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

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Abstract

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, on a flat gold film, and on 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 interpret by the appearance of an additional emission channel induced by surface plasmon polaritons and their conversion to p-polarized radiation at the grating.

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Semiconductor nanowires are considered as important building blocks for nanoscale optical and optoelectronical devices.[1] The large shape anisotropy of the quasi-one dimensional structures combined with the strong refractive index contrast of semiconductors like InP results in highly polarized absorption and emission properties of individual nanowires.[2] Single nanowires have been used as polarization dependent emitters and sensors[2, 3, 4, 5], while collections of wires have been shown to exhibit artificial birefringence.[6] 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[7], the construction of nanowire ring resonators[8], and photonic networks.[9]

In this article, 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[10], quantum dots[11, 12], quantum wells[13, 14], and rare earth ions.[15] Emphasis was put on the enhancement of emission efficiency via modification of the local density of states. 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.[12]

In this study we use n-doped InP:S nanowires with a length between 2 and 3 μm an 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. A argon-ion laser operating at 457.9 nm was focused onto a spot of several μm in diameter using a 50×, 0.75 N.A. objective. The excitation intensity was maintained at a low value (i.e. below $\sim$100 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 $\eta_{\rm abs} = I_{\rm abs_\parallel}/I_{\rm abs_\perp}$, taking the maximum peak intensities of the two spectra. For the particular nanowire under study, an absorption ratio $\eta_{\rm abs}$ 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. [2] In Fig. 1(b), we display the photoluminescence spectra of the same nanowire of (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 $\eta_{\rm em} = I_{\rm em_\parallel}/I_{\rm em_\perp}$ reaches for this nanowire a value of 7.3. In their explanation of the photoluminescence anisotropy Wang et al. [2] did not consider separately $\eta_{\rm em}$ and $\eta_{\rm abs}$, but explained the anisotropy using the quasistatic polarizability of a cylinder.

A refinement in this description has been made by Ruda et al. [16], who included the random distribution of the emission dipoles in the anisotropy. The resulting difference between absorption and emission anisotropies is in agreement with 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 (d) display the histograms of the absorption (c) and emission (d) anisotropy. We find a mean $\eta_{\rm abs}$ of 21.5 with a standard deviation of 12, and a mean $\eta_{\rm em}$ of 6.9 with a standard deviation of 1.9. The large standard deviation in $\eta_{\rm abs}$ and $\eta_{\rm em}$ are attributed to variations in the nanowire diameter.

Metal gratings were fabricated using 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 θ. 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 surface plasmon polaritons, 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 [17],

$$\mathbf{k}_0 \sin\theta + n\mathbf{G} = \pm\mathbf{k}_{\rm SPP} \ , \tag{1}$$

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

vector of the SPP, being $\epsilon_{\rm m}$ 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. Figure 2(b,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 in the figure indicate the polarization direction of the excitation beam (being this direction 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.

We have measured for each configuration (nanowires oriented parallel and perpendicular to the grating) the polarization ratio of the emitted light from 25 single nanowires. The resulting distributions of $\eta_{\rm em}$ are shown in Fig. 3(a,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 similar distribution as for the wires on silica in Fig. 1(b).

The strong difference in polarization anisotropy between the nanowires oriented parallel and perpendicular to the grating grooves indicates that the grating significantly modifies

the emission properties of the nanowires. The nanowire emission generates SPPs via the quasistatic 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 wire axis, which leads to a reduction of the polarization contrast η_{em} to 3.7. For the configuration where the nanowires are oriented perpendicular to the grooves, the SPP add to the already strong polarization component parallel to the wire axis, resulting in an increase of η_{em} to 11.4. From these values it appears that the amount of light generated via SPP is a significant fraction of the total emitted intensity. To discard a possible effect of the excitation on the anisotropy of the photoluminescence, we measured the absorption anisotropy ratio η_{abs} for wires oriented parallel and perpendicular to the grating. In these measurements (not shown here) we did not observe any difference in η_{abs} for different nanowire orientations.

According to Eq. 1, coupling of SPPs and light using a metal grating does not only modify the polarization of the emitted light, it is also expected to result in an angular redistribution of this emission. To check this redistribution, we measured the polarization anisotropy of individual nanowires with different numerical apertures NA in the optical microscope. If SPPs are coupled out by the grating at larger angles the acceptance angle defined by the NA of the objective, they are not collected and detected, affecting 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 comparison, we

have normalized the three curves to their values at the collection angle of 64.15° (NA 0.9). For the planar film, the ratio is independent of the collection angle of the detector, as is expected since the nanowires can be considered as a small dipole source in at least two of its dimensions. For the nanowire on the 630-nm grating, we observe only a small deviation around 15° (NA 0.25). This corresponds well to the expected reradiaton angle of the SPP around $\theta \simeq 20^o$ for $\lambda \simeq 850$ 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° (see Eq. 1). Indeed, we observe a more gradual variation of the emission contrast 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 surface plasmon polaritons 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.

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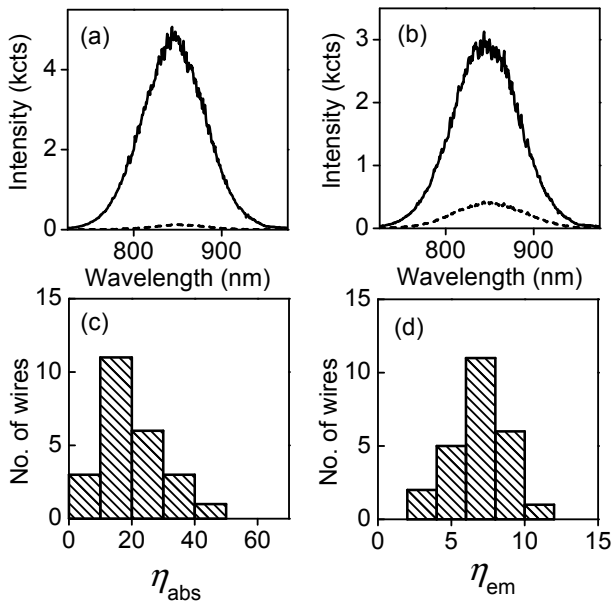

FIG. 1: (a) Photoluminescence spectra of a single InP nanowire on silica for polarizations of the excitation light parallel (solid line) and perpendicular (dashed line) to the nanowire axis. (b) Spectra of the same wire excited with a polarization parallel to its axis and detected with a polarization 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.

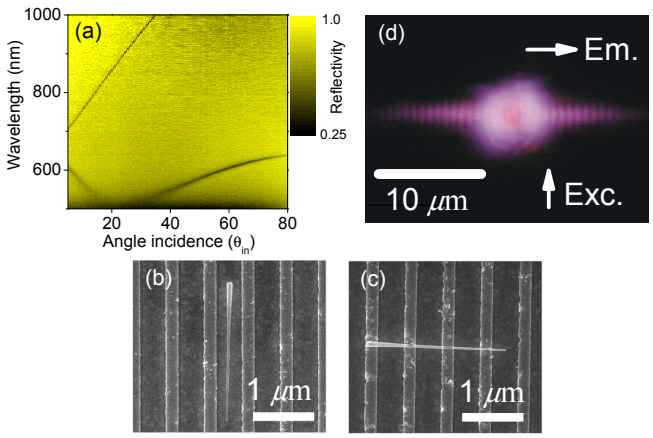

FIG. 2: (color online) (a) Angle-dependent, p-polarized reflectivity from a gold grating, showing coupling of light to SPPs (dark regions). (b,c) Scanning Electron Microscopy images of individual nanowires aligned parallel (b) and perpendicular (c) to the grooves of a gold grating. (d) Image of the photoluminescence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

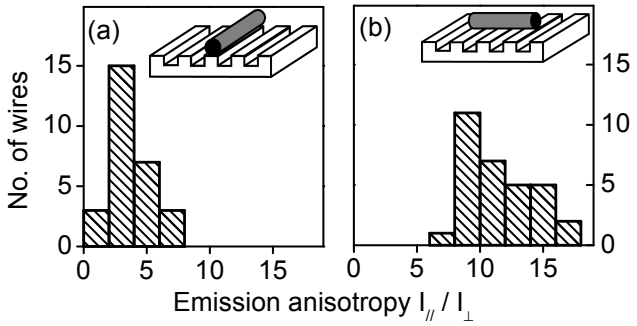

FIG. 3: Distributions of the emission anisotropy ratio $\eta_{\rm em}$ for nanowires deposited on a gold grating with their axes parallel to the grooves (a), and on a grating perpendicular to the grooves (b) (Configurations drawn as inset).

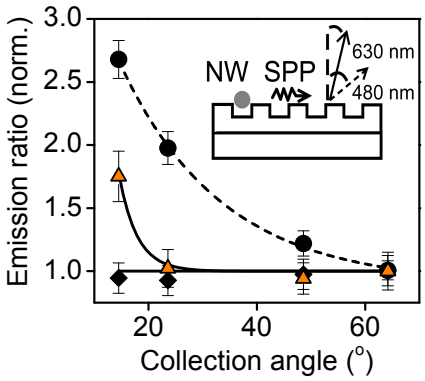

FIG. 4: Normalized emission anisotropies as a function of collection angle for single nanowires parallel to the grooves of a $d = 480$ nm grating (circles), parallel to the grooves of a $d = 630$ nm grating (triangles), and on a planar gold film (diamonds). The lines are guides to the eye. Inset: schematic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