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Time-resolved buildup of two-slit-type interference from a single atom

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A photoelectron forced to pass through two atomic energy levels before receding from the residual ion shows interference fringes in its angular distribution as a manifestation of a two-slit-type interference experiment in wave-vector space. This scenario was experimentally realized by irradiating a rubidium atom by two low-intensity continuous-wave lasers [Pursehouse *et al.*, *Phys. Rev. Lett.* **122**, 053204 (2019)]. In a one-photon process, the first laser excites the $5p$ level whereas the second uncorrelated photon elevates the excited population to the continuum. This same continuum state can also be reached when the second laser excites the $6p$ state, and the first photon then triggers the ionization. As the two lasers are weak and their relative phases uncorrelated, the coherence needed for generating the interference stems from the atom itself. Increasing the intensity or shortening the laser pulses enhances the probability that two photons from both lasers act at the same time, and hence, the coherence properties of the applied lasers are expected to affect the interference fringes. Here, this aspect is investigated in detail, and it is shown how tuning the temporal shapes of the laser pulses allows for tracing the time dependence of the interference fringes. We also study the influence of applying a third laser field with a random amplitude, resulting in a random fluctuation of one of the ionization amplitudes and discuss how the interference fringes are affected.

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I. INTRODUCTION

In a typical double-slit experiment, interference fringes are formed on a screen placed behind the slits which are then traversed by particles of suitable wavelengths. By blocking one of the slits, the spatial interference pattern disappears. In a recent wave-vector space double-slit experiment [1], a photoelectron wave packet receding from a single atom is forced to pass through two energy levels within the atom so that the levels play the role of the double-slit. As illustrated in Fig. 1, this is achieved by two low-intensity continuous-wave lasers which excite the $5p$ and $6p$ states of a rubidium (Rb) atom. The infrared light field can concurrently ionize the $6p$ state, and the blue laser can ionize the $5p$ state. We image the interference in wave-vector space by scanning the photoelectron angular distribution. The interference pattern disappears if only one state is excited, demonstrating that the phase relationship between the interfering waves is imprinted by the atom. The two lasers are not phase locked.

The $5p$ and $6p$ states thus represent slits, which can be closed by detuning the respective laser fields, leading to nonresonant excitation and damped occupation of the specific intermediate state. This damping is, however, different from thermal damping as it does not swiftly destroy the coherence. The interference in this case is affected because the two interfering amplitudes have largely different strengths.

In the experiment, the interference term and the associated difference in the phases of the amplitudes were recorded by essentially three measurements. First, both laser fields were set resonant to the $5p$ and $6p$ dipolar excitations leading to an ionization amplitude $t_1 + t_2$ [cf. Fig. 1(a)]. In the second and third measurements, one of the two laser fields was detuned to close one of the ionization pathways [cf. Fig. 1(b)] to extract

the individual amplitudes $t_1(t_2)$. The ground-state energy of the $5s$ Rb state is -4.177 eV whereas $E_{6p} = -1.589$ eV and $E_{5p} = -2.950$ eV. Thus, no other states in the bound spectrum of Rb are accessible by single-photon processes. An interesting point is that in the experiment the two continuous-wave (cw) lasers (cf. Fig. 1) were not phase locked and were weak. The calculations show that a random phase between these two lasers does not affect the interference. Thus the observed interference has to stem from the atom, whereas the photoelectron wave is propagating out to the detectors at infinity.

The question we are considering here is how the interference behaves when the number of photons (the laser intensity) increases, and hence one would expect an increase in the probability that a blue and red photon are absorbed at the same time. The same process applies when we consider much shorter pulses. In this case, one would expect the phase relation between the laser pulses to be important, and it then becomes possible to access the timescale on which the interference pattern builds up and evolves. Unfortunately, due to experimental limitations, we are currently not in a position to investigate these ideas in the laboratory, and therefore, the current paper is mostly theoretical.

In the final sections of this paper, we discuss mechanisms for controlling and manipulating the interference phenomena between both ionization pathways. Atomic units are used throughout the paper.

II. PROPAGATION ON A SPACE-TIME GRID

Within the single-particle picture the rubidium atom is well described by the angular-dependent model potential

FIG. 1. (a) Schematic of the atomic double-slit experiment in rubidium. The slits are represented by the bound and initially unoccupied $5p$ and $6p$ intermediate states which are resonantly excited by the infrared and blue laser pulses. The two lasers subsequently ionize the intermediate levels, giving rise to two ionization pathways t_1 and t_2 . The photoelectron is transferred into the continuum with a finite kinetic-energy E_f without knowing which pathway was taken. (b) Closing one slit by detuning one of the two lasers which then inhibits the occupation of the respective intermediate state (in this example, the $6p$ state). We hence obtain a conventional two-color photoionization process via a single-ionization pathway. The energy level of the final state is now shifted down due to the detuning, effectively decreasing $\varepsilon_f = \hbar\omega_{BL} + \hbar\omega_{IR} + \varepsilon_{5s}$.

introduced by Marinescu *et al.* [2],

$$V_\ell(r) = -\frac{Z_\ell(r)}{r} - \frac{\alpha_c}{2r^4} [1 - e^{-(r/r_c)^6}], \quad (1)$$

where α_c is the static dipole polarizability of the positive-ion core and the effective radial charge $Z_\ell(r)$ is given by

$$Z_\ell(r) = 1 + (z - 1)e^{-a_1 r} - r(a_3 + a_4 r)e^{-a_2 r}, \quad (2)$$

with the nuclear charge z and the cutoff parameter r_c as well as the parameters $a_1 - a_4$ fitted to experimental values. The optimized parameters are tabulated in Ref. [2].

On a very fine space grid ($\Delta r = 0.005$ arb. units) the radial wave functions of the atomic eigenstates $\langle \mathbf{r} | \phi_i \rangle = R_{n_i, \ell_i}(r) Y_{\ell_i, m_i}(\Omega_r)$ are found by (numerical) diagonalization of the matrix corresponding to the time-independent Hamiltonian $\hat{H}_0 = -(1/2)\partial_r^2 + \ell(\ell+1)/(2r^2) + V_\ell(r)$. In the presence of solenoidal and moderately intense electromagnetic fields, the light-matter interaction Hamiltonian is given by $\hat{H}_{\text{int}}(t) = -\mathbf{A}(\mathbf{r}, t) \cdot \hat{\mathbf{p}}$ where $\mathbf{A}(\mathbf{r}, t)$ is the vector potential and $\hat{\mathbf{p}}$ is the momentum operator.

To account for all multiphoton and multipole effects, a numerical propagation of the ground-state wave function in the external vector potential is necessary, e.g., by the matrix iteration scheme [3]. Exploiting the spherical symmetry of the atomic system, the time-dependent wave function is decomposed in spherical harmonics, i.e., $\Psi(\mathbf{r}, t) = \sum_{\ell, m}^{\ell_{\text{max}}} b_{\ell, m}(r, t) Y_{\ell, m}(\Omega_r)$. We, therefore, have to propagate $(\ell_{\text{max}} + 1)^2$ channel functions $b_{\ell, m}(r, t)$ which are coupled through the corresponding (dipole) matrix elements $\langle \ell' m' | \mathbf{A} \cdot \hat{\mathbf{p}} | \ell m \rangle$. Initially, the ground-state channel is fully occupied by the $5s$ rubidium orbital, i.e., $\Psi(\mathbf{r}, t \rightarrow -\infty) = \langle \mathbf{r} | \phi_{5s} \rangle$ meaning $b_{0,0}(r, t \rightarrow -\infty) = R_{n_i=5, \ell_i=0}(r)$. Introducing a time T_{obs} where the external electro-magnetic-field perturbation is off, the wave-function $\Psi(\mathbf{r}, t)$ is propagated to a time

$t > T_{\text{obs}}$ where the photoelectron wave packet is fully formed. The radial grid is extended to 10^4 a.u. to avoid nonphysical reflections at the boundaries. Additionally, we implemented absorbing boundary conditions by using an imaginary potential. The resulting simulation shows that the electron density at the final grid point r_N is then smaller than the numerical error at the considered propagation times.

To obtain the scattering properties of the liberated electron, we project $\Psi(\mathbf{r}, T_{\text{obs}})$ onto a set of continuum wave functions which are given by the partial-wave decomposition,

$$\langle \mathbf{r} | \varphi_{\mathbf{k}}^{(-)} \rangle = \sum_{\ell, m} i^\ell R_{k\ell}(r) e^{-i\delta_\ell(k)} Y_{\ell, m}^*(\Omega_{\mathbf{k}}) Y_{\ell, m}(\Omega_{\mathbf{r}}). \quad (3)$$

Here, the kinetic energy is defined by $E_k = k^2/2$, $\delta_\ell(k)$ are the scattering phases, and $R_{k\ell}(r)$ are radial wave functions satisfying the stationary radial Schrödinger equation for positive energies in the same pseudopotential $V_\ell(r)$ which is used for obtaining the bound spectrum. The scattering phases $\delta_\ell(k) = \arg[\Gamma(1 + \ell - i/k)] + \eta_\ell(k)$ consist of the well-known Coulomb phases (first term) and phase $\eta_\ell(k)$ characterizing the atomic-specific short-ranged deviation from the Coulomb potential. The radial wave functions are normalized to $\langle R_{k\ell} | R_{k'\ell} \rangle = \delta(E_k - E_{k'})$. Finally, the projection coefficients are given by

$$a_{\ell, m}(k) = e^{i(E_k T_{\text{obs}} + \delta_\ell(k) - \ell\pi/2)} \int_{r > r_a} dr b_{\ell, m}(r, T_{\text{obs}}) R_{k\ell}(r). \quad (4)$$

Here, we introduce the core radius r_a and ensure that the integration region is outside the residual ion. The photoionization probability [differential cross section (DCS) in the following] is defined as:

$$\begin{aligned} \text{DCS} &= \frac{d\sigma}{d\Omega_{\mathbf{k}}} (E_k, \Omega_{\mathbf{k}}) \\ &\propto \sum_{\ell, \ell'} \sum_{m, m'} a_{\ell', m'}^*(k) a_{\ell, m}(k) Y_{\ell', m'}^*(\Omega_{\mathbf{k}}) Y_{\ell, m}(\Omega_{\mathbf{k}}). \end{aligned} \quad (5)$$

whereas the total cross section is $\sigma(E_k) \propto \sum_{\ell, m} \sigma_{\ell, m}(E_k)$ with $\sigma_{\ell, m}(E_k) = |a_{\ell, m}(k)|^2$. Note that this treatment gives explicit insight into the population of the individual angular channels and their contributions to the photoelectron wave packet.

In the following two-color ionization of rubidium, the electric fields of both pulses are modeled according to:

$$E_p(t) = \epsilon_p \mathcal{E}_p \Omega(t + \Delta_p) \cos[\omega_p(t + \Delta_p) + \phi_p], \quad (6)$$

with $p = \text{IR, BL}$ standing for the infrared and blue laser pulses, respectively. The polarization vectors are ϵ_p , and the temporal envelope is given by $\Omega(t) = \cos^2(\pi t/T_d^p)$ for $t \in [-T_d^p/2, T_d^p/2]$ with the pulse duration $T_d^p = 2\pi n_p/\omega_p$ determined by the number of optical cycles n_p . Furthermore, we introduce a temporal difference Δ_p and a phase difference ϕ_p to account for both laser fields originating from different sources and which are, hence, not phase locked. Without loss of generality, we set $\phi_{\text{IR}} = 0$ and $\Delta_{\text{IR}} = 0$. Both laser fields are assumed to be linearly polarized in the z direction so that the azimuthal angular quantum number m is conserved. The number of angular channels then reduces to $\ell_{\text{max}} + 1$, and in the following treatment, we omit the subscript m for brevity. To balance the differences in the oscillator strengths between the $5s \rightarrow 5p$ and the $5s \rightarrow 6p$ channels we used the field

FIG. 2. Occupation numbers of the $5p$ and $6p$ intermediate states after laser excitation, which depend on the time delay Δ and the phase difference between both pulses. (a) shows the results of the numerical propagation for six optical cycles. (b) corresponds to ten optical cycles. Dots indicate the results for $\phi = 0$ radians and crosses belong to $\phi = 2\pi/3$.

strengths $\mathcal{E}_{\text{BL}} = 0.05$ and $\mathcal{E}_{\text{IR}} = 0.007$ a.u. in the following simulations.

The aim here is to determine the influence of the phase differences on the resulting photoionization and occupation probabilities. In Fig. 2, we present the occupation numbers of the intermediate $5p$ and $6p$ states for two different laser configurations at a time T_{obs} after laser excitation, meaning both light pulses are completely extinguished and the photoelectron wave packet propagates freely in the Coulomb field of the residual rubidium ion. Panel (a) corresponds to the case where both laser pulses have a length of six optical cycles. We see that both occupation numbers show a strong dependence on the temporal difference Δ as well as on a random-phase difference ϕ . The dots belong to $\phi = 0$ whereas the crosses indicate the results for a nonzero phase difference. Here, we show the occupation numbers for $\phi = 2\pi/3$, which show a large difference from the case of $\phi = 0$. In addition, we repeated the simulation for other numbers of the random-phase difference and obtained similarly pronounced discrepancies. The situation changes completely when increasing the number of optical cycles as shown in panel (b). For ten optical cycles, the influence of both the temporal and the phase differences on the resulting occupation numbers is drastically reduced, pointing to the transition into the *cw* limit.

Already, from the *bounded* properties, we suspect the rather fast convergence of the photoionization process into a description within the frequency domain, which is characterized by infinitely long laser pulses characterized by δ distributionlike bandwidths. We can underline this observation by looking at the characteristics of the ejected photoelectron wave packet. In Fig. 3(a), we present the ionization probability of the angular channel $\ell = 2$ characterized by the partial cross-section $\sigma_2 = |a_{\ell=2}(k)|^2$. As expected, the probability curve sharpens under an increase in the pulse lengths of both laser fields. Whereas, for $n_p = 6$, a manifold of energy states in the continuum is excited, from $n_p = 10$ upwards, we see clearly the unfolding of a Gaussian-like peak around the final energy $E_f = E_{5s} + \hbar\omega_{\text{IR}} + \hbar\omega_{\text{BL}}$. In the case of 35 optical cycles, the FWHM of the probability peak is around 0.07 eV.

It is also interesting to study the evolution of the quantum phase associated with the photoelectron wave packet, which can be expressed as $\varphi(k, \vartheta_k) = \arg[\sum_{\ell} a_{\ell}(k) Y_{\ell,0}(\Omega_k)]$. For

FIG. 3. Properties of the ejected photoelectron. (a) Ionization probability of the angular channel $\ell = 2$ for different pulse lengths of the incident IR and blue laser fields. (b) Angular variation of the averaged quantum phase $\bar{\varphi}(\vartheta_k)$ in dependence on the number of optical cycles.

six optical cycles, the angular channels with $\ell = 0$ and $\ell = 2$ already represent the dominant contributions to the photoelectron at the final energy around 0.4 eV as expected for a two-color photoionization process of an initial *s* state. The quantum phase is a result of interference between both partial waves, and for the long pulse limit it may be mathematically expressed by [4]

$$\varphi(k, \vartheta_k) = \arctan \left[\frac{\sum_{\ell=0,2} S_{\ell}(k, \vartheta_k) \sin[\phi_{\ell}(k)]}{\sum_{\ell=0,2} S_{\ell}(k, \vartheta_k) \cos[\phi_{\ell}(k)]} \right], \quad (7)$$

with $S_{\ell}(k, \vartheta_k) = |a_{\ell}(k)|^2 |Y_{\ell,0}(\Omega_k)|^2$ and $\phi_{\ell}(k) = \arg[a_{\ell}(k)] \simeq \delta_{\ell}(k) - \ell\pi/2$. The quantum phase, hence, depends crucially on the scattering phases $\delta_{\ell}(k)$ and on the ratio between the transition strengths into $\ell = 2$ and $\ell = 0$ angular channels, respectively. We note that, in principle, $\varphi(k, \vartheta_k)$ is an experimentally accessible quantity since it can be recovered by integrating the Wigner time delay in photoionization defined as $\tau_W(E_k, \vartheta_k) = (d/dE_k)\varphi(k, \vartheta_k)$. This can be extracted from delay measurements that are possible due to recent experimental advances within the attosecond time frame [5,6]. The measured atomic time-delay τ_a consists of an “intrinsic” contribution (Wigner time-delay τ_W) upon the absorption of an XUV photon which can be interpreted as the group delay of the outgoing photoelectron wave packet due to the collision process. As mentioned above, it contains information about the internal quantum phase. A second term τ_{cc} arises from continuum-continuum transitions due to the interaction of the laser probe field with the Coulomb potential and depends crucially on the experimental parameters. Hence, the difference $\tau_W = \tau_a - \tau_{\text{cc}}$ provides access to the phase information $\varphi(k, \vartheta_k)$.

In Fig. 3(b), it is shown how the phase develops by increasing the pulse lengths (number of optical cycles). Here, we show the phase averaged over the ionization probability [total cross section $\sigma(E_k)$]: $\bar{\varphi}(\vartheta_k) = \int dE_k \sigma(E_k) \varphi(k, \vartheta_k) / \int dE_k \sigma(E_k)$. Interestingly, for very short pulses ($n_p = 6$) where the cross section is far from being centered around a final energy $E_f = E_{5s} + \hbar\omega_{\text{IR}} + \hbar\omega_{\text{BL}}$, the shape of the phase matches that extracted from simulations with longer pulses. As anticipated from earlier results, from ten cycles upwards, the results converge quickly into the *cw* limit. As an example, this is seen since the discrepancy

between the quantum phase for 12 and 35 optical cycles is smaller than 5%.

III. FROM SHORT PULSES TO THE CW LIMIT

By considering the two-color ionization process using perturbation theory, we express the time-dependent wave function as $\Psi(\mathbf{r}, t) = \sum_{v\ell} d_{v\ell}(t) e^{-iE_{v\ell}t} \phi_{v\ell}(\mathbf{r})$. Note that the quantum number v includes both the bound and the continuum states. The first-order amplitude is given by

$$d_{0 \rightarrow f}^{(1)}(t) = -\frac{1}{i} \sum_{\lambda=\pm 1} [\langle f|D_{\text{IR}}|n\rangle \mathcal{E}_{\text{IR}} F_{\omega_{0f}}^{(1)}(t, \lambda\omega_{\text{IR}}, \Delta = 0) + e^{i\lambda\Delta\omega_{\text{BL}}} \langle f|D_{\text{BL}}|n\rangle \mathcal{E}_{\text{BL}} F_{\omega_{0f}}^{(1)}(t, \lambda\omega_{\text{BL}}, \lambda\Delta)], \quad (8)$$

where $D_i = \hat{\epsilon}_i \cdot \hat{\mathbf{d}}$ ($i = \text{IR}, \text{BL}$) is the dipole operator, $\omega_{0f} = E_f - E_0$ and

$$F_{\omega_{0f}}^{(1)}(t, \omega, \Delta) = \int_{-\infty}^t \Omega(t' + \Delta) e^{i(\omega_{0f} + \omega)t'} dt'. \quad (9)$$

Without loss of generality, we assume both laser fields are described by the same temporal function $\Omega(t) = \cos^2[\pi t/T_p]$ so that both pulses have the same pulse length $T_p = 2n_p\pi/\omega_{\text{IR}}$. In the following discussion, the number of optical cycles n_p , hence, refers to the infrared laser field. For the squared-cosine-shaped envelope $\Omega(t)$ of the pulses, the function $F_{\omega_{0f}}^{(1)}(t, \omega, \Delta)$ can be obtained analytically and converges against $F_{\omega_{0f}}^{(1)}(t, \omega, \Delta) \rightarrow \delta(\omega_0 - \omega)$ for $T_p \rightarrow \infty$. The second-order amplitude yields the following expression:

$$d_{0 \rightarrow f}^{(2)}(t) = -\sum_{i,j} \sum_{\lambda, \lambda'=\pm 1} \sum_n e^{i(\lambda\Delta_j\omega_j + \lambda'\Delta_i\omega_i)} \mathcal{E}_j \mathcal{E}_i \times \frac{\langle f|D_j|n\rangle \langle n|D_i|0\rangle}{\omega_{0n} - \omega_i} \times F_{\omega_{nf}, \omega_{0n}}^{(2)}(t, \lambda\omega_j, \lambda'\omega_i, \lambda\Delta_j, \lambda'\Delta_i), \quad (10)$$

where again $i, j = \text{IR}, \text{BL}$. The second-order temporal function is defined as

$$F_{\omega_{nf}, \omega_{0n}}^{(2)}(t, \omega_j, \omega_i, \Delta_j, \Delta_i) = (\omega_{0n} - \omega_i) \times \int_{-\infty}^t dt' \Omega(t' + \Delta_j) e^{i(\omega_{nf} + \omega_j)t'} F_{\omega_{0n}}^{(1)}(t', \omega_i, \Delta_i). \quad (11)$$

A closed expression for the second-order temporal function $F^{(2)}$ cannot be obtained analytically. However, a solution for $t > T_p/2$ (time of switch off) can be found and investigated for $T_p \rightarrow \infty$ (the continuous-wave limit). It follows that, in the many optical cycle limit, we find energy conservation, i.e.,

$$\lim_{n_p \rightarrow \infty} F_{\omega_{nf}, \omega_{0n}}^{(2)}(t, -\omega_j, -\omega_i, \Delta_j, \Delta_i) = \frac{3\pi}{4} \delta_{\omega_{0f}, \mathcal{E}_i + \mathcal{E}_j} (\omega_{0f} - \omega_j - \omega_i), \quad (12)$$

where δ is the Kronecker-Delta. Furthermore, in this limit, the individual temporal shifts Δ_j and Δ_i have no influence on the (second-order) transition amplitude. Thus, in the long (cw) pulse limit and for energies around $E_f = E_0 + \hbar\omega_{0f}$, this behavior allows us to rewrite the resulting second-order

transition amplitude,

$$d_{0 \rightarrow f}^{(2)} \xrightarrow{n_p \rightarrow \infty} \frac{3\pi}{4} \mathcal{E}_{\text{IR}} \mathcal{E}_{\text{BL}} e^{i\Delta\omega_{\text{BL}}} \sum_n \left[\frac{\langle f|D_{\text{IR}}|n\rangle \langle n|D_{\text{BL}}|0\rangle}{\omega_{n0} - \omega_{\text{BL}}} + \frac{\langle f|D_{\text{BL}}|n\rangle \langle n|D_{\text{IR}}|0\rangle}{\omega_{n0} - \omega_{\text{IR}}} \right], \quad (13)$$

which is similar to the traditional form of the two-photon matrix element [7]. From here we see directly that a random-phase $\phi = \Delta\omega_{\text{BL}}$ does not play any role (especially when analyzing the cross-sections $\sim |d_{0 \rightarrow f}^{(2)}|^2$).

Let us now come back to the two-color ionization process in the rubidium atom. It is crucial for the first- and second-order amplitudes to precisely evaluate the dipole matrix elements [8],

$$\langle f|D_i|n\rangle = (-1)^{m_f + \ell_n} \begin{pmatrix} \ell_f & 1 & \ell_n \\ -m_f & 0 & m_n \end{pmatrix} \sqrt{\ell_n} \langle f||\hat{\mathbf{d}}||n\rangle, \quad (14)$$

where $\ell_n = \max(\ell_f, \ell_n)$ and the reduced radial matrix element $d_{\ell_f \ell_n} = \langle f||\hat{\mathbf{d}}||n\rangle = \int_0^\infty dr R_f(r) D_i R_n(r)$. One way to account, at least, partially, for the interaction between the valence and the core electrons [9] is to modify the operator $\hat{Q}_L$ as

$$\hat{Q}_L \rightarrow \hat{Q}_L \left[1 - \frac{a_c^{(L)}}{r^{2L+1}} (1 - e^{-(r/r_c)^{2L+1}}) \right], \quad (15)$$

where $a_c^{(L)}$ is the 2^L tensor core polarizability and r_c' is an empirical cutoff radius (for Rb $r_c' = 4.339773$ a.u. [2]). The physical picture behind the corrections is roughly that the valence electron with a dipole moment $\mathbf{d}$ induces (by virtue of its field) a (core) dipole moment $-\alpha_c \mathbf{d}/r^3$ (and higher multipoles). Then, the complete dipole moment becomes $\mathbf{d}(1 - \alpha_c/r^3)$. Note that, in our case, the dipole operator $\hat{\mathbf{d}} = \hat{Q}_1$. Using the modified dipole operator delivers very accurate matrix elements near the ionization threshold in comparison with experiments and more-sophisticated theoretical models [10].

We learn from Eq. (13) that changing the electric-field amplitudes $\mathcal{E}_{\text{IR}}$ and $\mathcal{E}_{\text{BL}}$ will not balance any differences between the matrix element products $\langle f|D_{\text{IR}}|n\rangle \langle n|D_{\text{BL}}|0\rangle$ and $\langle f|D_{\text{BL}}|n\rangle \langle n|D_{\text{IR}}|0\rangle$. Thus, to reach equipollent ionization pathways $E_{5s} \rightarrow E_{5p} \rightarrow E_f$ and $E_{5s} \rightarrow E_{6p} \rightarrow E_f$, we have to detune the laser frequency corresponding to the stronger bound-bound transition. Given the reduced matrix elements $\langle 5p||\hat{\mathbf{d}}||5s\rangle = -5.158$ and $\langle 6p||\hat{\mathbf{d}}||5s\rangle = 0.468$, we have to detune the infrared field as shown in Fig. 4(a). The curves are obtained from a numerical integration of Eq. (10). For $n = 35$ optical cycles, a detuning of $\delta\omega_{\text{IR}} = +0.15$ eV is required to allow the second-order transition amplitudes within each pathway to have the same magnitudes. Note that the value of $\Delta\omega_{\text{IR}}$ decreases by increasing the pulse lengths. In this vein, we performed the simulation for 75 optical cycles and found that a detuning of only 0.07 eV is needed to reach equipollent ionization pathways (not shown for brevity).

Let us consider the time-dependent first-order probability $\delta\omega_{\text{IR}} = +0.15$ eV which reads explicitly,

$$P_{0 \rightarrow f}^{(0)}(t) = |\langle f|D_{\text{IR}}|n\rangle \mathcal{E}_{\text{IR}} F_{\omega_{0f}}^{(1)}(t, -\omega_{\text{IR}}, 0)|^2 + |\langle f|D_{\text{BL}}|n\rangle \mathcal{E}_{\text{BL}} F_{\omega_{0f}}^{(1)}(t, -\omega_{\text{BL}}, \Delta_{\text{BL}})|^2$$

FIG. 4. (a) Comparison between photoionization pathways via $5p$ and $6p$ intermediate states in the case of detuning $\delta\omega_{\text{IR}} = 0.15$ eV of the infrared field. The curves are extracted from Eq. (10). (b) Time dependence of the first-order product $\Re\{[F_{\omega_0}^{(1)}(t, -\omega_{\text{IR}}, 0)]^* F_{\omega_0}^{(1)}(t, -\omega_{\text{BL}}, \Delta_{\text{BL}})\}$ for different final energies. (c) Time-dependent occupation numbers $c_{5p}(t)$ and $c_{6p}(t)$ extracted from perturbation theory and a full numerical treatment. The gray curve indicates the temporal variation of the IR laser field.

$$+ 2\mathcal{E}_{\text{IR}}\mathcal{E}_{\text{BL}}\langle f|D_{\text{IR}}|n\rangle\langle f|D_{\text{BL}}|n\rangle \\ \times \Re\{F_{\omega_0}^{(1)}(t, -\omega_{\text{IR}})[F_{\omega_0}^{(1)}(t, -\omega_{\text{BL}}, \Delta_{\text{BL}})]^*\}. \quad (16)$$

Here, we have to emphasize that all terms containing $+\omega_i$ are negligibly small which we refer to as the rotating-wave approximation. The last term in the third line might look like a two-photon process, but a closer inspection of the function $F_{\omega_0}^{(1)}(t, -\omega, \Delta)$ reveals that it sharply peaks around $\omega_0 - \omega = 0$ even for times close to $T_p/2$. Since ω_{f0} is fixed, the product between both $F^{(1)}$ functions is zero so that

$$\lim_{T_p \rightarrow \infty} F_{\omega_0}^{(1)}(T_p/2, -\omega_{\text{IR}}, 0)[F_{\omega_0}^{(1)}(T_p/2, -\omega_{\text{BL}}, \Delta_{\text{BL}})]^* \\ = \delta(\omega_{f0} - \omega_{\text{BL}})\delta(\omega_{f0} - \omega_{\text{BL}}). \quad (17)$$

As confirmation, in Fig. 4(b), we show the time-dependent product of the functions $F_{\omega_0}^{(1)}(t, -\omega_{\text{IR}}, 0)$ and $F_{\omega_0}^{(1)}(t, -\omega_{\text{BL}}, \Delta_{\text{BL}})$ for different values of ω_{f0} . As stated above, all situations have in common that the product is zero at the time when the pulse is switched off, representing energy conservation. Finally, the first-order transitions for $t = T_p/2$ are given by

$$d_{0 \rightarrow f}^{(1)}(t > T_p/2) = -\frac{1}{2i}[\mathcal{E}_{\text{IR}}\langle f|D_{\text{IR}}|0\rangle\delta(\omega_{f0} - \omega_{\text{IR}}) \\ + e^{i\Delta_{\text{BL}}}\mathcal{E}_{\text{BL}}\langle f|D_{\text{BL}}|0\rangle\delta(\omega_{f0} - \omega_{\text{BL}})]. \quad (18)$$

The energies of the blue and red photons are not sufficiently high to reach the continuum, and only the two-photon matrix element developed in Eq. (10) gives insight into the properties of the photoelectron.

As a consequence, the amplitude $d^{(1)}$ describes the photoexcitation process of the intermediate $f = 5p$ and $f = 6p$ states. However, due to the sharp laser pulses, the same final-state f cannot be excited by both photons. Hence, in the resulting photoexcitation probabilities $c_f = |d_{0 \rightarrow f}^{(1)}(t > T_p/2)|^2$, the random-phase $\phi = \Delta_{\text{BL}}$ does not play a role, which confirms the full-numerical results shown in Fig. 2(b). In panel 4(c), we present the occupation numbers $c_{5p}(t) = |\langle \phi_{5p}|\Psi(t)\rangle|^2$ and $c_{6p}(t) = |\langle \phi_{6p}|\Psi(t)\rangle|^2$ extracted from the perturbative treatment (PT) in Eq. (8) and the numerical simulation scheme developed in Sec. II. We find a remarkable agreement between

the PT results and the full-numerical treatment, demonstrating the transition into the cw limit and the validity of the perturbative treatment of the two-color ionization problem. Due to the detuning of the IR field, the $c_{5s}(t)$ term decreases at the end of the pulse whereas the $c_{6p}(t)$ term belongs to resonant excitation ($5s \rightarrow 6p$) in the blue laser field. This is the reason for the nearly monotonous increase that is seen. We note that the corresponding Rabi frequencies Ω_{5s-5p} and Ω_{5s-6p} are very small which means the occupation numbers in Fig. 4(c) represent only the first segment of the first Rabi cycle which can be identified by the characteristic quadratic dependence on the time.

IV. INTERFERENCE BETWEEN BOTH IONIZATION PATHWAYS

As revealed by the two-photon matrix element in Eq. (13), both laser fields act simultaneously to produce the same final photoelectron state, and so we have to deal with two transition amplitudes t_1 and t_2 which are presented by the two terms. As already demonstrated in Sec. II, the final state $|f\rangle$ is mainly described by a superposition of two angular channels $\ell = 0$ and $\ell = 2$. Thus, we may write

$$d_{0 \rightarrow f}^{(2)}(\vartheta_{\mathbf{k}}) = t_1(\vartheta_{\mathbf{k}}) + t_2(\vartheta_{\mathbf{k}}) \\ = \underbrace{S_{\ell=0}^{(t_1)}(\vartheta_{\mathbf{k}})e^{i\phi_{\ell=0}(k_f)} + S_{\ell=2}^{(t_1)}(\vartheta_{\mathbf{k}})e^{i\phi_{\ell=2}(k_f)}}_{t_1=|t_1|e^{i\varphi_1}} \\ + \underbrace{S_{\ell=0}^{(t_2)}(\vartheta_{\mathbf{k}})e^{i\phi_{\ell=0}(k_f)} + S_{\ell=2}^{(t_2)}(\vartheta_{\mathbf{k}})e^{i\phi_{\ell=2}(k_f)}}_{t_2=|t_2|e^{i\varphi_2}}. \quad (19)$$

The final state is in the continuum and is defined by Eq. (3). Consequently, we can define

$$S_{\ell_f}^{(t_i)}(\vartheta_{\mathbf{k}}) = (-1)^{2+\ell_f/2}\sqrt{1+\ell_f/2}\begin{pmatrix} \ell_f & 1 & 1 \\ 0 & 0 & 0 \end{pmatrix}\begin{pmatrix} 1 & 1 & 0 \\ 0 & 0 & 0 \end{pmatrix} \\ \times \sum_n \left[\frac{\langle E_f \ell_f || \hat{d} || n \rangle \langle n || \hat{d} || 5s \rangle}{\omega_{0n} - \omega_1} \right] \\ \times \mathcal{E}_{\text{IR}}\mathcal{E}_{\text{BL}}F_{\omega_{nf}, \omega_{0n}}^{(2)}(t \rightarrow \infty, -\omega_2, -\omega_1, 0, 0) \\ \times Y_{\ell_f, 0}(\vartheta_{\mathbf{k}}). \quad (20)$$

FIG. 5. (a) DCS when the blue laser field is off-resonance thereby effectively closing the second ionization pathway. Comparison between two theoretical models (PT:red and propagation:gray) and experimental results. For the propagation, the blue field was detuned by $\delta\omega_{\text{BL}} = 0.13$ eV. Furthermore, the infrared field was slightly detuned by $\delta\omega_{\text{IR}} = 0.04$ eV to manipulate the ratio between both two-photon amplitudes (see discussions in Sec. III). (b) The same as in (a) with the infrared field completely off-resonance, effectively closing the first ionization pathway. The detuning amount is $\delta\omega_{\text{IR}} = 0.14$ eV. (c) Both laser fields resonant with associated pathways. To achieve pathway amplitudes that are comparable with experiment, the infrared field was slightly detuned by $\delta\omega_{\text{IR}} = 0.04$ eV. The inset shows a polar plot of the DCS. All theoretical curves are scaled by the same factor and are shifted by a constant offset of 0.1 to aid comparison with the experimental data. The data points are reproduced from Ref. [1].

Here, without loss of generality, we set the time-delays $\Delta_{\text{IR/BL}}$ to zero since we are in the *cw* limit. For pathway t_1 , $\omega_1 = \omega_{\text{IR}}$ and $\omega_2 = \omega_{\text{BL}}$, whereas, for pathway t_2 , the opposite is required. Furthermore, $\phi_\ell(k) = \delta_\ell(k) - \ell\pi/2$ whereas the corresponding pathway phases φ_i are already defined in Eq. (7). Note that $|t_1(\vartheta)|^2$ and $|t_2(\vartheta)|^2$ define the DCS for the individual pathways whereas the interference term between both pathways is given by

$$\begin{aligned} \text{DCS}_{\text{interf.}} &= t_1(\vartheta_{\mathbf{k}})t_2^*(\vartheta_{\mathbf{k}}) + t_2(\vartheta_{\mathbf{k}})t_1^*(\vartheta_{\mathbf{k}}) \\ &= |t_1(\vartheta_{\mathbf{k}}) + t_2(\vartheta_{\mathbf{k}})|^2 - [|t_1(\vartheta_{\mathbf{k}})|^2 + |t_2(\vartheta_{\mathbf{k}})|^2]. \end{aligned} \quad (21)$$

The *phase difference* related to $\text{DCS}_{\text{interf.}}$ is given by

$$\Delta\varphi_{12}(\vartheta_{\mathbf{k}}) = \cos^{-1} \left[\frac{t_1(\vartheta_{\mathbf{k}})t_2^*(\vartheta_{\mathbf{k}}) + t_2(\vartheta_{\mathbf{k}})t_1^*(\vartheta_{\mathbf{k}})}{2|t_1(\vartheta_{\mathbf{k}})||t_2(\vartheta_{\mathbf{k}})|} \right]. \quad (22)$$

The interference term is the result of differences in the ratios $S_{\ell=2}^{(t_1)}/S_{\ell=0}^{(t_1)}$ and $S_{\ell=2}^{(t_2)}/S_{\ell=0}^{(t_2)}$ between the *s*- and the *d*-partial waves associated with the individual pathways t_1 and t_2 . These ratios depend crucially on the reduced bound-continuum dipole matrix elements $\langle E_f, \ell || \hat{d} || 5p \rangle$ and $\langle E_f, \ell || \hat{d} || 6p \rangle$ as well as the bound-bound dipole matrix elements $\langle 5p || \hat{d} || 5s \rangle$ and $\langle 6p || \hat{d} || 5s \rangle$.

The perturbative treatment of the two-pathway ionization process shares some parallels with the theoretical description of the recently developed attosecond measurement techniques [11]. For instance, the occurrence and spectral characteristics of the $2q$ th sideband in the reconstruction of attosecond beating by interference of two-photon transitions scheme [12] stem from the interference between two ionization pathways: the absorption of harmonic H_{2q-1} or H_{2q+1} plus absorption or emission of a laser photon with $\hbar\omega$. Hence, similar to the effects studied in this paper, the measured intensity of the sideband depends on the phase difference between the

quantum paths. In contrast to typical attosecond experiments, we create a bound wave packet upon absorption of the first photon. This case was studied in photoionization of potassium where the spectral properties of the initially created bound wave packet was used to eliminate the influence of the dipole phase in the angle-integrated photoelectron spectrum making it possible to fully characterize the attosecond pulses [13]. Similar to the investigated experiment by Pursehouse *et al.* [1], the underlying physical principle is the quantum interference of pathways corresponding to ionization from different energy levels. Moreover, a realistic many-body treatment revealed that correlation effects have only a minor influence in alkali atoms which supports our theoretical treatment in this paper.

In Fig. 5, we present the individual: (a) t_1 DCS, (b) t_2 DCS, and (c) the DCS corresponding to the coherent summation $t_1 + t_2$. In all panels, we compare the ionization probabilities extracted from the full-numerical and perturbative treatment with experimental results from measurements performed by Pursehouse *et al.* [1]. In the experiment, the individual pathway cross sections are obtained by the appropriate detuning of the respective laser fields: To extract $|t_1|^2$ ($5s \rightarrow 5p \rightarrow E_f$), the blue laser beam is detuned to block occupation of the $6p$ state. To obtain the $|t_2|^2$ amplitude ($5s \rightarrow 6p \rightarrow E_f$), the infrared laser field is tuned to be off-resonant to the $5p$ transition. To obtain the total amplitude $|t_1 + t_2|^2$ both laser pulses are on-resonance to the respective $5s \rightarrow np$ transitions. As explained in Sec. III, the infrared laser field is always slightly detuned by a fixed $\delta\omega_{\text{IR}}$ so that both two-photon amplitude t_1 and t_2 are of similar magnitude.

In panel 5(a), we show the DCS of the individual pathway 1. For the full-numerical treatment with a number of 70 optical cycles, we used a blue detuning of $\delta\omega_{\text{BL}} = 0.13$ eV whereas the red field detuning amounts to $\delta\omega_{\text{IR}} = 0.04$ eV. In the perturbative treatment ($n_p \rightarrow \infty$), we need a much smaller detuning (in the range of meV). Both theoretical models agree

FIG. 6. (a) Interference term $DCS_{\text{interf.}}$ for two theoretical models and experimental data. Parameters are the same as in Fig. 5. (b) Phase difference φ_{12} between both ionization pathway amplitudes $t_1(\vartheta_k)$ and $t_2(\vartheta_k)$. The black line presents a fit to the experimental data to the symmetry-adapted function $f(\vartheta_k) = \sum_{n=0}^2 a_{2n} \cos^{2n}(\vartheta_k)$. The data points are reproduced from Ref. [1].

extraordinary well with the experiment in general, whereas minor discrepancies can be found around the maxima. A possible explanation is the shift of the final energy by nearly 0.17 eV in the full-numerical propagation (finite number of optical cycles) due to the detuning of both fields which changes slightly the ratios $S_{\ell=2}^{(t_1)}/S_{\ell=0}^{(t_1)}$. In comparison with the experiment, the theoretical models predict the correct shape of the DCS. They underestimate the data around $\vartheta_k = 90^\circ$ and $\vartheta_k = 270^\circ$, however, agree well at $\vartheta_k = 0^\circ$ and $\vartheta_k = 180^\circ$ (along the polarization vectors). The same observations apply for the DCS of the second ionization pathway $|t_2|^2$ in panel 5(b). Figure 5(c) shows the photoionization probability $|t_1 + t_2|^2$ when both laser fields are set to resonance with the respective $5s \rightarrow np$ transitions. Here, the infrared field is again detuned by a small $\delta\omega_{\text{IR}}$ so that both amplitudes t_1 and t_2 have the same magnitude. In addition, we show here the result for rather short laser pulses with 12 optical cycles. Surprisingly, the DCS extracted from the short-pulse calculations agree very well with the smaller maxima around $\vartheta_k = 90^\circ$ and $\vartheta_k = 270^\circ$. However, this rather accidental agreement must be considered within the experimental uncertainties.

In Fig. 6(a), we present the interference term $DCS_{\text{interf.}}$ for the two developed theoretical models and the experimental data. We see that the amplitude of the interference term is clearly nonzero and varies from 13% to 55% of the normalized signal shown in Fig. 5(c). In Fig. 6(b), we present the corresponding phase difference $\varphi_{12}(\vartheta_k)$ between the two-photon transition pathways t_1 and t_2 . In comparison to all amplitudes, the agreement is less satisfactory which can be explained

FIG. 7. (a) Interference cross section and (b) phase difference $\varphi_{12}(\vartheta_k)$ for different intermediate-state pairs extracted from the two-photon matrix element in Eq. (13). The IR field is detuned to reach equipollent pathway strengths. The numbers in panel (a) represent the ratio $\nu = (d_{\ell=2}^{(1)}/d_{\ell=0}^{(1)}) : (d_{\ell=2}^{(2)}/d_{\ell=0}^{(2)})$.

by the relatively large uncertainties due to error propagation through the arccos function [1]. Interestingly, the average value of the phase shift is accurately reproduced by both calculations. Under these conditions, the predicted angular variation is not very pronounced and ranges from 110° to 122° . Surprisingly, the models do not agree as well as for the DCS in Fig. 5, which points to the extreme sensitivity of the quantum phase to small changes in the transition matrix elements. The black curve represents a fit of the experimentally obtained phase difference to the symmetry-adapted function $\sum_{n=0}^2 a_{2n} \cos^{2n}(\vartheta_k)$. It highlights the agreement with the theoretical prediction with respect to the general shape.

In the next section, we will present mechanisms to manipulate, decrease, and increase this modulation.

V. MANIPULATION OF THE QUANTUM INTERFERENCE

A. Role of the energy gap

In Fig. 7, we present interference studies on different intermediate-state pairs defining the two-color ionization amplitudes t_1 and t_2 . The blue and the red curves present the interference between $5p/7p$ ($\Delta E = 1.88$ eV) and $5p/8p$ ($\Delta E = 2.13$ eV) states which have an increasing energy gap ΔE between them. The quantity $\nu = (d_{\ell=2}^{(t_1)}/d_{\ell=0}^{(t_1)})/(d_{\ell=2}^{(t_2)}/d_{\ell=0}^{(t_2)})$ represents the ratios of the bound-continuum reduced radial matrix elements between both pathways. As expected for higher Rydberg states, this ratio converges quickly to the same number, i.e., the coupling of the $7p$ and $8p$ states to the continuum is similar. The interference for both state pairs is hence comparable. Note that the final energy $E_f = E_{5s} +$

FIG. 8. (a) Addition of a third laser pulse (orange) with amplitude $\mathcal{E}_3$ creates an additional third (direct) ionization pathway $t_{\text{contr.}}$ to the final energy E_f . The blue and IR pulses are the same as in Fig. 1. (b) Interference term $\text{DCS}_{\text{interf.}}$ obtained from Eq. (23) for different strengths of the amplitude $t_{\text{contr.}}$ relative to $t_1 + t_2$. (c) Angular dependence of the associated interference phase φ_{12} between t_1 and t_2 . All results are obtained by numerical calculation with pulse lengths of 75 optical cycles whereas the infrared and blue laser fields have the same field amplitudes and detunings as in the previous sections.

$\hbar\omega_1 + \hbar\omega_2$ is higher in comparison to the $5p/6p$ case (0.84 and 1.09 eV). Interestingly, $\text{DCS}_{\text{interf.}}$ has a different shape and changes its sign in both cases, which has an impact on the angular modulation of the phase difference φ_{12} . Surprisingly, although the amplitude is smaller in comparison with the $5p/6p$ case shown in Fig. 6, the angular variation of the phase is significantly more pronounced, ranging from 80° to 100° . This highlights the change in sign of the interference term (negative sign of the argument of the arccos function means a phase larger than 90°).

The green and orange curves represent cases when we decrease the energy gap. We chose the intermediate-state pairs $6p/7p$ ($\Delta E = 0.51$ eV, $E_f = 2.22$ eV) and $7p/8p$ ($\Delta E = 0.25$ eV, $E_f = 2.97$ eV). Intriguingly, the interference cross section remains unaffected when changing to higher-lying state pairs. The amplitudes of $\text{DCS}_{\text{interf.}}$ range in both cases from 5% to 50% and is negative. However, the angular modulation of the phase difference φ_{12} decreases drastically. We address this development with the dipole matrix element ratio ν which converges rapidly to 1, meaning the ratio between the $\ell = 0$ and the $\ell = 2$ angular channels is nearly the same for both ionization pathways t_1 and t_2 . According to Eq. (7), the individual phase shapes are then comparable.

From these results, we learn that the angular modulation of the phase difference depends critically on the quantity ν , whereas the overall amplitude of the interference term ($\text{DCS}_{\text{interf.}}$) is more robust and reveals a dependence on the energy gap between the intermediate-state pair defining t_1 and t_2 .

B. Laser-driven perturbation of ionization pathways

Another method for dynamic control of the interference phenomena is the addition of a third control pulse. As represented in the scheme in Fig. 8(a), the corresponding parameters are chosen in a way that initiates a weak one-photon process directly into the continuum so that $\hbar\omega_{\text{contr.}} = (E_f - E_{5s})/\hbar$. The field amplitude $\mathcal{E}_{\text{contr.}}$ is, hence, chosen in a way

that the corresponding transition amplitude $t_{\text{contr.}}$ is of the same magnitude as that of the two-color pathways t_1 and t_2 . As indicated in the modified scheme in Fig. 8(a), in the presence of all fields, the total amplitude is given by the coherent sum $t_{\text{all}} = t_1 + t_2 + t_{\text{contr.}}$. To access the desired interference term $\text{DCS}_{\text{interf.}} = t_1 t_2^* + t_1^* t_2$, one has to then perform four different measurements: (i) with all fields on-resonance to the intermediate and final states, respectively, (ii) with the blue field to the $6p$ state off-resonance, (iii) with the red field to the $5p$ state off-resonance, and (iv) with both red and blue fields off-resonant (thereby blocking both pathways t_1 and t_2). The interference term is then given by

$$\text{DCS}_{\text{interf.}} = |t_{\text{all}}|^2 - (|t_{\text{(ii)}}|^2 + |t_{\text{(iii)}}|^2 - |t_{\text{(iv)}}|^2), \quad (23)$$

with $t_{\text{(ii)}} = t_1 + t_{\text{contr.}}$, $t_{\text{(iii)}} = t_2 + t_{\text{contr.}}$, and $t_{\text{(iv)}} = t_{\text{contr.}}$.

In Fig. 8(b), we present the interference term for different strengths of the perturbation by the third (control) laser field. As expected, the additional one-photon ionization route has a large impact on the quantum interference between t_1 and t_2 . Here, the field amplitude $\mathcal{E}_{\text{contr.}}$ has to be very low so that the associated $t_{\text{contr.}}$ is of the same magnitude as the two-photon pathways t_1 and t_2 . In comparison to the unperturbed case shown in Fig. 6(a), the magnitude of the interference is increased in the presence of the control field. In strong contrast to the previous findings, even the sign of the $\text{DCS}_{\text{interf.}}$ can be changed by the effect of the additional one-photon process when the field amplitude is sufficiently large. It is, therefore, not surprising that the large impact seen here is directly transferred to the phase φ_{12} associated with the quantum interference. As shown in Fig. 8(c), the angular variation is drastically increased due to the action of the controlling field. In the original experiment and theoretical treatment, the modulation in the polar angle was smaller than 20° . Now, we obtain strongly pronounced phase peaks and a rather complex angular structure of φ_{12} with a variation covering more than 140° .

The addition of the third laser field helps to emphasize that the interference effect is not robust to statistical fluctuations

FIG. 9. Stochastic nature of the interference effect. In this example, one of the ionization pathways is closed by slight detuning of the laser field. We hence find interference between the two-photon amplitude (pathway 1) and the one-photon transition initiated by the third control field $\mathcal{E}_{\text{contr.}}(t)$ [panel (a)]. (b) The stochastic nature of the interference effect: The amplitude of the one-photon process is randomly varied between $-1.0t_1$ and $+1.0t_1$. The curves show the dependence on the number of random amplitudes with reference to $t_{\text{contr.}} = 1.0t_1$ (blue line). Numbers in the parentheses denote the number of amplitudes used for the statistical average (see the text for details).

but is unique for every set of laser parameters. For this purpose, we slightly detuned the blue laser field so that the $6p$ state was not excited [cf. Fig. 9(a)]. The resulting interference phenomenon in the ionization channel then stems from the superposition of the two-photon pathway 1 (via $5p$ photoexcitation) and the one-photon direct photoionization amplitude mediated by $\mathcal{E}_{\text{contr.}}(t)$. By tuning the parameters of the third field so that the transition strength of the amplitude $t_{\text{contr.}}$ is equal to t_1 , we obtain a characteristic interference as shown by the dark blue curve in Fig. 9(b). We then varied the amplitude of the control field in a way that $t_{\text{contr.}} \in [-1.0t_1, 1.0t_1]$ by use of a random number generator. The additional curves in the figure show a statistical average based on the total number of random amplitudes input to the model. One can clearly see a trend that increasing the number of random events decreases the interference effect. This shows that, for an infinite number of measurements, the resulting interference would disappear.

VI. CONCLUSIONS

In this paper, we have presented a theoretical investigation of experimental interference studies in a single rubidium atom. We have systematically demonstrated the transition from the short-pulse into the continuous-wave regime and

the evolution of the occupation numbers, photoionization probability, and quantum phase under an increase in the pulse lengths. We find that for pulse lengths of more than ten optical cycles, the theoretical description of the ionization scheme via the two-photon matrix element in the frequency regime is sufficient and that this provides all the physical information required for interference studies.

Our theoretical model provides generally good agreement with the experimental data and predicts a pronounced interference amplitude $\text{DCS}_{\text{interf.}}$, whereas the angular variation of the associated phase difference φ_{12} is relatively weak. In this treatment, we have developed various strategies to manipulate the quantum interference between both photoionization pathways t_1 and t_2 . As an example, we can change the populations of the intermediate states by laser detuning which introduces an imbalance between both pathways so as to change the interference phenomena. Furthermore, by choosing different state pairs n_1p and n_2p , we change the energy difference and coupling to the continuum, which again markedly changes the quantum interference. A method which does not change the intermediate-state pairs and the parameters of the blue and infrared laser fields is the addition of a third control laser field which perturbs the original transition pathways t_1 and t_2 . An appropriate tuning of the corresponding one-photon transition amplitude into the continuum can even invert the sign of the interference amplitude as well as produce a much more pronounced angular variation of the interference phase. There is no analogy to this third control in the conventional double-slit experiments.

As well as the addition of a third pulse, there are several other possibilities to explore the behavior of the interference phenomenon. As an example, one can show that, in alkali atoms, quadrupole transitions into the continuum reveal Cooper minima at kinetic energies below 1 eV. Thus, one can choose intermediate-state pairs which lead to final energies in the region of such Cooper minima whereas such quadrupole transitions at low intensity are generally accessible by structured light fields [14]. Further work will, hence, be dedicated to studying these two-pathway interference effects with inhomogeneous light-induced quadrupole transitions.

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