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Disciplines
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Incorporating polaritonic effects in semiconductor nanowire waveguide dispersion

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We present the calculated and measured energy-propagation constant (E-β) dispersion of CdS nanowire waveguides at room temperature, where we include dispersive effects via the exciton-polariton model using physical parameters instead of a phenomenological equation. The experimental data match well with our model while the phenomenological equation fails to capture effects originating due to light-matter interaction in nanoscale cavities. Due to the excitonic-polaritonic effects, the group index of the guided light peaks close to the band edge, which can have important implications for optical switching and sensor applications. © *2010 American Institute of Physics*. [doi:10.1063/1.3479896]

Semiconducting nanowires are frequently used as active waveguide elements at the nanoscale in applications, such as sensing,[1] light generation,[2] light detection,[3] and photovoltaics.[4] Guiding of optical waves in nanowires resembles that in conventional microscale optical fibers but with the key difference that a large dielectric contrast between the nanowire and its surrounding provides the necessary optical confinement at such small lengthscales.[5] Therefore, popular choices of semiconductor nanowires for active waveguiding applications are ZnO, ZnSe, CdS, GaN, and SnO_2, all semiconductors exhibiting a relatively high refractive index, a direct electronic band gap and the formation of electron-hole pairs (excitons) as dictated by their relatively large exciton binding energies.[6]

Optical transport in waveguides or fibers is characterized by the energy-propagation constant dispersion (E-β), and the group velocity, which can both be obtained by analytically or numerically solving Maxwell's equations with appropriate boundary conditions if the dielectric functions $\varepsilon(\omega)$ of the core and cladding materials are known.[7–9] A common approach to include this material dispersion into waveguide dispersion calculations is by using a phenomenological Sellmeier type equation in which the coefficients are obtained by numerical fitting to dielectric dispersion obtained from measurements on macroscopic crystals.[10] Although this approach gives satisfactory results at energies much lower then the electronic band gap, deviations at energies close to the band gap occur, particularly if excitons are present.[8] Importantly, excitons have been detected at room temperature in bulk CdS crystals[11] and it is known that due to the substantially larger oscillator strength of excitons than that of free electron and hole recombination, excitons can strongly couple to the light field resulting in the formation of exciton-polaritons[11] which manifests in the formation of anticrossing upper and lower polariton branches (UPB and LPB) and drastic changes to the dielectric function.

It is through this polaritonic coupling mechanism, as well as giant exciton oscillator strength and super radiance effects,[12,13] that finite crystal sizes comparable to the optical wavelengths can result in material dispersion that is significantly different from that of macroscopic crystals.[14] Therefore, in order to describe the energy-propagation constant dispersion (E-β) and group velocity in these nanowire waveguides accurately, it is highly desirable to explicitly include physical quantities such as the transverse and longitudinal exciton resonance frequencies and their damping constants in the analysis so that the size effects can be readily incorporated.

In this paper, we show how in CdS nanowires the waveguide dispersion is altered due to the presence of excitons, which strongly couple to the confined photonic waveguide modes. After calculating the waveguide E-β dispersion for the purely photonic modes using a Sellmeier type equation, we introduce electronic resonance effects into the calculations via the polaritonic contributions to the dielectric function. Next, these calculations are compared with experimental data obtained on CdS nanowires and finally, we briefly discuss the implications of the strongly modified dispersion on photonic switching and sensing with nanowires.

CdS nanowires were obtained by the vapor-liquid-solid method using evaporation of CdS powder (99.999% Sigma Aldrich) and 5 nm Au/Pd covered silicon substrates.[15] After synthesis, the nanowires were transferred to Si/SiO_2 substrates containing markers so that individual wires could be characterized by both scanning electron microscopy (SEM) and optical microscopy. Optical experiments were carried out using a home-built microscope as described elsewhere.[9]

To calculate the confined photonic modes in the CdS nanowires, we simplified the wire geometry to that of a cylindrical step-index fiber of radius, r, with a CdS core [refractive index $n_{co}(\lambda)$] and air cladding ($n_{cl}=1$). It was previously shown that this simplification does not markedly influence the correspondence of the calculated modes with the experimental data.[8,9,16] Generally, the waveguide modes of cylindrical waveguides are of a hybrid nature, that is the electric and magnetic fields can have components in the propagation direction, z (HE and EH modes). The transverse electric (TE) and or transverse magnetic (TM) modes can be considered special cases of the hybrid modes where either the electric or the magnetic fields in the propagation direction vanish. From Maxwell's equations and the boundary condition of continuous tangential fields at the fiber surface,

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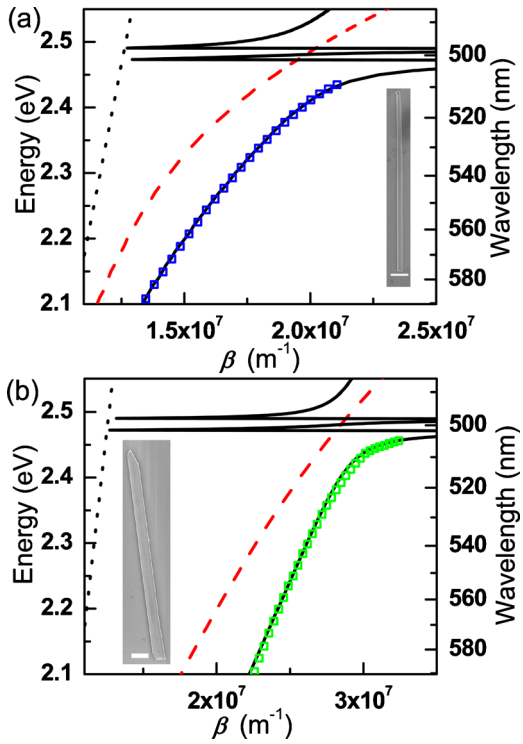

FIG. 1. (Color online) Waveguide dispersions for (a) the TM_{01} mode of a CdS nanowire with a radius of 120 nm and (b) the HE_{12} mode of a CdS nanowire with a radius of 255 nm, calculated using a Sellmeier equation (see Ref. 8) for material dispersion (dashed red line) and the polaritonic model (full black line). (Insets) SEM images of the wires, scale bars 500 nm. Black dotted lines: dispersion of light in air. Square data points determined from Fig. 2.

exact eigenvalue equations can be formulated for the various modes where the subscript ν denotes the order and the subscript m denotes the m-th root[7]

$$\mathrm{HE}_{\nu m}\mathrm{EH}_{\nu m}:\left[\frac{J_\nu'(U)}{UJ_\nu(U)}+\frac{K_\nu'(W)}{UK_\nu(W)}\right]\left[\frac{J_\nu'(U)}{UJ_\nu(U)}+\frac{n_{cl}^2}{n_{co}^2}\frac{K_\nu'(W)}{UK_\nu(W)}\right]=\left(\frac{\nu\boldsymbol{\beta}}{kn_{co}}\right)^2\left(\frac{V}{UW}\right)^4, \quad (1)$$

$$\mathrm{TE}_{0m}:\frac{J_1(U)}{UJ_0(U)}+\frac{K_1(W)}{UK_0(W)}=0, \quad (2)$$

$$\mathrm{TM}_{0m}:\frac{n_{co}^2J_1(U)}{UJ_0(U)}+\frac{n_{cl}^2K_1(W)}{UK_0(W)}=0. \quad (3)$$

With $U=r\sqrt{k^2n_{co}^2-\boldsymbol{\beta}^2}$, $V=rk\sqrt{n_{co}^2-n_{cl}^2}$, $W=r\sqrt{\boldsymbol{\beta}^2-k^2n_{cl}^2(\lambda)}$ and J is the Bessel function of the first kind, K is the modified Bessel function of the second kind, r is the nanowire radius, k is the free-space wave vector, and λ is the free-space wavelength. Subsequently these eigenvalue equations were numerically solved for the propagation constant, β, at each wavelength using a core refractive index defined by a Sellmeier equation obtained from macroscopic CdS crystals.[10] The results of these calculations are plotted in Figs. 1(a) and 1(b) as red dashed lines for two CdS nanowires with radii of 120 nm [(a), TM_{01} mode] and 255 nm [(b), HE_{12} mode] as determined from SEM imaging [inset in Figs. 1(a) and 1(b)], together with the dispersion of light in air (dotted). The calculated modes are to the right of the light line, showing that these modes are indeed confined to the nanowire core and are therefore waveguide modes.

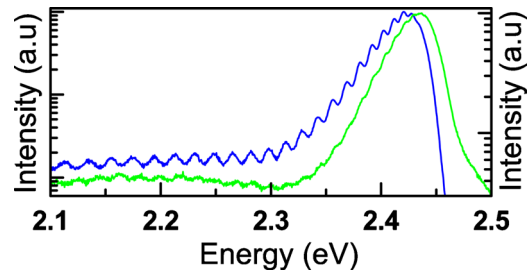

FIG. 2. (Color online) Emission spectra of guided photoluminescence light for a 9.59 μm long wire with a radius of 120 nm and (top curve) and a 9.10 μm long wire with a radius of 255 nm (bottom curve).

Next, to include dispersive effects due to the presence of excitonic resonances and polariton formation, we introduce a dispersive core refractive index $n_{co}(\lambda)$ that is taken as the real part of the square root of the dielectric function, where the dielectric function in the vicinity of the CdS excitons A and B can be described by a coupled oscillator model for two closely spaced resonances[17]

$$\varepsilon(\omega)=\varepsilon_b\left(1+\frac{\omega_{BL}^2-\omega_{AT}^2}{\omega_{BT}^2-\omega_{AT}^2}\frac{\omega_{AL}^2-\omega_{AT}^2}{\omega_{AT}^2-\omega^2-i\omega\Gamma_A}+\frac{\omega_{AL}^2-\omega_{BT}^2}{\omega_{AT}^2-\omega_{BT}^2}\frac{\omega_{BL}^2-\omega_{BT}^2}{\omega_{BT}^2-\omega^2-i\omega\Gamma_B}\right), \quad (4)$$

where ε_b is the background dielectric constant, ω_{AT} and ω_{AL} are the A-exciton transverse and longitudinal resonance frequencies, ω_{BT} and ω_{BL} are the B-exciton transverse and longitudinal resonance frequencies, Γ_A the A-exciton damping and Γ_B the B-exciton damping. We have omitted spatial dispersion since in our investigated energy range the UPB is severely damped and only modes on the LPB propagate along the nanowire.[18] Numerically solving the mode eigenequations, which now include the resonance effects, with bulk parameters $\varepsilon_b=8$,[18] $\hbar\omega_{AT}=2.4696$ eV,[19] $\hbar\omega_{AL}=2.4715$ eV,[18] $\hbar\omega_{BT}=2.4882$ eV,[19] $\hbar\omega_{BL}=2.4895$ eV,[18] $\hbar\Gamma_A=\hbar\Gamma_B=10$ μeV leads to the dispersions plotted as black solid lines in Figs. 1(a) and 1(b). At low energies the mode mimics a purely photonic mode but closer to the resonances the dispersion flattens out and resembles the dispersion of the electronic resonances. This is due to the dual nature of the exciton-polaritons, having more electronic or photonic character depending on the energy and propagation constant. It must be noted that the crystal dimensions of our nanowires are much larger (smallest dimension 240 nm) than the exciton Bohr radius in CdS (~2.8 nm) (Ref. 20) so that electronic quantum size effects that can alter the transverse resonance energies and the transition (oscillator) strength from the bulk values can be excluded. The nanowire crystal dimensions are such however, that the aforementioned super radiance and giant oscillator strength effects can be significant, and in the model they are incorporated by the longitudinal-transverse splitting $(\omega_{AL}^2-\omega_{AT}^2)$ which is proportional to the oscillator strength of the excitonic transition.[17]

To verify our calculations experimentally, CdS nanowires where excited nonresonantly across the band gap at one end whereas the waveguided photoluminescence was collected at the other end of the wire. The collected spectra (Fig. 2) consist of a strong peak of near band-edge emission

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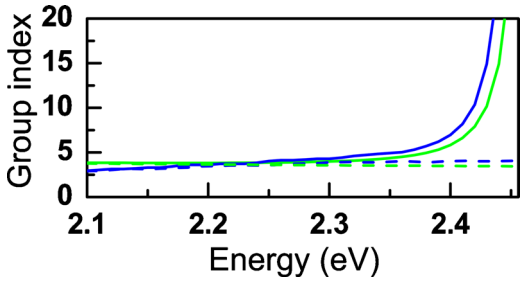

FIG. 3. (Color online) Group index vs energy for photonic (dashed lines) and polaritonic (solid lines) modes of the two nanowires shown in Fig. 1. The thinner nanowire (120 nm radius) shows higher group index in comparison to thicker wire (255 nm radius) especially at energies close to the band-gap.

which is periodically modulated. It has been previously demonstrated that these modulations are the result of standing wave formation inside the nanowire[2,21] with cavity modes equidistantly spaced in reciprocal space at integer multiples of π/L, with L the nanowire length. Thus the interference peaks in the waveguided photoluminescence spectrum can be used to reconstruct the waveguide dispersion.[8,14,18] The extracted peak positions for the spectra of the two wires are plotted in Figs. 1(a) and 1(b). A striking resemblance between the experimental data and the calculated polaritonic dispersions can be seen, whereas the correspondence with the modes calculated using the Sellmeier equation is poor. This demonstrates that this method of incorporating polaritonic effects into nanowire waveguide dispersion is valid and more appropriate than the phenomenological approach for its accuracy and flexibility toward incorporating real physical parameters.

The strong coupling of light with matter while still maintaining a propagation length of at least twice the nanowire length as evidenced by the Fabry–Perot interference peaks offers interesting opportunities. For instance, the strong curvature of the dispersion implies that the group index defined as $(dE/d\beta)_{vacuum}/(dE/d\beta)_{mode}$ dramatically goes up, reducing signal velocity, which can be beneficial for the sensitivity of a nanowire optical sensor due to an increased interaction time.[22] In Fig. 3, the group index of the photonic (dashed lines) and polaritonic (solid lines) modes are shown for the same nanowires. The photonic modes show a relatively constant low group index over the investigated energy range whereas the polaritonic modes reach up to a group index of 20. Furthermore, since the slowing of the signal velocity is dependent on the presence of excitons, it can be expected that the signal retardation can be switched by using low modulation intensities since the exciton resonances can be bleached by pumping up to the CdS exciton Mott density, causing the waveguide dispersion to revert back to the purely photonic one.

In conclusion, we have shown that close to the electronic band edge in CdS nanowires, polaritonic contributions to the dielectric function need to be taken into account in order to accurately describe the experimentally observed E-β dispersion of the confined waveguide modes. Furthermore, we include these effects properly by using a physical model of coupled oscillators which takes into account the different excitons that are present in the system. The coupled oscillator model has the advantage that it fits with basic exciton oscillator parameters such as their transverse and longitudinal resonance energies, which are directly related to the oscillator strength of the transitions, and their damping. The polaritonic effect in CdS nanowires at room temperature slowed the signal propagation velocity by a factor of 7 more when compared to photonic propagation, making polaritonic nanowires promising candidates for sensing and photonic switching applications.

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