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Antireflection coating for miniband transport and Fabry–Pérot resonances in GaAs/AlGaAs superlattices
High refractive index in wurtzite GaP measured from Fabry-Pérot resonances

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We investigate the optical emission of wurtzite GaP/Al0.4Ga0.6P core/shell nanowires (NWs) transferred to a SiOx substrate to demonstrate a high degree of waveguiding of the emitted photoluminescence (PL) signal. By analysing the Fabry-Pérot mode spacing in combination with calculations of the guided modes in the NWs, we calculate a very high refractive index of bulk WZ GaP of 4.2 at a wavelength of 600 nm. The measured quality factors up to 600 indicate the excellent optical quality of the nanowire resonator. Published by AIP Publishing.

Semiconductor nanowires (NWs) offer unique features such as the possibility to switch the crystal phase from zinc blende (ZB) into wurtzite (WZ).¹,² The resulting change in the atomic stacking sequence has a strong effect on the electronic band structure, with the indirect to direct band gap transitions in materials such as gallium phosphide,³ providing an enhanced light absorption that results in a strong efficiency increase in solar water splitting devices.⁴ Recent photoluminescence (PL) studies on WZ GaP NWs provided a deeper understanding on the band gap and excitonic emission in this material system.⁵,⁶ However, additional knowledge on the WZ GaP parameters such as the refractive index would be beneficial for the design of opto-electrical devices based on WZ GaP. In this work, we report a method to estimate the refractive index for materials which are available only in the nanowire geometry. From the Fabry-Pérot (FP) resonances observed in micro-photoluminescence (PL) measurements on a single WZ GaP/Al0.4Ga0.6P core/shell nanowire, we extract an effective refractive index of \( n_{\text{WZ}}^{\text{eff}} = 3.8 \) at 600 nm. We assign the HE11 fundamental mode as the guided mode in the nanowires, from which we estimate the refractive index of bulk WZ GaP to be \( n_{\text{WZ}} = 4.2 \) at a wavelength of 600 nm. The presence of the wurtzite crystal structure results in one of the highest values for the refractive index in the visible part of the spectrum, among the III-V semiconductors.

The NWs used in this work were grown according to the recipe provided in our earlier publications,⁵,⁶ resulting in the WZ GaP/Al0.4Ga0.6P core/shell NWs with lengths above 6 \( \mu \)m and diameters of approximately 220 nm. For micro-PL measurements, the NWs were transferred to a thermally oxidized Si wafer with gold markers and cooled down to 4 K. The optical data were collected in the backscattering geometry using a NA = 0.7 Nikon 50× CR objective and a 0.30 m spectrometer equipped with 1200 g/mm grating (500 nm blazing angle). A 405 nm CW-laser was used for the excitation in combination with an f = 500 mm cylindrical lens, resulting in a rectangular spot of around 1 \( \mu \)m × 10 \( \mu \)m at the sample, while the PL images were collected with a Water CCD camera. After optical measurements, the NWs were studied by the scanning electron microscopy (SEM) to determine the wire length and the diameter.

The SEM image in Fig. 1(a) shows a WZ GaP/Al0.4Ga0.6P core/shell nanowire with the core diameter of 200 nm and the shell thickness of 10–15 nm. The length is L = 18.5 \( \mu \)m and the nanowire is transferred onto a SiOx substrate. Under laser excitation, the PL image in Fig. 1(b) is obtained where the two bright emission spots are visible at the two ends of the nanowire, which indicate that the nanowire acts as a transparent waveguide for the emitted photoluminescence.⁷–⁹

Strong modulation in the PL spectra of the nanowires due to the Fabry-Pérot (FP) interferences as a result of the multiple reflections between the wire’s end facets¹⁰ is shown in Fig. 2(a). The presence of the Al0.4Ga0.6P shell strongly enhances the FP modes in the cavity, reaching the quality factors Q in the range of 100–600, as shown by the comparison with a WZ GaP nanowire (black curve in Fig. 2(a)). The mode spacing \( \Delta \lambda \) reduces for increasing the wire length L and increases with the emission wavelength \( \lambda \), as shown in the data set covering 11 different nanowires in Fig. 2(b). The typical FP-dependence

\[
\Delta \lambda = \frac{1}{L} \frac{\lambda^2}{n_G},
\]

where \( n_G \) is the group index.

FIG. 1. (a) SEM image of a WZ GaP/Al0.4Ga0.6P core/shell nanowire of \( \sim 220 \) nm diameter and 18.5 \( \mu \)m length transferred on SiOx. The gold droplet is visible in the right end facet of the wire. (b) The PL image of the nanowire showing two bright emission spots at the end facets, while the bright region at the centre is related to the excitation spot.
with the group index $n_G$ related to the mode index $n_E$ of the nanowire waveguide mode using\(^{11}\)

$$n_G = n_E - \frac{\lambda}{\Delta \lambda},$$

is shown in the inset of Fig. 2(b).

In case that the dispersion relation for $n_E$ is known, the mode spacing in the FP cavity could provide information on the mode index $n_E$ of the WZ GaP. The Sellmeier equation\(^{7,12}\) can be used to give an approximate dispersion relation for $n_E$:

$$n_E^2 = A + B \cdot \frac{\lambda^2}{\lambda^2 - C},$$

where $A$, $B$, and $C$ are the Sellmeier coefficients. By fitting the mode spacing $\Delta \lambda$ as a function of the wavelength $\lambda$ using the Sellmeier equation, we can estimate the mode index $n_E$ for WZ GaP wires. In Fig. 3, the measured values for $n_E$ of the WZ GaP wires as a function of the wavelength are shown.

FIG. 2. (a) PL spectra of three WZ GaP/Al$_{0.4}$Ga$_{0.6}$P core/shell nanowires measured at 4 K with different lengths showing a change in the FP modes spacing. The black curve refers to a WZ GaP nanowire. (b) Plot of the FP modes spacing as a function of the emission wavelength for 11 nanowires with different lengths. Inset: plot of the FP modes spacing as a function of $1/L$ at $\lambda = 600$ nm.

FIG. 3. Plot of the refractive index $n$ as a function of wavelength at $T = 4$ K. The measured refractive index of GaP/Al$_{0.4}$Ga$_{0.6}$P core/shell nanowires is labelled as $n_{WZ}^{EXP}$, while the bulk WZ GaP refractive index estimated under the assumption of relevant HE$_{11}$ or TE$_{01}$ modes is labelled as $n_{WZ}^{HE11}$ and $n_{WZ}^{TE01}$, respectively. The dashed curves indicate the error in the obtained index. The red line indicates the refractive index $n_{ZB}$ of the bulk ZB GaP at $T = 300$ K.

(Blue spheres) together with the refractive index for the ZB GaP bulk (red line). The error bars are derived from the fit on the set of 11 wires studied. The values of the mode index $n_E$ for the WZ wires are found to be ranging from 3.6 to 4.0. However, the measured value for the mode index of the WZ wires is only a lower limit for the actual bulk refractive index. The measured mode index corresponds to the waveguide mode in the wires to which the emission has coupled. However, this mode is not fully confined inside the nanowire and also extends partially into the surrounding air,\(^{13}\) while also the AlGaP shell may reduce the $n_E$, as will be discussed later. Depending on the specific mode excited in the wire, the mode index changes, with values always lower than the refractive index of the bulk material.\(^{14}\)

From the measured effective mode index, we can estimate the value of the bulk index for WZ GaP. We use the transcendental equation for waveguide modes supported by a cylinder with a radius $a$\(^{15}\)

$$m^2 = \frac{\lambda}{\Delta \lambda} \left( \frac{1}{\lambda^2} - \frac{1}{u^2} \right)^2,$$

where $\lambda$ is the vacuum wavelength, $n_E$ is the mode index, and $n_1, n_2, u, v, \mu_1, \mu_2$ are the material indices, the mode parameters, and magnetic permeabilities of the nanowire and the surrounding, respectively. $J_m$ and $H_m$ are the Bessel and Hankel functions of the first kind. The mode parameters are defined as $u^2 = (2\pi n_1/\lambda)^2 - n^2_E$ and $v^2 = (2\pi n_2/\lambda)^2 - n^2_E$. The parameter $m$ denotes the mode order. The solutions to this
equation are the hybrid electric modes $\text{HE}_{11}$ and the transverse electric $\text{TE}_{01}$ and transverse magnetic $\text{TM}_{01}$ modes, where $i$ is the order of the solution. For each measured $n_i(\lambda)$, we fit the given equation numerically, assuming the surrounding to be vacuum and using the bulk refractive index $n_1$ as a fitting parameter. For the fundamental $\text{HE}_{11}$ mode, the electric field is tightly confined in the nanowire, having a maximum field in the nanowire’s center and an electric field polarized perpendicular to the nanowire, allowing for efficient coupling with the emitting dipoles due to the wurtzite selection rules.\(^6\) The next tightly confined mode is the $\text{TE}_{01}$ mode with a field intensity equal to zero along the nanowire axis and thus limited spatial overlap with emitting dipoles close to the nanowire’s center. In Fig. 3, the bulk refractive indices of WZ GaP are plotted considering either the $\text{HE}_{11}$ or the $\text{TE}_{01}$ as the excited mode. If we would assume the $\text{TE}_{01}$ to be the excited mode, the bulk refractive index would decrease with energy below the band gap, which would be unprecedented for bulk materials. Similar considerations hold for higher order excited modes. The overlap with the emitting dipoles (located in the core) is also larger for the $\text{HE}_{11}$ mode than the $\text{TE}_{01}$ mode. Therefore, we conclude that the fundamental $\text{HE}_{11}$ mode is most probably excited in the WZ GaP nanowires under study. Under this assumption, the deduced refractive index of bulk WZ GaP at 4 K ranges between 4.1 and 4.4 in the visible wavelength range as shown in Fig. 3. The large difference in the refractive index induced by the change in the crystalline structure is remarkable, with an increase from $n_{\text{WZ}} = 3.4$ to $n_{\text{WZ}} = 4.2$ at 600 nm. Due to the low stacking fault density in the nanowires ($\sim$1 SF/μm), with a negligible defect volume with respect to the nanowires (less than 1%), we do not expect defects to have any effect on the field confinement. By considering the temperature dependence of the refractive index,\(^{16,17}\) the 4 K value of the refractive index of ZB GaP is $n_{\text{ZB}}^\lambda = 3.3$ at 600 nm, providing an even larger difference with the WZ value. In addition, we note that the presence of the $\text{Al}_x\text{Ga}_y\text{As}_z\text{P}_w$ shell slightly reduces the measured refractive index of WZ GaP ($n_{\text{WZ}}^{\text{Al}_x\text{Ga}_y\text{As}_z\text{P}_w} = 3.24$, while $n_{\text{ZB}}^{\text{Al}_x\text{Ga}_y\text{As}_z\text{P}_w} = 3.37$ at 600 nm);\(^{18}\) therefore, the derived bulk refractive index represents only a lower limit. The high refractive index of WZ GaP suggests the availability of many different waveguide modes even for small nanowire radii, which is due to the strong confinement enabled by the high refractive index. Due to the confinement of the mode’s electric field inside the nanowire, the substrate underneath the nanowire has a negligible effect on the refractive index calculation, with variations smaller than 1%. Recently, a lower refractive index for WZ GaP nanowires with respect to ZB bulk was indirectly estimated from the reflection measurements in the UV range,\(^{19}\) leading to a similar behaviour in the visible range due to the positive sign of $\frac{dn_i}{d\lambda}$. However, a value for the WZ refractive index smaller than the ZB refractive index does not match with the experimentally observed FP mode spacing in the present work. The origin of the discrepancy is not clear, and the additional measurements on a wider wavelength range may clarify this.

The very high value for the refractive index of 4.1–4.4 for WZ GaP in the visible wavelength range demonstrated in this work is one of the highest values among other III-V semiconductor materials. Additionally, we measure high values for the quality factor of the Fabry-Perot modes up to 600. These properties would make WZ GaP a beneficial material for opto-electronic applications, enhancing light confinement and absorption, which can provide higher efficiency nanowire-based (multi-junction) solar cells,\(^{20}\) photoelectrochemical cells,\(^5\) and photodetectors.\(^{21,22}\) In addition, the transparency of WZ GaP for wavelengths above 569 nm (Ref. 6) combined with the extremely high value of the refractive index may provide applications as a 1D metamaterial,\(^{23–25}\) where a control of the optical path of the light without absorption is required.

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The authors declare no competing financial interest.

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17. See http://www.ioffe.ru/SVA/NSM/Semicond/ for information about the temperature-dependence of the refractive index in ZB GaP.