

Orientation-Dependent Optical-Polarization Properties of Single Quantum Dots in Nanowires**

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Semiconductor quantum dots (QDs) are sources of single^[1,2] and entangled photons^[3–5] and single electrons^[6] and are naturally integrated with modern semiconductor electronics. Incorporating QDs in semiconducting nanowires^[7–10] brings additional unique features such as natural alignment of vertically stacked QDs^[11] and an inherent 1D channel for charge carriers. Furthermore, the unprecedented material and design freedom makes them very attractive for novel optoelectronic devices^[12–17] and for quantum information science in general.^[18–20] Electrical contacting of single nanowires is straightforward in a horizontal geometry^[8,14–17] and vertically contacted nanowires with a wrap-around gate

have also been shown.^[21–23] However, the nanowire geometry strongly affects the polarization of photons emitted or absorbed by a nanowire QD, and thus becomes the main obstacle for applications based on intrinsic spin or polarization properties of QDs such as an electron-spin memory^[24] or generation of entangled photons.^[3] It has been shown that luminescence of pure nanowires is highly linearly polarized and the polarization direction is parallel to the nanowire axis.^[12,16,25] In this Communication we demonstrate that by directing light along the nanowire axis we can access the intrinsic polarization properties of an exciton confined in a QD in a nanowire. We introduce a theoretical model that intuitively explains our experimental findings and shows how the polarization dependence in absorption is affected by various parameters such as nanowire diameter, dielectric constant of the surroundings, and photon wavelength. As an example, we analyze the polarization properties of a Zeeman split exciton in a QD along two different directions with respect to the nanowire axis.

For our experiments we used single InAs_{0.25}P_{0.75} QDs embedded in InP nanowires grown by means of metal–organic vapor-phase epitaxy.^[26–28] A sample with a low density of wires, each containing a single dot, could be grown under appropriate growth conditions.^[8] In Figure 1a we show a scanning electron microscopy (SEM) image of an as-grown sample with a low density of nanowires, enabling optical study of a single nanowire QD. The nanowire length is about 2 μm. A transmission electron microscopy (TEM) image of a nanowire QD is shown in Figure 1c. The dot is typically 10-nm high with a diameter of 30 nm as determined by energy dispersive X-ray analysis in a TEM (Figure 1d). To simplify TEM studies, the InP section following dot growth was reduced, while for the samples used in our experiments, the dot was centered in the nanowire.

As-grown samples with vertically oriented nanowires, referred as standing nanowires, were used for experiments where the excitation (emission) was directed (measured) along the nanowire. We also transferred nanowires from the same batch to a Si substrate with a 290-nm SiO₂ top layer, referred to as lying nanowires, for the experiments where light was directed perpendicular to the nanowire. Both samples were studied under the same experimental conditions. Micro-photoluminescence (PL) studies were performed at 4.2 K. The samples were excited with a 532-nm continuous wave (cw) laser focused to a spot size of 0.6 μm using a microscope objective with a numerical aperture (NA) 0.85. The PL signal was collected

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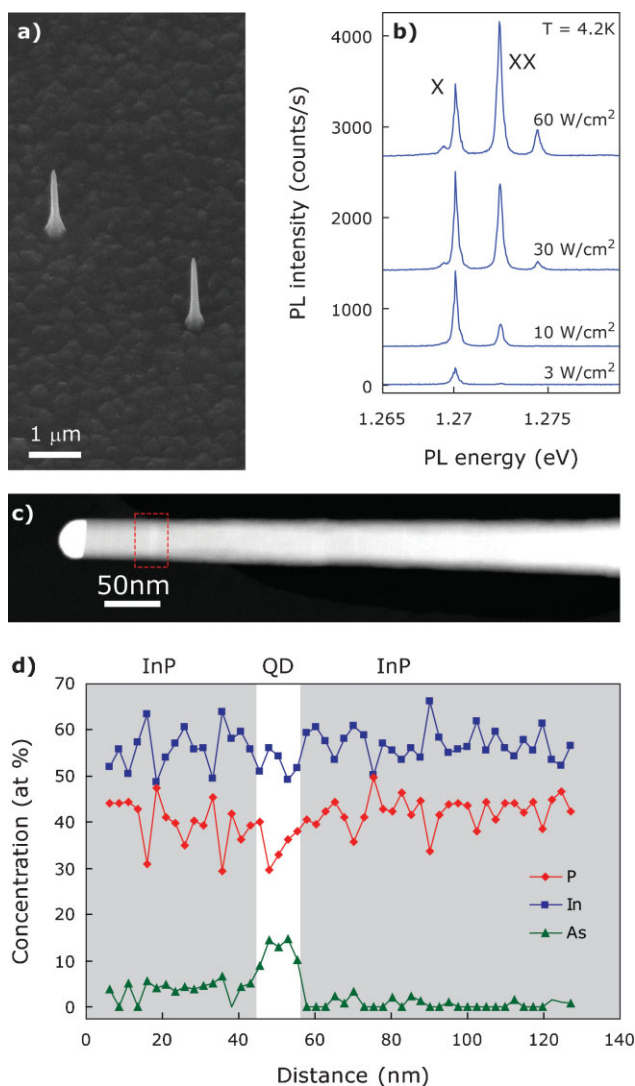


Figure 1. a) SEM image of an as-grown sample with a low density of standing nanowires. b) PL spectra of a single standing nanowire QD for various excitation powers. The two emission lines denoted with X and XX correspond to the exciton and biexciton emission, respectively. c) TEM image of a nanowire QD. The dot is indicated by the red rectangle. The low contrast of the dot is due to the low As concentration. d) Energy dispersive X-ray linescans measured in a TEM along the nanowire axis in atomic percentage. The QD is indicated in white.

using the same objective and was dispersed by a spectrometer onto a nitrogen-cooled silicon array detector, enabling 30 μeV resolution. Linear and circular polarizations were determined with a fixed polarizer together with a half- and quarter-waveplate, respectively. For absorption polarization measurements the incident light polarization was varied, while the detector was insensitive to polarization. For emission polarization measurements the incident light polarization was set parallel to the nanowire axis, while the analyzer was rotated.

In Figure 1b we show PL spectra of a single standing nanowire QD for various excitation powers. At low excitation power only one emission line is visible at 1.27 eV, denoted as X. With increasing excitation power a second emission line, denoted as XX, emerges 2.5 meV above X. The PL intensities of X and XX as a function of excitation power show that X (XX)

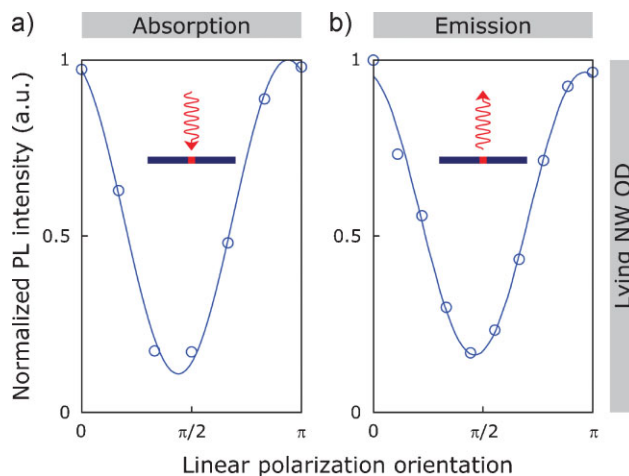


Figure 2. Integrated PL intensity of the exciton in a lying nanowire QD a) as a function of the orientation of the linear excitation polarization and b) by probing the orientation of the linear emission polarization. Zero angle of orientation was chosen to match the nanowire axis, determined by imaging. The solid curves are \cos^2 -fits used to extract the degree of linear polarization from minima and maxima of the fit. Fit parameters are amplitude, offset, and phase.

increases linearly (quadratically) with excitation power and saturates at high excitation powers. This behavior is typical for the exciton and biexciton under cw excitation. The spectral linewidths for various dots in our samples are in the range of 50–200 μeV , demonstrating high optical quality of the dots. Similar spectral characteristics are also found for lying nanowire QDs.

In Figure 2 we show the integrated PL intensity of the exciton in a lying nanowire QD as a function of linear excitation and emission polarization orientation. In Figure 2b the absorption is maximized by setting the excitation polarization parallel to the nanowire axis. However, the emission polarization is independent of the excitation polarization. We fit this data to a \cos^2 -function with amplitude, offset, and phase as fit parameters. The degree of linear polarization, $\rho = (I_{\max} - I_{\min}) / (I_{\max} + I_{\min})$, is obtained from the maxima and minima of the fit. Clearly, lying nanowire QDs show a large degree of linear polarization parallel to the nanowire for both absorption and emission.

Polarization-sensitive PL measurements were performed on a number of dots, both lying and standing, which are shown in Figure 3a. The upper two graphs show the statistics of absorption and emission for lying nanowire QDs, for which absorbed (emitted) light is directed (measured) perpendicular to the nanowire axis. The average degree of linear polarization is $\rho_{\text{abs}} = 0.78 \pm 0.06$ ($\rho_{\text{emi}} = 0.72 \pm 0.08$) for absorbed (emitted) light. This can be understood intuitively using classical electromagnetic theory by considering the dielectric properties of the nanowire QD and its surroundings.^[12] Electric fields perpendicular to the nanowire axis are reduced inside the nanowire since its diameter is much smaller than the wavelength of the light, thereby reducing the effective dielectric constant towards the dielectric constant of vacuum. Electric fields parallel to the nanowire axis are not reduced inside the nanowire, since its length is comparable or longer than the wavelength of the light. Contrary to Reference ^[29], no effect of the wurtzite structure on the polarization is observed.

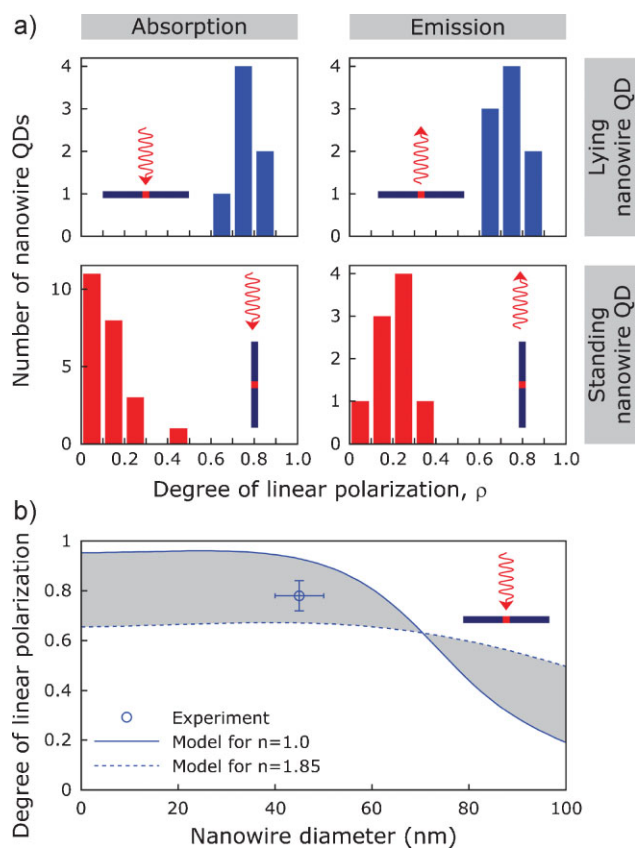


Figure 3. a) Statistics of the degree of linear polarization for lying and standing nanowire QDs. The lying nanowire QDs (shown in blue) show a high degree of linear polarization, about 0.8 and 0.7, for absorbed and emitted light, respectively. In contrast, the degree of linear polarization is low for standing nanowire QDs (shown in red), about 0.1 and 0.2 for absorbed and emitted light, respectively. Insets illustrate the nanowire orientations with respect to the light direction. b) Calculated degree of linear polarization in absorption as a function of nanowire diameter. In this case incident light is directed perpendicular to the nanowire axis as for the lying nanowire QDs. For the solid (dashed) curve an effective refractive index of $n = 1$ ($n = 1.85$) is used.

The same measurements were performed on standing nanowire QDs, for which light absorbed (emitted) by the dot is directed (measured) parallel to the nanowire axis. The results are shown in the lower two graphs of Figure 3a and give a degree of linear polarization for absorbed (emitted) light of $\rho_{\text{abs}} = 0.1$ ($\rho_{\text{emi}} = 0.2$). This low value is expected for the symmetric structure of the nanowires for light incident or emitted in the direction along the nanowire axis. In addition, the measured degree of circular polarization of the dot emission is also low (0.1).

To explain our experimental results for absorption by a lying nanowire QD more quantitatively, we use Mie theory for light scattering on dielectrics of cylindrical shape.^[30,31] The nanowire is modeled as an infinite cylinder. This approximation is valid as long as the nanowire diameter is much smaller than its length, as in our case. Furthermore, the effect of the dot is omitted in the model, since the experiments show that the polarizing effect from the nanowire dominates. We

calculate the scattering and absorption of light using the scalar wave equation in cylindrical coordinates. We use the dielectric function of InP, $\epsilon = 3.682 + 0.416i$, at the laser wavelength of 532 nm. We consider two cases: the incident electric field parallel or perpendicular to the nanowire axis. We extract the scattered field for both polarizations from the wave equation using the boundary conditions at the interface of two different dielectrics,^[32] that is, at the nanowire surface. From this field, we calculate for both cases the cross sections Q_{sca} and Q_{ext} for scattering and extinction. The absorption cross section is obtained by $Q_{\text{abs}} = Q_{\text{ext}} - Q_{\text{sca}}$ for both polarizations. To take into account the illumination of the nanowire through a microscope objective, we integrate the average absorption cross section over angles of incidence with respect to the nanowire axis comprised between $\arccos(\text{NA})$ and $\pi/2$.

Mie theory assumes the surrounding of the nanowire as a homogeneous medium, which differs from our situation where the nanowire is lying on a substrate. Therefore, to approximate the effect of the substrate, we consider the nanowire as being embedded in a medium with an effective refractive index, that is, an average of the refractive indices of the different media surrounding the nanowire: vacuum, SiO_2 , and Si. The outcome of the calculations, assuming an effective refractive index of $n_{\text{eff}} = 1.85 = 0.5n_{\text{vacuum}} + 0.25n_{\text{SiO}_2} + 0.25n_{\text{Si}}$ is represented by the dashed curve in Figure 3b. As an upper limit we consider the nanowire in vacuum, thus ignoring the substrate, which is represented by the solid curve in Figure 3b. For the calculations we use our actual excitation wavelength of 532 nm. Our experimental value lies in between the two curves, having a surrounding refractive index consisting of a mixture of vacuum, Si, and SiO_2 . As can be seen in Figure 3b, one can increase the degree of linear polarization by measuring nanowire QDs in vacuum or decrease it by increasing the nanowire diameter. However, in the latter case the advantage of the 1D channel of the device is reduced as well.

The nanowire geometry is not the only source of polarization anisotropy. Calculations by Niquet and Mojica^[33] show that the polarization properties are strongly affected by the aspect ratio of the QD dimensions due to strain originating from the lattice mismatch between the nanowire and the dot. However, in our case the strain is probably negligible due to the low phosphorus content and the main contribution to polarization anisotropy comes from the nanowire geometry.

The low degree of linear polarization for vertical nanowire QDs implies that the nanowire does not affect the polarization of photons radiated from the dot, thus allowing access to intrinsic polarization properties of nanowire QDs. Measurements in this geometry revealed a fine structure splitting of the neutral exciton, originating from anisotropic electron–hole exchange interaction.^[34] Moreover, to experimentally demonstrate the difference between the two geometries unambiguously, we measure the polarization of photons emitted from a Zeeman split exciton in a QD. In Figure 4a and b we show PL of a standing nanowire QD exciton under external magnetic field $B = 9\text{T}$ parallel to the nanowire axis, while Figure 4e and f shows PL of the same dot without external magnetic field $B = 0\text{T}$. Clearly, the exciton undergoes a Zeeman splitting. From these measurements we extract the exciton g -factor, $g = 1.3 \pm 0.1$, which is typical for our sample. In particular, polarization

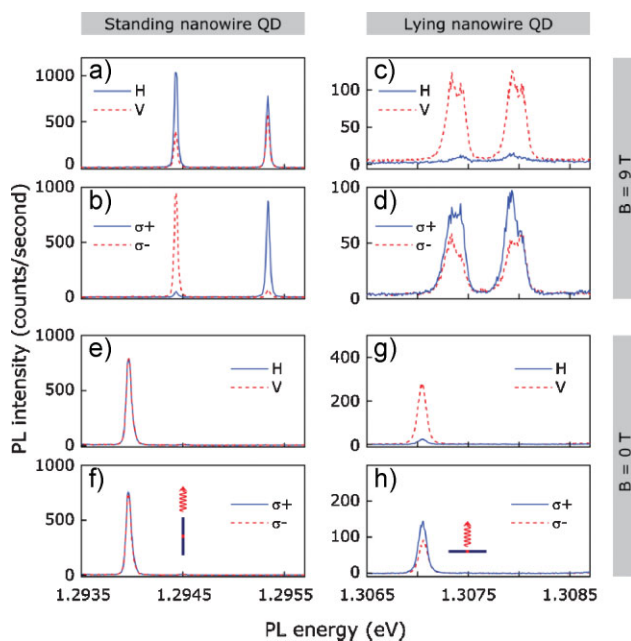


Figure 4. a) Linear and circular polarization dependence of excitonic emission from a standing nanowire QD under $B = 9\text{T}$ (a) and (b), respectively), and $B = 0\text{T}$ (e) and (f), respectively). c) (g) and d) (h) show the linear and circular polarization dependence of exciton emission from a lying nanowire QD under $B = 9\text{T}$ ($B = 0\text{T}$).

properties of the split exciton lines at 9 Tesla are shown in Figure 4a and b. As expected, the high-energy emission line is right-hand circularly polarized, while the lower-energy emission line is left-hand circularly polarized. These two polarization states, $\sigma+$ and $\sigma-$, correspond to the two different spin states of the exciton, $\downarrow\uparrow$ and $\uparrow\downarrow$, respectively, where $\uparrow(\uparrow)$ represents a spin-up electron (hole) and $\downarrow(\downarrow)$ represents a spin-down electron (hole). The reason for the difference in intensity of the horizontally and vertically polarized emission for the lower energy peak is unclear. However, in Reference [34] we show that the emission polarization of such nanowire QDs can be fully circularly polarized. At $B = 0\text{T}$ the excitonic emission is unpolarized (Figure 4e and f). These results show that in this case the emission polarization of a QD is not dominated by the nanowire geometry, and that electron–hole spin states in a dot are properly converted to the polarization states of the emitted photons. In contrast, measurements on a lying nanowire QD under external magnetic field $B = 9\text{T}$, represented in Figure 4c and d, show that the two emission lines of a Zeeman split exciton are strongly linearly polarized with respect to the nanowire axis, as expected. At $B = 0\text{T}$ the exciton emission shows the same linear polarization, as can be seen in Figure 4g and h. The exciton g -factor, $g = 1.2 \pm 0.1$, is similar to the standing case. We note that the Zeeman split emission lines possibly consist of two peaks. These could arise from dark excitons, due to symmetry-breaking, [35] or due to heavy-light hole-mixing effects, [36] since the magnetic field B is perpendicular to the symmetry axis in this case. This is currently under investigation.

To conclude, we have correlated the polarization of light absorbed and emitted by a QD embedded in a nanowire with its propagation direction with respect to the nanowire axis. We

found that the polarization of the absorbed (emitted) light, when directed (measured) perpendicular to the nanowire axis, is affected by the nanowire geometry and is strongly linearly polarized along the nanowire. In contrast, this is not the case for the configuration where the absorbed (emitted) light is directed (measured) parallel to the nanowire axis. In this configuration the intrinsic polarization of the nanowire QD can be accessed. We have demonstrated this by measuring the polarization of photons radiated from a Zeeman split exciton in a nanowire QD with the optical path aligned along the nanowire axis as well as perpendicular to the nanowire axis.

Keywords:

excitons · nanowires · photoluminescence · quantum dots

- [1] P. Michler, A. Kiraz, C. Becher, W. V. Schoenfeld, P. M. Petroff, L. Zhang, E. Hu, A. Imamoglu, *Science* **2000**, *290*, 2282.
- [2] C. Santori, M. Pelton, G. Solomon, Y. Dale, Y. Yamamoto, *Phys. Rev. Lett.* **2001**, *86*, 1502.
- [3] N. Akopian, N. H. Lindner, E. Poem, Y. Berlatzky, J. Avron, D. Gershoni, B. D. Gerardot, P. M. Petroff, *Phys. Rev. Lett.* **2006**, *96*, 130501.
- [4] R. J. Young, M. R. Stevenson, P. Atkinson, K. Cooper, D. A. Ritchie, A. J. Shields, *New J. Phys.* **2006**, *8*, 29.
- [5] R. Hafenbrak, S. M. Ulrich, P. Michler, L. Wang, A. Rastelli, O. G. Schmidt, *New J. Phys.* **2007**, *9*, 315.
- [6] A. Zrenner, A. Beham, S. Stufler, F. Findeis, M. Bichler, G. Abstreiter, *Nature* **2002**, *418*, 612.
- [7] M. T. Borgström, V. Zwiller, E. Muller, A. Imamoglu, *Nano Lett.* **2005**, *5*, 1439.
- [8] E. D. Minot, F. Kelkensberg, M. van Kouwen, J. A. van Dam, L. P. Kouwenhoven, V. Zwiller, M. T. Borgström, O. Wunnicke, M. A. Verheijen, E. P. A. M. Bakkers, *Nano Lett.* **2007**, *7*, 367.
- [9] M. Tchernycheva, G. E. Cirlin, G. Patriarche, L. Travers, V. Zwiller, U. Perinetti, J. C. Harmand, *Nano Lett.* **2007**, *7*, 1500.
- [10] J. Renard, R. Songmuang, C. Bougerol, B. Daudin, B. Gayral, *Nano Lett.* **2008**, *8*, 2092.
- [11] T. Lundström, W. Schoenfeld, H. Lee, P. M. Petroff, *Science* **1999**, *286*, 2312.
- [12] J. Wang, M. S. Gudiksen, X. Duan, Y. Cui, C. M. Lieber, *Science* **2001**, *293*, 1455.
- [13] M. H. Huang, S. Mao, H. Feick, H. Yan, Y. Wu, H. Kind, E. Weber, R. Russo, P. Yang, *Science* **2001**, *292*, 1897.
- [14] X. Duan, Y. Huang, Y. Cui, J. Wang, C. M. Lieber, *Nature* **2001**, *409*, 66.
- [15] X. Duan, Y. Huang, R. Agarwal, C. M. Lieber, *Nature* **2003**, *421*, 241.
- [16] H. Pettersson, J. Trägårdh, A. I. Persson, L. Landin, D. Hessman, L. Samuelson, *Nano Lett.* **2006**, *6*, 229.
- [17] J. Bao, M. A. Zimmler, F. Capasso, X. Wang, Z. F. Ren, *Nano Lett.* **2006**, *6*, 1719.
- [18] M. T. Björk, C. Thelander, A. E. Hansen, L. E. Jensen, M. W. Larsson, L. R. Wallenberg, L. Samuelson, *Nano Lett.* **2004**, *4*, 1621.
- [19] Z. Zhong, Y. Fang, W. Lu, C. M. Lieber, *Nano Lett.* **2005**, *5*, 1143.
- [20] A. Pfund, I. Shorubalko, R. Leturcq, K. Ensslin, *Appl. Phys. Lett.* **2006**, *89*, 252106.
- [21] H. T. Ng, J. Han, T. Yamada, P. Nguyen, Y. P. Chen, M. Meyyappan, *Nano Lett.* **2004**, *4*, 1247.
- [22] T. Bryllert, L. E. Wernersson, T. Lowgren, L. Samuelson, *Nanotechnology* **2006**, *17*, S227.
- [23] V. Schmidt, H. Riel, S. Senz, S. Karg, W. Riess, U. Gösele, *Small* **2006**, *2*, 85.
- [24] M. Kroutvar, Y. Ducommun, D. Heiss, M. Bichler, D. Schuh, G. Abstreiter, J. J. Finley, *Nature* **2004**, *432*, 81.

- [25] O. L. Muskens, J. Treffers, M. Forcales, M. T. Borgström, E. P. A. M. Bakkers, J. G. Rivas, *Opt. Lett.* **2007**, *32*, 2097.
- [26] K. Hiruma, T. Katsuyama, K. Ogawa, M. Koguchi, H. Kakibayashi, G. P. Morgan, *Appl. Phys. Lett.* **1991**, *59*, 431.
- [27] L. J. Lauhon, M. S. Gudiksen, D. Wang, C. M. Lieber, *Nature* **2002**, *420*, 57.
- [28] M. A. Verheijen, G. Immink, T. de Smet, M. T. Borgström, E. P. A. M. Bakkers, *J. Am. Chem. Soc.* **2006**, *128*, 1353.
- [29] A. Mishra, L. V. Titova, T. B. Hoang, H. E. Jackson, L. M. Smith, J. M. Yarrison-Rice, Y. Kim, H. J. Joyce, Q. Gao, H. H. Tan, C. Jagadish, *Appl. Phys. Lett.* **2007**, *91*, 263104.
- [30] C. F. Bohren, D. R. Huffman, *Absorption and Scattering of Light by Small Particles*, Wiley-VCH, Weinheim, Germany **1998**.
- [31] H. E. Ruda, A. Shik, *J. Appl. Phys.* **2006**, *100*, 024314.
- [32] J. D. Jackson, *Classical Electrodynamics*, Wiley, New York **1999**.
- [33] Y. M. Niquet, D. C. Mojica, *Phys. Rev. B* **2008**, *77*, 115316.
- [34] M. H. M. van Weert, N. Akopian, U. Perinetti, M. P. van Kouwen, R. E. Algra, M. A. Verheijen, E. P. A. M. Bakkers, L. P. Kouwenhoven, V. Zwiller, *Nano Lett.* **2009**, *9*, 1989-1993.
- [35] M. Bayer, G. Ortner, O. Stern, A. Kuther, A. A. Gorbunov, A. Forchel, P. Hawrylak, S. Fafard, K. Hinzer, T. L. Reinecke, S. N. Walck, J. P. Reithmaier, F. Klopf, F. Schäfer, *Phys. Rev. B* **2002**, *65*, 195315.
- [36] K. F. Karlsson, V. Troncale, D. Y. Oberli, A. Malko, E. Pelucchi, A. Rudra, E. Kapon, *Appl. Phys. Lett.* **2006**, *89*, 251113.

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