

# Modification of the photoluminescence anisotropy of semiconductor nanowires by coupling to surface plasmon polaritons

O. L. Muskens,<sup>1</sup> J. Treffers,<sup>1</sup> M. Forcales,<sup>1</sup> M. T. Borgström,<sup>2</sup> E. P. A. M. Bakkers,<sup>2</sup> and J. Gómez Rivas<sup>1,\*</sup>

<sup>1</sup>FOM Institute for Atomic and Molecular Physics AMOLF, c/o Philips Research Laboratories, High Tech Campus 4, 5656 AE, Eindhoven, The Netherlands

<sup>2</sup>Philips Research Laboratories, High Tech Campus 4, 5656 AE, Eindhoven, The Netherlands

\*Corresponding author: [rivas@amolf.nl](mailto:rivas@amolf.nl)

Received April 12, 2007; accepted May 21, 2007;  
posted May 29, 2007 (Doc. ID 81968); published July 18, 2007

We demonstrate efficient modification of the polarized light emission from single semiconductor nanowires by coupling this emission to surface plasmon polaritons on a metal grating. The polarization anisotropy of the emitted photoluminescence from single nanowires is compared for wires deposited on silica, a flat gold film, and a shallow gold grating. By varying the orientation of the nanowire with respect to the grating grooves, the large intrinsic polarization anisotropy can be either suppressed or enhanced. This modification is interpreted by the appearance of an additional emission channel induced by surface plasmon polaritons and their conversion to *p*-polarized radiation at the grating. © 2007 Optical Society of America  
OCIS codes: 240.6680, 050.1950, 260.3800.

Semiconductor nanowires are considered important building blocks for nanoscale optical and optoelectronic devices [1,2]. The large shape anisotropy of the quasi-one-dimensional structures combined with the strong refractive index contrast of semiconductors, such as InP results in highly polarized absorption and emission properties of individual nanowires [3]. Single nanowires have been used as polarization-dependent emitters and sensors [3–6], while collections of wires have been shown to exhibit artificial birefringence [7]. Recently, there have been developments toward the incorporation of nanowires into more complex photonic systems aimed at the modification control of the emission properties of the single nanowires. Examples are integration of nanowires with optical waveguides and one-dimensional photonic crystals [8], the construction of nanowire ring resonators [9], and photonic networks [10].

In this Letter, we demonstrate an efficient route for the modification of the nanowire emission properties via coupling to surface plasmon polaritons (SPPs) on a metal grating. By varying the orientation of the nanowire with respect to the grating grooves, we show that the large intrinsic polarization anisotropy of single nanowires can be either suppressed or enhanced. We also investigate the redirection of the emission of the nanowire grating coupled system. Metal films supporting SPPs have been used to modify the photoemission from dyes [11], quantum dots [12,13], quantum wells [14,15], and rare earth ions [16]. Since SPPs are longitudinal surface waves, their conversion to photons on a one-dimensional grating results in *p*-polarized radiation. Therefore, SPP-enhanced luminescence results in modification of the polarization distribution of the photoemission [13].

In this study we use *n*-doped InP:S nanowires with a length between 2 and 3  $\mu\text{m}$  and a diameter of 20 nm at their top and about 50 nm at their base. Nanowires were randomly deposited onto a substrate

and studied with an optical microscope equipped with a high-efficiency CCD-spectrometer. An argon-ion laser operating at 457.9 nm was focused onto a spot of several  $\mu\text{m}$  in diameter using a 50 $\times$ , 0.75 N.A. objective. The excitation intensity was maintained at a low value (i.e., below  $\sim 100 \text{ W/cm}^2$ ) to avoid spurious effects due to nanowire heating. The photoluminescence was collected using the same objective and imaged onto a spectrometer. The polarization state of both the excitation and emission was controlled independently using polarization filters.

We start by investigating the polarization anisotropy of nanowires deposited on a planar silica substrate. Figure 1(a) shows two photoluminescence spectra of a single InP nanowire on silica, in which the polarization of the incident beam was oriented parallel (solid line) and perpendicular (dashed line) to the nanowire axis, while the luminescence for both polarizations was detected. We define the absorption anisotropy ratio  $\eta_{\text{abs}} = I_{\text{abs}_{\parallel}} / I_{\text{abs}_{\perp}}$ , taking the maximum peak intensities of the two spectra. For the particular nanowire under study, an absorption ratio  $\eta_{\text{abs}}$  of 36 was obtained. The strong polarization anisotropy of absorption reproduces the results of Wang *et al.* obtained on a single InP nanowire of 20 nm diameter [3]. In Fig. 1(b), we display the photoluminescence spectra of the same nanowire of Fig. 1(a) but now excited with a polarization parallel to the nanowire axis and measured for a polarization parallel (solid line) and perpendicular (dashed line) to the wire axis. The emission anisotropy ratio  $\eta_{\text{em}} = I_{\text{em}_{\parallel}} / I_{\text{em}_{\perp}}$  for this nanowire reaches a value of 7.3. In their explanation of the photoluminescence anisotropy Wang *et al.* [3] did not separately consider  $\eta_{\text{em}}$  and  $\eta_{\text{abs}}$ , but explained the anisotropy using the quasi-static polarizability of a cylinder. A refinement in this description has been made by Ruda *et al.* [17], who included the random distribution of the emission dipoles in the anisotropy. The resulting difference between absorption and emission anisotropies is in agreement with

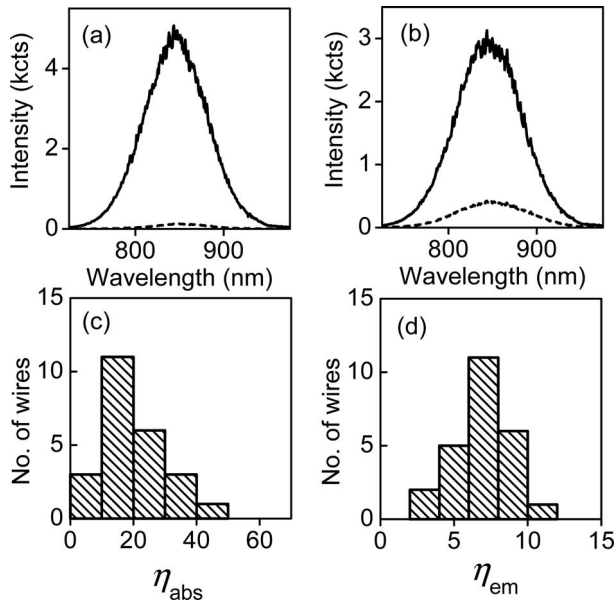


Fig. 1. (a) Photoluminescence spectra of a single InP nanowire on silica for polarizations of the excitation light parallel (solid line) and perpendicular (dashed line) to the nanowire axis. (b) Spectra of the same wire excited with a polarization parallel to its axis and detected with a polarization parallel (solid line) and perpendicular (dashed line). (c) and (d) Distributions of the absorption and emission anisotropy ratios of 25 single InP nanowires on a silica substrate.

the experimental observations. To obtain average values of the photoluminescence anisotropies, we have measured 25 different nanowires from the same batch. Figures 1(c) and 1(d) display the histograms of the absorption [Fig. 1(c)] and emission [Fig. 1(d)] anisotropy. We find a mean  $\eta_{\text{abs}}$  of 21.5 with a standard deviation of 12 and a mean  $\eta_{\text{em}}$  of 6.9 with a standard deviation of 1.9. The large standard deviations in  $\eta_{\text{abs}}$  and  $\eta_{\text{em}}$  are attributed to variations in the nanowire diameter.

Metal gratings were fabricated using electron-beam (e-beam) lithography and lift-off on top of a 300 nm layer of gold. A shallow gold grating of 30 nm height was produced with a period,  $d$ , of 630 nm. We verified the grating assisted coupling of light to SPPs by measuring the specular reflection of a collimated beam from a halogen lamp at different angles. These measurements are shown in Fig. 2(a), where a contour plot of the specularly reflected intensity onto the grating normalized by the reflection of a silver mirror is plotted as a function of the wavelength and the angle of incidence  $\theta$ . In these measurements the incident beam was  $p$ -polarized. The dark bands in Fig. 2(a) are due to the coupling of diffracted orders by the grating to SPPs, leading to the concomitant reduction of the specular reflection. This coupling occurs when the  $k$ -vector of the diffracted wave equals the  $k$ -vector of the SPP [18],

$$\mathbf{k}_0 \sin \theta + n\mathbf{G} = \pm \mathbf{k}_{\text{SPP}}, \quad (1)$$

where  $\mathbf{k}_0$  is the wave vector of the incident wave,  $\mathbf{k}_{\text{SPP}} = \mathbf{k}_0 \sqrt{\epsilon_m / (\epsilon_m + 1)}$  is the wave vector of the SPP,

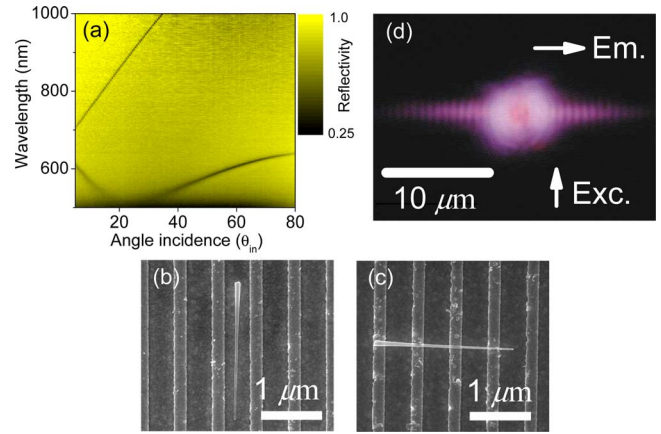


Fig. 2. (Color online) (a) Angle-dependent,  $p$ -polarized reflectivity from a gold grating, showing coupling of light to SPPs (dark regions). (b) and (c) SEM images of individual nanowires aligned parallel (b) and perpendicular (c) to the grooves of a gold grating. (d) Image of the photoluminescence of a nanowire on a gold grating.

being  $\epsilon_m$  the permittivity of gold,  $\mathbf{G} = 2\pi/d$  is the reciprocal lattice vector and  $n$  is an integer indicating the diffractive order. Next to the  $n = -1$  order above 700 nm, the  $n = 1$  and  $-2$  appear at short wavelengths and at small and large angles, respectively. As we will see the photoluminescence around 850 nm of InP nanowires on top of gratings couples to the  $n = -1$  mode.

The nanowires were deposited onto Au gratings. Figures 2(b) and 2(c) show scanning electron microscopy (SEM) images of two different nanowires oriented respectively parallel and perpendicular to the grating grooves. In Fig. 2(d) an image of the photoluminescence of a nanowire aligned parallel to the grating is shown. The arrows indicate the polarization direction of the excitation beam (this direction is parallel to the nanowire axis) and of the detection. The SPPs generated by the nanowire and coupled by the grating into radiation are identified as the lateral emission around the central bright spot, which corresponds to the direct emission from the nanowire.

For each configuration (nanowires oriented parallel and perpendicular to the grating) we have measured the polarization ratio of the emitted light from 25 single nanowires. The resulting distributions of  $\eta_{\text{em}}$  are shown in Figs. 3(a) and 3(b). The two configurations clearly yield very different distributions with mean anisotropies (and standard deviations) of  $3.7(\pm 1.7)$  and  $11.4(\pm 2.7)$  for the parallel and perpendicular configurations, respectively. Additional measurements of the polarization ratio of nanowires on a planar gold film (not shown) show a distribution similar to the wires on silica in Fig. 1(b).

The modification of the emission properties of the nanowires by the grating is explained as follows. The nanowire emission generates SPPs via the quasi-static near-field interaction with the free-electrons in the metal. The SPPs are scattered by the grating into  $p$ -polarized radiation, which adds to the direct luminescence from the nanowire. For a nanowire parallel to the grating grooves, this contribution adds to the weak direct emission polarized perpendicular to the

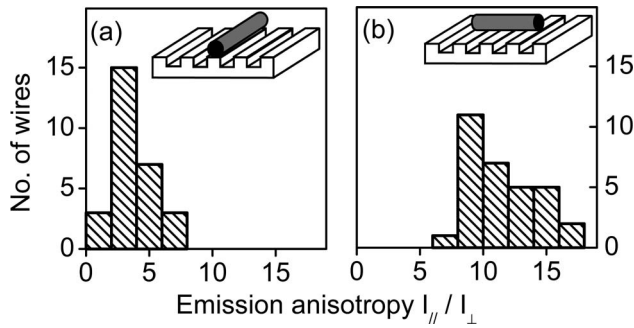


Fig. 3. Distributions of the emission anisotropy ratio  $\eta_{em}$  for nanowires deposited on a gold grating with their axes parallel to the grooves (a), and on a grating perpendicular to the grooves (b) (Configurations drawn as inset).

wire axis, which leads to a reduction of the polarization contrast  $\eta_{em}$  to 3.7. For the configuration where the nanowires are oriented perpendicular to the grooves, the SPP adds to the already strong polarization component parallel to the wire axis, resulting in an increase of  $\eta_{em}$  to 11.4. To discard a possible effect of the excitation on the anisotropy of the photoluminescence, we measured the absorption anisotropy ratio  $\eta_{abs}$  for wires oriented parallel and perpendicular to the grating. In these measurements (not shown here) we did not observe any difference in  $\eta_{abs}$  for different nanowire orientations.

According to Eq. (1), coupling of SPPs and light using a metal grating modifies the polarization of the emitted light and is expected to result in an angular redistribution of this emission. To check this redistribution, we measured the polarization anisotropy of individual nanowires with different NAs in the optical microscope. If SPPs are coupled out by the grating at larger angles than the acceptance angle—defined by the NA of the objective—light is not collected or detected, which affects the measured emission anisotropy. Figure 4 shows results for three different configurations: a nanowire parallel to a grating with period of 630 nm (triangles), a nanowire parallel to a grating with period of 480 nm (circles), and a nanowire on a plane metal film (diamonds). For compari-

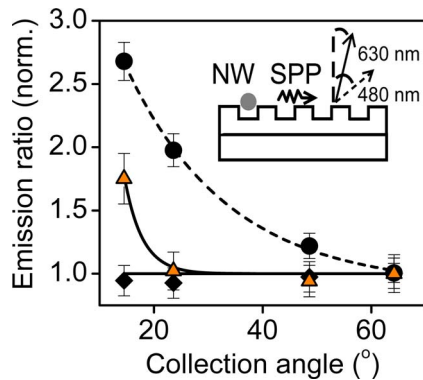


Fig. 4. (Color online) Normalized emission anisotropies as a function of collection angle for single nanowires parallel to the grooves of a  $d=480$  nm grating (circles), parallel to the grooves of a  $d=630$  nm grating (triangles), and on a planar gold film (diamonds). The lines are guides for the eyes. Inset: schematic drawing of the reradiation of SPP for the two grating periods.

son, we have normalized the three curves to their values at the collection angle of  $64.15^\circ$  (NA 0.9). For the planar film, the ratio is independent of the collection angle. For the nanowire on the 630 nm grating, we observe only a small deviation around  $15^\circ$  (NA 0.25). This corresponds well to the expected reradiation angle of the SPP around  $\theta \approx 20^\circ$  for  $\lambda \approx 850$  nm in the dispersion relation of Fig. 2(a). For the 480 nm grating, the outcoupling of SPP occurs at a larger angle of around  $50^\circ$  [see Eq. (1)]. Indeed, we observe a more gradual variation of the emission ratio for the nanowire placed on the 480 nm grating.

In conclusion, we have shown control over the emission properties of single InP nanowires using coupling to SPPs on a metal grating. Since nanowires are important new building blocks in nanophotonics, control over their optical properties will be of importance for future applications.

We thank J. A. Sánchez-Gil and V. Giannini for the fruitful discussion and G. Immink and B. Ketelaars for their technical assistance. This work was supported by the Netherlands Foundation “Fundamenteel Onderzoek der Materie (FOM)” and the “Nederlandse Organisatie voor Wetenschappelijk Onderzoek (NWO)” and is part of an industrial partnership program between Philips and FOM.

## References

1. Y. Li, F. Qian, J. Xiang, and C. M. Lieber, *Mater. Today* **9**, 18 (2006).
2. P. J. Pauzauskis and P. Yang, *Mater. Today* **18**, 36 (2006).
3. J. Wang, M. K. Gudiksen, X. Duan, Y. Cui, and C. M. Lieber, *Science* **293**, 1455 (2001).
4. H. Pettersson, J. Trägårdh, A. I. Persson, L. Landin, D. Hessman, and L. Samuelson, *Nano Lett.* **6**, 229 (2006).
5. Z. Fan, P. Chang, J. G. Lu, E. C. Walter, R. M. Penner, C. Lin, and H. P. Lee, *Appl. Phys. Lett.* **85**, 6128 (2004).
6. O. Hayden, R. Agarwal, and C. M. Lieber, *Nat. Mater.* **5**, 352 (2006).
7. O. L. Muskens, M. T. Borgström, E. P. A. M. Bakkers, and J. Gómez Rivas, *Appl. Phys. Lett.* **89**, 233117 (2006).
8. C. J. Barrelet, J. Bao, M. Loncar, H.-G. Park, F. Capasso, and C. M. Lieber, *Nano Lett.* **6**, 11 (2006).
9. P. J. Pauzauskis, D. J. Sirbuly, and P. Yang, *Phys. Rev. Lett.* **96**, 143903 (2006).
10. M. Law, D. J. Sirbuly, J. C. Johnson, J. Goldberger, R. J. Saykally, and P. Yang, *Science* **27**, 1269 (2004).
11. P. Andrew and W. L. Barnes, *Phys. Rev. B* **64**, 125405 (2001).
12. K. T. Shimizu, B. R. Fisher, H. J. Eisler, and M. G. Bawendi, *Phys. Rev. Lett.* **89**, 117401 (2002).
13. J. Zhang, Y.-H. Ye, X. Wang, P. Rochon, and M. Xiao, *Phys. Rev. B* **72**, 201306 (2005).
14. I. Gontijo, M. Boroditsky, E. Yablonovitch, S. Keller, U. K. Mishra, and S. P. DenBaars, *Phys. Rev. B* **60**, 11564 (1999).
15. K. Okamoto, I. Niki, A. Shvartser, Y. Narukawa, T. Mukai, and A. Scherer, *Nat. Mater.* **3**, 601 (2004).
16. J. Kalkman, C. Strohhofer, B. Gralak, and A. Polman, *Appl. Phys. Lett.* **83**, 30 (2003).
17. H. E. Ruda and S. Shik, *Phys. Rev. B* **72**, 115308 (2005).
18. S. C. Kitson, W. L. Barnes, and J. R. Sambles, *Opt. Commun.* **122**, 147 (1996).