conductive substrate with copper sheets laminated (with heat) onto one or both sides of the substrate. A PCB layout is designed then used to define the tracks onto the copper sheets via a 3-step process: exposure, developing, and etching.

Steps:

- PCB Design: The PCB layout is based on the circuit design drawn up in an electronic design computer application (e.g. Eagle by Autodesk or KiCad).

- Printing: The PCB layout is printed in all black on transparent paper to allow for the desired layout to be exposed on the copper sheets of the PCB.

- Exposing: This is done by exposing the PCB layout onto the copper sheets. The PCB layout is laid directly on top of the board and exposed to UV light for about 90 seconds.

- Developing: The PCB is immediately placed in a developing solution for about 30-45 seconds.

- Etching: The PCB is placed in an etching solution until the desired layout is the only remaining copper on the PCB.

- Cleaning: Since the copper sheets have a resist layer on top, the PCB is then cleaned using Methanol.

- Trimming: The PCB is trimmed using a guillotine to the desired size.

- Drilling: The desired holes for the electronic components are drilled.

- Soldering: Electronic components are soldered onto the printed circuit board (PCB).

- Testing: Final PCB is tested for shorts, faulty joints, etc.

5.3 The Power Supply Units

The power supply units (PSUs) were designed and built in-house, equipped with back panel interface boards for supplying the voltage rails via D25 cables directly to the variable voltage supply units. One unit houses the power supplies used for the electron gun voltage supply boards, Faraday cup voltage supply, current monitor board, and the Arduino-based control board. Another unit houses the power supplies needed for both energy analyzer voltage supply boards, analog-to-digital converter boards, and the Arduino-based control boards. The stepper motor control boards and their power supplies, along with the LabVIEW data acquisition breakout board, are all housed in an additional unit.
5.3.1 The Power Supply Circuitry and Electronics

The number of elements making up the electron gun (as shown in fig. 4.4) and electron energy analyzers (fig. 4.5) determined the required number of variable voltage supplies, which in turn determined the number of voltage rails needed to power the devices onboard the variable voltage supply circuit boards. Elements referenced to other elements (e.g. the analyzer deflectors ADX/Y were referenced to the analyzer mean AM voltage), required floating voltage supplies, which in turn required dedicated power supplies (voltage rails). Furthermore, voltage supplies referenced to a common potential point could share a power supply, only if the current drawn meets the electronic component specifications. Determining the number of voltage rails required was the first step in designing the power supply circuitry. The electron gun and Faraday cup voltage supplies required the following voltage rails:

- $\pm 15V + 5V$ for Grid, Gun Fine.
- $-5V + 30V$ for Gun Fine.
- $\pm 12V + 5V$ for GL1B, GL2B.
- $\pm 12V + 5V$ for Gun coarse supply.
- $+120V$ for Anode, GL1C.
- $\pm 15V \pm 5V$ for Anode, GL1C.
- $\pm 15V + 5V$ for GD1X.
- $\pm 15V + 5V$ for GD2X, GD3X.
- $\pm 15V + 380V$ for Faraday Cup.
- $\pm 15V \times 2$ for Current Monitor.
- $+5V$ for Arduino Control Board.

Each electron energy analyzer voltage supply unit required the following voltage rails:

- $+120V$ for ARSE, AL1B.
- $\pm 15V \pm 5V$ for ARSE, AL1B, AAM, AIH, AOH.
- $\pm 15V + 5V$ for ADX, ADY.
- $+30V + 60V$ for AAM, AIH, AOH.
- $\pm 15V + 5V$ for ADC Boards.
- $+5V$ for Arduino Control Boards.
The power supply printed circuit boards (PCBs) are fundamentally the same in design. The circuit design (as shown in fig. 5.3) is based on a step-down transformer able to supply the required amount of current, the voltage of which is then rectified, split, regulated, and then routed as an output voltage rail to the appropriate pin on the back panel boards. The output voltage rails are carried over via D25 cables to their respective pins on the voltage supply unit’s back panel boards. The amount of current and the voltage rails required, determine the type of transformer and voltage regulators needed for a particular power supply circuit board. The voltage rails listed above were used to design the circuits detailed in the sections below.

**Figure 5.3:** Block diagram of the principle circuit design for the power supply printed circuit boards.

The ±15 V, ±12 V, and ±5 V Power Supply Circuitry

The power supply schematic shown in fig. 5.4 is based on the principle circuit design shown in fig. 5.3. The power supply schematic can be modified to output one of the following sets of DC voltages:

- ±15 V and +5 V
- ±15 V and ±5 V
- ±12 V and ±5 V

**Figure 5.4:** The ±15 V, ±12 V, and ±5 V power supply circuitry. The capacitor and resistor values are: $C1 = C2 = 6800 \mu F$, $C3 = C4 = C5 = C6 = C7 = C8 = 0.1 \mu F$, and $R1 = R2 = 4.7 \, k\Omega$. See text for details.
Following the four stages shown in fig. 5.3, the required components for the power supply schematic shown in fig. 5.4 are identified:

1. The step-down transformer used to output any of the desired sets of DC voltages is the same across all three circuits. For this particular design, the transformer used has the following specifications:
   - Secondary voltage rating: 15 V AC.
   - Primary voltage rating: 230 V AC.
   - Power rating: 3.2 VA.
   - Number of outputs: 2.
   The two outputs of the step-down transformer are connected to a bridge rectifier.

2. The bridge rectifier used here is an integrated circuit composed of four 1N4007 diodes as shown in fig. 5.4. A bridge rectifier converts an alternating current (AC) to a direct current (DC) by 'rectifying' the mains AC input to a DC output. The two 6800 μF electrolytic capacitors C1 and C2 are required to reduce the ripple voltage. The 0.1 μF tantalum bypass capacitors C3 and C4 are used to eliminate high frequency noise. The + and - DC voltages are fed to the inputs the positive and negative voltage regulators respectively.

3. The voltage regulators in fig. 5.4 are labeled as VRegx where x is a number used to identify the component in the schematic. Each voltage regulator contains a four digit identifier. The 78xx family of linear voltage regulators produce a positive voltage relative to a common ground. The last two digits (xx) are replaced with numbers that indicate the output voltage. For example, a 7815 linear voltage regulator will produce a +15 V at its output. The 79xx family of linear voltage regulators have a similar numbering system, but produce a negative voltage at their output. For example, a 7905 linear voltage regulator will produce −5 V at its output. So for the set of output DC voltages ±15 V and ±5 V, the 7815, 7915, 7805, and 7905 linear voltage regulators are required. A bypass 0.1 μF capacitor at the output of each of the voltage regulators is required. These capacitors should be rated at a minimum of 30 V. For example, these capacitors for a ±15 V and ±5 V power supply would be capacitors C5-C8 in fig. 5.4. Heatsinks are required for the 78xx family of linear voltage regulators. These heatsinks allow the voltage regulators to operate at their limits by dissipating the thermal energy caused by power dissipation of the device. The output of each voltage regulator is then routed to its appropriate output header pin.
4. A DC voltage from each voltage regulator is routed to a header pin to be carried over to a back panel interface board via crimped wires. The common 0 V is also routed to one or more of these header pins. These DC voltages provide the voltage rails required to power up the electronic components onboard the voltage supply circuit boards.

The +30 V and +60 V Power Supply Circuitry

The schematic for the +30 V and +60 V power supplies is shown in fig. 5.5. It is similar in design to the circuit shown in fig. 5.4, with the exception of the step-down transformer, the electrolytic capacitors, and voltage regulators used. This power supply schematic can be modified to produce +30V and/or +60V at its output. The step-down transformer used has the following specifications:

- Secondary voltage rating: 24 V AC.
- Primary voltage rating: 230 V AC.
- Power rating: 3.2 VA.
- Number of outputs: 2.

The bridge rectifier, and the two bypass capacitors C3 and C4 are the same to those used in the circuit shown in fig. 5.4. The two electrolytic capacitors C1 and C2 are 1000 μF 100 V each. The voltage regulator used here is the TL783 3-terminal adjustable positive voltage regulator [99]. This voltage regulator is capable of producing a voltage range of 1.25 V

![Figure 5.5: The +30 V and +60 V power supply circuitry. The capacitor and resistor values are: C1 = C2 = 1000 μF 100 V, C3 = C4 = C5 = C6 = 0.1 μF, R1 = R5 = 82 Ω, and R2 = R3 = R4 = 3.9 kΩ. See text for details.](image-url)
to 125 V by connecting the adjustment (ADJ) pin to two external resistors (as a resistor divider) to set the desired voltage $V_O$:

$$V_O = V_{ref} \left( 1 + \frac{R_{prog}}{R_{load}} \right) \quad (5.1)$$

where $V_{ref} = 1.25 \text{ V}$ is generated by the internal reference between the output (OUT) and adjacent (ADJ) pins. The resistor $R_{prog}$ is the programming resistor used to adjust the output to the desired voltage. The resistor $R_{load}$ ($R_1$ and $R_5$ in fig. 5.5) requires a recommended value of 82 $\Omega$ which provides the minimum load current of 15 mA. The required resistor value for $R_{prog}$ for a +60 V output can be found using eq. (5.1):

$$R_{prog} = \left( \frac{60 \text{ V}}{1.25 \text{ V}} - 1 \right) \times 82 \Omega = 3854 \Omega \quad (5.2)$$

which can be achieved by using a 3900 $\Omega$ resistor for $R_2$. For a +30 V output, the required resistor value for $R_{prog} \approx 1950$. This can be achieved by using two 3900 $\Omega$ resistors in parallel for $R_3$ and $R_4$. The recommended values for the bypass capacitors C5 and C6 is 0.1 to 1 $\mu$F. The voltage regulator outputs are routed to header pins and carried over to the back panel interface boards via crimped wires.

The +120 V Power Supply Circuitry

The schematic for the +120 V power supply is shown in fig. 5.6. It is similar in design to the circuit shown in fig. 5.5. The only exceptions are the two 1N4007 diodes instead of the bridge rectifier, and the resistor values used for the programming resistor $R_{prog}$ which adjusts the output to the desired voltage. A 3900 $\Omega$ resistor is placed in series with a 10 k$\Omega$ potentiometer which is trimmed to produce +120 V at the output.

![Figure 5.6: The +120 V power supply circuitry. The capacitor and resistor values are: $C_1 = C_2 = 1000 \mu$F 100 V, $C_3 = C_4 = C_5 = 0.1 \mu$F, $R_1 = 82 \Omega$, $R_2 = 3.9 \text{k}\Omega$, and $R_3 = 10 \text{k}\Omega$.](image-url)
The Faraday Cup ±15 V and +380 V Power Supply Circuitry

The Faraday cup voltage supply board requires voltage rails of ±15 V and +380 V. The design for a dedicated ±15 V is based on the circuit shown in fig. 5.4. The schematic for the +380 V power supply is shown in fig. 5.7. The design is based on a 240 V 20 VA chassis type mains step-down transformer, a UD6KB80 6 A 800 V bridge rectifier, and various capacitors across the bridge rectifier’s ‒ and + outputs.

![Figure 5.7: The +380 V power supply circuitry. The capacitor and resistor values are: C1 = C2 = 220 µF 400 V, C3 = C4 = 1 µF 400 V, and R1 = 33 kΩ 1 W. See text for details.](image)

The capacitor and resistor values are as follows:

- C1 and C2: 220 µF 400 V.
- C3 and C4: 1 µF 400 V.
- R1: 33 kΩ 1 W

This power supply and a ±15 V are built on the same printed circuit board and share a common 0 V.

The Current Monitor ±15 V Power Supply Circuitry

The current monitor ±15 V power supply is based on the circuit design shown in fig. 5.4, and is modified to supply two separate ±15 V outputs. The step-down transformer used for each supply has the following specifications:

- Secondary voltage rating: 15 V AC.
- Primary voltage rating: 230 V AC.
- Power rating: 1.0 VA.
- Number of outputs: 2.

The bridge rectifiers used are the same as those used in the circuit shown in fig. 5.4, and the capacitors C1 and C2 that follow the bridge rectifier are both 1000 µF. The bypass capacitors C3-C6 are all 0.1 µF.
The +5 V Power Supply Circuitry

This design supplies +5V to the Arduino MEGA 2560 based control boards. It relies on a 7805 linear voltage regulator and a 9 VAC/230 VAC/5 VA step-down transformer. The schematic is also based on the circuit shown in fig. 5.4.

The schematics shown in figs. 5.4 to 5.7 were used to create printed circuit board (PCB) layouts (a process explained in section 5.2) for all required power supplies detailed on page 95.

5.3.2 The Power Supply Output Interface Boards

The voltage rails produced by the power supplies detailed above, are carried over to a set of printed circuit boards (PCBs) mounted on the back panel of the power supply units. The back panel printed circuit boards (shown in fig. 5.8 and fig. 5.10) are equipped with adaptors for use with D25 serial cables between the power supply and variable voltage supply units. The back panels shown in fig. 5.9 and fig. 5.11 are equipped with a mains plug and power switch. The back panel of the electron gun power supply unit is comprised of the three output interface boards shown in fig. 5.8. The three electron gun DC output interface boards are:

- Board A: Grid, Gun Fine, Gun Coarse, GL1B, and GL2B.
- Board B: Faraday Cup and Current Monitor.
- Board C: Anode, GL1C, and Gun X/Y Deflectors.

The back panel of the electron energy analyzers’ power supply unit is comprised of two identical output interface boards shown in fig. 5.10. These boards are:

- Board A: Electron energy analyzer 1 elements.
- Board B: Electron energy analyzer 2 elements.

5.3.3 Physical Layout of the Power Supply Units

The power supply electronics are consolidated into two units located inside a 19-inch rack cabinet, the size of which is an industry standard for mounting electronic equipment. As mentioned in the beginning of this chapter, one unit houses the power supply units used for the electron gun voltage supply boards, Faraday cup voltage supply, current monitor board, and the Arduino control interface (as shown in fig. 5.12). A second unit houses the power supply units needed for both energy analyzer voltage supply boards, analog-to-digital converter boards, and the Arduino control interface (as shown in fig. 5.13).
Figure 5.8: The electron gun, Faraday cup, and current monitor power supply back panel interface boards. Refer to tables A.1 to A.3 in appendix A.1 for voltage pin assignments.

Figure 5.9: Layout of the electron gun, Faraday cup, and current monitor power supply back panel interface boards.
Figure 5.10: The electron energy analyzer power supply back panel interface boards. The design for this board is identical for both electron energy analyzers. Refer to table A.4 in appendix A.2 for voltage pin assignments.

Figure 5.11: Layout of the electron energy analyzer power supply back panel interface boards.
Figure 5.12: Photograph of the electron gun, Faraday cup, and current monitor power supply unit (a top-down view). The labeled boards: (A1) to (A3) the back panel output interface boards A, B, and C, (B1) to (B3) +5 V power supplies, (C1) power supplies for GL1B, GL2B, gun energy coarse, gun energy fine, and grid (C2) power supplies for GD1X/Y, GD2X/Y, GD3X/Y, GL1C, and Anode, (D) the Faraday cup power supply, and (E) the current monitor power supply.
Figure 5.13: Photograph of the electron energy analyzers power supply unit (a top-down view). The labeled items are: (A1) and (A2) power supplies for electron energy analyzer 1 and 2 respectively, (B1) and (B2) the back panel output interface boards A and B, and (C1) to (C3) +5 V power supplies.
5.4 The Variable Voltage Supply Units

The variable voltage supply units are explained by considering their individual components. The voltage rails from the power supply units are fed through the back panel of the variable voltage supply units in the same manner; through back panel interface boards that mirror their respective boards on the back panel of the power supply units. The voltage rails are carried over via crimped wires to the voltage supply boards.

The voltage supply boards can be considered as two different units: the digital-to-analog electronics and the variable voltage supply electronics. The principle design for the digital-to-analog side of the electronics is identical for all boards that require computer control. The design for the variable voltage side of the electronics depends on the output voltage required by different elements inside the (e,2e) spectrometer. The variable voltage supply electronics use operational amplifiers (or similar components), to amplify the output voltage from the digital-to-analog side. The output voltages are then carried over to back panel interface boards via crimped wires, and are then delivered to the (e,2e) spectrometer through the vacuum system’s feedthroughs. Several designs for the variable voltage side of the boards are reused if the desired output voltage is the same (e.g. the deflectors for the analyzers and electron gun require the similar voltages and thus similar circuits.)

The remaining circuit designs are for various boards such as the Faraday cup, current monitor, and Arduino control boards, all of which are covered in the sections below. The voltages required to power the electronic components on these boards are supplied via back panel boards that interface to their respective boards on the back panel of the power supply units.

The variable voltage supplies are housed in 19-inch rack units. The variable voltage supply units are more sophisticated in function in comparison with the power supply units, and so further details of these are given in the sections below.

5.4.1 The Variable Voltage Supply Input Interface Boards

As mentioned above, the voltage rails supplied by the power supply units, are fed through the back panel input interface boards of the voltage supply units via D25 serial cables. These input interface boards deliver the voltages necessary to power up the digital-to-analog and variable voltage supply electronics. The back panel of the electron gun voltage supply unit is equipped with the three input interface boards shown in fig. 5.14 and fig. 5.15.
The three electron gun DC input interface boards are:

- Board A: Grid, Gun Fine, Gun Coarse, GL1B, and GL2B.
- Board B: Faraday Cup and Current Monitor.
- Board C: Anode, GL1C, and Gun X/Y Deflectors.

**Figure 5.14:** The electron gun, Faraday cup, and current monitor variable voltage supply back panel input interface boards. Refer to tables A.5 to A.7 in appendix A.3 for voltage pin assignments.

**Figure 5.15:** Layout of the electron gun, Faraday cup, and current monitor variable voltage supply back panel input interface boards.

The back panel of the analyzers voltage supply unit is comprised of two identical input interface boards shown in fig. 5.16 and fig. 5.17. These boards are:

- Board A: Electron energy analyzer 1 elements.
- Board B: Electron energy analyzer 2 elements.
5.4.2 Arduino Control Electronics

This section details the hardware side of the Arduino-based control boards used to control the digital-to-analog and variable voltage supply electronics covered in section 5.4.3 and section 5.4.4 respectively. The Arduino MEGA 2560 [57] shown in fig. 5.18 is based on the ATmega2560 micro-controller chip by Atmel [100]. The board is equipped with the following components and interfaces:

- 54 digital input/output pins (15 of which can be used as pulse width modulation (PWM) outputs)
- 16 analog inputs with 10-bit resolution
- 4 UARTs (hardware serial ports)
5.4 The Variable Voltage Supply Units

- 1 16 MHz crystal oscillator
- 1 USB connection for serial communication and/or power
- 1 power jack
- 1 ICSP header
- 1 reset button

![Arduino MEGA 2560 micro-controller](image)

**Figure 5.18:** The Arduino MEGA 2560 micro-controller. It was used to design the control boards for the electron gun and electron energy analyzer variable voltage supply units. See text for details.

The Arduino MEGA 2560 is powered using an external power supply or via a USB connection, and the power source is automatically selected by the Arduino itself. The recommended operating voltage range for the Arduino is 7 to 12 V. The ATmega2560 has 128 KB of flash memory (124 KB of which are programmable and 4 KB are used for the boot-loader). It also has 8 KB of SRAM and 4 KB of EEPROM (for data storage) with a 100,000 write limit. The Arduino MEGA 2560 has a number of built-in facilities for communicating with a computer, another Arduino, or other micro-controllers. The ATmega2560 chip supports Serial Peripheral Interface (SPI) and Inter-Integrated Circuit (I2C) communication. These methods of communication allow for seamless control of components such as a digital-to-analog converter device or an external liquid crystal display (LCD).

The Arduino MEGA 2560 can be programmed with the Arduino software; it runs machine code compiled from C or C++. Programs written for Arduino micro-controllers are divided
into three main parts: structure, values (variables and constants), and functions, which makes it incredibly easy to write efficient, minimal, and effective code. More details on the Arduino’s digital functions, algorithms, and programming methods and tools can be found in section 6.3.

Three Arduino MEGA 2560 based control boards were designed and constructed; one for each electron energy analyzer, and one for the electron gun. Each control board requires the following components:

- An Arduino MEGA 2560 [57].
- ATtiny85 micro-controllers [100].
- Alps’ 12 mm incremental rotary encoders with push buttons (EC12E Series) [101].
- An Adafruit 2.2” TFT LCD [102].
- Switches and push-buttons.

The ATtiny85 [100] is a small micro-controller in the ATMEL AVR series of chips, and comes in an 8-pin DIP package for through-hole based electronics. The ATtiny85 is programmable with 8 KB of flash memory, and contains 6 input/output (I/O) pins which can be used to perform a number of functions. The ATtiny85 chips are used here to increase the efficiency of the rotary encoders, and reduce the number of digital I/O pins used on the Arduino MEGA 2560. Each rotary encoder has a dedicated ATtiny85 chip, all of which are daisy chained together using their input/output pins. The first and last ATtiny85 chips are wired to a pair of serial TX (transmit) and RX (receive) pins on the Arduino MEGA 2560. This configuration allows the Arduino MEGA 2560 to transmit and receive a rotary encoder event (such as an incrementing or a decrementing turn) along with its rotary encoder ID, quickly and efficiently. Each ATtiny85-enabled rotary encoder is assigned to an (e,2e) spectrometer element and its variable voltage supply.

The Adafruit 2.2” TFT LCD [102] has 320x240 color pixels and a TFT driver (ILI9340) which can display full 16-bit color. It uses the serial peripheral protocol (SPI) to communicate with the Arduino MEGA 2560 and has its own built-in pixel-addressable frame buffer. The Adafruit 2.2” TFT LCD is used to display the values of the set and measured voltages for a particular variable voltage supply unit. Three of these displays were used; one for each electron energy analyzer, and one for the electron gun.

The switches used are single-pole double-throw (SPDT) switches which alternate between two states; open or closed. The push-buttons are momentary which remain active as long as they are pressed. The functions these switches and push-buttons performed were replaced by the push-buttons on the rotary encoders.
The Arduino MEGA 2560 micro-controller and the components detailed above, were used to design this control board to perform a number of operations, which collectively control the variable voltage supplies’ computer and manual controls. Specifically, an Arduino-based control printed circuit board is designed to do the following:

- Send and receive digital data (in the form of serial commands) to and from the computer running LabVIEW.
- Send a clocked digital signal using the Serial Peripheral Interface (SPI) protocol to the variable voltage supply digital-to-analog converters (DACs).
- Receive digital signals from rotary encoders on the front panel for manual control of the voltage supplies.
- Receive digital signals from switches and push-buttons on the front panel that can be assigned to perform different operations (e.g. send an SPI command to all DACs to set voltages).
- Send the set (and measured) voltages to be displayed on 2.2’’ 18-bit color TFT LCDs on the front panel of the variable voltage supply unit.

The central component to this control board is the Arduino MEGA 2560 programmable micro-controller (as shown in fig. 5.19); it is mounted onto the back side of the board (the inner side facing the voltage supplies). The Arduino MEGA 2560 is connected to several components and pins via the PCB’s tracks, most of which are on the same side as the Arduino; those components are the ATtiny85 micro-controller chips, and various pins for connection with the TFT LCD, switches, and push-buttons. Rotary encoders for manual control of the voltage supplies, are soldered to the front (outer) side of the board.

The fundamental circuit design for the Arduino-based control board is shown in fig. 5.19. The dash-outlined boxed portion of the circuit shown in fig. 5.20 is cloned to meet the
voltage supply requirements, whether it is for control of an electron energy analyzer or the electron gun. The number of ATtiny85-based rotary encoders required for each electron energy analyzer and for the electron gun, are 7 and 13 respectively. Two push-buttons were added for saving and loading the set voltages onto the Arduino’s EEPROM (more details can be found in chapter 6). Two switches were initially incorporated into the design, for fine/coarse adjustments and to lock the rotary encoders, but these became redundant as their functions became accessible via the rotary encoder push-buttons. One additional switch is used for switching the Arduino’s auto-reset feature ON and OFF. The Arduino-based control printed circuit boards (PCBs) shown in fig. 5.21 were all made by hand and mounted on the front panels of the variable voltage supply units. The front panel gives access to the rotary encoders and the Adafruit 2.2” TFT LCD. The back side of this board faces the digital-to-analog and variable voltage supply electronics and contains the Arduino MEGA 2560, the ATtiny85 micro-controllers, and header pins used for breakout purposes, the Arduino reset switch, and the Adafruit 2.2” TFT LCD.
The Variable Voltage Supply Units

5.4.3 Digital-to-Analog Electronics

The voltage supply boards for the electron gun and electron energy analyzers are all computer controlled. The digital-to-analog electronics are fundamentally the same for all circuits that require computer control, and are broken down into the three stages shown in fig. 5.22.

![Block Diagram of Digital-to-Analog Electronics](image)

**Figure 5.22:** A block diagram of the digital-to-analog electronics.

The digital-to-analog electronics shown in fig. 5.23 are optically isolated from the digital side to allow the use of different 0 V voltage references. An opto-isolator acts as an isolation (protective) barrier between two separate circuits by means of a light sensitive optical interface. The device is comprised of a light-emitting diode (LED), a light-sensitive receiver such as a photo-transistor, and a transparent medium (e.g. glass, plastic, or air) between the first two components, resulting in electrical isolation of up to 10 kV. The optocoupler used to isolate the computer system from the voltage supplies is the HCPL-2630 [103] shown in fig. 5.24. The device is an 8-Pin DIP Dual-Channel High Speed 10 MB/s Logic Gate Output Optocoupler. This opto-isolator contains two input channels that are useful in this case because of the number of digital signals required to control the voltage supplies. The two opto-isolators shown in fig. 5.23 handle SPI signals from an Arduino MEGA 2560 micro-controller. The first opto-isolator uses a single input for a chip select
Figure 5.23: The digital-to-analog electronics converts digital signals to analog voltages for use with one of the variable voltage circuits detailed in section 5.4.4. The resistor and capacitor values are as follows: $R_1 = R_2 = R_3 = 820 \, \Omega$ (see text for details), $R_4 = R_5 = R_6 = R_7 = 1 \, k\Omega$, and $C_1 = C_2 = C_3 = C_4 = 1 \, \mu F$. See text for additional details on this circuit.

Figure 5.24: The HCPL-2630 pinout diagram. See text for details.

(CS) pin, while the other two opto-isolator inputs are used for the digital data (SDI) and clock (SCK) signals. It is important to note that the data (or clock) signals are common to all opto-isolators used in a master circuit containing one or more of the circuits shown in fig. 5.23. For this reason, the resistors $R_2$ (or $R_3$) are wired in parallel to other resistors that share a common data (or clock) digital signal. To meet the HCPL-2630 opto-isolator specifications, the input current $I_{\text{input}}$ for a high logic level ($V_{\text{high}}$) of 5 V must be between 6.3 mA and 15 mA. Hence, the resistor values for $R_2$ (or $R_3$) vary and are chosen based on how many there are in a master circuit. Eight and five opto-isolator inputs are used by the electron gun and each analyzer variable voltage supply respectively. For example, to
achieve an input current $I_{\text{input}}$ of around 8 mA (which is above the minimum threshold of 6.3 mA), the resistor value for $R_2$ (or $R_3$) is simply:

$$R_2 = R_3 = \frac{V_{\text{high}}}{I_{\text{input}}} = \frac{5 \text{ V}}{0.008 \text{ A}} = 625 \Omega \quad (5.3)$$

which can be achieved by using a 680 Ω resistor. The Arduino MEGA 2560 specifications require a 40 mA current draw from each pin, and a total of 200 mA from all pins. Therefore, the current drawn by an $n$ number of opto-isolators with 680 Ω resistors at their inputs is $\approx 59$ mA and 37 mA, for the electron gun and each analyzer respectively. To prevent the opto-isolated variable voltage supplies from exceeding the current draw limit, a 2N3704 transistor is used as a current source between them and the data (or clock) pin (as shown in fig. 5.20).

The output signals of the optocouplers are fed through a SN74F04N hex (6 channel) inverter (NOT gate), the outputs of which are the opposite digital logic-level to their inputs. Inverters are required due to the logic inversion (negation) caused by the optocouplers.

The final stage is a digital-to-analog converter (DAC) which takes the digital signal and converts it to an analog voltage, that can be used with the variable voltage stage of the circuit. The DAC used here was the MCP-4822 by Microchip [100]; a dual 12-Bit voltage output DAC. The 12-bit resolution defines the number of steps (increments) between the minimum and maximum DAC output voltage. The device has a built-in Serial Peripheral Interface (SPI) with 20 MHz clock support, rail-to-rail output, a 2.048 V high precision internal voltage reference, a single-supply operation of 2.7 V to 5.5 V, and extended temperature range of $-40^\circ C$ to $+125^\circ C$. The DAC can be configured to the full-scale range of the device (2.048 V or 4.096 V) by setting the GAIN selection option bit (1 or 2). Each DAC channel can also be operated in ACTIVE or SHUTDOWN mode.

The MCP4822 takes in a binary command as a string of bits (2 bytes of information per write, where 1 byte = 8 bits) from one end, and outputs a voltage on the other end of
the device. The MCP4822 shown in fig. 5.25 has 8 pins, with each having the following function:

1. **$V_{DD}$** (Supply Voltage Input of 2.7 V to 5.5 V): this pin is used to supply power to the DAC. It is recommended to use a bypass capacitor of 0.1 μF to 1 μF to ground.

2. **$\overline{CS}$** (Chip Select Input): this pin’s state (HIGH or LOW) determines if a command is being written or not. The bar (over-line) indicates that the CS pin is in active LOW by default; an active LOW is required to enable serial clock and data functions.

3. **$SCK$** (Serial Clock Input): this pin’s state determines the clocking speed of the written command.

4. **SDI** (Serial Data Input): a binary command is fed to this pin in a series of bits into the DAC’s input registers.

5. **$\overline{LDAC}$** (Latch DAC Input): the state of this pin transfers input latch registers to their corresponding DAC registers. When this pin is LOW, the $V_{OUT_A}$ and $V_{OUT_B}$ pins are updated simultaneously. This pin can be tied to LOW (with the $V_{SS}$ pin) if the $V_{out}$ update is desired at the rising edge of the $\overline{CS}$ pin.

6. **$V_{OUT_B}$**: an analog voltage output for DAC channel B.

7. **$V_{SS}$**: ground reference point for all circuitry on the device.

8. **$V_{OUT_A}$**: an analog voltage output for DAC channel A.

The output voltage for either channel (DAC A or DAC B) can be easily calculated using:

$$V_{out} = \frac{V_{ref} \cdot G}{2^n} \cdot \sum_{i=1}^{n} D_{n-i}$$

(5.4)

where $V_{ref} = 2.048$ V is the voltage reference, $n$ is the resolution, $D$ is the DAC input code (a digital value determined by the summation of the values of the $n$ bits that make up the binary command; $D$ ranges from 0 to $2^{n-1}$), and $G$ is the gain (1 or 2).

Each write command is made up of 16 bits: 4 configuration bits and 12 data bits. A write command is initiated once the $\overline{CS}$ is set to LOW, and stays LOW for the duration of the write command. The 4 configuration bits and the 12 data bits are clocked on the rising edge of the SCK pin and fed to the SDI pin. The $\overline{CS}$ pin is then raised to allow the data to be latched onto the selected DAC input registers. If the $\overline{LDAC}$ pin is LOW, the data moves from the DAC’s input registers to the output registers $V_{OUT}$ to update $V_{OUT_A}$ and $V_{OUT_B}$. These outputs are then used by the variable voltage electronics detailed in the next section.
5.4 The Variable Voltage Supply Units

5.4.4 Variable Voltage Electronics

This stage of the voltage supply circuit is where the digital-to-analog converter (DAC) output is buffered then amplified through one or more operational amplifiers. The type of operational amplifier used is dependent on the voltage desired at the output. For output voltages in the range of 0 V to 15 V, the TL084 \cite{99} was used. The OPA-445 \cite{99} operational amplifier is used for output up to 60 V. The LTC-6090 \cite{104} operational amplifier (surface mount) was used for supplies that required a 120 V output. For the elements requiring the high voltages of 500 V and 1000 V, Tracopower’s THV 12-500P and THV 12-1000P DC/DC converters \cite{105} were used, respectively. These miniature high voltage regulated power modules required smoothing capacitors on their outputs to dampen switching noise.

There is a total of 37 electrostatic lens elements making up the (e,2e) spectrometer, each of which requiring its own voltage: 19 voltages for the electron gun and Faraday cup, and 9 voltages for each electron energy analyzer. The voltages required to supply all elements of the (e,2e) spectrometer can be achieved with four fundamental circuits, each of which is designed to output the following variable voltage(s):

- $\pm 15$ V and $-15$ V: for the electron energy analyzer deflectors and the electron gun grid element.
- +30 V and +60 V: for the electron energy analyzer AM, OH, IH supplies, and the electron gun energy fine control.
- +120 V: for the electron energy analyzer RSE and L1B supplies, and the electron gun GL1C and Anode supplies.
- +500 V and +1000 V: for the electron gun coarse, and GL1B and GL2B supplies.

The $\pm 15$ V and $-15$ V Variable Voltage Circuity

This circuit relies on a +5 V voltage reference and eight operational amplifiers. This is achieved with an 8-Lead SOIC MCP1501 4.096 V high-precision voltage reference \cite{100}, and two TL084 JFET-Input Quad Operational Amplifiers (as seen in fig. 5.26) \cite{99}. The circuit has four outputs (two pairs of +/- polar opposite voltages), so is required by the electron gun and electron energy analyzer deflectors: one supply is required for the each electron energy analyzer and three are required for the electron gun. The potentiometers R14 and R21 in fig. 5.26 are trimmed so that the magnitudes of the DX+ and DY+ outputs are equal to those of the DX- and DY- outputs.

The circuit shown in fig. 5.27 is needed for the electron gun’s grid element which has to vary between 0 V and $-15$ V with respect to the filament potential. It requires only one
Figure 5.26: The deflectors’ ±15 V variable voltage supply circuitry. The resistor and capacitor values are as follows: \( R_1 = R_5 = R_6 = R_7 = R_8 = R_{11} = R_{18} = 1\,\text{k}\Omega, \quad R_2 = 22\,\text{k}\Omega, \quad R_3 = R_4 = R_{13} = R_{15} = R_{19} = R_{22} = 100\,\text{Ω}, \quad R_9 = R_{16} = 4.7\,\text{k}\Omega, \quad R_{10} = R_{17} = 15\,\text{k}\Omega, \quad R_{12} = R_{20} = 900\,\text{Ω}, \quad R_{14} = R_{21} = 100\,\text{Ω}, \quad C_1 = C_2 = 2.2\,\mu\text{F}, \) and \( C_3 = C_4 = C_5 = C_6 = 10\,\mu\text{F}. \)

Figure 5.27: The grid −15 V variable voltage supply circuitry. The resistor and capacitor values are as follows: \( R_1 = R_2 = 100\,\Omega, \quad R_3 = 4.7\,\text{k}\Omega, \quad R_4 = 47\,\text{k}\Omega, \quad R_5 = 15\,\text{k}\Omega, \) and \( C_1 = C_2 = 10\,\mu\text{F}. \)

TL084 JFET-Input quad operational amplifier. Since the grid requires a negative voltage, the 'positive' terminal of this circuit is connected to the common 0 V rail. Since the tuning conditions for the grid element varied between −4 V and 0.1 V, a ±12 V power supply was used.
The +30 V and +60 V Variable Voltage Circuitry

This circuit shown in fig. 5.28 relies on TL084 and OPA445 operational amplifiers. The first operational amplifier is responsible for buffering and inverting the DAC voltages. The two resulting outputs are used for fine and coarse adjustments with a 10:1 ratio across the two, thereby allowing 24-bit control of the output voltage. This is achieved by placing a resistor at each of the outputs with the value required to achieve this 10:1 ratio. These output voltages are then summed by connecting them to the same input of the OPA445 operational amplifier. Depending on the desired gain and the maximum voltage rail used, this circuit will supply +30 V or +60 V. This supply is used for the following:

- A1AM and A2AM: electron energy analyzer mean elements.
- A1IH and A2IH: electron energy analyzer inner hemispheres.
- A1OH and A2OH: electron energy analyzer outer hemispheres.
- GUNE Fine: electron gun energy fine control supply.

Figure 5.28: The +30 V or +60 V variable voltage supply circuitry for the electron gun fine control, and the electron energy analyzer mean, and inner and outer hemispheres. The resistor and capacitor values are as follows: R1 = R2 = 100 Ω, R3 = R4 = R6 = R7 = 1 kΩ, R5 = 4.7 kΩ, R8 = 47 kΩ, R9 = 33 kΩ for +30 V voltage gain at $V_{out}$ or R9 = 68 kΩ for +60 V voltage gain at $V_{out}$, C1 = C2 = 10 μF, C3 = 0.1 nF, and C4 = C5 = 10 μF.

The +120 V Variable Voltage Circuitry

The circuit shown in fig. 5.29 uses one TL084 and two LTC6090 high voltage operational amplifiers. The TL084 quad operational amplifier acts as a buffer for the two DAC output channels. Each inverted output addresses an LTC6090 operational amplifier with the appropriate gain to achieve the desired voltage of 0 V to +120 V. This circuit is used for the following elements:

- A1RSE and A2RSE: electron energy analyzer residual energies.
• A1L1B and A2L1B: electron energy analyzer lens element.
• Anode: electron gun’ anode.
• GL1C: electron gun lens element.

**Figure 5.29:** The 120 V variable voltage supply circuitry for the electron gun GL1C and Anode, and electron energy analyzer RSE and L1B elements. The resistor and capacitor values are as follows: $R_1 = R_2 = 100\,\Omega$, $R_3 = R_7 = 4.7\,k\Omega$, $R_4 = R_8 = 2.2\,k\Omega$, $R_5 = R_9 = 68\,k\Omega$, $R_6 = R_{10} = 1\,k\Omega$, $C_1 = C_2 = 10\,\mu F$, $C_3 = C_6 = 0.1\,nF$, and $C_4 = C_5 = C_7 = C_8 = 0.1\,\mu F$.

The +500 V and +1000 V Variable Voltage Circuitry

This supply uses one TL084 and a Tracopower high voltage power module (THV series). The TL084 operational amplifier is used to buffer the DAC voltage. One circuit uses a THV 12-500P module, while the other uses two THV 12-1000P modules. The circuit (as seen in fig. 5.30) that uses the THV 12-500P module is designed to produce a maximum output of 500 V, but upon testing the absolute maximum output voltage was found to be 415 V. The second circuit (as seen in fig. 5.31) buffers two DAC output channels to address two independent THV 12-1000P modules. These are designed to output a maximum of 1 kV but upon testing, the absolute maximum these modules could deliver varied between 831 V and 835 V. The circuit using the THV 12-500P module is used for the electron gun.
coarse energy supply, while the circuits using the THV 12-1000P modules are used for the electron gun lens elements GL1B and GL2B. Noise of approximately 1 V was measured on the output of both modules (THV 12-500P and THV 12-1000P). This was minimized to sub-mVs by using a bank of 'smoothing' capacitors across the outputs of the modules to reduce the noise.

**Figure 5.30:** The electron gun coarse control 500 V variable voltage supply circuitry. The resistor and capacitor values are as follows: \( R_1 = 100 \, \Omega \), \( R_3 = 1 \, k\Omega \), \( R_4 = 220 \, k\Omega \), \( C_1 = C_2 = 10 \, \mu F \), \( C_5 = 4700 \, pF \) 6 kV, and \( C_6 = C_7 = C_8 = 1 \, \mu F \) 400 V.

**Figure 5.31:** The GL1B and GL2B 1 kV variable voltage supply circuitry. The resistor and capacitor values are as follows: \( R_1 = R_2 = 100 \, \Omega \), \( R_3 = R_5 = 1 \, k\Omega \), \( R_4 = R_6 = 220 \, k\Omega \), \( C_1 = C_2 = C_4 = C_9 = 10 \, \mu F \), \( C_5 = C_10 = 4700 \, pF \) 6 kV, and \( C_6 = C_7 = C_8 = C_11 = C_12 = C_13 = 1 \, \mu F \) 400 V.
5.4.5 Faraday Cup Voltage Supply Circuitry

This circuit (fig. 5.32) relies on a REF02 voltage reference [99], two OPA277 operational amplifiers, a MOSFET transistor, and a 1N4007 rectifier diode. A dedicated power supply (as seen in fig. 5.7) delivers the required voltage rails of ±15 V and +380 V. The voltage is not computer-controlled and is adjusted manually. The Faraday cup voltage is monitored via $V_{\text{Mon}}$ using an external voltmeter.

![Faraday Cup Voltage Supply Circuit](image)

Figure 5.32: The Faraday cup voltage supply circuitry. The voltage is not computer-controlled and is adjusted manually. The Faraday cup voltage is monitored via $V_{\text{Mon}}$ using an external voltmeter. The resistor and capacitor values are as follows: $R_1 = R_2 = R_3 = R_4 = 12 \, \text{k} \Omega$, $R_5 = R_6 = R_7 = R_9 = 1 \, \text{M} \Omega$, $R_8 = 10 \, \text{k} \Omega$, $R_{10} = 56 \, \text{k} \Omega$, $R_{11} = R_{16} = 10 \, \text{k} \Omega$, $R_{12} = R_{13} = 820 \, \Omega$, $R_{14} = 910 \, \text{k} \Omega$, $R_{15} = 82 \, \text{k} \Omega$, $C_1 = C_2 = C_3 = C_6 = C_7 = C_8 = 1 \, \mu\text{F}$, and $C_4 = C_5 = 100 \, \text{nF}$ 400 V.

5.4.6 Current Monitor Circuitry

This circuit (fig. 5.33) uses an ISO124 precision isolation amplifier [99], and an OPA277 operational amplifier. The dedicated power supply (detailed in section 5.3.1 on page 100) delivers the two required independent ±15 V voltage rails. This circuit is fed the Faraday cup voltage $V_{\text{Fcup}}$ across an external resistor $R_{\text{Fcup}}$. The output is a voltage proportional to the Faraday cup current by a factor of 10. This voltage is sent to the PCI6221 DAQ card to be measured by the (e,2e) LabVIEW computer application [56].

5.4.7 The Variable Voltage Supply Output Interface Boards

The outputs of the voltage supplies are routed to a set of back panel interfacing printed circuit boards and are carried over via D25 cables to the (e,2e) spectrometer through several
5.4 The Variable Voltage Supply Units

The Faraday cup current monitor circuitry. This circuit is fed the Faraday cup voltage $V_{\text{FCup}}$ across an external resistor $R_{\text{FCup}}$. The output is a voltage proportional to the Faraday cup current by a factor of 10, which is sent to the PCI6221 DAQ card to be measured by the (e,2e) LabVIEW computer application. The resistor and capacitor values are as follows: $R_{\text{FCup}} = 1\, \text{M}\Omega$ and is external to this circuit, $R_1 = R_4 = 1\, \text{k}\Omega$, $R_2 = 4.75\, \text{k}\Omega$, $R_3 = 9.76\, \text{k}\Omega$, $C_1 = C_2 = C_3 = C_4 = C_7 = C_8 = 1\, \mu\text{F}$, $C_5 = 1\, \text{nF} \, 400\, \text{V}$, and $C_6 = 220\, \text{pF} \, 400\, \text{V}$.

Feedthroughs. To prevent internal rewiring to the elements of the (e,2e) spectrometer, the voltages were mapped according to the old pin assignments. Figures 5.34 to 5.37 detail the mapping of the output voltage pins for the electron energy analyzers, electron gun, and Faraday cup.

5.4.8 Physical Layout of the Variable Voltage Supply Units

The voltage supply electronics are consolidated into two units that fit inside a 19-inch rack cabinet. One unit houses the voltage supplies used for the electron gun, the Faraday cup voltage supplies, the current monitor board, and the Arduino-based control board. The other unit houses the voltage supplies needed for both energy analyzers, and the Arduino-based control board. Mounted on the back panels of both units are the DC-input (voltage rails) boards and the output voltage interface boards. Photographs of the voltage supply units are shown in figs. 5.38 to 5.40.
Figure 5.34: The electron gun, Faraday cup, and current monitor variable voltage supply back panel output interface boards. Refer to table A.9 in appendix A.5 for voltage pin assignments.

Figure 5.35: A drawing showing the layout of the electron gun, Faraday cup, and current monitor variable voltage supply back panel output interface boards. The faded boards are the input interface boards shown in fig. 5.15.
5.4 The Variable Voltage Supply Units

Electron Energy Analyzer 1
Board C
(Output to \((e,2e)\))

Electron Energy Analyzer 2
Board D
(Output to \((e,2e)\))

Figure 5.36: The electron energy analyzer variable voltage supply back panel output interface boards for analyzer 1 (left) and analyzer 2 (right). Refer to table A.10 in appendix A.6 for voltage pin assignments.

Electron Energy Analyzer 1
Board A
(DC Input)

Arduino 1 USB

Electron Energy Analyzer 2
Board B
(DC Input)

Arduino 1 Reset Switch

Figure 5.37: A drawing showing the layout of the electron energy analyzer variable voltage supply back panel output interface board. The faded boards are the input interface boards shown in fig. 5.17.
**Figure 5.38:** Photograph of the electron gun, Faraday cup, and current monitor variable voltage supply unit (a top-down view). The labeled boards: (A1) variable voltage supplies for GL1B, GL2B, gun energy coarse, gun energy fine, and grid, (A2) variable voltage supplies for GD1X/Y, GD2X/Y, GD3X/Y, GL1C, and Anode, (B1) to (B3) the back panel input interface boards A, B, and C, (C1) to (C3) the back panel output interface boards D, E, and F, (D) smoothing capacitor bank for high voltage supplies, and (E) the current monitor board.

**Figure 5.39:** Photograph of the electron energy analyzers variable voltage supply unit (a top-down view). The labeled items are: (A1) and (A2) variable voltage supplies for electron energy analyzer 1 and 2 respectively, (B1) and (B2) the back panel input interface boards A and B, and (C1) and (C2) ADC boards for analyzers 1 and 2 respectively, and (D) Arduino USB ports.
Figure 5.40: Photograph of the front panel of the electron gun (top) and electron energy analyzers' (bottom) variable voltage supply units. The labeled items are: (A) inline current monitor sockets for the electron gun GL1B, GL2B, GL1C, grid, and anode elements (clockwise starting with top left socket), (B) the Faraday cup manual adjustment potentiometer and voltage monitor socket, (C) rotary encoders for manual control of voltage supplies, (D) switches and push-buttons, and (E) an Adafruit 2.2” TFT LCD.
5.5 The Stepper Motor Control and Data Acquisition Unit

A further unit was built to house a new stepper motor control and interlocks system, and a LabVIEW data acquisition breakout board (CB-68LP) [56], a block diagram of which is shown in fig. 5.2. The stepper motor control and interlocks systems were modified mainly for compatibility with the new LabVIEW experimental control application. The unit houses an Arduino MEGA 2560 based control board, a stepper board for three L298N dual H-bridge driver modules, a stepper board power supply, and a PCI6221 LabVIEW DAQ breakout board (CB-68LP).

5.5.1 The Arduino-based Stepper Motor Control Board

The Arduino-based stepper motor control board shown in fig. 5.41 uses its digital I/O pins, USB serial, and the serial peripheral interface (SPI) protocol to perform a number of functions:

- The micro-controller (Arduino MEGA 2560) reads the position of the potentiometers that move to the angles of the electron energy analyzers and electron gun, and reports the angles to the Arduino control program.
- Receives angles from the LabVIEW-based computer and calculates appropriate number of steps from the difference between set and measured angles.
- Controls the switches of the H-Bridges inside the L298N motor drivers [106].
- To prevent the (e,2e) spectrometer’s elements from running into each other, it monitors the opto-isolators and stops the stepper motors if an interlock is triggered (the stepper motors can also be emergency stopped via the serial line, and maximum and minimum angles can be set in the firmware via LabVIEW commands if so desired). See section 4.3.3 for more details.

5.5.2 The Stepper Motors

The old stepper motors for both electron energy analyzers were replaced due to aging and their unstable (noisy) holding current. The electron gun does not use a stepper motor for two reasons: the electron gun is usually manually moved and locked in place for days or weeks at a time, and controlling the electron gun angle would require the holding current to be applied continuously.

The new stepper motor (as seen in fig. 5.42) is the RS PRO Unipolar Stepper Motor [107]. It has a step angle accuracy of 1.8°, a holding torque of 0.42 Nm, a voltage rating of 5.7 V, and a current rating of 1 A. It is a 6-wire stepper used in a 4-wire mode with H-bridge drivers located on the L298N-based stepper board.
5.5 The Stepper Motor Control and Data Acquisition Unit

5.5.3 The L298N-based Stepper Board and Stepper Motors

The slave L298N-based stepper board receives digital signals from the Arduino-based stepper motor control board, which in turn controls the rotation direction of the stepper motors. The stepper board consists of three L298N dual H-Bridge motor drivers (as seen in fig. 5.43), one for each electron energy analyzer; the third driver is currently unused.

The L298N drivers have digital inputs, and since the 6-wire stepper motors are used in a 4-wire mode (full step drive with four steps per cycle as shown in fig. 5.44), only four of the digital inputs of the L298N breakout board are used. The remaining two digital
Figure 5.42: Photograph of the new RS PRO stepper motor; 1.8°, 0.42 Nm, 5.7 Vdc, 1 A, 6 Wires.

Figure 5.43: Photograph of the L298N dual H-Bridge motor driver.

(ENABLE) inputs are left unused to allow the stepper motor to move at maximum speed. Each L298N module consists of two H-Bridges, and each stepper motor consists of two coils (one H-Bridge per stepper motor coil). An H-Bridge is essentially composed of four transistors arranged in the shape of the letter H. Turning on the different transistors
(switches), controls the direction of the current flow through a stepper motor coil, thus controlling the rotation direction of the stepper motors. The direction of the current flow through a stepper motor coil is best understood in the four steps it takes to complete one cycle (as seen in table 5.1). The numbers 1 and 0 can be seen as HIGHs and LOWs, respectively. A 1 or a HIGH drives the current one way through a coil, while a 0 or a LOW drives the current in the opposite direction; a sequence that causes the magnetic field to flip. The speed is determined by the rate, which is controlled by how fast the driver cycles through the step pattern. Once a move has been completed (an electron energy analyzer has been positioned), the H-Bridges are switched off to stop the current flow through the coils and prevent the coils and the L298N driver boards from overheating. The connections from the three potentiometers for the electron gun and analyzers were completely rewired.

![Phases of a 4-wire stepper motor](image)

**Figure 5.44:** Phases of a 4-wire stepper motor. The stepper motor is composed of two coils A and B and a central permanent magnetic stepper rotor. The L298N H-Bridge drivers turn each of the coils into a temporary electromagnet by pulsing current through each of those coils. Each step corresponds to a phase, and all four phases correspond to one complete cycle. Each electromagnetic coil attracts the central stepper rotor towards it causing the rotor to move (step through) in a clockwise (or counterclockwise) fashion.

<table>
<thead>
<tr>
<th>Phase</th>
<th>Step Pattern</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>1 0 0 1</td>
</tr>
<tr>
<td>2</td>
<td>1 1 0 0</td>
</tr>
<tr>
<td>3</td>
<td>0 1 1 0</td>
</tr>
<tr>
<td>4</td>
<td>0 0 1 1</td>
</tr>
</tbody>
</table>

**Table 5.1:** A table showing the direction of the current flow through a stepper motor coil. It is best understood in the four steps it takes to complete one cycle. The numbers 1 and 0 can be seen as HIGHs and LOWs, respectively. A 1 or a HIGH drives the current one way through a coil, while a 0 or a LOW drives the current in the opposite direction; a sequence that causes the magnetic field to flip.
5.5.4 The Stepper Board Power Supply

A ±12 V and ±5 V power supply was designed to provide voltage rails for the Arduino-based stepper motor control and the L298N-based stepper board. The power supply shown in fig. 5.41 uses the VTX-120-4206-215, a 6 VA PCB-mount isolation transformer with 230 VAC primary and dual 15 VAC secondary. The power supply design was based on the circuit shown fig. 5.4.

5.5.5 The PCI6221 LabVIEW DAQ and CB-68LP Breakout Board

The PCI6221 data acquisition (DAQ) card by National Instruments [56] is a multifunction I/O device (as seen in fig. 5.45). It offers analog I/O, digital I/O, two 32-bit counters/timers, and digital triggering through its CB-68LP breakout board. The PCI6221 LabVIEW DAQ card is attached to a PCI slot (a 32-bit computer bus), that can be accessed and addressed from within the LabVIEW environment. The PCI6221 device is used to measure the following:

- Time-to-amplitude (TAC) pulse via an onboard high speed analog-to-digital converter (ADC).
- Time-to-amplitude (TAC) valid start.
- Analyzer 1 rate-meter counts.
- Analyzer 2 rate-meter counts.
- Faraday cup current.
- Vacuum system pressure.

5.5.6 The Stepper Motor Back Panel Output Board

The stepper motor back panel output board shown in fig. 5.41 is used to connect the interlocks system inside the vacuum chamber and the stepper motor potentiometers to the Arduino-based stepper motor control board. The L298N driver outputs are carried to the stepper motors through a D15 adaptor that is mounted to the back panel of the stepper motor control and data acquisition unit.
5.6 Filament Constant Current Supply and Current Boost Circuitry

The filament constant current supply is a 3 A constant current supply as seen fig. 5.46. This is responsible for feeding current to the cathode in the electron gun. More details can be found in the (e,2e) Coincidence Spectrometer operation and reference manual [4].

5.7 The (e,2e) Coincidence Detection Electronics

5.7.1 The Channel Electron Multiplier Pulse Electronics

Single electron detection begins with Mullard X919BL channel electron multipliers (CEM), with one channel electron multiplier per energy analyzer. Each channel electron multiplier used a dedicated Brandenberg EHT 5kV high voltage supply (as seen in fig. 5.47). The channel electron multipliers collect and amplify single (energy selected) electrons from the energy analyzer exit apertures, so as to produce a current pulse at their output that is
correlated to the single electron that is detected. These channel electron multipliers use an activated lead glass which has a high coefficient of secondary electron emission when a high voltage (around 2000V–3000V) is applied across the multiplier. For a single electron that enters the input cone, the channel electron multiplier can yield around $1 \times 10^8$ electrons in the output current pulse. More details on the channel electron multiplier and their EHT high voltage supplies can be found in section 4.3.2 and the (e,2e) manual [4].

![Figure 5.47: Photograph of Brandenberg EHT 5kV high voltage supplies for electron energy analyzer 1 (top) and 2 (bottom).]

5.7.2 The External Pulse Detection Electronics

The external pulse detection electronics consist of several modules (as shown in fig. 5.48 and fig. 5.49), that are responsible for signal filtering, coincidence detection, and counting and monitoring of the pulses.

As explained in section 4.3.2, the 25 to 40 mV negative going pulses from the channel electron multipliers require amplification prior to transmission to the constant fraction discriminators (CFDs). Two Phillips Scientific 6954 100X amplifiers (one for each energy analyzer) are used to amplify the channel electron multiplier pulses. The slow NIM outputs from the two ORTEC 473A discriminators pass to two ORTEC 441 ratemeters for pulse counting and monitoring. The fast NIM pulses from the two discriminators address the START and STOP inputs of an ORTEC 567 time-to-amplitude converter (TAC) (as shown in fig. 5.48); one of the signals is first passed through a delay generator before it reaches
the **STOP** input of the TAC. This delay allows the coincidence signal to fall within the time interval set by the time-to-amplitude converter.

The time-to-amplitude converter (TAC) measures the time difference between the pulses that arrive at its **START** and **STOP** inputs, converts this time interval into a proportional analog voltage amplitude, and then sends it out as an **OUTPUT** pulse. The full width of this time interval is set using the front panel TAC 567 range and multiplier selectors. The range selector can be set to 50, 100, or 200 ns, which can be multiplied by a factor of 1, 10, 100, 1 k, or 10 k using the multiplier selector. The experiments detailed in this thesis use a 200 ns full width window in addition to the \( \sim 100 \text{ ns} \) delay by the pulse that arrives at the **STOP** input of the TAC.

The CB-68LP connector block and the PCI6221 DAQ shown in fig. 5.45 are used for measurement and detection:

- The ratemeters feed the two PCI6221 32-bit counters.
- The TAC **START** pulse is fed to a PCI6221 digital pin (PF14: pin 41). This is configured as a clock pulse trigger.
- The TAC **OUTPUT** pulse is fed to a PCI6221 analog pin of the high speed analog-to-digital converter (ADC) input (A10: pin 68).

**Figure 5.48**: Block diagram of the coincidence timing pulse detection electronics.
The TAC output pulse is sampled over 1000 channels for a full width window of 200 ns and a bin width of \( \sim 0.2 \text{ ns} \). A channel is incremented if the measured time difference between the detected electrons falls within the selected full width window of 200 ns. The electrons that are detected in coincidence with each other, will increment channels that correspond to the delay between the \textit{START} and \textit{STOP} pulses of the TAC. These counts are accumulated over a period of time, the result of which is an \((e,2e)\) coincidence spectrum. These spectral scans are carried out over a range of energies and kinematics using the new \((e,2e)\) LabVIEW computer application, and analyzed using the new \((e,2e)\) data analysis Python library, both of which are detailed in chapter 6.

The pulse electronic modules (constant fraction discriminators, ratemeters, delay generator, and time-to-amplitude converter) are located in a single NIM crate in the same 19-inch rack as the experimental hardware units.

5.8 Conclusion

In this chapter, the modernized experimental hardware was explained in detail. The power supply units for the electron gun and both electron energy analyzers, and the Arduino control, digital-to-analog, and variable voltage electronics that make up the variable voltage supply units were all described. The power supply and variable voltage supply back panel
interface boards and their pin assignment tables were covered. The stepper motor and data acquisition unit was described as well as the pulse detection electronics used in timing coincidence signals. The process by which printed circuit electronic boards are made was also covered. The next chapter will cover the new experimental software used to control, perform, and analyze data generated by (e,2e) coincidence experiments.
CHAPTER 6

Experimental Software

6.1 Introduction

The (e,2e) coincidence experiment required the design of new computer applications and programs for experimental control, variable voltage supply control, communication, and data exchange, extraction, analysis, and visualization. LabVIEW by National Instruments [56], was used to develop a user-friendly (e,2e) computer application for communicating with and controlling the variable voltage supply and stepper motor control units, and measuring, collecting and formatting data. Arduino MEGA 2560 micro-controllers [57] were used for direct control of the variable voltage supply and stepper motor control units, and as a medium between the (e,2e) LabVIEW computer application and the (e,2e) spectrometer. The Python programming language [58] and the Jupyter notebook environment by Project Jupyter [62], were used to extract, analyze, and visualize the data collected and formatted by the (e,2e) LabVIEW computer application. The development of the computer applications and programs were the result of a collaborative effort with Dr. Matthew A. Harvey, who joined University of Manchester’s AMO physics group in January of 2017.

6.2 The (e,2e) LabVIEW Computer Application

The (e,2e) computer application shown in fig. 6.1 was designed and built using LabVIEW (Laboratory Virtual Instrument Engineering Workbench) by National Instruments. LabVIEW is a development environment that uses a visual programming language to create user-friendly computer applications that are primarily used for hardware control, measurement, and data analysis. The (e,2e) computer application itself communicates with the four Arduino MEGA 2560 micro-controllers mentioned in chapter 5, and the National Instruments PCI-6221 DAQ card via the CB-68LP breakout board located inside the stepper motor control and data acquisition unit (detailed in section 5.5).
6.2.1 The LabVIEW-Arduino Communication

The communication between the (e,2e) LabVIEW computer application and the four Arduino MEGA 2560 micro-controllers, is handled via LabVIEW modules that perform the following functions:

- Analyzer 1 control board: to set and measure the electrostatic element voltages for analyzer 1.
- Analyzer 2 control board: to set and measure the electrostatic element voltages for analyzer 2.
- Electron gun control board: to set the voltages for the electron gun’s electrostatic elements. A measurement analog-to-digital (ADC) board can be added to the electron gun’s voltage supply unit to report voltage measurements back to the (e,2e) LabVIEW computer application.
- Stepper motors’ control board: to monitor the position of the electron gun and analyzers 1 and 2, and position analyzers 1 and 2 at angles instructed by the (e,2e) LabVIEW computer application.

The Arduino MEGA 2560 micro-controllers are programmed with a robust serial communications protocol which performs and sends a checksum calculation at the end of each
message. To prevent the system from using messages containing errors or incomplete commands, the receiving end performs a recalculation and a comparison with that checksum. In the event of an error, the receiver requests the message to be sent again. This feature is a preventive method that ensures that incorrect voltages and angles can not be set. More details on the code behind the Arduino MEGA 2560 micro-controllers are found in section 6.3.

6.2.2 The LabVIEW-PCI6221 Communication

The PCI-6221 data acquisition (DAQ) card and the CB-68LP breakout board are used to send the following signals to the (e,2e) LabVIEW computer application:

- Analyzer 1 rates: these rates are fed to a counter/timer pin from the ratemeter output of analyzer 1.
- Analyzer 2 rates: these rates are fed to a counter/timer pin from the ratemeter output of analyzer 2.
- Time-to-amplitude (TAC) valid start: this is for triggering the reading of the TAC’s voltage amplitude signal.
- Time-to-amplitude (TAC) output: this is for measuring the pulse amplitude which gives the time that the TAC was stopped by an event, which is then summed to a histogram in the (e,2e) LabVIEW computer application for building an (e,2e) coincidence signal.
- Vacuum chamber pressure: this is an analog voltage from the pressure gauge, proportional to the pressure (calculated in LabVIEW) inside the vacuum chamber.
- Faraday cup current: this is an analog voltage from the current monitor board’s isolated operational amplifier that measures the voltage across a 1 MΩ resistor (as seen in section 5.4.6).

6.2.3 The (e,2e) LabVIEW Application Structure

The (e,2e) LabVIEW computer application (like any program designed in LabVIEW) consists of portions of code termed virtual instruments (VIs) which could contain other virtual instruments called subVIs. Virtual instruments are essentially routines which can be used to perform a number of functions. Each virtual instrument is made up of three components; a block diagram, a front panel, and a connector pane, as seen in fig. 6.2, fig. 6.3, and fig. 6.4, respectively.

- The block diagram is essentially the back panel (hidden from the user) of the LabVIEW program which contains the graphical source code.
A

B

C

Figure 6.2: An example of a block diagram for a subVI called SVIGenerateEnergyArray in the (e,2e) LabVIEW computer application. This subVI is used to set the parameters for a binding energy scan. It is fed the electron gun energy’s start, stop, and increment size, as well as the desired time for each step. The subVI outputs an array of the linearly spaced energies as well as the total time required to perform the binding energy scan. The labeled items are: (A) controls, (B) indicators, and (C) which is a for loop similar in function to those used in any modern computer programming language.

Figure 6.3: An example of a front panel in LabVIEW for the subVI SVIGenerateEnergyArray shown in fig. 6.2. This front panel takes in inputs from the controls labeled as (A), passes them to the block diagram shown in the previous figure, and outputs the results in the indicators labeled as (B).

- The front panel is what the user interacts with, and is composed of controls (inputs) and indicators (outputs) that appear as modules with terminals in the back panel.
- The connector pane is a visual representation of the virtual instrument (VI or subVI) and its terminals which correspond to the VI’s controls and indicators (inputs and outputs). The connector pane assigns the number of inputs and outputs that can
6.2 The (e,2e) LabVIEW Computer Application

Figure 6.4: A layout of all possible connector panes used in LabVIEW. The connector pane assigns the number of inputs and outputs that can be wired to a virtual instrument (VI or subVI). The connector pane uses the input terminals to receive data, then passes the data to the block diagram which then sends the results back to the connector pane’s output terminals. Image is courtesy of National Instruments [56].

be wired to a virtual instrument (VI or subVI). The connector pane uses the input terminals to receive data, then passes the data to the block diagram which then sends the results back to the connector pane’s output terminals.

The (e,2e) LabVIEW computer application consists of approximately 100+ VIs and subVIs. The application itself consists of the five parallel loops described below.

1. Front panel event structure loop: this loop is always paused and only responds to user inputs via the front panel’s controls. An example is the START button used to set up a scan based on the front panel control inputs found in the various tabs (such as energy scan, angular scan, settings, etc). See fig. 6.5.

2. Analyzer count rates loop: this loop runs every 100 ms to provide the user with the rates for analyzers 1 and 2 through two types of indicators: numeric and chart. The chart provides the user with up to two minutes worth of rate history for both analyzers; helpful for tuning the electron gun and analyzers before setting up the scan parameters. See fig. 6.6.

3. Voltage supply monitor loop: this loop runs four times per second to poll the voltage supply Arduino MEGA 2560 micro-controllers. This loop requests the latest set and measured voltages from the three Arduinos controlling the electron gun, analyzer
1, and analyzer 2, voltage supplies. This keeps the (e,2e) LabVIEW computer application up to date, if any of the voltages were changed using the voltage supply front panel rotary encoders. The only measured voltages reported are for analyzers 1 and 2 (the electron gun is not currently equipped with an analog-to-digital converter measurement board). In addition, the loop also updates the front panel with the status of whether the Nelder-Mead [108] optimization routines are running on either analyzer. The Nelder-Mead method is also known as the downhill simplex method for finding the minimum or maximum of a function in a multidimensional space. This method is used to optimize the number of counts for either analyzer by tuning three parameters: the analyzer lens element L1B and the analyzer deflectors Dx and
Dy. The search ends when the three parameters converge to the optimal number of analyzer counts. See fig. 6.7.

Figure 6.7: The LabVIEW block diagram of the voltage supply monitor loop.

4. Analyzer position loop: this loop runs ten times per second to request the stepper motor controller Arduino for position (angle) updates of the electron gun and both analyzers, as well as the status of the interlocks’ opto-isolator interrupters. The loop also displays a 3D model of the (e,2e) spectrometer showing the positions of the electron gun and analyzers. In addition, the loop estimates and updates the front panel with the projected time remaining on a scan. See fig. 6.8.

Figure 6.8: The LabVIEW block diagram of the analyzer position loop.

5. Main state handler loop: this loop runs twenty times per second, and contains a
state machine that controls the experiment. The multi-channel analyzer histogram is continuously updated whenever the (e,2e) spectrometer is in an idle state (no scan is being performed). If a scan is being carried out, the typical pattern would be:

- Position the analyzers.
- Setup the optimization routine by getting initial Nelder-Mead vertex information.
- Step through the Nelder-Mead routine until an end condition is met.
- Repeat optimization if it is set to run more than once.
- Log the vacuum chamber pressure and Faraday cup current.
- Reset the TAC’s MCA histogram.
- Keep updating the MCA until the desired step time has elapsed.
- Save the data to the save file.
- Repeat all of the above until all the steps have been completed or the user decides to hit the stop button.

The experiment can be stopped at any time. If this is carried out the remaining states are deleted, and the loop returns to an idle state where the multi-channel analyzer histogram is continuously updating. See fig. 6.9.

![LabVIEW block diagram of the main state handler loop.](image)
6.3 The Programmable Arduino Micro-controllers

Arduino [57], conceived at the Ivrea Interaction Design Institute, is a completely open-source electronics platform based on a variety of inexpensive, easy-to-use, and extensible hardware and software for prototyping and building electronics projects. The many advantages that the Arduino platform offers, made its programmable micro-controller boards the most sensible choice for developing the new computer-controlled electronics for the (e,2e) coincidence experiment. These advantages include:

- Affordability: Arduino boards are extremely inexpensive compared to other micro-controller boards. The Arduino integrated development environment (IDE) software is free.
- Cross-platform: the Arduino software (IDE) runs on Windows, MacOS, and Linux.
- Simplicity, flexibility: the Arduino board and its companion software (IDE) are easy-to-use, and flexible enough for complicated electronics projects such as the computer-controlled voltage supplies.
- Extensibility: the software and hardware can be extended using C++ libraries and modular circuit boards called shields, respectively.
- Open-source: both hardware and software are open-source.

6.3.1 The Arduino Hardware and Software

An Arduino also refers to two components: a physical programmable circuit board (also known as a micro-controller) and an integrated development environment (IDE) software [57]. As detailed in chapter 5 the particular Arduino board used to control the variable voltage supply and stepper motor units is the Arduino MEGA 2560 (as seen in fig. 5.18) [57].

The Arduino software is an integrated development environment (IDE) (seen in fig. 6.10) which allows the user to write programs using Arduino’s programming language and C/C++, and upload them to compatible boards. Arduino’s programming language consists of its built-in functions, values composed of variables and constants, and the structure which contains the raw C/C++ portion of the code [57].

6.3.2 The Arduino Program Structure

The structure of every Arduino program can be broken down into four parts; library imports, value declarations, the setup() function, and the loop() function (seen in fig. 6.10):
Figure 6.10: The Arduino integrated development environment (IDE) software.

- Library imports: this section of the program is used to import external C/C++ libraries, which are essentially collections of functions that can be called from the main program.
- Value declarations: this section of the program is used to declare all variables and constants used in the main program.
- The setup() function: this function is called once when the Arduino program runs for the first time (after an Arduino is powered up or following an Arduino board reset). This function is used to initialize variables, call functions, set pin modes, make use of libraries, and more.
- The loop() function: this function loops consecutively, allowing the Arduino program to respond and change. Inputs passed as arguments to functions are one example of how an Arduino program can respond and change. This continuous loop function is what allows the user to control the Arduino board.

6.3.3 The (e,2e) Variable Voltage Supply Arduino Code

This master program was written and uploaded to the three Arduinos that control the electron gun and energy analyzer voltage supply units. The program is assigned a value
The Programmable Arduino Micro-controllers

6.3 The Programmable Arduino Micro-controllers

called mainID (1, 2, or 3 for analyzer 1, analyzer 2, and electron gun, respectively) before it is uploaded to the Arduino; an ID which identifies the voltage supply unit to be controlled and which of the portions of the code to use and ignore. The program performs four primary functions: send and receive serial messages to and from the (e,2e) LabVIEW computer application, responds to rotary encoder events and send Serial Peripheral Interface (SPI) signals to the voltage supply digital-to-analog converters (DACs), read the measured analog-to-digital voltages (for analyzers 1 or 2 only), and write to and update a 2.2" TFT LCD screen. The (e,2e) voltage supply Arduino program structure is detailed below:

1. The program uses the following external libraries, some of which have been written and developed particularly for this program, while others were external and Arduino IDE built-in libraries:

   - **HardwareSerial**: this library is used to access the three serial ports on the Arduino MEGA 2560 (Serial1, Serial2, and Serial3).
   - **Adafruit GFX**: this is Adafruit’s core graphics library for all of their displays. The library is used in conjunction with another hardware-specific library for Adafruit’s 2.2" TFT LCD screen (Adafruit ILI9340 library).
   - **Adafruit ILI9340**: this library is specific to the Adafruit 2.2" TFT LCD screen; it is designed to handle the lower level functions.
   - **SPI**: this library allows the Arduino (as the master device) to communicate with (slave) peripheral devices such as the digital-to-analog converters (DACs) using the serial peripheral interface (SPI) protocol.
   - **EEPROM**: this library is for accessing, reading, writing, and updating the Arduino’s EEPROM (electrically erasable programmable read-only memory). The Arduino’s EEPROM, is used to store the last known voltage supply settings in the event of a power failure, an Arduino reset, or a new code upload.
   - **MillisTimer**: this library sets up a timer which allows the caller to check how much time has elapsed since it was last polled and report whether a minimum interval has been passed.
   - **ADCHandler**: this library is used to handle the communication between the ADC boards and the Arduino. It is primarily used to continuously read the measured voltages for the analyzers’ electrostatic element.
   - **calcChecksum**: this simple library calculates a checksum based on a message sent over serial between the LabVIEW and an Arduino board.
2. Several variables and constants are declared to help perform a number of functions:

- **TFT screen settings**: pin assignments, the position coordinates and text for the electrostatic element titles, their set and measured voltages, and any information that may appear on the screen whilst controlling the voltage supply unit.

- **SPI settings**: digital pin assignments, mapping of the DACs addressed by the Arduino, the number of electrostatic elements used, and other values and constant used in SPI communication.

- **Rotary encoder settings**: contains a mapping of the rotary encoders, rotary encoder rates for fine and coarse adjustments, and rotary encoder push button settings.

- **Calibration values**: contains the minimum and maximum voltages (and DAC voltages) for all electrostatic elements. These values are used to calibrate the set and DAC voltages with each other. These values were obtained by directly measuring the absolute minimum and maximum output voltages for each DAC and operational amplifier responsible for supplying an actual voltage to an \((e, 2e)\) spectrometer element.

- **Switch and button settings**: pin assignments, initial states, and other values used with EEPROM related functions.

- **ADC settings**: ADC voltage measurement mappings, and ADCHandler and MilliTimer settings.

3. The setup() function runs once and initializes the following:

- **Serial Peripheral Interface (SPI)**: initializes the SPI bus with pre-defined settings.

- **Voltage supply initialization**: the mainID assigned in the program, initializes the appropriate digital pins, calibration values, TFT position coordinates, rotary encoder mappings, and SPI settings.

- **TFT initialization**: initializes the TFT display with the electrostatic element names, and their initial set voltages. The live ADC voltage measurements are also initialized.

- **Voltage supply ID**: a function called supplyID() uses the assigned mainID to send the voltage supply ID over the serial line to the \((e, 2e)\) LabVIEW computer application.
4. The loop() function calls the following functions (many of which contain nested functions):

- checkSerial(): this function is designed to read messages over serial (from LabVIEW) one character at a time, starting with a starting character ($). It reads the message until it reaches the termination character (&). If the message is 3 or more characters long (including the starting and termination characters), then it calls the calcChecksum() function to calculate the checksum using the message and the message length. The checksum calculation is performed to handle errors so as to ensure that only sensible messages are passed and used. If the checksum calculation is successful, the parseMessage() is then called to analyze and identify the command within the message. The (e,2e) voltage supply Arduino program uses the following commands:
  - $ID&: this command is used to call the supplyID() function which prints and sends the voltage supply unit ID (A1, A2, or EG) to LabVIEW over serial.
  - $RATE&: this command calls the encodersRateFunction() function which toggles encodersRate between true and false. When encodersRate is true the step size for each rotary encoder turn changes according to the turn rate. This is useful for changing between fine and coarse adjustments. If encodersRate is false, the step size is fine and remains unchanged.
  - $LOCK&: this command calls the encodersLockFunction() function which toggles encodersLock between true and false. This is used to lock and unlock the front panel rotary encoders to prevent them from being inadvertently changed during a scan.
  - $ADC&: this command calls the adcDisplayOnTftFunction() function which toggles adcDisplayOnTftState between true and false. When adcDisplayOnTftState is true, the ADC measurement is engaged and the values are updated on the TFT screen.
  - $VMDA&: this command calls the voltageMesuredVSDacAdcFunction() function which toggles vmVSdaState between true and false. When vmVSdaState is false the DAC and ADC values are shown on the TFT screen, and when vmVSdaState is true, the set and measured voltages are displayed.
  - $RBLV&: this command calls the reportBackToLVFunction() function which toggles reportBackToLVstate between true and false. When report-
BackToLVtate is **true**, serial communication to LabVIEW is allowed, and not allowed when reportBackToLVtate is **false**.

- **$G\{\text{Value Type}\}\{\text{Element Name}\} &:** this command is used for getting (reporting) the current (last known) voltages back to LabVIEW. The $G$ in the command is for GET, the **Value Type** is one of four characters (D, V, A, or M, for DAC, Set Voltage, ADC, or Measured Voltage, respectively), and the **Element Name** is two characters long representing the electrostatic element. To request **All Elements** from LabVIEW, then **AE** is used for the **Element Name**.

- **$S\{\text{Value Type}\}\{\text{Element Name}\}\{\text{Value}\} &:** this command is for setting a voltage for an electrostatic element. The $S$ in the command is for SET, the **Value Type** is one of two characters (D or V, for DAC or Set Voltage, respectively), the **Element Name** is two characters long representing the electrostatic element (e.g. IH for Inner Hemisphere, AN for Anode, etc), and the **Value** is a number used to set the voltage for that electrostatic element.

If the checksum calculation was a mismatch, or if a message was the incorrect length, a **NAK** (Negative Acknowledge) is sent over serial to LabVIEW, and the checkSerial() function returns back to the beginning of the loop awaiting the next serial message.

- **rotaryEncoders():** this function polls the ATtiny85-based rotary encoders (discussed in section 5.4.2) for events. A rotary encoder event can be triggered via a turn or a push button. An event reports back the rotary encoder’s address and the result for that particular event. The address is used to map the rotary encoder to an electrostatic element. The result returns an action such as incrementing or decrementing a value (a voltage), or performing a specific operation in the event that a rotary encoder’s push button is pressed, held, or double-clicked. A few of the rotary encoder push buttons are programmed to do the following in the event of a button press:
  - Lock the rotary encoders by calling the encodersLockFunction() function.
  - Change the rotary encoder rate by calling the encodersRateFunction() function.
  - Engage or disengage the ADC board measurement updates on the TFT screen by calling the adcDisplayOnTftFunction() function.
– Toggle the type of values (set and measured voltages versus DAC and ADC
values) displayed on the TFT screen by calling the voltageMesuredVSDA-
cAdcFunction() function.

– Toggle serial communication back to LabVIEW ON and OFF by calling
the reportBackToLVFunction() function.

If the event is a rotary encoder turn, the electrostatic element is located using
the address, the rotation direction (clockwise or anti-clockwise) increments or
decrements the last known value for that electrostatic element, then the value
is used to set the electrostatic element’s DAC voltage using the Arduino’s SPI
protocol.

• updateScreenVoltages(): this is the primary function for continuously updating
the TFT screen with the last known settings for all electrostatic elements. It
cycles through all elements and updates a value in the event of a value change.
Nested within the updateScreenVoltages() function are other TFT-related func-
tions which are responsible for updating the TFT screen appropriately.

• ADCBoardA.update(), ADCBoardB.update(), and other ADC-related functions:
these functions are responsible for updating the last known voltage measurements
from the ADC boards, and displaying them on the TFT screen.

The entire (e,2e) voltage supply Arduino code and its libraries are available as a Github
repository [109].

6.3.4 The (e,2e) Stepper Motor Arduino Code

This program was written and uploaded to one Arduino MEGA 2560 micro-controller, for
controlling the stepper motors, measuring the stepper motor potentiometers for determining
the position of the electron gun and analyzers, and for reading the status of the interlocks’
opto-isolator interrupters. The (e,2e) stepper motor Arduino program structure is detailed
below:

1. The program uses the following external libraries, some of which have been written
and developed particularly for this program, while others were external and Arduino
IDE built-in libraries:

• Stepper: this library was written specifically for the controlling (e,2e) stepper mo-
tors and measuring their potentiometers, and preventing the (e,2e) spectrometer
elements from running into each other.
• MicroTimer: this library sets up a timer which allows the caller to check how much time has elapsed since it was last polled and report whether a minimum interval has been passed. This allows events to be setup to occur at minimum set intervals within a block of code.

• MCP3208: this library is used in conjunction with Microchip’s SPI-enabled MCP3208 8-Channel 12-Bit analog-to-digital converter. The MCP3208 is used to read the stepper motor potentiometers, and report them back as digital signals to the Arduino using the SPI protocol.

2. Several variables and constants are declared to help perform a number of functions:

• Communication settings: contains variables and constants used in serial communication acknowledgement, parsing serial messages, and calculating checksums.

• MCP3208 and stepper motors’ settings: for the initialization of the steppers for the electron gun and analyzers, and the ADC for measuring the stepper motor potentiometers.

• Serial communication values: variables and constants used in serial communication.

3. The setup() function runs once and initializes the following:

• Serial: initializes serial data transmission by setting the data rate in bits per second.

• ADC and stepper motors’ initialization: using the MCP3208 and stepper libraries, the ADC and stepper motor functions are initialized.

• Opto-isolator flags’ initialization: using Arduino’s built-in pinMode() function, several digital pins are configured to behave as INPUTS to read the status of the opto-isolator flags. See table 6.1 for the opto-isolator digital pin assignments.

4. The loop() function calls the following functions (many of which contain nested functions):

• checkSerial(): this is identical to the checkSerial() function used in the (e,2e) voltage supply Arduino program, with the exception being how the messages are parsed. Similar commands such as S and G are used for setting and getting the position of the analyzers and the electron gun.

• optoFlags() and stopMotor(): these functions are used to halt the stepper motors if the opto-isolator flags are engaged.
run(): this function is called to engage a stepper motor and move a spectrometer element to a new position.

<table>
<thead>
<tr>
<th>Digital Pin</th>
<th>Opto-isolator Flag</th>
</tr>
</thead>
<tbody>
<tr>
<td>D19</td>
<td>A2 Min</td>
</tr>
<tr>
<td>D18</td>
<td>A2 MaxOP</td>
</tr>
<tr>
<td>D15</td>
<td>A2 Max</td>
</tr>
<tr>
<td>D14</td>
<td>A2: A12 Right</td>
</tr>
<tr>
<td>D9</td>
<td>Gun in plane</td>
</tr>
<tr>
<td>D7</td>
<td>A2: A12 Left</td>
</tr>
<tr>
<td>D6</td>
<td>A1 MaxOP</td>
</tr>
<tr>
<td>D3</td>
<td>A1 Max</td>
</tr>
<tr>
<td>D2</td>
<td>A1 Min</td>
</tr>
</tbody>
</table>

Table 6.1: The opto-isolator flag Arduino digital pin assignments. The first column shows the numbers for each of the digital pins on the Arduino MEGA 2560 board used to engage the opto-isolator flags (shown in the second column).

6.4 Data Analysis with Python

The final step in an (e,2e) coincidence experiment is the analysis of the data collected by the (e,2e) LabVIEW computer application. The data analysis portion of the experiment was designed to handle datasets of any of the possible scan types, quickly and efficiently. The data were handled using several methods in the following order of steps.

1. LabVIEW: this portion is where the data is collected and prepared to be extracted, analyzed, and visualized in Python.
   - Data collection: handled by the (e,2e) LabVIEW computer application.
   - Data formatting and exchange: the data collected by the (e,2e) LabVIEW computer application is saved to a file using the JSON format (for storing, exchanging, and transporting data) [95]. The JSON format is one of the most sensible formats for the (e,2e) coincidence experiment, because it is human readable, and its data objects are organized in attribute-value pairs and array data types. The JSON format structure makes data extraction easier and quicker in Python.

2. Python: the Python programming language [58] and external libraries were used to extract, analyze, and visualize the JSON-formatted datasets. The Jupyter Notebook environment [62] was used to write and run the Python program(s) that perform the following:
• Data extraction: this is handled by several functions, which can be called independently or from within other functions. As standalone functions, they can extract information such as a scan’s summary (which contains a scan’s settings), dataframes (for each step through a scan) containing the voltage supply settings for all electrostatic elements, and dataframes containing the analyzers’ count rates.

• Data analysis: this portion analyzes the extracted datasets by performing a number of operations and calculations to generate the relative differential cross-sections for a particular scan.

• Data visualization: the extracted and/or analyzed datasets are passed through 'visualization’ functions. Datasets containing information such as cross-sections, or voltage supply settings per step or increment, are visualized in the form of plots and tables.

6.4.1 Python and Jupyter Notebooks

Python is an interpreted, object-oriented, high-level, general-purpose programming language, with a simple and easy to learn syntax, making the programming language a powerful tool for scripting and developing small or large-scale programs [110]. Python 3, the most recent version, was used to develop the (e,2e) data analysis library, inside the Jupyter Notebook environment. A Jupyter notebook is essentially a web-based interactive document composed of input and output cells, each of which can contain code (such as Python), Markdown-based text, rich media, or mathematics (see fig. 6.11 for an example Jupyter notebook) [96].

6.4.2 The (e,2e) Data Analysis Python Library

1. The (e,2e) data analysis library was built using the following core libraries and modules:

   • pandas: this is a powerful Python package for handling data using flexible data structures to easily manipulate and analyze data.

   • json: a Python built-in package for encoding and decoding JSON-formatted data. This package is used to extract summaries and datasets generated by the (e,2e) LabVIEW computer application.

   • csv: a Python built-in module for reading and writing csv (comma separated values) files. This module is used to write and save output datasets (such as cross-sections) to a csv-formatted file.
Jupyter Notebook: An Example

This is a brief demonstration on how a Jupyter notebook is used.

Python Examples

```python
In [7]:
getaAngles = True
analyzer1angle = 20
analyzer2angle = 45
if getaAngles:
    print('Analyzer 1 is at ' + str(analyzer1angle) + ' degrees and analyzer 2 is at ' + str(analyzer2angle) + ' degrees')

Analyzer 1 is at 20 degrees and analyzer 2 is at 45 degrees

In [8]:
for i in range(3):
    print(2**i - 1)

0
1
3

Mathematics Example

In [9]:
x = 5
y = 10
print('x + y = ' + str(x+y), ' and x * y = ' + str(x*y))
x + y = 15 and x * y = 50

Plotting Example

```python
In [5]:
import matplotlib.pyplot as plt
import numpy as np
fig = plt.figure()
analyzer1angle = 20
analyzer2angle = 45
x = np.linspace(0, 10, 1000)
x.plot(x, np.sin(x));
```

Figure 6.11: An example of a Jupyter notebook.

- **numpy**: this is a widely used library for computing scientific and mathematical data in Python.

- **matplotlib**: a python library for producing publication quality plots and figures in a variety of formats. This library is used to generate plots showing cross-sections, voltage settings over time, etc.

Additional libraries were used to access files and streams, and interface with the underlying operating system Python is running.
2. An extensive library of functions were written to perform data extraction, data analysis, and data visualization. The (e,2e) data analysis library is imported as a module into an empty Jupyter notebook. Once imported, the notebook’s cells are used to call individual functions from the (e,2e) data analysis library. Below is a list of functions and what they return:

- **GunVoltages()**: this function finds the most recent or current scan’s JSON file, and extracts the voltage supply settings for the electron gun’s electrostatic elements for each angular or energy step. The function can return the voltage supply settings for a single or all electrostatic element(s).

- **A1Voltages(), A2Volatges() and analysersMeasuredVoltages()**: these functions behave in the same way as the **GunVoltages()** function, but with the voltage supply settings for analyzers 1 and 2. The functions can return the settings for a single or all electrostatic element(s), and since the analyzer voltage supply units are equipped with ADC boards, the function can return set or measured values as well.

- **FindFileNameOfCurrentRun()**: this function finds the filename for the most recent or current scan.

- **checkIfItemsInListAreIdentical()**: this function compares two lists containing data such as analyzer angles, and compares them to see if they are identical or not. This is useful for determining which of the analyzers are fixed and/or mobile in angular-based scans.

- **CheckIfItemsAreEqual()**: this function iterates through a list and checks if items are equal.

- **ExtractSummaryFromJSON()**: this function opens a scan’s output JSON-formatted file, and extracts the scan’s summary. The extracted summary details contain the following information:
  - **StartDate**: a scan’s start date
  - **NumberOfSteps**: the number of energy or angular steps in a scan
  - **StepTime**: the time per energy or angular step (in seconds)
  - **Species**: the name of the target molecule
  - **Energies**: a list containing the scan’s electron gun energies
  - **Angles**: lists containing the scan’s angles for analyzers 1 and 2
– **ExperimentType**: the type of scan performed (e.g. energy scan, symmetric angular scan, etc.)

– **CountType**: the type of counts for the experiment (rates or coincidence).

– **TAC WindowStart (ns)**: used as a marker position to identify the start of a peak.

– **TAC WindowStop (ns)**: used as a marker position to identify the end of a peak.

– **TAC NumberOfBins**: the total number of bins used in a spectrum.

- **plotTitle()**: when called, this function generates and returns the correct title based on extracted information.

- **UsableSetToJSON()**: this function opens a JSON-formatted scan file, and writes a new object, labeling the file as usable. This is useful for identifying successful scans for future reference.

- **WriteToSummaryOfJSON()**: this is a general function that can add a custom attribute-value pair to the JSON-formatted file’s summary.

- **SlackUpdate()**: this function is used to send a PNG image to a Slack channel. Slack is a team collaboration software with a chat module [97]. During an experiment, certain functions can be called to generate the latest cross-sections’ plot, which in turn can be sent (as an update on the experiment) to mobile phones running the Slack application.

- **DetermineFixedAnalyserOrIndices()**: this function returns which analyzer is fixed in an asymmetric angular scan. It also creates lists of indices for identifying the start and end of a sweep in an angular-based scan.

- **GenerateCrossSections()**: this functions returns a dataframe containing the cross-sections for a scan.

- **BuildOverview()**: this functions builds an overview table displaying information on all the scans performed for a particular experiment. The overview will contain information such the filename of the scan’s results, the initial settings, and the type of scan.

- **GeneratePlots()**: this function generates a plot for one or all usable scans.

- **GenerateMasterDataSet()**: besides the cross-sections, the dataframe returned can include additional data such as voltage supply settings per step, Faraday cup current, and vacuum chamber pressure.
• GenerateSetForNormalisation(): this function returns a normalized dataset from the dataset returned by the GenerateMasterDataSet() function.

• GeneratePlotFromMasterDataSet(): this function generates a plot from the dataset returned by the GenerateSetForNormalisation() function.

• FindPeakFromEnergyScan(): this function returns the peak energy for an energy scan.

• SweepIdentifier(): this function returns the total number of sweeps and filename for a scan.

• GenerateSweepsFromRawCounts(): this function returns a dataset containing the raw analyzer counts.

• InitialSettings(): this function returns the initial voltage supply settings by calling the GunVoltages(), A1Voltages(), and A2Voltages() functions.

The (e,2e) data analysis Python library was designed and written specifically for the modernized (e,2e) coincidence experiment. However, since the library is broken down into functions, they can be easily modified to accommodate any changes to the JSON data file generated by LabVIEW, or to include new methods for analysis, normalization, or visualization. The (e,2e) data analysis Python library is available for download as a Github repository (see [111]).

6.5 Conclusion

This chapter covered the new experimental applications, routines, and scripts used to control, perform, and analyze data generated by (e,2e) coincidence experiments. The new (e,2e) LabVIEW computer application used for controlling the variable voltage supply units, and the stepper motor and data acquisition unit, was described in detail. The programs behind the Arduino micro-controllers and their functions were covered. The new (e,2e) Python data analysis library for data extraction, analysis, and visualization was described. The next chapter will cover the experimental procedures used to operate the (e,2e) spectrometer and experimental hardware, as well as the procedural steps for performing spectral scans using the (e,2e) LabVIEW computer application for, and the data analysis methods using the new (e,2e) Python data analysis library.
Experimental Procedures

7.1 Introduction

This chapter covers the experimental procedures for preparing the vacuum system, operating and optimizing the (e,2e) spectrometer, performing (e,2e) coincidence and rates' scans, and analyzing the data generated by the (e,2e) coincidence experiment. More details on the operation and maintenance of the (e,2e) apparatus can be found in the (e,2e) coincidence spectrometer operation and reference manual [4].

7.2 Preparation of the (e,2e) Spectrometer and Vacuum System

The preparation of the (e,2e) spectrometer and vacuum system for an (e,2e) spectral scan involves the pumping system (covered in chapter 4), the experimental hardware (covered in chapter 5), and a few components of the (e,2e) LabVIEW computer application (covered in chapter 6). The procedural steps explained below assume that: the (e,2e) spectrometer has been lowered into the vacuum chamber, the (e,2e) vacuum chamber is at atmospheric pressure, and all experimental hardware and electronics are powered off. The following steps for preparing the (e,2e) spectrometer and vacuum system are detailed below:

1. Pumping down: this step assumes that the (e,2e) spectrometer has been lowered into the vacuum chamber, that the top flange has been securely aligned with the top rim of the vacuum chamber, that the target gas needle valve is shut off, and all experimental hardware is powered off. Pumping down is done by switching on the roughing pump and then the Balzers TPU-510 500 l/s turbo-molecular pump mounted from the underside of the vacuum chamber. This is done by plugging in the D-type adaptor (seen in fig. 7.1), which is configured to activate the pump when plugged in. The turbo-molecular pump is left overnight to evacuate the vacuum chamber to a base pressure of $\approx 1 \times 10^{-7}$ torr. The vacuum pressure can be monitored externally via the ionization gauge controller’s LCD, and/or from the (e,2e) LabVIEW computer application. If left for longer periods, the pumping system can achieve a background working pressure of $\approx 1 \times 10^{-8}$ torr.
2. Experimental hardware: at this stage, the experimental hardware can be switched on in the following order:

a) Detection electronics: this includes the Phillips Scientific 100x preamplifiers, the two ORTEC 473A constant fraction discriminators, the two ORTEC 441 ratemeters, an ORTEC 416A gate & delay generator, and the ORTEC 567 time-to-amplitude converter (TAC). All detection electronic modules are located in and powered by one NIM (Nuclear Instrumentation Module) crate.

b) Channel electron multiplier EHT supplies: the dials for setting the EHT supplies must be set to a minimum (0 kV) before switching on. The supplies are switched on and slowly adjusted to ≈ 2.3 kV - 2.8 kV, depending on the age of the channel electron multipliers.

c) Power and variable voltage supply units: these supplies are for the electron gun, electron energy analyzers, and Faraday cup. The two power supply units (which deliver the voltage rails necessary to power up the variable voltage supply units) are then switched on using a dedicated switch located on the back panel. The two respective variable voltage supply units are live once the power supply units are switched on. The electron gun, electron energy analyzers, and Faraday cup, are tuned to the initial settings seen in table 7.1 and table 7.2.

d) Filament constant current supply: the supply must be at 0 A before switching
on. The filament supply is switched on and slowly brought up, until the desired current is reached.

<table>
<thead>
<tr>
<th>Gun Energy</th>
<th>Grid</th>
<th>Anode</th>
<th>GL1B</th>
<th>GL1C</th>
<th>GL2B</th>
<th>F Cup</th>
</tr>
</thead>
<tbody>
<tr>
<td>70.0</td>
<td>-1.0</td>
<td>70.0</td>
<td>588.0</td>
<td>70.0</td>
<td>230.0</td>
<td>300</td>
</tr>
<tr>
<td>GD3Y</td>
<td>GD3Y</td>
<td>GD3Y</td>
<td>GD3Y</td>
<td>GD3Y</td>
<td>GD3Y</td>
<td>GD3Y</td>
</tr>
<tr>
<td>0.0</td>
<td>0.0</td>
<td>0.0</td>
<td>0.0</td>
<td>0.0</td>
<td>0.0</td>
<td>0.0</td>
</tr>
</tbody>
</table>

Table 7.1: The initial settings for the electron gun electrostatic elements and the Faraday cup. The deflectors are initially left at 0.0V to be adjusted during the tuning of the (e,2e) spectrometer. Elements such as the Grid, Anode, GL2B, are also used in tuning of the (e,2e) electron gun. The desired Gun Energy changes according to the type of experiment being performed. The electrostatic elements GL1B and GL1C are rarely adjusted and are left unchanged. All settings are in Volts (V).

<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>10.0</td>
<td>47.8</td>
<td>0.0</td>
<td>0.0</td>
<td>15.0</td>
<td>31.0</td>
<td>7.6</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>A2 Residual Energy</th>
<th>A2L1B</th>
<th>A2DX</th>
<th>A2DY</th>
<th>A2AM</th>
<th>A2IH</th>
<th>A2OH</th>
</tr>
</thead>
<tbody>
<tr>
<td>10.0</td>
<td>50.0</td>
<td>0.0</td>
<td>0.0</td>
<td>15.0</td>
<td>29.1</td>
<td>7.6</td>
</tr>
</tbody>
</table>

Table 7.2: The initial settings for both electron energy analyzers. The deflectors are initially left at 0.0V to be adjusted during the tuning of the (e,2e) spectrometer. The lens elements A1L1B and A2L1B are used in tuning of the (e,2e) electron energy analyzers. The desired Residual Energy for both analyzers change according to the type of experiment being performed. The electrostatic elements AM, IH, and OH for both electron energy analyzers are only adjusted when the resolution of the analyzers is to be changed. All settings are in Volts (V).

3. Target gas and line evacuation: introducing or changing a target gas into the vacuum chamber involve the following series of steps:

   a) Close and ensure that the needle valve entering the vacuum chamber through the top flange is shut off.

   b) Attach a gas regulator to the desired gas bottle (e.g. Helium, Nitrogen, etc) and keep the gas bottle switched off.

   c) Open all valves between the gas bottle and the purging pump. In other words, open the gas regulator valve so that the line right up to the gas bottle’s own valve can be evacuated. This prevents air contaminating the line between gas bottle and gas regulator from when it was attached. Ensure the that gas bottle is still switched off.

   d) Shut off the valve that vents the purging pump to atmospheric pressure.

   e) Switch on the purging pump.

   f) Evacuate the line for 30 seconds.
g) Shut off the valve to the purging pump.

h) Open the gas bottle.

i) Shut off the gas bottle.

j) Evacuate line for 30 seconds.

k) Repeat from steps (g) through (j) two to three times.

l) Shut off the valve to the purging pump.

m) Turn off the roughing pump, open the valve that vents the purging pump to atmospheric pressure, and let air in to prevent oil from the purging pump bleeding back in to the gas line over time (due to vacuum pressure).

n) Open the gas bottle.

o) Open the needle valve until the vacuum chamber reaches the desired pressure of \( \approx 1 \times 10^{-5} \) torr. At this point, the \((e,2e)\) spectrometer is ready for tuning. The needle valve must be opened slowly while monitoring the vacuum pressure via the ionization gauge controller’s LCD or from the \((e,2e)\) LabVIEW computer application, since the pressure in the chamber will only change slowly with time as the gas is fed into the system.

4. **Tuning of the \((e,2e)\) spectrometer:** at this stage the electron gun and electron energy analyzers are tuned in preparation for an \((e,2e)\) scan.

   a) **Tuning of the electron gun:** the electron gun energy is set according to the desired experiment. The electron gun is initially tuned by maximizing the current through the Faraday cup. This is done by adjusting The GRID, ANODE, and GL2B, and working through the deflectors GD1X, GD1Y, GD2X, GD2Y, GD3X, and GD3Y, until the current is maximized.

   b) **Tuning of the electron energy analyzers:** the analyzer residual energies A1RSE and A2RSE are set according to the desired experiment. The electron energy analyzers are tuned by maximizing the count rates (seen on the ratemeters and from the \((e,2e)\) LabVIEW computer application) by adjusting the elements A1L1B and A2L1B, and deflectors A1DX, A1DY, A2DX, and A2DY.

Once the vacuum chamber has been evacuated, the experimental hardware has been switched on, the target gas line has been evacuated, the target gas has been introduced into the system, and the \((e,2e)\) spectrometer has been tuned, the \((e,2e)\) apparatus is ready for performing a scan.
7.3 Performing (e,2e) Scans

An (e,2e) scan can be performed once the (e,2e) apparatus has been prepared (as covered in section 7.2). The (e,2e) LabVIEW computer application (the inner workings of which are detailed in section 6.2) is accessed via its front panel (shown in fig. 7.2) which contains a tab selector window surrounded by a set of indicators that display information regardless of which tab the user is on. The LabVIEW indicators display the angular positions for the electron gun and electron energy analyzers, the status of the interlocks’ opto-isolator flags, the Faraday cup current, the vacuum pressure, and scan information and status (number of steps, the scan time in HH:MM, and scan completion date and time). The tab selector window consists of the following tabs, each of which is capable of performing one or more function(s):

- TAC and Counters: this tab provides real-time feedback on the (e,2e) coincidence signal, and the electron energy analyzer count rates (shown in fig. 7.2).
- Electron Gun: this tab shows the current set voltages for all electron gun electrostatic elements. It also allows the user to set the voltages for one or more element(s).
- Analyzers: this tab shows the current set and ADC measured voltages for all electron energy analyzer electrostatic elements. It also allows the user to set the voltages for

![Figure 7.2: The (e,2e) LabVIEW computer application TAC and counters front panel tab. The tabs allow the user to tune and optimize the (e,2e) spectrometer, set the electrostatic element voltages, position the electron energy analyzers, and perform several types of scans.](image_url)
one or more element(s).

- **Angle Control**: this tab shows a 3D model of the current positions of the electron gun and electron energy analyzers. It allows the user to set the angular positions for one or both electron energy analyzer(s).

- **Scan tabs**: the following tabs are used for performing (e,2e) scans.

  - **Energy Scan**: this tab allows the user to perform an energy scan. The expected parameters are the start energy (eV), the stop energy (eV), and the increment or step size (eV). For feedback, the tab creates and displays a list of the energies based on the entered experimental parameters. In addition, a real-time plot of the energy scan is displayed and updated per completed energy step.

  - **Analyzer Scan**: this tab is unfinished and could be implemented in the future. It will allow the user to scan the analyzer energies while holding the electron gun energy fixed. This is similar but opposite to an energy scan.

  - **Tracking Scan**: this tab is unfinished and could be implemented in the future. It will allow the user to simultaneously scan both the electron gun and electron energy analyzers.

  - **Angular Scan**: this tab allows the user to perform various types of angular scans. The user selects the desired count type (rates or coincidence). If the desired count type is **Coincidence**, then the user must select the **Angular scan type** as symmetric or asymmetric; symmetric angular scans step through the same angles for both electron energy analyzers, and asymmetric angular scans are for scans where one of the electron energy analyzers is fixed. The **Analyzer Angles Generation** will create an **Analyzer Angles List**: a sweep of angles generated by using the **Start Angle** (°), the **Stop Angle** (°), and the **Increment** (°) parameters. Appending these parameters more than once will create additional sweeps. The **Mirror Angle Method** is used for scans that perform more than one sweep; the mirroring method determines if an appended list of angles are in an ascending or descending order. The **Analyzer Optimization** gives the option for the electron energy analyzer elements L1B and deflectors DX and DY, to be optimized per angular step using the Nelder-Mead optimization [108] routine (the parameters of which are included in the NM Optimization tab). Lastly, a real-time plot of the differential cross-sections is generated per completed angular step of the scan.

  - **Arbitrary Scan**: this tab is unfinished and could be implemented in the future.
It will allow the user to perform any type of scan using a constructed 2D array where each step contains parameters for analyzer 1, analyzer 2, the electron gun, and analyzer angular positions.

- **Settings**: this tab is used to output and save the data collected during an (e,2e) scan to a JSON-formatted file. The parameters it takes, are the save file path, the name of the species (target gas), and an optional textual description of the (e,2e) scan. The data in the output file is extracted, analyzed, and visualized using the (e,2e) data analysis Python library detailed in section 6.4.

- **Summary**: this tab displays "summarized" plots of the vacuum pressure and the Faraday cup current over the duration of an (e,2e) scan.

- **NM Optimization**: this tab is used to enter the parameters and settings for the Nelder-Mead optimization routine; used to optimize the electron energy analyzer during angular scans.

- **PS Optimization**: this tab is unfinished and could be implemented in the future. It will allow the user to perform angular scans using a Particle Swarm optimization routine on the electron energy analyzer lens and deflector elements.

To perform an (e,2e) scan, the following tabs are used: The **Electron Gun Analyzers** tabs are optional but can be used to set the electrostatic element voltages. The **Angle Control** tab is used to position the electron energy analyzers; optional for angular scans and mandatory for energy scans or scans that do not require the electron energy analyzers to change position during a scan. One of the scan tabs and its parameters (mentioned above) is required to perform an (e,2e) scan. The **Settings** tab is required to output and save the data collected during an (e,2e) scan into a JSON-formatted file. Finally the **NM Optimization** tab is used to optimize the electron energy analyzers during angular scans.

To check the progress of an (e,2e) scan, the **TAC and Counters** tab can be used to monitor the coincidence signal and analyzer count rates in real-time, the **Summary** tab for monitoring the Faraday cup current and vacuum pressure over the duration of an (e,2e) scan, and the indicators surrounding the tab selector window (as mentioned in the beginning of this section) for other information pertaining to the experiment being performed.

### 7.4 Data Analysis Procedures using a Jupyter Notebook

The output JSON-formatted file generated by a complete (e,2e) scan is used along with the (e,2e) data analysis Python library. To better understand this section, refer to
section 6.4 to become familiar with the types of functions required to extract, analyze, and visualize the data generated and collected during an \((e,2e)\) scan.

### 7.4.1 Creating and Running an Empty Jupyter Notebook

To analyze an \((e,2e)\) scan output file using the \((e,2e)\) data analysis Python library, an empty Jupyter notebook running the Python 3 kernel is created and opened in a web browser (shown in fig. 7.3).

![Figure 7.3: An empty Jupyter notebook running the Python 3 kernel in a browser (e.g. Safari, Google Chrome, etc).](image)

### 7.4.2 Importing the \((e,2e)\) Data Analysis Python Library

The empty cells are used to execute code such as calling functions from the \((e,2e)\) data analysis Python library. The first cell must be used to import the \((e,2e)\) data analysis Python library (shown in fig. 7.4).

![Figure 7.4: A Jupyter notebook cell demonstrating how to import the \((e,2e)\) data analysis Python library.](image)
7.4.3 The BuildOverview Function

A new Jupyter notebook cell is used to call the `BuildOverview()` function to display a summary table of all (e,2e) scan (JSON) files found in the current working directory (shown in fig. 7.5). The summary table contains information such as the start date, experiment type, count type, species, step time, number of steps, etc. This is useful because particular items of information displayed in the summary table (e.g. filename and experiment type), can be used and passed as arguments into other functions. In other words, knowing the correct filename can be passed into a function to locate the desired file, extract and analyze the data, and generate a plot.

![Summary Table](image)

**Figure 7.5:** A Jupyter notebook cell demonstrating how to call the `BuildOverview()` function and output an up-to-date summary of all (e,2e) scan (JSON) files found in the current working directory.

7.4.4 The GeneratePlots Function

Another Jupyter notebook cell is used to call the `GeneratePlots()` function which opens an (e,2e) scan (JSON) file and extracts the data. The function accepts three arguments:

- the JSON filename (without the file extension) surrounded by single or double quotation marks.
- `SaveToCSV` boolean (True or False) which saves the cross-sections plot to the current working directory.
- `SendToSlack` boolean (True or False) which sends the generated cross-sections plot as a PNG image file via a Slack channel using the Slack messaging service to a mobile phone or device running the Slack application.
If the Count Type is Coincidence then the cross-sections are calculated for each spectrum (for an angular or energy step) such as the one shown in fig. 7.6, by performing the following:

- Locating the Peak Window containing the coincidence signal using the Peak Window Start and Peak Window Stop from the extracted Summary section of the JSON-formatted output file.
- Locating the background for each spectrum; the two portions of the spectrum outside of the Peak Window.
- Calculating the background mean using the two location portions of the spectrum.
- Subtracting the background mean from every bin containing coincidence counts; the bins in between the Peak Window Start and Peak Window Stop.
- Summing all resultant coincidence counts.
- Repeating the steps above for next the spectrum (angular or energy step) and appending the resultant coincidence count sums as cross-sections to a Python dataframe (a table).

The resulting cross-sections table is used to generate a plot with the x-axis for steps in electron gun energy or electron energy analyzer angles, and the y-axis for (e,2e) coincidence cross-sections. The GeneratePlots() function (shown in fig. 7.7) is intelligent enough to detect and keep track of multiple angular sweeps for angular scans; each additional sweep is appended as a new column to the cross-sections’ table, and displayed using a new color in the generated cross-sections plot.
7.4 Data Analysis Procedures using a Jupyter Notebook

7.4.5 The InitialSettings Function

The `InitialSettings()` function shown in ?? accepts one argument (a filename) and extracts the initial voltage supply settings for the electron gun and the electron energy analyzers. These settings represent the (e,2e) spectrometer’s initial tuning conditions before the start of an (e,2e) scan. These settings can be used to repeat an (e,2e) scan or as a reference.

7.4.6 The GenerateCrossSections Function

The `GenerateCrossSections()` function shown in ?? accepts two arguments: the filename and the `SaveToCSV` boolean (True or False) which saves the cross-sections plot to the current working directory. As a standalone function it outputs a cross-sections dataframe as a table to a Jupyter notebook output cell. As a nested function it passes the dataframe to be used for analysis and visualization, such as the case inside the `GeneratePlots()` function.
7.4.7 Additional Data Analysis Python Functions

A number of functions (mentioned in section 6.4 on page 155) can be used (and modified) to perform further analysis, such as the `GeneratePlotFromMasterDataSet()` and `GenerateSetForNormalisation()` functions. As an example, the `GeneratePlotFromMasterDataSet()` can locate all successful and completed (e,2e) coincidence scans with common parameters (such as experiment type and fixed analyzer angle), extract all sweeps, perform cross-section calculations, and visualize all individual sweeps into one plot (as seen in fig. 7.10). The `GenerateSetForNormalisation()` function takes this a step further, and finds the mean, standard error, and standard deviation for all sweeps, and outputs the result as a plot and a table as shown in fig. 7.11. Since normalization of the data is unique for each (e,2e) coincidence experiment, the resultant output table of the `GenerateSetForNormalisation()` function is saved as a CSV file, and imported into an Excel spreadsheet for normalization.

![Figure 7.10: A Jupyter notebook cell used to call the GeneratePlotFromMasterDataSet() function to output a plot of all successful sweeps for scans with common parameters such as the experiment type.](image)
7.5 Conclusion

The sections in this chapter are intended to give the reader a set of instructions for preparing the vacuum system and (e,2e) spectrometer, performing (e,2e) scans using the (e,2e) LabVIEW computer application, and extracting, analyzing, and visualizing experimental data using the (e,2e) data analysis Python library. More details on the operation and maintenance of the (e,2e) apparatus can be found in the (e,2e) coincidence spectrometer operation and reference manual [4]. The (e,2e) data analysis Python library can be downloaded as a Github repository [111].

This chapter is the last in a series of chapters on the (e,2e) apparatus. The next two chapters are a direct application of the modernized (e,2e) apparatus, which focus on the experimental studies carried out on $N_2$ and $CH_4$, along with their theoretical frameworks and published results.
CHAPTER 8

Experimental (e,2e) Studies on Molecular Nitrogen (N$_2$)

8.1 Introduction

This chapter gives an introduction to the nitrogen molecule, and then covers the (e,2e) coincidence studies on molecular nitrogen that resulted in two publications in the Journal of Physics B: Atomic, Molecular and Optical Physics, which can be found in [59] and [60]. The work discussed in those two publications are covered in section 8.3 and section 8.4.

Section 8.2 is a detailed introduction to molecular nitrogen; nitrogen’s electronic structure, molecular interactions between its atomic orbitals, bonding types, symmetries, states of N$_2$ and its ions, and potential energy curves.

Section 8.3 covers the work published in [59], which was the first (e,2e) coincidence experiment on the N$_2$ molecule during the course of this Ph.D. This experiment was performed using the setup of the old (e,2e) apparatus, and in parallel with the development of the modified and modernized experimental hardware and software. The experiment was performed with incident electron energies ~10 and ~20 eV above the ionization potential of the 3$\sigma_g$, 1$\pi_u$ and 2$\sigma_u$ states for N$_2$, using both equal and non-equal outgoing electron energies and a range of geometries from the coplanar to the perpendicular plane. The publication of this work was carried out in collaboration with Don Madison’s group from Missouri University of Science & Technology (USA). The work resulted in experimental and theoretical ionization triple differential cross-sections.

Section 8.4 covers the results published in [60], which was the second (e,2e) coincidence experiment performed on the N$_2$ molecule. The old (e,2e) apparatus setup was used again, since the modified and modernized experimental hardware and software were still in development. The experiment was performed with the incident electron beam in the scattering plane where $\psi = 0^\circ$, at symmetric energies ~20 and ~40 eV above the ionization potential for the 3$\sigma_g$ and 1$\pi_u$ states for N$_2$, and for a number of fixed angles for one of the outgoing electrons. This experiment used the same theoretical models from Don Madison’s group as was used in the experiment detailed in section 8.3. This experiment was an extension of the first, to further test the theoretical models and see if their predictions
improved at higher energies.

8.2 Molecular Nitrogen \( \text{N}_2 \)

Molecular nitrogen \( \text{N}_2 \), which makes up a large portion of the Earth’s atmosphere, is a homo-nuclear centro-symmetric diatomic molecule with molecular weight of 28.0134 g/mol. It is a colorless and odorless gas, it exhibits diamagnetism, and is known for its extremely strong triple bond [112]. Its triple bond comes from its 10 valence electrons (5 from each nitrogen atom), with 8 occupying the three \((2\sigma_g^2, 1\pi_u^1, 3\sigma_g^3)\) bonding orbitals, and 2 occupying the \(2\sigma_u^2\) anti-bonding orbital. When ionized from the ground state, the molecular nitrogen cation exhibits states of different symmetries and parities, where each state corresponds to the molecular orbital in which the ionization had occurred. A binding energy spectrum can help identify the ionized states and their respective ionization energies. The ground state of \( \text{N}_2 \) and the ionized states of \( \text{N}_2^+ \), can be represented by their respective potential energy curves and their vibrational and rotational energy levels. The probability of transitions and transition energies from the ground state of \( \text{N}_2 \) to the ion states of \( \text{N}_2^+ \) can be calculated using the eigenfunctions and eigen-energies of the vibrational levels in question.

8.2.1 Molecular Interactions

As two atoms approach each other, the interactions between their atomic orbitals (AOs) yield molecular orbitals (MOs) of different bonding types and symmetries. A molecular orbital diagram of molecular nitrogen (fig. 8.1) can help visualize and better understand these interactions.

Electrons in any atom can be classified as core or valence electrons. Even though core electrons in atomic orbitals do form hybridized/molecular orbitals, their contributions are minimal and are usually ignored in molecular orbital diagrams and molecular electron configurations, and for this reason valence shell electrons are the only interacting particles that are considered [113]. In molecular nitrogen, there are 10 valence electrons in total (5 from each nitrogen atom) participating in molecular interactions. Molecular nitrogen in its ground state has the following electron configuration:

\[
2\sigma_g^2 2\sigma_u^2 1\pi_u^1 3\sigma_g^3 1\Sigma_g^+ 
\]

(8.1)

with orbitals that correspond to those shown in the molecular orbital diagram in fig. 8.1. The symbols and subscripts in an electron configuration represent molecular orbital symmetries, while the superscripts denote the number of electrons occupying a molecular orbital. Occasionally, for anti-bonding orbitals, an additional right superscripted asterisk is written.
Figure 8.1: Molecular orbital energy level diagram of $N_2$.

In the case of the electron configuration above, the anti-bonding orbital term is $2\sigma_u^2$. The different molecular bonding types and symmetries are discussed in the sections below.

The $\Sigma_g^+$ at the end of the electron configuration is a molecular term symbol; it is a characterization of the state of the molecule. The left superscript is known as the spin multiplicity $2S + 1$ or $2\Sigma + 1$ (not to be confused with the $\Sigma$ in the molecular term symbol). The spin multiplicity is essentially the number of possible orientations of spin $S$ or $\Sigma$ angular momenta. The second component of the term symbol is the projection of the
orbital angular momentum along the internuclear axis denoted by $\Lambda$ which can take on values $\Lambda = 0, 1, 2, 3, \ldots$; values that respectively correspond to the greek capital letters $\Sigma$, $\Pi$, $\Delta$, $\Phi$, etc. The third component is the parity, written as a right subscript $g$ or $u$ (gerade or ungerade). Lastly, is the reflection symmetry along an arbitrary plane containing the internuclear axis, which is written as a right superscript $+$ or $-$ in the molecular term symbol. In some texts, an additional right subscript $\Omega$ is included; it represents the projection of the total angular momentum along the internuclear axis [114]. The different excited and ionic states would then be reflected in different electron configurations and appropriate molecular term symbols.

**Bonding Types**

The interaction between atomic orbitals yield molecular orbitals of two bonding types: bonding and anti-bonding. Atomic orbitals that do not contribute to the overall interaction, result in non-bonding molecular orbitals. The energy of the resultant molecular orbitals with respect to their atomic orbitals is shown in fig. 8.2; with bonding molecular orbitals lower in energy, non-bonding molecular orbitals with the same energy, and anti-bonding orbitals with higher energy than the atomic orbitals that formed them.

**Figure 8.2:** Energy of molecular orbitals.

Bonding molecular orbitals are a result of constructive (in-phase) interactions between atomic orbitals (electrons that spend most of their time in between the nuclei, with each pair forming a bond), while out-of-phase atomic orbitals yield destructive interactions that
8.2 Molecular Nitrogen $N_2$

form anti-bonding molecular orbitals of paired electrons that negate bonding (since the probability of finding electrons between the nuclei is lower). The number of electrons that participate in bonding and anti-bonding orbitals determine the bond order and the bond strength of the molecule [115]. The bond order of a molecule can be found by:

$$\frac{\text{[number of bonding } e - \text{ number of anti-bonding } e]}{2}$$

and so for the nitrogen molecule, the bond order is $(8 - 2)/2 = 3$; the strong triple bond that molecular nitrogen is known for.

Symmetries

The two other molecular interactions to consider are the symmetries of the molecular orbitals and parities that are exclusive to centro-symmetric molecules. The first set of symmetries arise from the overlap of atomic orbitals; from which we get symmetries labeled $\sigma$, $\pi$, $\delta$, $\phi$, etc; because they reflect the symmetries of the atomic orbitals they were formed from. For molecular nitrogen, the symmetries are the result of $\sigma$ and $\pi$ bonds only. A $\sigma$ bond is an end-to-end bond formed from symmetric atomic orbitals and a $\pi$ bond is formed from a sideways overlap, with the $\sigma$ bond being the strongest covalent chemical bond of the two.

Since molecular nitrogen is homo-nuclear, it is also a centro-symmetric molecule; a molecule that exhibits a type of symmetry (parity) based on an inversion with respect to the centre of the internuclear axis. This type of symmetry is of two kinds: gerade (even) and ungerade (odd/uneven), which are denoted by right subscripts $g$ and $u$. Molecular orbitals of gerade ($g$) symmetry are the result of molecular orbitals with the same phase, while molecular orbitals of ungerade symmetry are the result of a phase change in the molecule orbitals following an inversion through the centre of symmetry of the molecule [115],[114].

8.2.2 States of $N_2$ and $N_2^+$

A binding energy spectrum identifies the ionized states and their respective ionization energies. A binding energy spectrum can be constructed using an (e,2e) coincidence experiment; in which an electron gun fires electrons at a target (e.g. $N_2$) and two electron energy analyzers detect the scattered and ejected electrons with equal energies. Since each molecular orbital has its own ionization energy, the energy of an ejected electron reflects the ionization energy of the molecular orbital it was ejected from. For this reason, the electronic structure of the ion $N_2^+$ will have various electronic configurations. Depending on the energy, the most dominant electronic configurations converge to the states: $X^2\Sigma_g^+$, $A^2\Pi_u$, $B^2\Sigma_u^+$, $C^2\Pi_u$. 
and $B^2\Sigma_u^+$ which respectively correspond to "holes" in the $3\sigma_g$, $1\pi_u$, and $2\sigma_u$ molecular orbitals. The electron configurations for those states with the number of electrons in each orbital can be seen in table 8.1 below [116].

<table>
<thead>
<tr>
<th>Molecule</th>
<th>State</th>
<th>$1\pi_u$</th>
<th>$3\sigma_g$</th>
<th>$1\pi_g$</th>
<th>$3\sigma_u$</th>
<th>Other</th>
</tr>
</thead>
<tbody>
<tr>
<td>$N_2$</td>
<td>$X^1\Sigma_g^+$</td>
<td>4</td>
<td>2</td>
<td>0</td>
<td>0</td>
<td></td>
</tr>
<tr>
<td>$N_2^+$</td>
<td>$X^2\Sigma_g^+$</td>
<td>4</td>
<td>1</td>
<td>0</td>
<td>0</td>
<td></td>
</tr>
<tr>
<td></td>
<td>$A^2\Pi_u$</td>
<td>3</td>
<td>2</td>
<td>0</td>
<td>0</td>
<td></td>
</tr>
<tr>
<td></td>
<td>$B^2\Sigma_u^+$</td>
<td>4</td>
<td>2</td>
<td>0</td>
<td>0</td>
<td>$-2\sigma_u$</td>
</tr>
</tbody>
</table>

Table 8.1: Electron configurations for molecular nitrogen. The numbers in columns 3 through 7 reflect the number of electrons occupying the molecular orbital in that column.

8.2.3 Potential Energy Curves for Molecular Nitrogen

Since nitrogen is a diatomic molecule, it will exhibit molecular vibration and rotation, and the potential between the two nuclei will vary with different ionic states. The Morse potential [117] can be used to approximate the potential between the two nuclei, the dissociation limits, and the vibrational and rotational levels. The Morse potential in terms of the internuclear distance is of the form:

$$V(R) = D_0 \left( e^{-2\alpha(R-r_e)} - 2e^{-\alpha(R-r_e)} \right)$$

(8.2)

where $D_0$ is the dissociation energy with respect to the minimum of the potential, $r_e$ the internuclear equilibrium distance, and $\alpha_e$ which is a constant that depends on the force constant $k$. The additional constants needed to find the energy of the vibrational and rotational levels can be seen in table 8.2 below [116],[118],[119].

<table>
<thead>
<tr>
<th>Molecule</th>
<th>State</th>
<th>$r_e$ (Å)</th>
<th>$\alpha_e$ (m$^{-1}$)</th>
<th>$B_e$ (eV)</th>
<th>$D_e$ (eV)</th>
<th>$\omega_e$ (rad/s)</th>
<th>$\omega_e\chi_e$ (rad/s)</th>
<th>$D_0$ (eV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$N_2$</td>
<td>$X^1\Sigma_g^+$</td>
<td>1.09708</td>
<td>1.7318</td>
<td>2.4771E-4</td>
<td>7.1410E-10</td>
<td>4.4430E14</td>
<td>2.6981E12</td>
<td>9.9017</td>
</tr>
<tr>
<td></td>
<td>$X^2\Sigma_g^+$</td>
<td>1.1642</td>
<td>1.881</td>
<td>2.3950E-4</td>
<td>7.5630E-10</td>
<td>4.1572E14</td>
<td>3.0327E12</td>
<td>8.8803</td>
</tr>
<tr>
<td>$N_2^+$</td>
<td>$A^2\Pi_u$</td>
<td>1.17364</td>
<td>2.000</td>
<td>2.1672E-4</td>
<td>6.9430E-10</td>
<td>3.5856E14</td>
<td>2.8275E12</td>
<td>7.7123</td>
</tr>
<tr>
<td></td>
<td>$B^2\Sigma_u^+$</td>
<td>1.07772</td>
<td>2.000</td>
<td>2.5701E-4</td>
<td>7.6500E-10</td>
<td>4.5581E14</td>
<td>4.3682E12</td>
<td>5.6921</td>
</tr>
</tbody>
</table>

Table 8.2: Vibrational and rotational constants for $N_2$ and $N_2^+$. The second column contains the electronic state. The remaining columns contain the following constants: $r_e$ is the internuclear equilibrium distance, $\alpha_e$ is a rotational constant (first term), $B_e$ is the rotational constant in equilibrium position, $D_e$ is the centrifugal distortion constant, $\omega_e$ is a vibrational constant (first term), $\omega_e\chi_e$ is a vibrational constant (second term), and $D_0$ is the dissociation energy with respect to the minimum of the potential.

The eigen-energies for vibrational levels (assuming a Morse potential) can be found by
inputting the constants $\omega_e$ and $\omega_e\chi_e$ from table 8.2 into the following [43]:

$$E_v = \hbar \left[ \omega_e \left( v + \frac{1}{2} \right) - \omega_e\chi_e \left( v + \frac{1}{2} \right)^2 \right]$$  \hspace{1cm} (8.3)

where $v$ corresponds to the vibrational levels with $v = 0, 1, 2, ..., v_{\text{max}}$. Using the Morse potential, and the appropriate eigenenergies and constants, the potential energy curves can then be plotted for the ground state of $N_2$ and the three ionic states of $N_2^+$ (as shown in fig. 8.3).

Figure 8.3: Morse potential energy curves and the first vibrational energy levels for the ground state of $N_2$ and the three ionic states of $N_2^+$.

The Franck-Condon principle [120] is then used to describe the intensities of vibronic transitions (or photon emission or absorption). This principle states the following: due to the relative size of the nuclei with respect to the electrons, the speed of the transition of those electrons (in a process such as ionization) is faster than the reaction time of the nuclei, and for this reason the nuclear configuration in a molecule experiences no significant change. However, following a transition such as ionization, the nuclei have to undergo realignment with the new electronic configuration; a realignment which manifests as molecular vibration. The calculated probability of transitions from the ground states of $N_2$ to any of the vibrational levels of the ionic states of $N_2^+$ can be found by calculating
the square of the Franck-Condon factors:

\[
P_{v=0,v'} = \left[ \int_{R=0}^{\infty} \psi_{v=0}(R)\psi_{v'}(R)dR \right]^2
\]

where the eigenfunctions using a Morse potential are given by [43]:

\[
\psi_v(R) = A_v e^{-\alpha(R-re)} e^{-\alpha(R-re)} \sum_{k=0}^{v} c_k \chi_k(R-re)
\]

where \(A_v\) is the normalization constant, \(\lambda = \frac{1}{2\chi_e} - \frac{v}{2}\), and the constants \(c_k\) are found from the recursive formula [43]:

\[
c_{k+1} = \frac{c_k}{\chi_e} \left( \frac{k-v}{(k+1)(k+\frac{1}{2} - 2v)} \right), \quad c_0 = 1
\]

The probability amplitudes and the energy transitions are shown in table 8.3.

<table>
<thead>
<tr>
<th>State</th>
<th>(FCF_{v=0,v'})</th>
<th>(E_{v=0,v'})</th>
<th>(E_{v,v'})</th>
<th>(E_{v=5,v'})</th>
<th>(E_{v=6,v'})</th>
</tr>
</thead>
<tbody>
<tr>
<td>(X^2 \Sigma_g^+)</td>
<td>0.95582 0.04177</td>
<td>1.03154 1.30118</td>
<td>2.15949 2.39177</td>
<td>4.21368 4.50795</td>
<td>9.8372E-7 2.1328E-7</td>
</tr>
<tr>
<td>(A^2 \Pi_u)</td>
<td>0.48161 0.29498</td>
<td>0.56683 1.56683</td>
<td>2.62034 2.62034</td>
<td>4.79647 4.79647</td>
<td>9.8372E-7 2.1328E-7</td>
</tr>
<tr>
<td>(B^2 \Sigma_u^+)</td>
<td>0.95933 0.03679</td>
<td>1.56683 1.56683</td>
<td>2.62034 2.62034</td>
<td>5.07924 5.07924</td>
<td>5.62752 5.62752</td>
</tr>
</tbody>
</table>

Table 8.3: Franck-Condon factors and Energy of Transitions.

The energy transitions that occur within a molecule are essentially the result of changes to the electronic, vibrational, and rotational states. In other words, the energy of a molecule’s transition from one electronic state to another is dependent upon molecular vibration, and the very vibrational transitions associated with rotational change; changes that are known as rovibronic transitions. As shown above, eigen-energies for specific vibrational levels \(v\) can be found using eq. (8.3):

\[
E_v = \hbar \left[ \omega (v + \frac{1}{2}) - \omega \chi_e \left( v + \frac{1}{2} \right)^2 \right]
\]

However, to take into account the rotational change that occurs during rovibronic transitions, we need an approximation with additional rotational terms that depend on the internuclear distance which depends on \(v\). These constants are the rotational constant \(B\) and the
centrifugal distortion constant $D$. The rotational constant $B$ is provided via rotational spectroscopy; a value the depends on the moment of inertia of the molecule. The centrifugal constant $D$ is related to the rotational constant $B$, and the reason for this is because of the centrifugal force that the molecule experiences during rotation. In a linear diatomic molecule such as $N_2$, the moment of inertia of the molecule increases as the centrifugal force pulls the atoms apart from each other. These constants have been calculated and experimentally found using different methods for a large number of molecules, and are widely available online. The constants for $N_2$ are labeled as $B_e$ and $D_e$ and are found in table 8.2. Another term needed for the approximation of transition energies is $J_{\text{max}}$, where $J$ is the molecular rotational quantum number; $J$ represents a given rotational level, in the same way that $v$ represents a given vibrational level. To find the peak $J_{\text{max}}$, a representation of the population of the rotational states is needed which can be found via the Boltzmann distribution [121]:

$$\frac{n_i}{n_0} = g_i e^{-E_i/k_B T}$$

(8.7)

where $k_B$ is the Boltzmann constant, $T$ is the temperature, $g_i$ is the degeneracy of state $i$, $n_i$ and $n_0$ are the population of the states, and $E_i$ is the energy of state $i$. The Boltzmann distribution is then maximized to find the most populated state, which can be seen as the most intense peak in a rovibrational spectrum. The equation then becomes:

$$\frac{n_i}{n_0} = (2J + 1)e^{-B_v J(J+1)/k_B T}$$

(8.8)

This equation can be used to display the population of rotational levels (as rotational lines) in a spectrum for a given state. For example, using the rotational constant $B_v$ (from table 8.2) for the ground state $X^1 \Sigma_g^+$ of $N_2$ and setting $T = 300K$, we obtain the populations shown in fig. 8.4.

From fig. 8.4, the peak is predicted at $J_{\text{max}} = 7$; this can be found mathematically by taking the equation used to find the population of rotational states and setting its first derivative to zero, then solving for $J_{\text{max}}$.

$$J_{\text{max}} = \frac{1}{2} \sqrt{\frac{2k_B T}{B_v}} - \frac{1}{2} \approx \sqrt{\frac{k_B T}{2B_v}}$$

(8.9)

$J_{\text{max}}$ for the $X^1 \Sigma_g^+$ state of $N_2$ and the $X^2 \Sigma_g^+$, $A^2 \Pi_u$, and $B^2 \Sigma_u^+$ states of $N_2^+$ were calculated and tabulated in table 8.4.

Now that we have established what the rotational and distortion constants and the $J_{\text{max}}$
values are (for the $X^1\Sigma_g^+$ state of $N_2$ and the $X^2\Sigma_g^+$, $A^2\Pi_u$, and $B^2\Sigma_u^+$ states of $N_2^+$), we can then introduce the additional terms to rewrite the approximation for the eigen-energies as

$$E_{v,J} = \hbar \left[ \omega_e \left( v + \frac{1}{2} \right) - \omega_e \chi_e \left( v + \frac{1}{2} \right)^2 \right] + B_v J(1 + J) - D_v J^2 (1 + J)^2 \quad (8.10)$$

Note that $E$ now depends on $v$ and $J$. From this new approximation, the transition energies

![Image of Figure 8.4: Population of rotational states for the ground state $X^1\Sigma_g^+$ of $N_2$.]

<table>
<thead>
<tr>
<th>$J_{max}$</th>
<th>$X^1\Sigma_g^+$</th>
<th>$X^2\Sigma_g^+$</th>
<th>$A^2\Pi_u$</th>
<th>$B^2\Sigma_u^+$</th>
</tr>
</thead>
<tbody>
<tr>
<td>7.2237</td>
<td>7.3465</td>
<td>7.7309</td>
<td>7.0891</td>
<td></td>
</tr>
</tbody>
</table>

**Table 8.4:** Calculated $J_{max}$ values.
from the ground state of \( N_2 \) can be calculated by

\[
\Delta E_{v,J} = E(v',J') - E(v,J) = \hbar \left[ \omega_e \left( v' + \frac{1}{2} \right) - \omega_e \chi_e \left( v + \frac{1}{2} \right) \right]^2 + B_v'J'(J' + 1) - D_v'J'^2(J' + 1)^2
\]

where \( J = J_{\text{max}} \) for the given states. The energy of these transitions have been calculated and are shown in table 8.3. Wolfram Mathematica [122] (a computational and technical software) was used to find the Morse potential energy curves, the eigenfunctions, the Franck-Condon factors, the eigen-energies of the vibrational levels, and the population of the rotational levels for all four states.

8.3 (e,2e) Studies on \( N_2 \) in Coplanar and Non-Coplanar Geometries

The first (e,2e) coincidence experiment on the \( N_2 \) molecule was carried out using the setup of the old (e,2e) apparatus, and in parallel with designing and building the modernized (e,2e) hardware and software. This experiment resulted in the peer-reviewed publication in the Journal of Physics B: Atomic, Molecular and Optical Physics [59]. The experiment was done in collaboration with theoreticians Sadek Amami and Don Madison from Missouri University of Science & Technology (USA). Using a range of geometries from the coplanar to the perpendicular plane and the neutral \( N_2 \) molecule as the experimental target, experimental and theoretical ionization triple differential cross-sections were determined and compared to each other.

The experiment was performed with incident electron energies ~10 and ~20 eV above the ionization potential of the \( 3\sigma_g, 1\pi_u \) and \( 2\sigma_u \) states for \( N_2 \), using both equal and non-equal outgoing electron energies. The data were obtained with the incident electron beam in the scattering plane \( \psi = 0^\circ \), at 45° to this plane and orthogonal to the plane \( \psi = 90^\circ \). This series of experiments resulted in a set of nine inter-normalized measured ionization triple differential cross-sections, with each at a given outgoing electron energy (4.6 eV ± 0.5 eV, 9.7 eV ± 0.5 eV, and 4.6 eV ± 0.5 eV and 14.5 eV ± 0.5 eV). Experimental results were compared against new calculations using various distorted wave methods:

- The molecular three-body distorted wave (M3DW) approximation.
- The distorted wave Born approximation (DWBA).
- The DWBA using the Ward–Macek (WM) approximation.
Further details on the mentioned theoretical models are found in chapter 2.

8.3.1 Experimental Procedures

As mentioned above, the ground state electronic configuration of $N_2$ is $2\sigma_g^22\sigma_u^21\pi_u^43\sigma_g^21\Sigma_g^+$. Ionization can occur from each of the orbitals, leading to a number of $N_2^+$ ions in different final states. The target’s ionized states are the $X^2\Sigma_g^+$, $A^2\Pi_u$, and $B^2\Sigma_u^+$ states, which respectively correspond to "holes" in the $3\sigma_g$, $1\pi_u$, and $2\sigma_u$ molecular orbitals. The first step to performing an (e,2e) coincidence experiment is to identify the energies of the target states. This was carried out by performing a binding energy spectral scan over a range of incident electron energies from 23 eV to 29 eV, in steps of 0.125 eV. For the first set of energy scans, both electron energy analyzers were tuned to detect outgoing electron energies of $4.6 \pm 0.5$ eV at a forward angle of $45^\circ$ to the z-axis shown in fig. 1.1. Similar energy scans were carried out for each of the data sets that were measured. The binding energy spectral scans were carried out in all three geometries (in the scattering plane $\psi = 0^\circ$, at $45^\circ$ to this plane and orthogonal to the plane $\psi = 90^\circ$). The binding energy spectra (shown in fig. 8.5) showed that the energy for the states $3\sigma_g$, $1\pi_u$, and $2\sigma_u$ states for $N_2$ were approximately $24.7 \text{ eV}$, $26.1 \text{ eV}$, and $27.9 \text{ eV}$, respectively.

Figure 8.5 is a demonstration that the (e,2e) experimental apparatus can resolve each of the molecular orbital contributions. The binding energy spectra also show the dependency of the cross-section measurements on the ionized individual states. The operating conditions were kept the same for each binding energy scan (for every angle $\psi$ with respect to the detection plane). The results were inter-normalized to the $3\sigma_g$ state in a coplanar geometry after it was normalized to unity. The results (including the additional data sets at any given energy) were further inter-normalized through the common point at $\theta_1 = \theta_2 = 90^\circ$.

Using the results obtained from these binding energy scans, the (e,2e) spectrometer was setup to perform angular-based scans. The electron energy analyzers were tuned to detect the following outgoing electron energies:

- symmetric energies: $4.6 \text{ eV} \pm 0.5 \text{ eV}$ for both analyzers 1 and 2.
- symmetric energies: $9.7 \text{ eV} \pm 0.5 \text{ eV}$ for both analyzers 1 and 2.
- asymmetric energies: $4.6 \text{ eV} \pm 0.5 \text{ eV}$ for one analyzer, with the other tuned at $14.5 \text{ eV} \pm 0.5 \text{ eV}$.

For each of these tuning conditions, the incident electron beam angles were set to:

- $\psi = 0^\circ$ for coplanar geometry.
- $\psi = 45^\circ$ with respect to the detection plane.
8.3 \((e,2e)\) Studies on \(N_2\) in Coplanar and Non-Coplanar Geometries

Figure 8.5: Binding energy spectra for outgoing electron energies of 4.6 eV ± 0.5 eV taken with the electrons detected at a forward angle of 45° to the z-axis. The results are shown for (a) the coplanar geometry where \(\psi = 0°\), (b) for the incident electron at 45° to the detection plane, and (c) for the perpendicular geometry where \(\psi = 90°\). Gaussian curves were fitted to the data for each state, so that the relative contributions and their peak energies could be determined. The peak of the \(3\sigma_g\) state in a coplanar geometry was normalized to unity, and all data were then inter-normalized to this peak as discussed in the text.

- \(\psi = 90°\) for the perpendicular geometry.

Lastly, for each of these analyzer tuning conditions and spectrometer geometries, the electron gun was tuned to the three ionizing energies obtained from the binding energy spectra in fig. 8.5. This resulted in three master \((e,2e)\) coincidence data sets (a total of 27 individual angular-based triple differential cross-section (TDCS) measurements).

In addition to the analyzer and electron gun tuning conditions, and spectrometer geometries, the data were collected using up to 10 angular sweeps of the detection plane (from 35° to 125° in 2.5° increments), with \((e,2e)\) coincidence measurements taken for approximately 5000s at each scattering angle. The tuning conditions for the lens (L1B) and deflector elements (Dx and Dy) for both electron energy analyzers were optimized for
each new angle in any given angular sweep. The following additional settings were also used:

- Vacuum chamber: operating pressure of approximately \(2 \times 10^{-5}\) torr, and a base pressure of \(1 \times 10^{-7}\) torr.
- Incident electron beam current: \(\approx 200\) nA.
- Detection electronics: The time-to-amplitude converter’s timing window was set to 500 ns, with an added \(-300\) ns delay time to the stop signal. This positioned the \((e,2e)\) coincidence peak close to the center of the spectrum (for equal energy scans).

As mentioned earlier in this section, a symmetric geometry was chosen for all data sets where \(\theta_1 = \theta_2 = \theta\). The cross-section measurements at each angle \(\theta\) were averaged, and the standard error on the mean was used to calculate their respective uncertainties. As mentioned above, in each of the three data sets, the cross-section measurements were inter-normalized to the \(3\sigma_g\) peak in the coplanar geometry. Results for this series of experiments including comparison to different theories are covered in the next section.

8.3.2 Results

Data analysis procedures (extraction, analysis, and visualization) were carried out on the angular-based \((e,2e)\) coincidence data sets. The cross-section measurements were calculated for each angle by subtracting the background mean from the coincidence signal, and then summing all resultant coincidence counts within that window (coincidence peak window). This step was repeated for each angular step in an angular sweep (from \(35^\circ\) to \(125^\circ\) in \(2.5^\circ\) increments). For additional angular sweeps with the same tuning conditions and spectrometer geometries, the mean cross-section measurements for each angular step were calculated from all sweeps.

The complete data sets are shown in fig. 8.6, fig. 8.7, and fig. 8.8, with comparisons to the three different distorted wave models. As mentioned earlier, the electron energy analyzer tuning conditions for detecting the energy of outgoing electrons were symmetric \((E_1 = E_2)\), the results of which are seen in fig. 8.6 and fig. 8.7. The results shown in fig. 8.8 are for electron energy analyzers tuned with asymmetric energies \((E_1 \neq E_2)\). The following outgoing electron energies for all figures are:

- Figure 8.6: \(E_1 = E_2 = 4.6\) eV \(\pm 0.5\) eV
- Figure 8.7: \(E_1 = E_2 = 9.7\) eV \(\pm 0.5\) eV
- Figure 8.8: \(E_1 = 4.6\) eV \(\pm 0.5\) eV and \(E_2 = 14.5\) eV \(\pm 0.5\) eV
The cross-section measurements obtained from these experiments were not absolute. For this reason, the maximum peak for the $3\sigma_g$ state from the experimental data was normalized to its respective peak from the molecular three-body distorted wave (M3DW) calculations in a coplanar geometry at each energy. The remaining experimental data in each of the three data sets were then inter-normalized to this maximum. The red circle in the figures highlights the common point ($\theta_1 = \theta_2 = 90^\circ$) for all spectrometer geometries. For direct comparison, all theoretical cross-sections were calculated on an absolute scale.

As mentioned before, there is a total of nine sets of data for each figure. The results for each column in the figures are as follows:

- The first column: this shows the results from ionizing the $3\sigma_g$ state.
- The middle column: this shows the results from ionizing the $1\pi_u$ state.
- The last column: this shows the results from ionizing the $2\sigma_u$ state.

The rows in each figure correspond to a spectrometer geometry where:

- (a): $\psi = 0^\circ$.
- (b): $\psi = 45^\circ$.
- (c): $\psi = 90^\circ$.

The theoretical calculations in the figures are represented in the following manner:

- Solid black curve: this represents the molecular three-body distorted wave (M3DW) calculations.
- Red coarse-dashed curve: this represents the distorted wave Born approximation (DWBA) calculations.
- Blue finely-dashed curve: this represents the calculations for the DWBA model using the Ward–Macek (WM) approximation method; this then includes a term for post collision interactions (PCI), which are not included in the DWBA model.

It is important to note that the post collision interactions (PCI) between the electrons, force the triple differential cross-sections (TDCS) to be zero at $\theta = 0^\circ$ and $180^\circ$ when the outgoing electrons have equal energy (such as $E_1 = E_2 = E$ in fig. 8.6 and fig. 8.7).

Results for Symmetric Outgoing Electron Energies of $4.6\text{ eV} \pm 0.5\text{ eV}$

The results shown in fig. 8.6 are for outgoing electron energies of $4.6\text{ eV} \pm 0.5\text{ eV}$. The experimental data for the $3\sigma_g$ state indicate the following:
Figure 8.6: This data set shows normalized triple differential cross-sections (TDCS) for outgoing electron energies where $E_1 = E_2 = 4.6\, \text{eV} \pm 0.5\, \text{eV}$ in (a) a coplanar geometry where $\psi = 0^\circ$, (b) for the incident electron beam at $45^\circ$ to the detection plane, and (c) for the perpendicular geometry where $\psi = 90^\circ$. Normalization procedures were carried out by setting the peak for the $3\sigma_g$ state from the experimental data to its respective peak from the molecular three-body distorted wave (M3DW) calculations. In addition, the coplanar data for the $1\pi_u$ and $2\sigma_u$ states were then set relative to the $3\sigma_g$ state using the binding energy spectra shown in fig. 8.5. Inter-normalization of all data sets were carried out by using the common normalization point when $\theta_1 = \theta_2 = 90^\circ$. The normalized data sets are plotted on a logarithmic scale.

- The coplanar experimental data show that the triple differential cross-sections (TDCS) are dominated by forward scattering, until the incident electron beam is moved out of the detection plane. The cross-section peak is found at $\theta \sim 45^\circ$ with a minimum occurring at $\theta \sim 90^\circ$. The cross-section then increases at higher scattering angles.
- The experimental data for $\psi = 45^\circ$ with respect to the detection plane show that the electrons are back scattered at this energy, and uniform cross-sections with increasing scattering angles.
- The experimental data for the perpendicular geometry shows symmetry around $\theta = 90^\circ$.

The experimental data for the $1\pi_u$ state show the following:

- The experimental data for all geometries follow a similar trend to that of the $3\sigma_g$ state.
The coplanar geometry shows a maximum in the cross-sections in the forward direction.

The back scattering is slightly dominant for $\psi = 45^\circ$ with respect to the detection plane.

Overall, the forward and back scatter peaks are not as strong as those seen in the $3\sigma_g$ experimental data.

Lastly, the experimental data for the $2\sigma_u$ state indicate the following:

- There is very little change when the incident electron beam moves through the three geometries.
- The experimental data show uniform triple differential cross-section measurements with increasing scattering angles.
- Overall, there is no particular preference for either forward or backward scattering.

A different trend is seen upon comparison of the experimental data to the theoretical calculations. Since post collision interactions (PCI) are not included in the distorted wave Born approximation (DWBA), a large difference is seen between it and the experimental data; this points to the importance of including PCI. In all geometries and for all target states, there is an over-estimation of the cross-section measurements, and a prediction of features that are not observed in the experimental data.

There is a significant improvement in the theoretical results compared to the experimental data when the Ward–Macek (WM) interaction is included in the distorted wave Born approximation, but there is still a prediction of features that are not seen in the experimental data. However, the peaks produced by including the Ward–Macek (WM) interaction are in the same position as in the experimental data for non-coplanar geometries (when $\psi = 45^\circ$ and $\psi = 90^\circ$).

Surprisingly, the molecular three-body distorted wave (M3DW) calculations do not show any improvements in the results from those provided by the Ward–Macek (WM) theory, given that post collision interactions (PCI) are also included in the M3DW calculation. The features predicted by this model are in disagreement with the results in the data for all geometries and states. This theory predicts a dominant peak in the triple differential cross-section (TDCS) calculations near $\theta = 90^\circ$, as seen for helium at similar energies [26]. Compared to other ionizing scattering processes, the molecular three-body distorted wave (M3DW) theory seems to include post collision interactions (PCI) too strongly. Earlier studies on $H_2$ revealed an overestimation of the post collision interaction effects (PCI) when
using the molecular three-body distorted wave (M3DW) theory, and a better agreement between the Ward–Macek (WM) theoretical model and the experimental data [82]. Lastly, when the incident electron beam is perpendicular to the detection plane ($\theta = 90^\circ$), the molecular three-body distorted wave (M3DW) calculations predict a maximum, while the other two models (WM and DWBA) predict a minimum.

**Results for Symmetric Outgoing Electron Energies of 9.7 eV ± 0.5 eV**

![Figure 8.7](image)

**Figure 8.7:** This data set shows normalized triple differential cross-sections (TDCS) for outgoing electron energies where $E_1 = E_2 = 9.7\text{ eV} \pm 0.5\text{ eV}$ in (a) a coplanar geometry where $\psi = 0^\circ$, (b) for the incident electron beam at $45^\circ$ to the detection plane, and (c) for the perpendicular geometry where $\psi = 90^\circ$. Normalization procedures were carried out by setting the peak for the $3\sigma_g$ state from the experimental data to its respective peak from the molecular three-body distorted wave (M3DW) calculations at this energy. The inter-normalization procedures used were the same as those described in fig. 8.6.

The results shown in fig. 8.7 are for equal outgoing electron energies of $9.7\text{ eV} \pm 0.5\text{ eV}$. Relative to the previous data set, there is an improvement in the comparison between the theoretical calculations and experimental data, especially for the molecular three-body distorted wave (M3DW) calculations. The forward scattering is again dominant in a coplanar geometry for the $3\sigma_g$ and the $1\pi_u$ states. The experimental data for $\psi = 45^\circ$ for all states, show more features relative to those seen in the lower energy data set. The experimental data for the perpendicular plane geometry show little to no features across all scattering angles, despite the two broad peaks that appear around the minimum of $\theta = 90^\circ$ for the $1\pi_u$ state. There is little to no change seen in the experimental data for
the $2\sigma_u$ state across all scattering angles and geometries at this energy.

Comparison of the experimental data to the theoretical calculations show:

- The distorted wave Born approximation (DWBA) theory fails to predict key features in the data.
- The Ward–Macek (WM) approximation for post collision interactions improves the comparison greatly between theory and experiment.
- The molecular three-body distorted wave (M3DW) calculations are the most accurate in predicting the features seen in the coplanar geometry, especially for the $3\sigma_g$ and $1\pi_u$ states.

All three calculations fail to predict the results for the $2\sigma_u$ state for any of the incident electron angles. However, the calculations resemble those seen in the lower energy data set (shown in fig. 8.6) for when the incident electron beam is moved away from the detection plane.

**Results for Asymmetric Outgoing Electron Energies of 4.6 eV ± 0.5 eV and 14.5 eV ± 0.5 eV**

The results shown in fig. 8.8 relax the ‘doubly symmetric’ conditions of the experiment by tuning the electron energy analyzers to have unequal outgoing electron energies where $E_1 = 4.6 \text{ eV} \pm 0.5 \text{ eV}$ and $E_2 = 14.5 \text{ eV} \pm 0.5 \text{ eV}$. The experimental data reveal dominant forward scattering for the $3\sigma_g$ and the $1\pi_u$ states in the coplanar geometry, with the features being less pronounced than those seen with equal outgoing electron energies. The experimental results for all states when $\psi = 45^\circ$ reside somewhere between those seen in fig. 8.6 and fig. 8.7. The experimental data in the perpendicular plane show little to no features at all scattering angles. Experimental results for the $2\sigma_u$ state show little to no structure for all scattering angles and geometries.

Comparison of the experimental data to the theoretical calculations show similar trends as those seen in the previous data set. The distorted wave Born approximation (DWBA) calculations fail again to predict the experimental results, but an agreement is seen with the Ward–Macek (WM) calculations for the $3\sigma_g$ and the $1\pi_u$ states. There is an overestimation yet again of the post collision interaction (PCI) effects from the molecular three-body distorted wave (M3DW) calculations. This overestimation appears as a dominant peak when $\theta = 90^\circ$ and $\psi = 90^\circ$ in the calculations which is not seen in the experimental results. The coplanar and backward scattered cross-sections that were calculated using the Ward–Macek (WM) and the molecular three-body distorted wave (M3DW) models, are higher than those seen in the experimental results. The Ward–Macek (WM) calculations
Figure 8.8: This data set shows normalized triple differential cross-sections (TDCS) for outgoing electron energies where $E_1 = 4.6 \text{ eV} \pm 0.5 \text{ eV}$ and $E_2 = 14.5 \text{ eV} \pm 0.5 \text{ eV}$ in (a) a coplanar geometry where $\psi = 0^\circ$, (b) for the incident electron beam at $45^\circ$ to the detection plane, and (c) for the perpendicular geometry where $\psi = 90^\circ$. Normalization procedures were carried out by setting the peak for the $3\sigma_g$ state from the experimental data to its respective peak from the molecular three-body distorted wave (M3DW) calculations at this energy. The inter-normalization procedures used were the same as those described in fig. 8.6 and fig. 8.7.

are closer in agreement with the data obtained in non-coplanar geometries. All calculations fail to predict the results for the $2\sigma_u$ in both coplanar and non-coplanar geometries.

Results for Symmetric Outgoing Electron Energies of $20 \text{ eV} \pm 0.5 \text{ eV}$

The additional results shown in fig. 8.9 are for ionization from the $3\sigma_g$ state only (no experiments were done for the $1\pi_u$ and $2\sigma_u$ states) with an incident energy of $\sim 40 \text{ eV}$ above the ionization potential (with $E_1 = E_2 = 20 \text{ eV} \pm 0.5 \text{ eV}$). The reason for this additional set of experimental scans was to determine how each theoretical model compares to experimental results at a higher energy.

As seen in fig. 8.9, there is an overall improvement in the results obtained in the coplanar geometry when compared to the experimental results at lower energies. This can be seen for forward scattered cross-sections where both M3DW and WM models predict the position of the peak in the triple differential cross-sections (TDCS). However, as the scattering angles increase in this geometry, the agreement becomes weaker between the models and the experimental data. The DWBA model shows an agreement with the experimental
Figure 8.9: This data set shows normalized triple differential cross-sections (TDCS) for outgoing electron energies where $E_1 = E_2 = 20\,\text{eV} \pm 0.5\,\text{eV}$ in (a) a coplanar geometry where $\psi = 0^\circ$, (b) for the incident electron beam at $45^\circ$ to the detection plane, and (c) for the perpendicular geometry where $\psi = 90^\circ$. Normalization procedures were carried out by setting the peak for the $3\sigma_g$ state from the experimental data to its respective peak from the molecular three-body distorted wave (M3DW) calculations at this energy. The data are inter-normalized to the common point.

results for $\psi = 45^\circ$, while the other two models seem to underestimate the cross-sections. All models have a much better agreement with the experimental results for data taken in the perpendicular plane, despite the underestimation in the cross-sections. Overall, the accuracy of all three theoretical models is seen to improve with increasing incident energies.

8.3.3 Discussion and Conclusions

The experimental results presented in this section are for outgoing electron energies where $E_1 = E_2 = 4.6\,\text{eV} \pm 0.5\,\text{eV}$, $E_1 = E_2 = 9.7\,\text{eV} \pm 0.5\,\text{eV}$, and $E_1 = 4.6\,\text{eV} \pm 0.5\,\text{eV}$ and $E_2 = 14.5\,\text{eV} \pm 0.5\,\text{eV}$. An additional set of experimental scans were carried out at a higher incident energy of $\sim 40\,\text{eV}$ to ascertain the comparison between theory and experiment. The results were obtained by using a range of scattering angles in all three geometries.
(where $\psi = 0^\circ$, $\psi = 45^\circ$, and $\psi = 90^\circ$), and were normalized to a single and common point. Binding energy spectral scans were performed which allowed data obtained from the $3\sigma_g$, $2\sigma_u$, and $1\pi_u$ states to be inter-normalized. Overall, the experimental results on $N_2$ show that the cross-sections are very sensitive to the states from which the ionization occurs, as well as the incident energy from the post collisional interactions.

In addition to experimental results, calculations based on three theoretical models were presented, all of which show the importance and sensitivity of post collision interactions at these energies. It was seen in several cases that the molecular three-body distorted wave (M3DW) calculations overestimated the effects of post collision interactions, and how the Ward–Macek (WM) approximation was more accurate.

The experimental results and comparisons to all three theoretical models discussed in this section, were published in the Journal of Physics B: Atomic, Molecular and Optical Physics in 2016, which can be found in [59].

8.4 (e,2e) Studies on $N_2$ in Coplanar Geometry with Fixed Angles

Further (e,2e) coincidence experiments on the $N_2$ molecule were carried out using the setup of the old (e,2e) apparatus. This series of experiments resulted in a second publication in the Journal of Physics B: Atomic, Molecular and Optical Physics [60]. The experiment was again carried out in collaboration with theoreticians Sadek Amami and Don Madison from Missouri University of Science & Technology (USA). Experimental and theoretical ionization triple differential cross-sections were determined and compared with one another, using only a coplanar geometry at higher energies than those in section 8.3, and for fixed angles for one of the outgoing electrons.

The experiment was performed with incident electron energies $\sim 20$ and $\sim 40$ eV above the ionization potential for the $3\sigma_g$ and $1\pi_u$ states for $N_2$, using equal energies for the outgoing electrons. The data were obtained with the incident electron beam in the scattering plane where $\psi = 0^\circ$. This experiment resulted in six sets of measured ionization triple differential cross-sections, with fixed angles of $45^\circ$, $90^\circ$, and $125^\circ$ with respect to the incident electron beam for one of the outgoing electrons. Experimental results were compared against calculations using various distorted wave methods:

- The distorted wave Born approximation (DWBA).
- The molecular three-body distorted wave (M3DW) approximation.
- The M3DW using the Ward–Macek (WM) approximation for post collision interactions (PCI).

Further details on the theoretical models are found in chapter 2.
8.4.1 Experimental Procedures

In the experiment covered in the previous section, the experimental results for the ionization of \( \text{N}_2 \) were obtained in a doubly-symmetric geometry, where the tuning conditions for both electron energy analyzers were symmetric in scattering angles and energies (\( \theta_1 = \theta_2 \) and \( E_1 = E_2 \)). The experimental results were compared with calculations based on three theoretical models from the Missouri group of Don Madison. As seen in section 8.3, there were challenges performing the experiment using the doubly-symmetric geometry due to the sensitivity of the triple differential cross-sections to both initial and final states of the system. In addition, the theoretical models failed to predict the experimental results for low energies such as \( E_1 = E_2 \sim 4.6 \text{ eV} \), however the predictions improved for the higher energies \( E_1 = E_2 = 20 \text{ eV} \). Overall, the experiment covered in section 8.3 motivated further work (discussed here) to obtain cross-section measurements at higher energies to see if the theoretical models improve in predicting the experimental results. As mentioned in section 8.2, the ground state electronic configuration of \( \text{N}_2 \) is:

\[
2\sigma_g^22\sigma_u^21\pi_u^43\sigma_g^21\Sigma_g^+ \]

Ionization can occur from each of the mentioned orbitals, leading to a number of \( \text{N}_2^+ \) ions in different final states. The target ionized states for this experiment are the \( X^2\Sigma_g^+ \) and \( A^2\Pi_u \) states.

A binding energy spectrum was performed to identify the ionization energies required to target the \( 3\sigma_g \) and \( 1\pi_u \) molecular orbitals. The procedures for this experiment are similar to those in section 8.3.1, with the exception of the parameters (and constraints) chosen for this experiment; coplanar geometry, higher energies, and fixed angles for one of the outgoing electrons. The base and working pressure for the vacuum chamber, the incident electron beam current, and the detection electronics’ settings, were kept the same as those in the previous experiment (see section 8.3.1).

The binding energy spectral scans were performed over the \( 3\sigma_g \) and \( 1\pi_u \) states with \( \theta_1 = \theta_2 = 45^\circ \), for both electron energy analyzers at 10 eV and at 20 eV. The results from the binding energy scans were used to determine cross-section ratios of the states:

- \( (1\pi_u^{45^\circ} : 3\sigma_g^{45^\circ})^{10/10 \text{ eV}} = (49\% \pm 6\%) \)
- \( (1\pi_u^{45^\circ} : 3\sigma_g^{45^\circ})^{20/20 \text{ eV}} = (23\% \pm 3\%) \)

These ratios identified the relative strengths of the ionized states, which were then used to inter-normalize the experimental data. An example of a binding energy spectra obtained for the 10 eV outgoing electron energies can be seen in fig. 8.10.
Figure 8.10: An example of a binding energy spectra, obtained for the 10 eV outgoing electron energies at scattering angles of 45°. The $3\sigma_g$ and $1\pi_u$ states are well resolved, allowing the relative ratios of the peaks of those states to be determined and used in inter-normalization procedures.

The experimental constraints applied on the geometry, scattering angles, and the outgoing electron energies were as follows:

- The coplanar geometry where $\psi = 0^\circ$ was used so that the incident, scattered, and ejected electrons were detected in the same plane.
- A fixed angle with respect to the incident electron momentum $k_0$ was chosen for one of the emerging (scattered or ejected) electrons, and a varying angle around the interaction region was used for the other.
- Equal energies for the outgoing electrons were chosen so that $| k_1 | = | k_2 |$ and $E_1 = E_2 = (E_{inc} - IP)/2$.

The scattering angles for this experiment were set by fixing $k_1$ and moving $k_2$ around the plane, and then fixing $k_2$ and moving $k_1$ around the plane, as in fig. 8.11. The fixed angles chosen for this experiment were $\theta_1(\theta_2)^{Fixed} = 45^\circ$, $90^\circ$, and $125^\circ$. The measured triple differential cross-sections must be the same regardless of which of the scattering angles is fixed (thus proving the physical accuracy of the (e,2e) spectrometer). Since no differences were found in the measured cross-sections, the experimental results for a series of angular sweeps with similar parameters were averaged and used to produce the final results.

For obtaining the cross-section measurements, the electron energy analyzers were tuned to detect outgoing electrons of symmetric energies $(E_1, E_2) = (10\,\text{eV}, 10\,\text{eV})$ and $(E_1, E_2) = (20\,\text{eV}, 20\,\text{eV})$, and the incident electron beam energy was tuned to select either state ($3\sigma_g$ or $1\pi_u$). For each outgoing electron energy (10 eV and 20 eV), the experiments
produced results containing six individual data sets (three per state). The experimental data for each energy were inter-normalized to the common point, and since the measured cross-sections were not absolute, the experimental data was normalized once more to the molecular 3-body distorted wave (M3DW) calculations.

In addition to the experimental scans mentioned above, coplanar doubly-symmetric experiments 100 eV above the $3\sigma_g$ ionization potential were performed. The experimental data were taken with each electron energy analyzer at 50 eV and the same scattering angle ($\theta_1 = \theta_2$). The reason for this additional series of experiments, was to elucidate if the cross-section predictions made by the theoretical models improve at higher energies, which was found to be the case.

8.4.2 Results

Coplanar Measurements with Fixed Angles for Outgoing Electron Energies of 10 eV

The results shown in fig. 8.12 are for outgoing electron energies of 10 eV, at the fixed angles of 45°, 90°, and 125° for both $3\sigma_g$ and $1\pi_u$ states. The logarithmic scales was chosen for the plots to allow comparison of the experimental data to the three different theoretical models. The theoretical calculations have been convoluted with the (e,2e) spectrometer’s
Figure 8.12: Results for outgoing energies of 10 eV, for the $3\sigma_g$ state shown in (a)–(c) and the $1\pi_u$ state shown in (d)–(f). Since the experiments do not measure absolute cross sections, the experimental data are normalized to the peak of the molecular three-body distorted wave (M3DW) calculations in (a) as described in section 8.3 and in the published results in [59]. All other data are then inter-normalized at the angles shown. All calculations are convoluted with the angular resolution of the apparatus.

Angular resolution.

Similar to the approach mentioned in section 8.3 and [59], the data were again normalized to the maximum peak of the molecular three-body distorted wave (M3DW) calculations, because the experiments do not measure absolute cross-sections. The normalization point is shown in (a) of fig. 8.12. The data for the $3\sigma_g$ state shown in subfigures (a), (b), and
(c) of fig. 8.12, were inter-normalized through their common points to allow for reflection symmetry in the detection plane. The inter-normalization points can be better understood by looking at subfigure (b) in fig. 8.12 as an example; the left-hand inter-normalization point where \( \text{TDCS}(\theta_1 = 45^\circ, \theta_2 = 90^\circ) \equiv \text{TDCS}(\theta_1 = 90^\circ, \theta_2 = 45^\circ) \) in subfigure (a) of fig. 8.12, and the right-hand inter-normalization point where \( \text{TDCS}(\theta_1 = 90^\circ, \theta_2 = 125^\circ) \equiv \text{TDCS}(\theta_1 = 125^\circ, \theta_2 = 90^\circ) \) in subfigure (c) of fig. 8.12. The experimental results for the \( 1\pi_u \) state are shown in subfigures (d), (e), and (f) in fig. 8.12, are inter-normalized to the \( 3\sigma_g \) state using the obtained binding energy spectra ratios of the peaks. The inter-normalization procedure used for the \( 1\pi_u \) state is the same as that of the \( 3\sigma_g \), and the relative common points are shown in the same manner.

The experimental data of fig. 8.12 show that none of the theoretical models succeed in predicting the results for outgoing electrons with energies of 10 eV for all three fixed angles:

- Fixed angle of 45\(^\circ\): the experimental data for the \( 3\sigma_g \) state at the fixed angle of 45\(^\circ\) respectively show maxima and minima, at \( \theta = 45^\circ \) and \( \theta = 80^\circ \), in contrast to the predictions made by all theoretical calculations. Even though there is an improvement in the predictions for the \( 1\pi_u \) state at this angle, the models predict features that are not seen in the experimental results.

- Fixed angle of 90\(^\circ\): the calculations for \( 3\sigma_g \) and \( 1\pi_u \) states at this angle predict a broad minimum at \( \sim \theta = 90^\circ \), where the experimental results find a shallow minimum at \( \sim \theta = 90^\circ \) for the \( 1\pi_u \) state and at \( \sim \theta = 100^\circ \) for the \( 3\sigma_g \) state. Although the calculations and results show a better agreement, the calculations for the \( 3\sigma_g \) state are an order of magnitude larger than the normalized data.

- Fixed angle of 125\(^\circ\): the models fail to predict the cross-sections for both the \( 3\sigma_g \) and \( 1\pi_u \) states; calculated cross-sections are much larger than those measured by the experiment.

The theoretical models fail again (as seen in fig. 8.12 and in [59]) in predicting the experimental results.

**Coplanar Measurements with Fixed Angles for Outgoing Electron Energies of 20 eV**

The results shown in fig. 8.13 are for outgoing electron energies of 20 eV, again for fixed angles of 45\(^\circ\), 90\(^\circ\), and 125\(^\circ\) for both \( 3\sigma_g \) and \( 1\pi_u \) states. The data seen in fig. 8.13 are again inter-normalized and normalized to the M3DW calculated peak of the \( 3\sigma_g \) state. The relative common points between the data sets for each state are also shown.

Although a better agreement is seen between the results and the calculations at this energy, there still remains significant difference between experiment and theory:
Figure 8.13: Results for outgoing electron energies of 20 eV for the $3\sigma_g$ and $1\pi_u$ states. The data are normalized to the peak of the M3DW theoretical calculations in (a). All other data are then inter-normalized at the angles shown, as described in the text. All calculations are convoluted with the angular resolution of the apparatus.

- Fixed angle of 45°: the calculations for both states at this angle are closer in magnitude to the results, with the M3DW and WM calculations being better in agreement (due to significance of the post collision interactions) than those of the DWBA model. Similarly, the minima and maxima for the $3\sigma_g$ state predicted by the calculations disagree with the experimental results. However, the WM and M3DW calculation for the $1\pi_u$ state (shown in subfigure (d) of fig. 8.13) shows better agreement with
8.4 (e,2e) Studies on $N_2$ in Coplanar Geometry with Fixed Angles

experiment.

- Fixed angle of 90°: the best agreement at this angle is the M3DW calculation for the $3\sigma_g$ state (shown in subfigure (b) in fig. 8.13). However, this calculation predicts a minimum at $\sim 105^\circ$ in contrast with the measured minimum found at $\sim 70^\circ$. All three models show a better agreement with experiment for the $1\pi_u$ state for the forward direction. However, the minima predicted by theory do not agree with those found in the experimental results.

- Fixed angle of 125°: compared to the $1\pi_u$ state, the models show a better agreement for the $3\sigma_g$ state (shown in subfigure (c) in fig. 8.13). In addition, the experimental results do not show the theories’ predicted deep minima at $\sim 65^\circ$.

The disagreements (discussed above) between theory and experiment make it difficult to know how the models can be improved. Overall, calculations for the $1\pi_u$ state are in better agreement with experimental results, which could imply that the $1\pi_u$ wave-function used is better than the wave-function used for the $3\sigma_g$ state. The predicted features and magnitudes for all models are in poor agreement with the measured cross-sections. However, improvements in the calculations appear at higher energies which is consistent with the results discussed in section 8.3 and in [59].

Coplanar Doubly-Symmetric Experiments 100 eV above the $3\sigma_g$ IP

The results shown in fig. 8.14 are for coplanar doubly-symmetric experiments at 100 eV above the $3\sigma_g$ ionization potential. This additional experiment was performed to ascertain if the agreement between the theory and experiment improve at higher energies those described in this section and in section 8.3.

As mentioned, the incident electron energy was tuned to 100 eV the ionization potential for the $3\sigma_g$ state, and the electron energy analyzers were tuned with symmetric energies and scattering angles where $(E_1 = E_2 = 50 \text{ eV}; \theta_1 = \theta_2 = \theta)$. The electron gun was positioned at $\psi = 0^\circ$ and also at $\psi = 45^\circ$ with respect to the scattering plane. Similar normalization procedures as those mentioned in section 8.3 were adopted.

The results described here and from the experiments in section 8.3 show that the ionization cross-sections depend on the energy of the collisions and the state from which the ionization occurs. Subfigure (a) of fig. 8.14 shows the results and calculations for the $3\sigma_g$ state at 100 eV above the ionization potential; the agreement between the results and theory is much better in comparison to what was found at lower energies (as seen in section 8.3 and [59]). It is important to note that the best agreement is for cross-section measurements in the forward direction with those of the molecular three-body distorted
Figure 8.14: Results with incident electron energy 100 eV above the ionization potential for the $3\sigma_g$ state, in both (a) a coplanar doubly-symmetric geometry and (b) for the incident electron beam direction $k_0$ at an angle of 45° to the detection plane spanned by $k_1$ and $k_2$. A common normalization point exists between measurements when $\theta_1 = \theta_2 = 90°$. DWBA, M3DW and WM theories are shown which have been convoluted with the experimental angular resolution. The data are normalized to the M3DW calculation at the forward peak in a coplanar geometry, so that a comparison can be made between theory and experiment.

wave (M3DW) calculations. The cross-section measurements beyond $\theta = 90°$ do not agree with the any of the theoretical calculations.

In the geometry where $\psi = 45°$ as seen in subfigure (b) in fig. 8.14, the experimental results show two peaks and a plateau region:

- A decrease in the cross-section is seen with increasing scattering angles starting from the $35°$ to $75°$.
- The cross-sections show little structure in the region between scattering angles of $75°$ and $110°$.
- An increase in the cross-section measurements is seen for scattering angles higher than $110°$ (and up to $125°$).

The data for this geometry was normalized through the common point ($\theta_1 = \theta_2 = 90°$) to the coplanar data. The normalization points are seen in fig. 8.14 as dotted lines and circles.
All three models predict the two peaks and plateau region seen in the experimental results, with the exception of the predicted local minima which is deeper than those seen in the data. The plateau region is predicted to occur at \( \sim 60^\circ \) in comparison to the measured start of the plateau region which occurs at \( \sim 75^\circ \). The back-scattered cross-sections around 110\(^\circ\) show a closer agreement between theory and experiment, despite the underestimation in the cross-section calculations. The best comparison is seen from the distorted wave Born approximation (DWBA) for forward-scattered electrons. It is worth noting that back-scattered electrons require a strong interaction between them and the nuclei of the target, which implies that theory is underestimating the magnitude of the nuclear force in the interactions.

### 8.4.3 Discussion and Conclusions

The experimental and theoretical results for experiments at energies 20 and 40 eV above the ionization potential are seen to be in poor agreement with each other. The agreement of theory with experiment improved slightly as the energy is increased. However, and despite the thorough series of tests, the calculations predicted features and cross-section magnitudes not found in the experimental results. The measured and calculated results do not point to any definitive conclusions on the reasons behind these disagreements at the present time.

The measured and calculated results for experiments 100 eV above the 3\( \sigma_g \) ionization potential show that the ionization cross-sections depend on the energy of the collisions and the state from which ionization occurs. Compared to the work covered in section 8.3, better agreement is seen here between theory and experiment for the coplanar data for the 3\( \sigma_g \) state. The agreement is better with the molecular three-body distorted wave (M3DW) calculations for forward-scattered electrons. The calculated cross-sections at higher scattering angles predict a decrease in magnitude not seen in the experimental results. Similar results are seen when the incident electron beam is at \( \psi = 45^\circ \) with respect to the scattering plane; the agreement between theory and experiment is better for forward-scattered electrons than it is at higher scattering angles (back-scattered electrons).

A possibility for the disagreements between theory and experiment could be due to the averaging techniques (orientation averaged molecular orbital approximation) which average over all molecular orientations [123]. Several attempts were made to use a different (proper averaging) approach, which calculates the cross-sections for each molecular orientation, then averages the final result. However, the calculations using this proper averaging approach were found not to converge. The conclusion that can be drawn from these studies is that further work is required to indicate how the theoretical models might be improved.
The results discussed in this section, were published the Journal of Physics B: Atomic, Molecular and Optical Physics in 2018, which can be found in [60].

8.5 Conclusion

As discussed in section 8.3.3 and section 8.4.3, the theoretical calculations and the experimental results are in poor agreement. Despite the range of energies and kinematics used to test the models, only certain conditions (e.g. forward-scattered versus back-scattered electrons) showed an improvement in the theoretical predictions or in any of their agreements with experiment. No definitive conclusions were drawn on why the agreement between theory and experiment was so poor, except for the possible improvement that can be made by averaging correctly over the the orientation of molecules.
CHAPTER 9

Experimental (e,2e) Studies on Methane (\(CH_4\))

9.1 Introduction

Methane (\(CH_4\)) is the most common greenhouse gas emitted on earth from both human and animal activities, and is directly linked with global warming and climate change. The \(CH_4\) molecule is composed of a carbon atom at the center which shares 4 bonds with the hydrogen atoms, making the molecule the smallest hydrocarbon. The work discussed in this chapter studies the electron-impact ionization of methane (\(CH_4\)) at intermediate incident energies; an international collaboration which resulted in publication in the Journal of Chemical Physics [61].

The experiments and theoretical investigations were carried out on the ionization of the highest occupied molecular orbital (HOMO) \(1t_2\) and the next highest occupied molecular orbital (NHOMO) \(2a_1\) states of \(CH_4\) at an incident electron beam energy of 250 eV, for ejected electron energies of 50 eV and 30 eV. The fixed scattering angles (20°, 22.5°, 25°, 27.5°, and 30°) were used to perform the experiments detailed in this chapter. The measured triple differential cross-sections were compared to the experimental results of the Afyon group in Turkey. Both experimental results were also compared to theoretical calculations using the molecular 3-body distorted wave (M3DW) and generalized Sturmian function (GSF) models. In addition, both models calculated the theoretical triple differential cross-section (TDCS) for each molecular orientation; the M3DW model used the proper averaging (PA) approach for this additional step. Further details on these theoretical models can be found in chapter 2.

9.2 (e,2e) Studies on \(CH_4\) in Coplanar Geometry at Intermediate Energy

This section covers the experimental results from the work carried out on the ionization of the \(CH_4\) \(1t_2\) and \(2a_1\) orbitals (detailed in section 9.2.2), and a brief overview of the experimental results from the Afyon group (covered in section 9.2.1), including comparisons to theoretical calculations of both the molecular three-body distorted wave (M3DW) and generalized Sturmian function (GSF) models.
9.2.1 The Afyon Experiment and Results

The experiments carried out by the Afyon group used an electron-electron coincidence spectrometer, the details and applications of which can be found in [124–126]. The Afyon spectrometer is housed in a cylindrical stainless steel vacuum chamber, and is comprised of the following:

- An electron gun.
- Two hemispherical electron energy analyzers, each of which is equipped with a channel electron multiplier (CEM). The analyzers have an energy resolution of $\sim 0.6$ eV.
- Faraday cup.
- Gas jet.

During experimentation, the working pressure inside the vacuum chamber was maintained at $\sim 2 \times 10^{-6}$ torr. The electron gun was operated at a current of $\sim 1 \mu$A; it could produce a collimated beam (with a diameter of 2mm) with energies that could be varied between 10 eV and 450 eV. The electron gun and the gas jet were fixed in place, producing orthogonal beams that intersected at the collision center. The emerging (scattered and ejected) electrons were detected by the electron energy analyzers.

The Afyon experimental results and theoretical triple differential cross-section (TDCS) calculations are shown in fig. 9.1 and fig. 9.2. The results in both figures are for the ionization of the highest occupied molecular orbital (HOMO) state of $\text{CH}_4$, with an incident electron beam energy of 250 eV, and energies of 50 eV and 30 eV for the ejected outgoing electrons. The direction of the momentum transfer $+q$ and $-q$ are indicated by vertical arrows in both figures. The calculations and experimental data have been normalized to unity at the binary peak in each of the subfigures, because the experiment did not measure absolute cross-section values.

Figure 9.1 shows the results for $10^\circ$, $20^\circ$ [124], and $25^\circ$ fixed scattering angles, as well as the M3DW and GSF calculations, which are in good agreement with the experimental results. Upon comparison of each model with experiment, it is seen that:

- The binary peak (at $25^\circ$) predicted by the GSF calculation is symmetric about the momentum transfer direction.
- The peaks from experiment and the M3DW calculations for the scattering angles $10^\circ$ and $20^\circ$ are at an angle that is higher than that of the momentum transfer direction, which is possibly due to post collision interactions.
9.2 (e,2e) Studies on \( CH_4 \) in Coplanar Geometry at Intermediate Energy

Figure 9.1: Afyon’s results with incident electron energy of 250 eV for ionization of the \( 1t_2 \) orbital of \( CH_4 \), and 50 eV ejected electrons with different scattered electron angles of 10° in subfigure (a), 20° in subfigure (b), and 25° in subfigure (c). The Afyon group experimental results were normalized to and compared with the calculations of the GSF and M3DW models (in atomic units).

- The good agreement between experiment and the M3DW calculations is improved by the inclusion of the PCI terms (which are calculated to all orders of approximation).
- The small recoil peak at the scattering angle of 10° found in the experimental results agree reasonably well with the M3DW calculations.
Figure 9.2: The Afyon group results with an incident electron energy of 250 eV for ionization of the 1t_2 orbital of CH_4, and for 30 eV ejected electrons with different scattered electron angles (10° in subfigure (a), and 20° in subfigure (b)). The Afyon’s experimental results were normalized to and compared with the calculations of the GSF and M3DW models (in atomic units).

The results in fig. 9.2 are a comparison between the same models as above and measurements for 30 eV ejected electrons with fixed scattering angles of 10° and 20°. The experimental results are again in good agreement with both M3DW and GSF calculations. Upon comparison of each model with experiment, it is seen that:

- The binary peak predicted by the M3DW calculations found in the experimental results for \( \theta_a = 10^\circ \), are at higher scattering angles than that of the momentum transfer direction.
- The agreement of the M3DW calculations with the experimental results, are not as good for results with ejected electron energies of 50 eV.
- For the binary peak at \( \theta_a = 20^\circ \), the experimental results show that the amplitude of the smaller angle peak is larger than the amplitude of the "sister" peak. The magnitudes of the peaks are predicted to be equal by the GSF model, whereas the
higher angle peak is predicted to be larger by the M3DW calculations. The minimum for both ejected electron energies is close to the predicted momentum transfer direction. Overall, the agreement between theory and experiment for ejected electron energies of 50 eV and 30 eV is reasonably good. More details on the Afyon experimental procedures and results can be found in [61].

To gain a better understanding and additional experimental verification of the structure of the binary peak, a series of more detailed experiments were carried out using the Manchester (e,2e) coincidence spectrometer (as part of this Ph.D thesis). The experimental procedures and results of these experiments are detailed in the next section.

9.2.2 The Manchester Experiment and Results

The modernized (e,2e) apparatus was used to perform the series of experiments detailed below. The experimental procedures used for the operation, tuning, and optimization of the (e,2e) spectrometer, as well as the data acquisition and analysis procedures are detailed in chapter 7.

The following procedures were performed to prepare the (e,2e) apparatus for experimentation:

- Vacuum chamber: operating pressure of approximately $2 \times 10^{-5}$ torr, and a base pressure of $1 \times 10^{-7}$ torr.
- Incident electron beam current: $\approx 1 \mu$A was chosen, to obtain a good coincidence signal-to-noise ratio. This makes it easy to resolve the coincidence signal against the background noise.
- Faraday cup: a smaller Faraday cup was installed to allow the electron energy analyzers to reach the minimum forward angles of $\theta_{a,\text{min}} = 20^\circ$ and $\theta_{b,\text{min}} = 27.5^\circ$. The reason for this change was due to the limitations of the physical size of the electron gun, electron energy analyzers, and Faraday cup, which (in coplanar geometry) restricted the analyzers from reaching their minimum angles of $35^\circ$. The required higher incident energy of 250 eV (to perform this experiment) made it possible to use a smaller Faraday cup.

The gun and analyzer electrostatic voltages, incident beam current, and gas pressure were monitored, logged, and adjusted using the new (e,2e) LabVIEW computer application detailed in this thesis, to optimize the (e,2e) spectrometer and coincidence signal.

Since the results discussed in this section are on the ionization from the $1t_2$ and $2a_1$ molecular orbitals, energy binding spectral scans were performed over a range of incident
electron energies (from ~10 eV to 27 eV in steps of 0.125 eV) to identify the specific target orbitals. Figure 9.3 shows the results from one of these binding energy spectral scans; this scan clearly shows the 1t₂ and 2a₁ orbitals.

Figure 9.3: A binding energy spectral scan showing the well-resolved CH₄ 1t₂ and 2a₁ orbitals for analyzer angles θₐ = 30° and θₐ = 45°.

For the series of experiments discussed here, the electron energy analyzers were positioned in an asymmetric coplanar geometry, with:

- Analyzer (a) held at one of the fixed angles (20°, 22.5°, 25°, 27.5°, or 30°) to detect the high energy "scattered" electrons.
- Analyzer (b) swept around the detection plane from θₐ = 27.5° to 130° in 2.5° steps, to detect the low energy "ejected" electrons.

In addition to the analyzer and electron gun tuning conditions, the data were collected using several angular sweeps (up to 252 measurements) of the detection plane for each fixed angle θₐ. The (e,2e) coincidence measurements were obtained for 1000 s at each scattering angle. The tuning conditions for the lens and deflector elements for both electron energy analyzers were optimized for each new angle in any given angular sweep. The mean and standard errors were calculated for the data collected for each angle θₐ (in each set of fixed angle θₐ). The procedures and settings described above were used to carry out two experiments for the 1t₂ orbital for the outgoing electron energies of Eₗ = 50 eV and 30 eV, the results of which are shown in fig. 9.4 and fig. 9.5. An additional experiment was performed for the 2a₁ orbital with the fixed angle θₐ at 25° and for an outgoing electron energy Eₗ of 30 eV, the results of which are shown in fig. 9.6.

The experiments whose results are shown in fig. 9.4 and fig. 9.5 did not measure absolute
9.2 (e,2e) Studies on \( \text{CH}_4 \) in Coplanar Geometry at Intermediate Energy

Cross-sections and so the data were normalized to theory at a single point in the set, where the fixed angle was \( \theta_a = 20^\circ \) (since it was the strongest signal for both experiments). The data were then inter-normalized for different fixed angles \( \theta_a \).

Figure 9.4: Experimental results with incident electron energy of 250 eV for ionization of the \( 1\sigma_2 \) orbital of \( \text{CH}_4 \), and 50 eV ejected electrons with different scattered electron angles (20° in subfigure (a), 22.5° in subfigure (b), 25° in subfigure (c), 27.5° in subfigure (d), and 30° in subfigure (e)). The results were compared to the data from the Afyon group (in red) where possible, and to both the GSF and M3DW calculations.

The results in both figures include experimental data sets (highlighted in red) from the Afyon group where the kinematic conditions overlapped. Their data agree well with the data taken by the modernized Manchester (e,2e) coincidence spectrometer. The experimental
data sets (for the scattering angles 20° and 25°) for ejected electron energies of 50 eV (shown in fig. 9.4) are in excellent agreement with each other. The experimental data sets for 30 eV ejected electrons and the fixed angle \( \theta_a \) of 20°, agrees well on the location and amplitude of the higher angle peak, but not for the location of the forward-scattered angle peak. Overall, the experimental results are in good agreement with those from the Afyon group.

The predictions made by both theories, are consistent with the cross-section measurements found in the experimental results. The location and amplitude of the peaks found in the experimental results are in a good agreement with those from the M3DW calculations; the agreement is much better for the 50 eV ejected electrons than at 30 eV. Both experimental results show that the small angle peak is larger in intensity than that of the higher angle peak. The peak intensities predicted by the GSF calculations are symmetric and equal in magnitude; these are not consistent with the peak intensities found by experiment. A difference in the peak intensities is found in the M3DW calculation, but the higher angle peak is consistently overestimated.

The evolution of the two peaks was easily monitored due to the angular step size of 2.5° chosen for this series of experiments. The results for the 50 eV ejected electrons show the evolution of the structure from a single to a double peak between the angles of \( \theta_a = 20° \) and \( \theta_a = 25° \). As the scattering angle \( \theta_a \) increases, the higher angle peak decreases in magnitude relative to that of the small angle peak, until it is almost gone at \( \theta_a = 30° \). The evolution of the double peak structure with increasing scattering angles found by experiment is in good agreement with the M3DW predictions.

The experimental results seen in fig. 9.5 for the 30 eV ejected electrons, show that the magnitude of the higher angle peak is almost negligible by the angle \( \theta_a = 25° \), in contrast with the measurements for the 50 eV ejected electrons discussed above. The M3DW calculation shows that the higher angle peak is gone by \( \theta_a = 27.5° \). In addition, both models predict that the higher angle peak will disappear when the scattering angle reaches \( \theta_a = 30° \).

Both the M3DW and GSF calculations were convoluted using a Gaussian with a full width at half maximum (FWHM) that represents the angular acceptance angle of the apparatus (shown as dotted and dashed curves for the M3DW and GSF calculations, respectively). The reason for this additional step was to explore if the experimental angular resolution had an effect on the results, however as seen in the figures, this had little effect.

The experimental results (normalized to theory) shown in fig. 9.6 are for ionization from the \( 2a_1 \) orbital for 50 eV ejected electrons with \( \theta_a = 25° \). The error bars seen in the experimental data show the uncertainty in the spectrometer’s angular resolution (horizontal
Figure 9.5: Experimental results with incident electron energy of 250 eV for ionization of the 1\(\pi_2\) orbital of \(CH_4\), and 30 eV ejected electrons with different scattered electron angles (20° in subfigure (a), 22.5° in subfigure (b), 25° in subfigure (c), 27.5° in subfigure (d), and 30° in subfigure (e)). The results were compared to the data from the Afyon group (in red) where possible, and to both the GSF and M3DW calculations.
error bars), and the standard error of the mean of multiple measurements (vertical error bars). The location of the peak predicted by the GSF model is at a smaller angle, in contrast with the higher angle predicted by the M3DW model, however, both theoretical results are within the experimental uncertainty. No structure was seen by experiment and in theory, and since no additional information could be extracted under these conditions, no further measurements were made from the \(2a_1\) orbital.

9.3 Conclusion

This chapter covered the (e,2e) studies on the ionization of the \(CH_4\) \(1t_2\) and \(2a_1\) orbitals as published in [61], with an incident electron beam energy of 250 eV for ejected electron energies of 50 eV and 30 eV, at the fixed scattered electron angles of 20°, 22.5°, 25°, 27.5°, and 30°. The experimental measurements were compared to those from the Afyon group in Turkey, and the M3DW and GSF theoretical calculations. Overall, there is very good agreements between both experiment and theory. The structure and peak locations in the results for 50 eV ejected electrons are in good agreement with the theoretical calculations. However, the agreement at 30 eV was not as satisfactory. The M3DW calculations were slightly better in their predictions of the location and magnitude of the experimental peaks than the GSF model.

The evolution of the peak structure from a single peak (at \(\theta_a = 20^\circ\)) to a double peak (at \(\theta_a = 25^\circ\)) was seen for 50 eV ejected electrons. The higher angle peak decreased in magnitude and then almost disappeared at the scattering angle reached \(\theta_a = 30^\circ\). The evolution of the experimental double peak structure is in good agreement with the predictions made by both theoretical models. The experimental results for 30 eV ejected
electrons show the evolution of the peak structure from a double to a single peak with increasing scattering angles $\theta_a$. The results agree reasonably well with the theoretical predictions, with the exception of the magnitude of the higher angle peak which was found to decrease with increasing scattering angles, quicker than predicted by both theoretical models.

An additional experiment measured the triple differential cross-sections (TDCS) for ionization of the $2\alpha_1$ orbital for 50 eV ejected electrons and a fixed scattering angle of $\theta_a = 25^\circ$. The models predicted a single symmetric binary peak as seen in the experimental data, and so no additional measurements were taken due to the absence of any detailed structure found from the $2\alpha_1$ orbital.

Despite the many experimental measurements performed in the past [83, 84], this series of experiments is the first to clearly observe the double peak structure by electron impact single ionization of the $CH_4$ molecule. In addition to the publication (which can be found in [61]), the results were presented at the 20th International Symposium on Correlation, Polarization and Ionization in Atomic and Molecular Collisions (an official satellite meeting of the 31st 2019 ICPEAC) in Metz, France. Following from these experiments, further measurements have now been carried out for lower incident energies of 20 eV and 40 eV in both coplanar and non-coplanar geometries. These results have now been compared to the M3DW calculations from the Rolla group (Don Madison’s group in Missouri), and have now been submitted for publication.
CHAPTER 10

Conclusion and Future Work

10.1 Conclusions

The work presented in this thesis was divided into three main sections. The first is composed of two chapters which give an introduction to (e,2e) coincidence experiments and the theoretical frameworks used to describe (e,2e) reactions. The second section consists of five chapters which detail all components of the (e,2e) apparatus, including a comparison between the previous and modernized setup, as well as the experimental procedures for operating the new (e,2e) experimental hardware and software. The third section is comprised of two chapters which detail the experimental (e,2e) studies on both nitrogen ($\text{N}_2$) and methane ($\text{CH}_4$) molecules.

Chapter 1 introduced the kinematics of (e,2e) coincidence reactions by detailing the geometries and symmetries in which such experiments are carried out, and the kinematic constraints used to obtain triple differential cross-section measurements (TDCSs). The chapter then covered a brief history of different (e,2e) coincidence experiments carried out in the last fifty years; beginning with the first of those experiments by Ehrhardt et al. (Germany) [5] and Amaldi et al. (Italy) [10], through to various techniques and methods such as electron momentum spectroscopy developed by Weigold and McCarthy [7], and the Cold Target Recoil Ion Momentum Spectroscopy (COLTRIMS) technique developed in Frankfurt [8, 9]. This was then followed by an introduction to the Manchester (e,2e) coincidence spectrometer, and a brief overview of the experiments carried out using this apparatus since 1982 by Jones [18] through to the extensive studies and upgrades carried out by Murray and colleagues, examples of which can be seen in [21, 22, 25, 32, 37, 47, 48]. This was then followed by an overview of the new (e,2e) studies, and hardware and software developments carried out during the time period of this PhD. The introductory chapter concludes with a chapter-by-chapter layout of this thesis.

Chapter 2 gave an overview of electron collisions including the types of scattering found in atomic and molecular interactions, which includes elastic scattering, inelastic scattering, and the ionization process of atomic or molecular targets. Examples of such scattering
phenomena were detailed such as the gold foil experiment carried out by Rutherford in Manchester, which lead to the development of modern physics. The quantum processes and challenges of electron collisions were also discussed; quantum distortions to electron wavefronts, post collisional interactions (PCIs), multiple scattering, and polarization of the atomic or molecular target. The chapter then gave brief introductions to the theoretical models that were tested against the experimental results of (e,2e) studies on the nitrogen ($N_2$) and methane ($CH_4$) molecules discussed in chapter 8 and chapter 9 respectively. These include:

- The distorted wave Born approximation (DWBA).
- The molecular three-body distorted wave (M3DW) approximation.
- The orientation averaged molecular orbital (OAMO) approximation.
- The Ward–Macek (WM) approximation.
- The proper averaging (PA) approach.
- The Generalized Sturmian Functions (GSF) approach.

Chapter 3 gave an overview of the fundamental components making up the (e,2e) apparatus. These components are:

- The (e,2e) spectrometer and vacuum system;
- The experimental hardware;
- The experimental software;

as well as the experimental procedures for operating the (e,2e) coincidence experiment, all of which are detailed in chapters 4 to 7. This chapter gave a comparison between the previous and modernized (e,2e) apparatus setup, highlighted the modernized components of the (e,2e) apparatus, and the reasons for those modifications, replacements, and upgrades.

Chapter 4 gave an overview of the entire (e,2e) vacuum system, with details on the components and inner workings of the (e,2e) spectrometer’s electron gun, electron energy analyzers, Faraday cup, and gas jet. The details on the apparatus covered the vacuum chamber, the top flange of the vacuum system, the pumping system, and the outer and inner portions of the target gas line. In addition, the chapter also detailed the components of the (e,2e) spectrometer with descriptions and drawings of the non energy-selected electron gun and hemispherical electron energy analyzers. The channel electron multipliers and their pickup and low pass filter circuitry were also detailed. Lastly, the stepper motors used to move the electron energy analyzers were introduced, as well as details on the internal
interlocks’ system used for preventing any of the (e,2e) spectrometer components from running into each other.

Chapter 5 detailed all components, modules, and units making up the modernized experimental hardware. The chapter provided a set of instructions for making the printed circuit boards (PCBs). This was followed by detailed circuit designs and schematics used to build the (e,2e) spectrometer power supply units. The Arduino control electronics used to control the variable voltage supply units were discussed in detail. The details on the variable voltage supply boards were considered in two sections: the digital-to-analog electronics and the variable voltage electronics. The Faraday Cup voltage supply and the Faraday cup current monitor board were covered. The stepper motor control and data acquisition unit consisting of the Arduino stepper motor control board, the stepper motors, the L298N H-Bridge drivers used to drive the stepper motors, the stepper motor board power supply, and the PCI6221 LabVIEW DAQ and CB-68LP breakout board was discussed in detail. A brief overview was given on the hardware carried over from the previous control systems to the new setup; the constant current filament supply, the pulse detection electronics, and the channel electron multiplier EHT supplies. The coincidence timing pulse detection electronics was also explained in detail.

Chapter 6 detailed the new experimental applications, routines, and scripts used for experimental control, voltage supply control, communication, data exchange, extraction, analysis, and visualization. The three main sections in this chapter detailed the (e,2e) LabVIEW computer application, the programmable Arduino micro-controllers, and the use of Python and Jupyter notebooks for data analysis. The new (e,2e) LabVIEW computer application was covered in detail: the front panel event structure loop, the analyzer count rates loop, the voltage supply monitor loop, the analyzer position loop, and the main state handler loop. In addition, the communication between the LabVIEW program and the four Arduino MEGA 2560 micro-controllers, as well as the signals sent from the PCI-6221 data acquisition (DAQ) card to the (e,2e) LabVIEW computer application were also detailed. The programs and functions behind the Arduino MEGA 2560 micro-controllers for the variable voltage supply units and the stepper motor control board were covered in detail. The new (e,2e) Python data analysis library for data extraction, analysis, and visualization was also described here.

Chapter 7 was the last in a series of five chapters on the (e,2e) apparatus. This chapter detailed the experimental procedures used to prepare the vacuum system, operate and optimize the (e,2e) spectrometer, perform (e,2e) coincidence and rate scans, and analyze the data generated by the (e,2e) coincidence experiment. The preparation of the (e,2e) spectrometer and vacuum system cover the evacuation of the vacuum chamber and the
target gas line, initialization of the experimental hardware, introducing the target gas into
the system, and the tuning conditions for the (e,2e) spectrometer. This was followed by
the procedures for performing (e,2e) scans by detailing the (e,2e) LabVIEW computer
application’s front panel tab selector window along with the function(s) for each tab. This
was then followed by a detailed section on the extraction, analysis, and visualization of the
data collected by the (e,2e) LabVIEW computer application using imported functions from
the (e,2e) data analysis Python library into a Jupyter notebook environment. In addition,
procedures for obtaining cross-section measurements were also detailed in this section.

Chapter 8 introduced the nitrogen molecule, and then detailed the two published (e,2e)
coincidence studies that were carried out on molecular nitrogen [59, 60]. The chapter gave
a detailed introduction on molecular nitrogen; the electronic structure of nitrogen, the
molecular interactions between its atomic orbitals, the bonding types, the symmetries, the
states of \( \text{N}_2 \) as well as its ions, and the Morse potential energy curves.

This was followed by an (e,2e) study on molecular nitrogen with incident energies
above the ionization potential of the \( 3\sigma_g \), \( 1\pi_u \) and \( 2\sigma_u \) states for \( \text{N}_2 \). The experimental
results for this study were for outgoing electron energies where \( E_1 = E_2 = 4.6 \text{ eV} \pm 0.5 \text{ eV}, \)
\( E_1 = E_2 = 9.7 \text{ eV} \pm 0.5 \text{ eV}, \) and \( E_1 = 4.6 \text{ eV} \pm 0.5 \text{ eV} \) and \( E_2 = 14.5 \text{ eV} \pm 0.5 \text{ eV} \). These
results were compared to calculations using various distorted wave methods carried out
by Don Madison’s group from Missouri University of Science & Technology, USA. These
methods were:

- The molecular three-body distorted wave (M3DW) approximation.
- The distorted wave Born approximation (DWBA).
- The DWBA using the Ward–Macek (WM) approximation.

Additional results were presented for a set of experimental scans carried out at a higher
incident energy of \( \sim 40 \text{ eV} \), and over a range of scattering angles in all three geometries
(where \( \psi = 0^\circ, 45^\circ, \) and \( 90^\circ \)) which were all normalized to a single common point. This
additional set of spectral scans was carried out to ascertain the comparison between theory
and experiment. The experimental results on \( \text{N}_2 \) showed that the cross-sections are very
sensitive to the states from which the ionization occurs, as well as the sensitivity of post
collision interactions at these energies. It was seen in several cases that the molecular
three-body distorted wave (M3DW) calculations overestimated the effects of post collision
interactions, and how the Ward–Macek (WM) approximation was more accurate.

This was then followed by a second (e,2e) study where experimental and theoretical
ionization triple differential cross-sections were determined and compared with one another,
using only a coplanar geometry at higher energies than those in the first (e,2e) study, and for fixed angles for one of the outgoing electrons. The experiment was carried out with incident electron energies ~20 and ~40 eV above the ionization potential for the $3\sigma_g$ and $1\pi_u$ states for $N_2$, using equal energies for the outgoing electrons. The data were obtained with the incident electron beam in the scattering plane where $\psi = 0^\circ$. The results of this experiment were presented in six sets of measured ionization triple differential cross-sections, with fixed angles of $\theta_1(\theta_2)^{Fixed} = 45^\circ$, $90^\circ$, and $125^\circ$ with respect to the incident electron beam for one of the outgoing electrons. The experimental results were compared to Don Madison’s group calculations using the following various distorted wave methods:

- The distorted wave Born approximation (DWBA).
- The molecular three-body distorted wave (M3DW) approximation.
- The M3DW using the Ward–Macek (WM) approximation for post collision interactions (PCI).

The experimental and theoretical results for these experiments were seen to be in poor agreement with each other. The agreement of theory with experiment improved slightly as the energy is increased. However, despite the thorough series of tests, the calculations predicted features and cross-section magnitudes not found in the experimental results. The measured and calculated results do not point to any definitive conclusions on the reasons behind these disagreements at the present time.

Chapter 9 covered the electron-impact ionization studies on the methane ($CH_4$) molecule at intermediate incident energies, with comparisons to experimental results from another experimental group in Afyon, Turkey, and with the M3DW and GSF theoretical calculations of Don Madison’s group, and Ugo Ancarani’s group in France respectively. The chapter presented the experiments and theoretical investigations on the ionization of the highest occupied molecular orbital (HOMO) $1t_2$ and the next highest occupied molecular orbital (NHOMO) $2a_1$ states of $CH_4$ at an incident electron beam energy of 250 eV, for ejected electron energies of 50 eV and 30 eV. The fixed scattering angles ($\theta_1 = 20^\circ$, $22.5^\circ$, $25^\circ$, $27.5^\circ$, and $30^\circ$) were used to perform the experiments detailed in this chapter.

The results of this experiment showed very good agreement between both experiment and theory. The structure and peak locations in the results for 50 eV ejected electrons were in good agreement with the theoretical calculations. However, the agreement at 30 eV was not as satisfactory. The M3DW calculations were slightly better in their predictions of the location and magnitude of the experimental peaks than the GSF model. The work discussed in this chapter was published in the Journal of Chemical Physics [61].
10.2 Future Work

10.2.1 Future Experiments

The Manchester (e,2e) coincidence experiment was used to carry out electron impact ionization studies for this Ph.D. thesis. The experiment is currently funded by an EPSRC grant to receive further modifications that would allow for electron impact ionization studies from laser-excited atoms. The Manchester (e,2e) coincidence spectrometer will be adapted to allow for the delivery of laser radiation into the vacuum chamber and to the experiment. Several experiments have been proposed to test new theoretical models that were developed to test these interactions. In particular, the studies will be carried out for ionization of laser-excited $\text{Mg}$, $\text{Ca}$, and $\text{Sr}$ in both coplanar and non-coplanar geometries. The continuous wave (CW) lasers in the Photon Science Institute at the University of Manchester will provide long-term stable radiation required to carry out these experiments. The calculations will be carried out by the Missouri and Los Alamos groups in the USA, who will provide theoretical guidance and direction.

10.2.2 Standardization of Laboratory Hardware

The Atomic & Molecular Physics group which houses the (e,2e) spectrometers used in this body of work features several other experiments including an apparatus currently also configured to take (e,2e) measurements, a super-elastic scattering experiment featuring laser control and electron optics, and a cold atom trapping experiment used to study cold Rydberg atoms, also utilizing charged particle optics. Currently these experiments use a fragmented set of hardware and software to control all of their power and voltage supplies. There is a large overlap in required electronics, for example, all experiments require computer controlled power supplies for electron lenses and deflectors and all work in similar voltage ranges.

The lab could be modernized to use power supplies based on those detailed in this thesis so that all experiments then share one set of hardware and software designs. This would simplify the coding used by future PhD students and postdocs, and make it easier to diagnose and rapidly fix faulty units in future.

To aid with this modernization, and based on lessons learned during the development phase of this project, the power supplies would benefit from the following features:

- A single, consistent communications protocol based on opto-isolated asynchronous serial communications based on that developed in this project. This would aid and speed up future experimental development work.

- Modular circuit boards so that faulty supplies can be quickly swapped out and
diagnosed offline.

– Each board should have a single connector that either uses a socket such as one used in Eurocard based equipment or for example a PCI slot as used in a PC. This would minimize the chance of getting a faulty connection and allow for quick removal and replacement of the board.

– Bring the ADC voltage monitoring part on to each board so that they can operate independently. This would allow for faster feedback and settling of voltages and currents.

Such a design would greatly speed up the configuration of new experiments or even the reconfiguration of existing ones as requirements change.

10.3 Closing Remarks

This marks the end of this Ph.D. thesis. It took 91 days (432.5 effective hours) to write. The main matter of this body of work contains 44935 words (excluding captions, headers, sub-headers, and the front matter content). The back matter contains the bibliography and appendix sections which conclude this thesis.
Bibliography


### A Back Panel Interface Board Pin Assignment Tables

#### A.1 Electron Gun Power Supply Output Pin Assignment Tables

<table>
<thead>
<tr>
<th>Header Pin Group</th>
<th>Pin Voltage</th>
<th>Pin Color</th>
<th>Elements</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>+15 V</td>
<td>Red</td>
<td>Gun Coarse</td>
</tr>
<tr>
<td></td>
<td>$V_{Gun\ Fine}$</td>
<td>Green</td>
<td></td>
</tr>
<tr>
<td></td>
<td>−15 V</td>
<td>Blue</td>
<td></td>
</tr>
<tr>
<td></td>
<td>$V_{Gun\ Fine}$</td>
<td>Green</td>
<td></td>
</tr>
<tr>
<td></td>
<td>+5 V</td>
<td>Brown</td>
<td></td>
</tr>
<tr>
<td>B</td>
<td>+15 V</td>
<td>Red</td>
<td>GL1B GL2B</td>
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<tr>
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<td>Green</td>
<td>Gun Fine</td>
</tr>
<tr>
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<td></td>
</tr>
<tr>
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<td>0 V</td>
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<td></td>
</tr>
<tr>
<td></td>
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<td></td>
</tr>
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<td>−5 V</td>
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</tr>
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<td>0 V</td>
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<td></td>
<td>0 V</td>
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<td></td>
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<tr>
<td></td>
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</tr>
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<td>0 V</td>
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<td></td>
</tr>
<tr>
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**Table A.1:** Pin assignments for board (A) of the back panel of the electron gun power supply unit shown in fig. 5.8. The letters in the first column represent the header pins. The second column contains the assigned voltages. The third column contains the colors of the assigned voltage pins shown in fig. 5.8. The last column contains the assigned voltage supplies by element name.
<table>
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<th>Header Pin Group</th>
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<td></td>
</tr>
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<td>UNUSED</td>
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<td></td>
</tr>
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</tr>
<tr>
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</tr>
<tr>
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<td>Brown</td>
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<td>Blue</td>
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</tr>
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<tr>
<td>I</td>
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Table A.2: Pin assignments for board (B) of the back panel of the electron gun power supply unit shown in fig. 5.8. The letters in the first column represent the header pins. The second column contains the assigned voltages. The third column contain the colors of the assigned voltage pins shown in fig. 5.8. The last column contain the assigned voltage supplies by element name.
### Table A.3: Pin assignments for board (C) of the back panel of the electron gun power supply unit shown in fig. 5.8. The letters in the first column represent the header pins. The second column contains the assigned voltages. The third column contain the colors of the assigned voltage pins shown in fig. 5.8. The last column contain the assigned voltage supplies by element name.

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<td>Blue</td>
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<td></td>
<td>$V_{GL1C}$</td>
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<td></td>
</tr>
<tr>
<td></td>
<td>+5 V</td>
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</tr>
<tr>
<td>K</td>
<td>+15 V</td>
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<td></td>
</tr>
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</tr>
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</tr>
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## A.2 Analyzer Power Supply Output Pin Assignment Table

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<td>−15 V</td>
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<td></td>
<td>$V_{Analyzer\ Mean}$</td>
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</tr>
<tr>
<td></td>
<td>+5 V</td>
<td>Brown</td>
<td></td>
</tr>
<tr>
<td>B</td>
<td>+5 V</td>
<td>Red</td>
<td>Arduino Control</td>
</tr>
<tr>
<td></td>
<td>0 V</td>
<td>Green</td>
<td></td>
</tr>
<tr>
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<td>+15 V</td>
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<td>AM</td>
</tr>
<tr>
<td></td>
<td>0 V</td>
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<td>ARSE</td>
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<td></td>
<td>+30 V</td>
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<tr>
<td>E</td>
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**Table A.4:** Pin assignments for boards (A) and (B) of the back panel of the electron energy analyzers' power supply unit shown in fig. 5.10. The letters in the first column represent the header pins. The second column contains the assigned voltages. The third column contain the colors of the assigned voltage pins shown in fig. 5.10. The last column contain the assigned voltage supplies by element name.
### A.3 Electron Gun Variable Voltage Supply Input Pin Assignment Tables

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<td>$V_{\text{Gun Fine}}$</td>
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</tr>
<tr>
<td></td>
<td>+5 V</td>
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<tr>
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<tr>
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<td>0 V</td>
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</tr>
<tr>
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<td>+12 V</td>
<td>Red</td>
<td>Grid</td>
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<td></td>
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<td>Green</td>
<td></td>
</tr>
<tr>
<td></td>
<td>−12 V</td>
<td>Blue</td>
<td></td>
</tr>
<tr>
<td></td>
<td>0 V</td>
<td>Green</td>
<td></td>
</tr>
<tr>
<td></td>
<td>+5 V</td>
<td>Brown</td>
<td></td>
</tr>
<tr>
<td>D</td>
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<td>Brown</td>
<td>Gun Fine</td>
</tr>
<tr>
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<td></td>
</tr>
<tr>
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<td>Yellow</td>
<td></td>
</tr>
<tr>
<td>E</td>
<td>+15 V</td>
<td>Red</td>
<td>Gun Fine</td>
</tr>
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<td>Green</td>
<td></td>
</tr>
<tr>
<td></td>
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<tr>
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<td>Green</td>
<td></td>
</tr>
<tr>
<td></td>
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<td>Brown</td>
<td></td>
</tr>
<tr>
<td>F</td>
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<td>Red</td>
<td>GL1B GL2B</td>
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**Table A.5:** Pin assignments for board (A) of the back panel of the electron gun variable voltage supply unit shown in fig. 5.14. The letters in the first column represent the header pins. The second column contains the assigned voltages. The third column contains the colors of the assigned voltage pins shown in fig. 5.14. The last column contains the assigned voltage supplies by element name.
Table A.6: Pin assignments for board (B) of the back panel of the electron gun variable voltage supply unit shown in fig. 5.14. The letters in the first column represent the header pins. The second column contains the assigned voltages. The third column contains the colors of the assigned voltage pins shown in fig. 5.14. The last column contains the assigned voltage supplies by element name.

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<tr>
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<th>Pin Voltage</th>
<th>Pin Color</th>
<th>Elements</th>
</tr>
</thead>
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</tr>
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<td></td>
</tr>
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<td></td>
<td>−15 V</td>
<td>Blue</td>
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</tr>
<tr>
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<td></td>
<td>−15 V</td>
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<tr>
<td>J</td>
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Table A.7: Pin assignments for board (C) of the back panel of the electron gun variable voltage supply unit shown in fig. 5.14. The letters in the first column represent the header pins. The second column contains the assigned voltages. The third column contains the colors of the assigned voltage pins shown in fig. 5.14. The last column contains the assigned voltage supplies by element name.

<table>
<thead>
<tr>
<th>Header Pin Group</th>
<th>Pin Voltage</th>
<th>Pin Color</th>
<th>Elements</th>
</tr>
</thead>
<tbody>
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<td>Red</td>
<td>GD2X/Y</td>
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<td>$V_{GL1C}$</td>
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<td>GD3X/Y</td>
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<tr>
<td></td>
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<td>Blue</td>
<td></td>
</tr>
<tr>
<td></td>
<td>$V_{GL1C}$</td>
<td>Green</td>
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<td></td>
<td>+5V</td>
<td>Brown</td>
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<td>Anode</td>
</tr>
<tr>
<td></td>
<td>0V</td>
<td>Green</td>
<td>GL1C</td>
</tr>
<tr>
<td></td>
<td>−5V</td>
<td>Yellow</td>
<td></td>
</tr>
<tr>
<td>O</td>
<td>+15V</td>
<td>Red</td>
<td></td>
</tr>
<tr>
<td></td>
<td>0V</td>
<td>Green</td>
<td></td>
</tr>
<tr>
<td></td>
<td>−15V</td>
<td>Blue</td>
<td></td>
</tr>
<tr>
<td></td>
<td>0V</td>
<td>Green</td>
<td></td>
</tr>
<tr>
<td></td>
<td>+5V</td>
<td>Brown</td>
<td></td>
</tr>
</tbody>
</table>
### A.4 Analyzer Variable Voltage Supply Input Pin Assignment Table

<table>
<thead>
<tr>
<th>Header Pin Group</th>
<th>Pin Voltage</th>
<th>Pin Color</th>
<th>Elements</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>+5 V</td>
<td>Red</td>
<td>Arduino Control</td>
</tr>
<tr>
<td></td>
<td>0 V</td>
<td>Green</td>
<td></td>
</tr>
<tr>
<td>B</td>
<td>+5 V</td>
<td>Red</td>
<td>ADC</td>
</tr>
<tr>
<td></td>
<td>0 V</td>
<td>Green</td>
<td></td>
</tr>
<tr>
<td>C</td>
<td>+15 V</td>
<td>Red</td>
<td>ADX/Y</td>
</tr>
<tr>
<td></td>
<td>$V_{Analyzer Mean}$</td>
<td>Green</td>
<td></td>
</tr>
<tr>
<td></td>
<td>−15 V</td>
<td>Blue</td>
<td></td>
</tr>
<tr>
<td></td>
<td>$V_{Analyzer Mean}$</td>
<td>Green</td>
<td></td>
</tr>
<tr>
<td></td>
<td>+5 V</td>
<td>Brown</td>
<td></td>
</tr>
<tr>
<td>D</td>
<td>+30 V</td>
<td>Red</td>
<td>AM</td>
</tr>
<tr>
<td></td>
<td>0 V</td>
<td>Green</td>
<td></td>
</tr>
<tr>
<td></td>
<td>−5 V</td>
<td>Yellow</td>
<td></td>
</tr>
<tr>
<td>F</td>
<td>+30 V</td>
<td>Red</td>
<td>AOH</td>
</tr>
<tr>
<td></td>
<td>0 V</td>
<td>Green</td>
<td></td>
</tr>
<tr>
<td></td>
<td>−5 V</td>
<td>Yellow</td>
<td></td>
</tr>
<tr>
<td>H</td>
<td>+60 V</td>
<td>Red</td>
<td>AIH</td>
</tr>
<tr>
<td></td>
<td>0 V</td>
<td>Green</td>
<td></td>
</tr>
<tr>
<td></td>
<td>−5 V</td>
<td>Yellow</td>
<td></td>
</tr>
<tr>
<td>J</td>
<td>+120 V</td>
<td>Red</td>
<td>ARSEAL1B</td>
</tr>
<tr>
<td></td>
<td>0 V</td>
<td>Green</td>
<td></td>
</tr>
<tr>
<td></td>
<td>−5 V</td>
<td>Yellow</td>
<td></td>
</tr>
<tr>
<td>E, G, I, K</td>
<td>+15 V</td>
<td>Red</td>
<td>AM (E)</td>
</tr>
<tr>
<td></td>
<td>0 V</td>
<td>Green</td>
<td>AOH (G)</td>
</tr>
<tr>
<td></td>
<td>−15 V</td>
<td>Blue</td>
<td>AIH (I)</td>
</tr>
<tr>
<td></td>
<td>0 V</td>
<td>Green</td>
<td>ARSE (K)</td>
</tr>
<tr>
<td></td>
<td>+5 V</td>
<td>Brown</td>
<td>AL1B (K)</td>
</tr>
</tbody>
</table>

*Table A.8*: Pin assignments for boards (A) and (B) of the back panel of the electron energy analyzers’ variable voltage supply unit shown in fig. 5.16. The letters in the first column represent the header pins. The second column contains the assigned voltages. The third column contain the colors of the assigned voltage pins shown in fig. 5.16. The last column contain the assigned voltage supplies by element name.
### Table A.9: Pin assignments for boards (D), (E), and (F) of the back panel of the electron gun variable voltage supply unit shown in fig. 5.34. The first column correspond to the board name (D, E, or F), and its D-sub adaptor type (male or female). The letters in the second column represent the header pin groups. The third column contains the assigned voltages. The fourth column contains the pin assignments by element name. The last column contains the board’s assigned D-sub adaptor pin number (refer to first column for gender of D-sub adaptor).

<table>
<thead>
<tr>
<th>Board (D-sub)</th>
<th>Pin Group</th>
<th>Pin Voltage</th>
<th>Element</th>
<th>D25 Pin</th>
</tr>
</thead>
<tbody>
<tr>
<td>D (Female)</td>
<td>A</td>
<td>0 V to +30 V</td>
<td>Gun Energy Fine</td>
<td>NA</td>
</tr>
<tr>
<td></td>
<td>B</td>
<td>0 V to +30 V</td>
<td>Gun Energy Fine</td>
<td>NA</td>
</tr>
<tr>
<td></td>
<td>C</td>
<td>0 V to +500 V</td>
<td>Gun Energy Coarse</td>
<td>NA</td>
</tr>
<tr>
<td></td>
<td>D</td>
<td>0 V to +120 V</td>
<td>GLIC (Bottom Pin)</td>
<td>2, 5, 8, 10</td>
</tr>
<tr>
<td></td>
<td></td>
<td>0 V to +120 V</td>
<td>Anode (Top Pin)</td>
<td>4</td>
</tr>
<tr>
<td>E (Female)</td>
<td>E</td>
<td>0 V to +380 V</td>
<td>To Current Monitor</td>
<td>23</td>
</tr>
<tr>
<td></td>
<td>F</td>
<td>0 V to +380 V</td>
<td>Faraday Cup</td>
<td>23</td>
</tr>
<tr>
<td></td>
<td>G</td>
<td>0 V to +30 V</td>
<td>Gun Energy Fine</td>
<td>NA</td>
</tr>
<tr>
<td></td>
<td>H</td>
<td>0 V to +30 V</td>
<td>Gun Energy Fine</td>
<td>NA</td>
</tr>
<tr>
<td></td>
<td>I</td>
<td>0 V to +500 V</td>
<td>Gun Energy Coarse</td>
<td>3, 5, 6, 10-19, 21, 22, 24, 25</td>
</tr>
<tr>
<td></td>
<td>J</td>
<td>NA</td>
<td>Filament Supply</td>
<td>1, 2</td>
</tr>
<tr>
<td></td>
<td>K</td>
<td>NA</td>
<td>UNUSED</td>
<td>7, 8</td>
</tr>
<tr>
<td>F (Male)</td>
<td>L</td>
<td>0 V to +1 kV</td>
<td>GLIC Inline Current</td>
<td>21</td>
</tr>
<tr>
<td></td>
<td>M</td>
<td>0 V to +1 kV</td>
<td>GLIC (Bottom Pin)</td>
<td>21</td>
</tr>
<tr>
<td></td>
<td></td>
<td>0 V to +1 kV</td>
<td>GLIC (Top Pin)</td>
<td>19</td>
</tr>
<tr>
<td></td>
<td>N</td>
<td>0 V to +1 kV</td>
<td>GLIC Inline Current</td>
<td>19</td>
</tr>
<tr>
<td></td>
<td>O</td>
<td>0 V to −12 V</td>
<td>Grid Inline Current</td>
<td>22</td>
</tr>
<tr>
<td></td>
<td>P</td>
<td>0 V to −12 V</td>
<td>Grid</td>
<td>22</td>
</tr>
<tr>
<td></td>
<td>Q</td>
<td>±15 V to ±15 V</td>
<td>GDIX+, X-, Y+, Y-</td>
<td>1, 2, 3, 4</td>
</tr>
<tr>
<td></td>
<td>R</td>
<td>±15 V to ±15 V</td>
<td>GDIY+, X-, Y+, Y-</td>
<td>5, 6, 7, 8</td>
</tr>
<tr>
<td></td>
<td>S</td>
<td>±15 V to ±15 V</td>
<td>GDIY+, X-, Y+, Y-</td>
<td>9, 10, 11, 12</td>
</tr>
<tr>
<td></td>
<td>T</td>
<td>0 V to +120 V</td>
<td>GLIC (Right Pin)</td>
<td>NA</td>
</tr>
<tr>
<td></td>
<td></td>
<td>0 V to +120 V</td>
<td>Anode (Left Pin)</td>
<td>NA</td>
</tr>
<tr>
<td></td>
<td>U</td>
<td>0 V to +30 V</td>
<td>Gun Energy Fine</td>
<td>NA</td>
</tr>
<tr>
<td></td>
<td>V</td>
<td>0 V to +120 V</td>
<td>Anode Inline Current</td>
<td>NA</td>
</tr>
<tr>
<td></td>
<td>W</td>
<td>0 V to +120 V</td>
<td>GLIC Inline Current</td>
<td>NA</td>
</tr>
<tr>
<td></td>
<td>X</td>
<td>0 V to +120 V</td>
<td>GLIC (Right Pin)</td>
<td>NA</td>
</tr>
<tr>
<td></td>
<td></td>
<td>0 V to +120 V</td>
<td>Anode (Left Pin)</td>
<td>NA</td>
</tr>
</tbody>
</table>

**Table A.9:** Pin assignments for boards (D), (E), and (F) of the back panel of the electron gun variable voltage supply unit shown in fig. 5.34. The first column correspond to the board name (D, E, or F), and its D-sub adaptor type (male or female). The letters in the second column represent the header pin groups. The third column contains the assigned voltages. The fourth column contains the pin assignments by element name. The last column contains the board’s assigned D-sub adaptor pin number (refer to first column for gender of D-sub adaptor).
### A.6 Analyzer Variable Voltage Supply Output Pin Assignment Table

<table>
<thead>
<tr>
<th>Board (D-sub)</th>
<th>Pin Group</th>
<th>Pin Voltage</th>
<th>Element</th>
<th>D25 Pin</th>
</tr>
</thead>
<tbody>
<tr>
<td>C (Female)</td>
<td>A</td>
<td>0 V to +30 V</td>
<td>A1OH</td>
<td>5</td>
</tr>
<tr>
<td></td>
<td>B</td>
<td>0 V to +60 V</td>
<td>A1IH</td>
<td>4</td>
</tr>
<tr>
<td></td>
<td>C</td>
<td>0 V to +30 V</td>
<td>A1AM</td>
<td>3</td>
</tr>
<tr>
<td></td>
<td>D</td>
<td>0 V to +120 V</td>
<td>A1L1B (Top Pin)</td>
<td>1</td>
</tr>
<tr>
<td></td>
<td>E</td>
<td>±15 V to ±15 V</td>
<td>A1DX+, X-, Y+, Y-</td>
<td>9, 10, 11, 12</td>
</tr>
<tr>
<td>D (Male)</td>
<td>F</td>
<td>±15 V to ±15 V</td>
<td>A2DX+, X-, Y+, Y-</td>
<td>14, 15, 16, 5</td>
</tr>
<tr>
<td></td>
<td>G</td>
<td>0 V to +120 V</td>
<td>A2L1B</td>
<td>13</td>
</tr>
<tr>
<td></td>
<td>H</td>
<td>0 V to +30 V</td>
<td>A2OH</td>
<td>8</td>
</tr>
<tr>
<td></td>
<td>I</td>
<td>0 V to +60 V</td>
<td>A2AM</td>
<td>11</td>
</tr>
<tr>
<td></td>
<td>J</td>
<td>0 V to +30 V</td>
<td>A2IH</td>
<td>10</td>
</tr>
<tr>
<td></td>
<td>K</td>
<td>0 V to +120 V</td>
<td>A2RSE</td>
<td>1</td>
</tr>
</tbody>
</table>

**Table A.10:** Pin assignments for boards (C) and (D) of the back panel of the electron energy analyzers’ variable voltage supply unit shown in fig. 5.36. The first column correspond to the board name (C or D), and its D-sub adaptor type (male or female). The letters in the second column represent the header pin groups. The third column contains the assigned voltages. The fourth column contains the pin assignments by element name. The last column contains the board’s assigned D-sub adaptor pin number (refer to first column for gender of D-sub adaptor).