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Modal refractive index measurement in nanowire lasers - a correlative approach

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Abstract

We present a method to correlate multi-modal measurements – namely optical spectroscopy and electron microscopy – over large ensembles of randomly distributed single nano-objects. Using an algorithmic approach derived from astrometry, a marker-free method of uniquely associating nano-objects characterised using multiple techniques is described. This approach is applied to nanolasers, enabling an experimental calculation of modal refractive index in sub-micron diameter nanowires. By matching the lasing spectrum and electron microscopy image of 13 nanowire lasers, the refractive index of the TE₀₁ mode in GaAs/AlGaAs multiple-quantum well nanolasers is determined to be $n_g=4.7 \pm 0.3$.

Keywords

III-V Nanowire lasers, Multimodal characterisation

Introduction

A number of functional devices based on single semiconductor nanowires are now active topics of research, including quantum devices,^{1,2} single photon sources,^{2,3} detectors^{4,5} and nanolasers.^{6–14} As higher levels of structural complexity are demanded of the nanowire architecture, it is increasingly required that multiple characterisation approaches are taken; for instance, optical spectroscopy for electronic state information,¹⁵ time-resolved measurements for dynamics,¹⁶ Raman measurements for stoichiometry or doping,¹⁷ scanning electron microscopy for geometry, transmission electron microscopy for crystallographic information¹⁸ and device¹⁹ or non-contact electronic approaches^{20–23} for functional understanding. Correlating these single-wire approaches is challenging, as single nanowires have to be relocated in diverse experimental apparatus. While substrate marker-based techniques allow for reliable relocation of nanowires^{19,24} and highly complex measurements,^{25,26} where we need to understand specific functional behaviour in *low-yield* systems the placement of this marker needs to be done *after* characterisation and identification of nanowires of interest. This presents a great challenge for scaling-up of correlated multi-technique measurements.

Here, we report a novel approach based on a computational matching algorithm originally developed for astrophysics²⁷ which is able to uniquely identify *single nanowires* from within an ensemble of over $>15,000$, given the relative location of $\gtrsim 18$ neighbours. This approach is translation, rotation and scale invariant, and is robust in the case of a fraction of additional or missing nanowires (false positives or negatives), allowing for reliable identification of single wires and matching of measurements taken with multiple techniques. We demonstrate this approach for the specific case of semiconductor nanolasers fabricated from GaAs/AlGaAs with an active gain region of multiple radial quantum wells.^{7,28}

Experimental measurement of the modal refractive index for a given transverse laser mode confined within a semiconductor nanowire is a challenging task, as wire-to-wire variation in length, diameter and gain spectrum can mask any systematic variations. One approach requires longitudinal mode spacing observed in lasing spectra to be correlated with sub-wavelength resolution geometric measurements, typically through electron microscopy. By applying our matching algorithm to an ensemble of 15960 nanowires distributed on a substrate (located using a recently reported automated optical microscopy tool²⁸), we match spectroscopic information with electron microscopy images of 256 nanowires without the use of markers. By identifying 13 functional nanowire lasers with both optical spectroscopic and geometric information from SEM, we determine the TE01 modal refractive index to be $n_g = 4.7 \pm 0.3$, in agreement with previous reports.⁷

Methods

Nanowire Growth

GaAs/AlGaAs multiple quantum well nanowires were grown using metal organic vapour phase epitaxy (MOVPE) according to a previously published recipe,⁷ resulting in an ensemble of wires approximately 4 μm long and 460 nm diameter, with $8 \times \sim 5$ nm thickness radial GaAs quantum wells with $\text{Al}_x\text{Ga}_{1-x}\text{As}$ barriers (where the aluminium fraction $x \approx 0.4$). These nanowire were dispersed onto a quartz substrate by gentle rubbing for optical study (detail of the experimental conditions are provided in the **Supporting Information**).

Nanowire Characterisation

15960 nanowires were identified on the substrate using a home-built automated microscopy platform, as described previously.²⁸ In addition to bright-field images, optical spectroscopy was carried out on each nanowire sequentially, with measurements of photoluminescence spectra (and fluorescence images) measured under low-excitation conditions. A random set

of nanowires ($\sim 6\%$) were selected for power-dependant photoluminescence measurements under pulsed excitation with a defocussed excitation pulse ($\sim 15\ \mu\text{m}$ excitation spot diameter). These measurements were used to identify a lasing threshold - for approximately 50% of these, room-temperature lasing was observed, for which spectra and threshold were recorded. This resulted in around 500 nanowires with confirmed lasing; as each power dependant measurement required around 90 s to perform, we limited the total number of measurements. The sample was subsequently coated in 2–5 nm of Pt/Au, and imaged at $500\times$ magnification (around $250\ \mu\text{m}$ field-of-view) using a scanning electron microscope (SEM).

Nanowire Matching

Matching of nanowires was accomplished using an algorithmic point matching function developed by the *astrometry.net* project.²⁷ Full details are given in the supplementary information and example code is available online¹, however, the process is briefly described here. All 15960 nanowires positions identified from automated optical microscopy were used as points in a 2D space. From this point array, *quads* - arrangements of four non-collinear points - were randomly selected at a range of scales covering the field-of-view of the techniques used. Each quad is converted to a 4-byte hash code according to the approach outlined by Lang and colleagues,²⁷ and the hash and centre point of the quad is recorded. The process is repeated, generating $\sim 10^6$ **reference hashes**. These hashes have the property of being scale, rotation and translation invariant.²⁷ For each SEM image, nanowire positions are extracted, and a similar process is used to generate **sample hashes**. The sample hashes are individually matched to references hashes using a *kd*-tree approach (to allow for nearest neighbour matches arising from small location errors), and the matching hashes are then recorded. Where multiple hash-matches indicate a given alignment, the efficacy of the match is assessed using a point-by-point overlap. When a sufficient number of wires from each of the reference and sample sets overlap, the transformation is accepted and individual nanowires

¹MATLAB reference code is available from https://bitbucket.org/paparkinson1/nanowire_matching/

are matched. This approach is general for point matching, and requires $\gtrsim 18$ nanowires in the sample image to produce a good alignment (this number is dependant on the reference size and errors in determining position). Where no alignment is found, this is most often due to too small a number of nanowires in the sample image or obscuration in either source image (such as grease or dust).

Results

To illustrate the scale of the challenge for matching single nanowires - the nanowires were inhomogeneously spread over a 4.5 mm diameter region; for the working nanolasers characterised (~ 500), this means one nanolaser every $\sim 32000 \mu\text{m}^2$. For the given field of view for SEM, there should be approximately 1 – 2 characterised nanolasers for each SEM image (around 3%). 29 SEM images were taken at random across the substrate - 19 ($\sim 65\%$) were successful matched to their corresponding nanowire sets from optical microscopy and spectroscopy. Figure 1 shows the results of applying the matching algorithm, with the spatial distribution of nanowires identified by optical microscopy shown in blue, and matching nanowires identified from electron microscopy shown in red.

In total, 256 nanowires were matched. Two measurements were extracted from the SEM images; nanowire length, and nanowire orientation. It is expected that SEM imagery provides a more accurate measure of these parameters, due to the higher spatial resolution of the technique (250 nm at the given magnification). This is significantly better than from optical microscopy, where even diffraction-limited performance would be on the order of 700 nm for the optical system used. Figure 2(a-b) shows the nanowire length as determined by optical and electron microscopy techniques; this length was defined as the distance between the 50% contrast points along a line along the nanowire axis. It is noted that significant deviation is observed between the two microscopies, with nanowire lengths being routinely overestimated by $\Delta l = l_{\text{optical}} - l_{\text{SEM}} \approx 650 \text{ nm}$ when determined from optical microscopy.

Figure 1: (a) A spatial map of all 15960 nanowires identified by the automated microscopy procedure (blue dots). 256 nanowires matched to SEM images are indicated in red, with 17 tested, matched and functioning nanolasers depicted in green. The black dashed line shows the circular edge of the scan region. (b) A close view of the nanowire point array indicated by the black square in panel a).

This is expected, given the diffraction limited performance predicted. A significant spread was also observed in optical microscope measurements - this may be due to neighbouring nanowires being misidentified as single wires by the optical technique. The orientation of nanowires is better correlated, with a < 100 mrad (6°) deviation observed between the two approaches (Figure 2d). Taken together, these correlations are a further good indicator that the matching algorithm works as expected. Figure S4 (Supporting Information) shows matched imagery for over 200 nanowires.

We identified 13 nanowires which a) showed room-temperature lasing, b) showed > 2 longitudinal modes and c) were matched to SEM imagery, reduced from 17 due to the need for > 2 longitudinal modes to accurately calculate intermodal spacing. The imagery and spectroscopic data for these wires are shown in Figure 3. It is noted that the quality of the optical images is not always good - this is due to intermittent vibration in the microscopy apparatus, coupled with the high-speed of sequential measurement. It is reassuring to note that, where they exist, neighbouring nanowires are observed in both techniques (for instance, nanowire 120 and 184).

Figure 2: Correlations between geometric measurements taken using automated optical microscopy (μscope) and matched electron microscopy (SEM). (a) Nanowire length correlations (red line indicates a 1:1 relationship), (b) a histogram of deviation between optical and SEM measurement. Note the $\sim 2 \mu\text{m}$ spread, and a 650 nm offset indicating the limited resolution of the optical approach. (c) The orientation of the nanowire, and (d) the deviation between the two measurements, showing a smaller offset and good agreement.

Figure 3: Details for 13 matched nanolasers, showing (from top to bottom row) extracts from SEM images, bright-field (back-illuminated) optical microscope image, fluorescence image under low excitation density, photoluminescence spectra and lasing spectra. In some cases (nanowires 57, 228 and 252), the optical images are blurred due to system vibrations.

For each matched nanolaser, the lasing mode spacing is determined from a photoluminescence spectra taken when optically pumped just above their lasing threshold. A typical lasing spectra is shown in the inset to Figure 4. The difference between mode wavelengths, $\Delta\lambda$ is used; the standard deviation for all intermodal spacings for a given wire is used to calculate the uncertainty. Figure 4 shows the relationship between intermodal spacing and nanowire length (as measured from SEM imagery). These nanowires have been shown to exhibit Fabry-Pérot (rather than whispering gallery mode) type lasing;⁷ as such, the intermodal spacing is related to nanowire length by:

$$L = \left(\frac{\lambda_0^2}{2\Delta\lambda} \right) \left(n_g - \lambda_0 \left(\frac{dn_g}{d\lambda} \right) \right)^{-1} \quad (1)$$

In principle, tapering in the nanowires can lead to variation in the group index. However, for the nanowire structures studied with diameter ~ 460 nm and TE01 mode, any variation is expected to be small.⁷

Discussion

An accurate measure of the modal refractive index in nanowires is useful both in the design of nanowire structure, and, in general, for determination of the dominant lasing mode. The small deviation between data and model shown in Figure 4 indicates that all nanowires are lasing on the TE01 mode, confirming the modelling previously reported.⁷ In future, this approach might be used with nanowires which exhibit multiple transverse-mode lasing to aid a classification of nanolasers into those with different dominant transverse modes, or to observe more complex waveguiding effects across multiple nanowires.²⁹ It is noted that the present approach makes an assumption of a constant modal refractive index for a given nanowire; the extent to which this value may vary with carrier density (and hence pumping level) is not straightforward to understand analytically, and is not considered in the presented analysis.

Figure 4: The relationship between lasing mode separation and nanowire length (as measured from SEM images). Horizontal error bars reflect the standard deviation of inter-modal spacing for each wire, while vertical error bars are fixed at ± 250 nm. The solid line is a fit according to the model presented in the main text with $n_g = 4.7 \pm 0.3$, while the shaded area represents the 95% confident interval for the fit parameters. The inset shows a typical lasing spectrum with longitudinal modes identified.

More generally, the marker-free identification of randomly positioned nanowires (or other nano-materials) using multiple techniques provides great opportunities; for instance, where markers cannot be applied, or in cases such as that presented where the yield is sufficiently low that markers would need be applied after an initial survey scan. We have demonstrated that simply by recording the positions of nanowires using two different microscopy techniques, nanowires can be uniquely and reliably matched with $\sim 60\%$ yield. It is anticipated that through further development of the matching algorithm and identification of their key parameters required for a successful match, this yield may be increased in future.

Conclusion

We have presented a computational approach to allow unique identification and matching of single nanowires using the relative position of nearest neighbours. By using this approach, we have demonstrated the marker-free matching of nanowires imaged by two techniques (SEM and optical microscopy/spectroscopy) which were performed on the same nanowires by different researchers on different continents. Using the accurate geometric information provided by electron microscopy coupled with spectroscopy from optical approaches, we have demonstrated a proof-of-principle method to accurately determine the modal refractive index for a class of multiple-quantum well nanowire lasers. This pure-software approach enables a powerful new class of multi-modal characterisation techniques for nanotechnology and particularly correlative statistical approaches.^{26,28}

Author Contributions

The project was conceived by PP, and the lasing data was primarily taken and analyzed by JAA. The nanolasers were designed by DS and SM, and grown by NJ under the supervision of HHT and CJ. Scanning electron microscopy was performed by KP under the supervision of LF. The manuscript was primarily written by PP, with contributions from all authors.

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Supporting Information Available

A full description of the hash code generation, matching, and validation. Experimental arrangement for optical spectroscopy. Example code for MATLAB is provided at https://bitbucket.org/paparkinson1/nanowire_matching/.

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