One- and two-dimensional cavity modes in ZnO microwires

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Abstract. We demonstrate that one-dimensional (1D) Fabry–Pérot modes (FPM) and 2D whispering gallery modes (WGM) occur separately or simultaneously within a single, tapered ZnO microwire. Their observation is strongly correlated with the size and shape of the microwire cross-section. FPM can preferentially be observed in thick wires with an elongated wire cross-section without hexagonal symmetry. The formation of WGM otherwise requires hexagonal symmetry—they can be observed in thin wires having a regular hexagonal cross-section. All optical eigenmodes were unambiguously identified by their in- and out-of- (cross-sectional) plane photonic mode dispersion.

Contents

1. Introduction 2
2. Experimental details 2
3. Results and discussion 2
4. Conclusion 8
Acknowledgments 9
References 9

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1. Introduction

The increasing demand for photonic and optoelectronic devices fueled the investigation of lasers [1], optical sensors [2], switches [3] or waveguides [4] at the micro- and nano-scale and this has become one of the most prominent fields of research in the last two decades [5]. Small optical resonators are the centerpiece of these technical applications whose efficient operation requires low lasing thresholds and thus high $Q$-factors of the resonator eigenmodes. The formation of standing waves in resonators is determined by the resonator size and shape and the operating wavelength. ZnO microwires perfectly fulfill all the conditions for optical waveguiding due to their large range of possible diameters and short-wavelength emission. They have been subject to intensive studies on a variety of optical eigenmodes [6–10].

Here, we report on the influence of individual resonator shapes and resonator sizes on the formation of optical resonances in ZnO microwires. We show that given hexagonal symmetry is a key element for the presence of special resonant eigenmodes: one-dimensional (1D) resonances (laterally 1D propagating waves) occur in microwires with large diameters, whereas 2D eigenmodes (propagation in a plane) can be observed in thin wires. As the presence of optical eigenmodes in dielectric media is a major prerequisite for the achievement of stimulated emission, such as exciton or polariton lasing, this paper is of great importance since it provides a comprehensive insight into the formation and identification of optical eigenmodes.

2. Experimental details

All investigated ZnO microwires were fabricated by carbothermal reduction in a tube furnace. Details of fabrication can be found in [11]. Note that carbothermal reduction is a highly random growth process—the fabricated microwires can have different shapes and sizes. Experiments were carried out on hexagonal, tapered ZnO microwires with the $c$-axis being the wire axis. In the following, we define the wire axis as the $z$-direction, whereas $x$ and $y$ are in-plane directions perpendicular to the wire axis.

The fabricated microwires have diameters in the range of 1–50 $\mu$m—lengths are up to 10 mm. Microwires were transferred to substrate edges and attached with their thick part to the substrate in such a way that the thinner part protrudes over the substrate edge and possible coupling of light into the substrate is suppressed. Photoluminescence (PL) from microwires is then excited at room temperature using a HeCd laser (emission at 325 nm) operating in continuous-wave mode and detected by a 320 mm focal length grating monochromator with a nitrogen-cooled, back-illuminated charged-coupled device. Angular-resolved PL measurements were carried out using fixed excitation and movable detection arms equipped with ultraviolet (UV) lenses (300 mm focal distance). The resulting angular spread in detection and excitation is about $2^\circ$ with a laser spot size of about 50 $\mu$m on the wire.

3. Results and discussion

Figure 1 (left) shows the wire length versus the wire diameter of a tapered ZnO microwire. The wire diameter varies from 1 to 20 $\mu$m. Plane view, scanning electron microscopy (SEM) images of two positions on the microwire can be seen on the right side of figure 1. The corresponding wire cross-sections were reconstructed from a combination of atomic force microscopy (determination of top facet tilt and tilt angles between side facets) and electron
microscopy at angles of $-60^\circ$, $0^\circ$ and $60^\circ$. For a diameter of $2.6 \mu m$ (figure 1, bottom right), the microwire has a nearly perfect hexagonal cross-section, whereas for $d = 16.6 \mu m$ (figure 1, top right), the microwire shows a slightly different cross section: the side facets are still parallel to each other, but the lengths of the individual facets differ. In fact, the equilaterality, and thus the hexagonal symmetry, is lost for large diameters of the microwire. For both microwire positions, angular-resolved PL spectra were recorded and can be seen in figure 2 for $d = 16.6 \mu m$ (top) and $d = 2.6 \mu m$ (bottom), angles $\varphi_z$ (left) and $\varphi_y$ (right) and transverse-electric (TE-) polarization.

At room temperature, ZnO microwires typically show a narrow recombination in the near-band edge region (assigned to radiative recombination of free excitons, centered around 380 nm) and a broad emission band in the visible spectral range (caused by recombination of carriers at deep defect centers). In many cases, the free exciton transition is accompanied by a broad recombination band centered at 390 nm, which is most commonly attributed to the so-called P-band, caused by exciton–exciton scattering [12]. In the following, we will now focus on another spectral issue: all spectra are modulated by a multitude of peaks covering the visible and UV spectral range whose peak distance decreases with decreasing wavelength. This is a clear indication of the presence of resonant optical eigenmodes since the optical mode spacing depends on the index of refraction ($\Delta \lambda \propto n$).

In general, optical eigenmodes in hexagonal dielectric resonators can be observed when conditions for constructive interference of light waves are fulfilled by geometry. Once a standing wave is formed, its character (dimensionality and optical pathway) can directly be deduced from the photonic dispersion relation $E(k)$. The generalized detection angle $\varphi$ is related to the light wave vector $k$ [13] via

$$hck(\varphi) = E(\varphi) \sin \varphi. \tag{1}$$
If equation (1) is fulfilled for the y- and the z-direction, an in-plane dispersion curve ($E(k_y)$) of a photonic mode in a wire with hexagonal cross-section can be obtained by measuring the PL signal between the normals of side facets (angle $\varphi_y$) and an out-of-plane dispersion curve ($E(k_z)$) can be derived by measurement between the normal of a side facet and the wire axis (angle $\varphi_z$).

The top part of figure 2 shows the angular-resolved PL measurement for $d = 16.6 \mu m$ of the ZnO microwire from figure 1. Along $\varphi_z$ and $0 \leq \varphi_y \leq 30^\circ$ the observed cavity modes blueshift with increasing emission angle due to their photonic mode dispersion. Both dispersion curves (in the y- and the z-direction) qualitatively show the same curvature for angles smaller than $30^\circ$. From this, one can deduce two mode properties: (i) light waves do not propagate along zigzag or spiral pathways because they should otherwise exhibit different or no photonic dispersion curves in the y- and the z-direction. (ii) The presence of the dispersion curves can only be caused by lateral photonic confinement in both the y- and the z-direction and thus displays the one-dimensionality of the present cavity modes only propagating in x-direction. This means, constructive interference of light is achieved by waves being reflected between opposite hexagon facets—the so-called Fabry–Pérot modes (FPM). Possible optical pathways for FPM are displayed in figure 3(a). In this particular case, FPM most probably pass the elongated hexagon on pathway 1.

In principle, FPM can propagate along (between the wire end facets) or perpendicular to the wire axis (between the wire side facets). The formation of standing waves between both ends of an individual wire has been thoroughly investigated by several groups \[14, 15\]. However, FPM between the hexagon side facets are very unlikely due to low reflectivity of light waves at the ZnO/air interface ($\approx0.2$ for normal incidence) and have not been reported in the literature.
so far. Note that figure 2 shows an additional red shift of eigenmodes for $\varphi_y > 30^\circ$. This shift of modes is due to FPM with pathway 2 rather than pathway 1 in an elongated hexagon (see figure 3). For this case, the mode spacing is smaller because the inter-facet distance is larger ($\Delta\lambda \propto d(1 + \varepsilon)$). Here, the parameter $\varepsilon$ was introduced in order to account for the elongation of the cross section compared with a regular hexagon (see figure 3(a)).

The bottom part of figure 2 shows the angular-resolved PL measurement for $d = 2.6 \mu m$ of the ZnO microwire from figure 1. While the $\varphi_z$-scan exhibits a dispersion curve similar to that observed for FPM (both TE and TM contributions are visible although the measurement is TE polarized [16]), no dependence of the wave vector can be found for $\varphi_y$. This has already been observed by Trichet et al [17] and is a clear indication for the propagation of light waves only confined in the $z$-direction—within the wire cross-section—and is attributed to the formation of whispering gallery modes (WGM) that arise due to constructive interference of light waves passing a closed round trip inside the hexagon by total internal reflection [6]. At this point, we assume to have hexagonal WGM that undergo reflection at every single facet of the hexagon (see figure 3)—this assumption can be easily confirmed by mode calculations (see below). In principle, triangular WGM also have stable pathways in a regular hexagon, they can be excluded here due to different pathways and thus different mode spacings. We annotate that the PL spectra for $\varphi_z = 0$ and $\varphi_y = 0$ slightly differ in mode positions. Technically, these spectra should be identical. However, the wires are rotated by $90^\circ$ between measurements in different axial directions $z$ and $y$. This can cause a slight change of the focus position on the wire and thus of the diameter and subsequently a change of the mode positions.

Our results show that the formation of WGM is preferred in thin ZnO microwires with a given hexagonal symmetry according to [18, 19], but WGM cannot be formed when symmetry is broken as is the case for thick wires (figure 2). In contrast to that, FPM are unfavored for small diameters (cavity losses can be calculated to be $6500 \text{ cm}^{-1}$ for a $2.6 \mu m$ cavity and $380 \text{ nm}$ wavelength [20]) since they undergo a large number of reflections at the wire side facets for a given photon lifetime (typically several tens of femtoseconds) due to a shorter optical pathway. Elongating the optical pathway and decreasing the number of reflections leads to an increase of the probability of observing FPM (losses are only $1400 \text{ cm}^{-1}$ in the UV spectral range for $2R_i = 12.1 \mu m$). Note that in principle FPM should also be present for small wire diameters, but probably linewidths are too large to observe them or the incident light intensity is more easily captured by WGM than FPM.
Figure 4. Angular-resolved PL measurements at room temperature along the angles $\varphi_z$ (top) and $\varphi_y$ (bottom) as well as TE (left) and TM polarizations (right) of a ZnO microwire with $d = 6.5 \mu m$.

In order to verify our findings, we further investigated a ZnO microwire with an intermediate diameter of $d = 6.5 \mu m$—large enough for the formation of FPM—and a regular hexagonal cross-section, necessary for the establishment of WGM. As can be seen from figure 4, both 1D and 2D cavity modes are simultaneously present in the angular-resolved spectra showing a superposition of the phenomena from figure 2: FPM and WGM blue shift with increasing $\varphi_z$, but only FPM also shift for changing $\varphi_y$, whereas WGM are angle independent in the $y$-direction. Note that here FPM show the same dispersion for $\varphi_y > 30^\circ$ and $\varphi_y < 30^\circ$ due to a given cross-sectional, hexagonal symmetry.

In general, the dispersion of an optical eigenmode is given as $E = \hbar c \sqrt{k_x^2 + k_y^2 + k_z^2}$ and can be evaluated into a parabolic expression [21] (for small $k$) using a non-vanishing photonic mass $m_{ph} = \hbar n_{\perp/\parallel} N/cL$ with $L = 4R_i$ or $6R_i$ for FPM or WGM, respectively. Hereby, $n_{\perp/\parallel}$ is the refractive index for TE- ($\perp$) and TM-polarization ($\parallel$), respectively, $N$ is the mode number and $R_i$ is the inner radius of the hexagon (see figure 3). It reads

$$E(\varphi) \approx E^0 + \frac{\hbar^2 k(\varphi)^2}{2m_{ph}}.$$  \hspace{1cm} (2)

The energetic positions of FPM $E^0_{\text{FPM}}$ can be calculated via a simple plane-wave model which is valid for optical pathways much longer than the incident wavelength ($\lambda \ll d$) [19]. They are given by

$$E^0_{\text{FPM}} = \frac{\hbar c N_{\text{FPM}}}{2n_{\perp/\parallel}d_{\text{FPM}}}.$$  \hspace{1cm} (3)
Figure 5. Angular-resolved PL measurements (left) and calculated FPM mode dispersion (right) at room temperature along the angles $\varphi_z$ (top) and $\varphi_y$ (bottom) for TM polarization of a ZnO microwire with $d = 16.6 \, \mu m$ and $\varepsilon = 0.37$.

with $N_{\text{FPM}}$ being the FPM mode number and $d_{\text{FPM}}$ being the cavity length. For FPM between the end facets, $d_{\text{FPM}}$ equals the wire length—for FPM between the side facets, $d_{\text{FPM}} = 2R_i$. The energetic positions of hexagonal WGM $E_{\text{WGM}}^0$ can be calculated similarly to FPM via [6]

$$E_{\text{WGM}}^0 = \frac{hcN_{\text{WGM}}}{6n_{\perp/\parallel}R_i} + \frac{2hc}{n_{\perp/\parallel}R_i}\delta$$

with the WGM mode number $N_{\text{WGM}}$. In equation (4), $\delta = \arctan(\kappa_{\perp/\parallel}\sqrt{3n_{\perp/\parallel}^2 - 4})$, and $\kappa_{\perp} = n_\perp$ for TE- and $\kappa_{||} = 1/n_\parallel$ for TM-polarization, respectively. In comparison with FPM, the $\delta$-term arises due to phase shifts during total internal reflection.

Optical losses determine the linewidth $\Gamma$ of individual cavity modes. Note that in our case only modes with mode energies well below the ZnO bandgap were investigated in order to ensure that losses due to absorption can be neglected. In this case, optical losses for FPM are only given by mirror losses at the ZnO/air interface. They can be quantified using the reflection coefficients at the top $R_T$ and bottom facet $R_B$:

$$\Gamma_{\text{FPM}} = -\frac{hc}{d_{\text{FPM}}} \ln(R_T R_B).$$

The loss mechanisms for WGM and corresponding linewidths $\Gamma_{\text{WGM}}$ were addressed by Wiersig [19] and were experimentally evidenced by Czekalla et al [20]. They can be ascribed to boundary wave leakage and pseudo-integrable ray losses (for details see [19]). $\Gamma_{\text{WGM}}$ reads

$$\Gamma_{\text{WGM}} = -\frac{hc\lambda}{2\pi R_i^2} \left( \frac{3n_{\perp/\parallel}^2}{4\sqrt{3n_{\perp/\parallel}^2 - 4(n_{\perp/\parallel}^2 - 1)}} + \frac{4\pi\delta}{3n_{\perp/\parallel}} \right).$$
In the following, the photonic mode dispersions were calculated using a combination of equations (1) and (2) with equations (3) or (4) for FPM or WGM, respectively, by using Lorentzians with linewidths $\Gamma_{\text{FPM}}$ or $\Gamma_{\text{WGM}}$ from equations (5) and (6), respectively. The ZnO refractive index dispersion was taken from [22] including excitonic contributions.

Figure 5 shows angular-resolved, TM-polarized PL spectra for angles $\phi_z$ (top) and $\phi_y$ (bottom) and corresponding FPM calculations for a ZnO microwire with $d = 16.6 \, \mu m$. Note that the TM-polarized free exciton emission is notably redshifted compared to TE-polarization (see figures 2 and 5). This is due to the ZnO valence band ordering with strongly TE-polarized A- and B-excitons, whereas C-excitons are TM-polarized [23]. From SEM pictures, an inner radius of $R_i = (5.24 \pm 0.01) \, \mu m$ and an elongation of $\epsilon = (0.37 \pm 0.05)$ can be deduced. In order to fit the measured PL spectra at $\phi_{z,y} = 0^\circ$ and $\phi_y = 60^\circ$, the values of $R_i = 5.234 \, \mu m$ and $\epsilon = 0.379$ were chosen, in excellent agreement with the experimental data. Further, the observed PL mode dispersions are perfectly reproduced by our calculations. This is also the case for WGM, as can be seen in figure 6. Here, an experimental value of $R_i = (1.12 \pm 0.01) \, \mu m$ was determined and $R_i = 1.109 \, \mu m$ was chosen for the calculation.

4. Conclusion

In summary, we have demonstrated that both 1D FPM and 2D WGM can be observed separately and simultaneously within a single, tapered ZnO microwire. Their presence is strongly correlated to the shape of the wire cross-section. On the one hand, FPM are preferentially formed for large diameters and imperfect (elongated) hexagonal cross-section. On the other, WGM are stable for small wire diameters with hexagonal symmetry in the...
cross-sectional plane. For both kinds of cavity modes, unambiguous experimental evidence is due to their dispersion curves that display one- and two-dimensionality of FPM and WGM, respectively.

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