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Radiation Channels Close to a Plasmonic Nanowire Visualized by Back Focal Plane Imaging

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Abstract

We investigated the angular radiation patterns, a key characteristic of an emitting system, from individual silver nanowires decorated with rare earth ion-doped nanocrystals. Back focal plane radiation patterns of the nanocrystal photoluminescence after local two-photon excitation can be described by two emission channels: Excitation of propagating surface plasmons in the nanowire followed by leakage radiation and direct dipolar emission observed also in the absence of the nanowire. Theoretical modeling reproduces the observed radiation patterns which strongly depend on the position of excitation along the nanowire. Our analysis allows to estimate the branching ratio into both emission channels and to determine the diameter dependent surface plasmon quasi-momentum, important parameters of emitter-plasmon structures.

Keywords

Plasmonics; metallic nanowires; rare earth ions; back focal plane imaging; up-conversion

Propagating surface plasmon polaritons (SPPs) are electromagnetic waves bound to a metal-dielectric interface. They offer the distinguished possibility to concentrate light to subwavelength scales and transport this energy over a length several magnitudes larger.^{1–4} These properties provide the basis for plasmonics, a very active research area aiming at optical device miniaturization and the integration of optics and electronics on a single chip.^{5,6} Metallic nanowires (NWs) have drawn particular attention as plasmonic building blocks due to their successful implementation as waveguides,^{7–9} routers and logic gates.^{10–12} SPPs on metallic NWs have been investigated by direct visualization,¹³ using a scanning aperture probe,¹⁴ by electrical detection¹⁵ as well as by calculations.¹⁶ A key step in plasmonic applications of NWs is the coupling of the initial energy source to the NW and the contributing energy relaxation pathways.^{17,18} Importantly, subwave-length light confinement by the SPPs can be used to enhance the interaction between objects and light.¹⁹ This coupling and the excitation and propagation of SPPs have been experimentally visualized by leakage radiation microscopy^{20–22} combined with imaging of the back focal plane (BFP) for a variety of plasmonic structures and devices.^{23–25}

In this manuscript we studied the coupling of the emission from rare earth doped nanocrystals to SPP modes in silver NWs on glass. We use the ability of these nanocrystals to exhibit stable, non-bleaching upconverted photoluminescence (PL) on the anti-Stokes side

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of the laser energy²⁶ to avoid temporal intensity fluctuations and to exclude any background contribution from laser scattering, metal luminescence or the sample substrate. Importantly, since two-photon excitation requires high excitation densities it will only be efficient at the position of the laser focus. Two-photon excitation of NCs located outside of the focus along the NW mediated by propagating surface plasmons can be neglected due to limited light-plasmon coupling and plasmon damping. In addition, due to the extremely small oscillator strength of the nanocrystals at the emission wavelength reflected in a radiative lifetime of the f-f transitions in the μs -regime,²⁶ plasmon mediated coupling between nanocrystals located along the same NW can be neglected.⁹ With the help of the quantitative analysis of the recorded BFP patterns we were able to separate the contributions of two different radiation channels. Namely, the direct dipolar emission of the nanocrystals into the glass substrate, observed also in the absence of the NW, and the excitation of propagating SPPs in the NW. In addition to the branching ratio into these two channels we obtained the SPP quasi-momentum, important parameters of active emitter-plasmon structures.^{17,27}

RESULTS AND DISCUSSION

First, a sample area of $100\ \mu\text{m}^2$ was characterized by detecting the upconverted PL after excitation at 980 nm with a narrow band pass filter centered at 670 nm with a full width at half maximum of 10 nm in front of the avalanche photodiode used for signal detection (see Supporting Information, Figure 1). A representative PL image of a magnified sample area can be seen in Figure 1 (a). Subsequently the same sample area was mapped *via* atomic force microscopy to measure the topographic height of each investigated NW. In the PL map (Figure 1 (a)) the NWs appear as luminescent elongated structures with additional bright end spots or bright spots along the wire. In addition weaker luminescent spots are distributed all over the sample. The topography of the same sample area (Figure 1 (b)) shows the NWs at the same positions with lengths of several μm and heights between 100 and 250 nm as well as a number of smaller structures nearby (about 20 nm in height). The size of the smaller structures together with their luminescent behavior identifies them as single nanocrystals or small clusters of several crystals. From the PL signal (Figure 1 (a)) it is evident that nanocrystals deposited in the vicinity of the NWs show enhanced PL and that the plasmonic enhancement is most efficient at the NW ends. Emission enhancement can result from new radiation channels provided by the metal nanostructure corresponding to an increased local density of states.²⁸ In general, radiation channels are connected to characteristic angular distributions of emission that can be studied by BFP imaging^{29–31} (see Supporting Information, Figure 1).

To illustrate the change of the emission characteristics induced by the NW BFP patterns of NCs without NW were measured, filtering the detected signal for the upconverted PL (bandpass 670 nm). Figure 2 (a) shows a representative example. The patterns consist of a radially symmetric intensity distribution that increases from the largest detectable angle given by the NA of the microscope objective towards the critical angle. For smaller angles the pattern shows a more uniform and weaker intensity.

In the next step BFP patterns were recorded for different NWs at varying excitation positions along the NW, again filtering the detected signal for the upconverted PL. Figure 3(a) shows a representative example of the PL map of a single NW together with the excitation positions and the recorded BFP patterns (Figure 3 (b)-(d)). Other examples for nanowires of different diameters are shown in the Supporting Information, Figure 2. When the NW is excited in the middle the BFP image (Figure 3 (c)) shows a symmetric pattern oriented along the NW axis with two brighter lobes at the rims of the pattern. In contrast, the patterns recorded upon excitation of the NW ends (Figure 3 (b),(d)) exhibit an asymmetric intensity distribution with the lobes on the opposite side of the excitation position being

brighter than the other. In addition a fringe pattern is visible when the NW is excited at the ends. This indicates that the position of excitation strongly influences the angular emission characteristics of the NW.

In order to analyze the measured BFP patterns and to extract the contributing radiation channels we start from the two apparent signal contributions.

The first signal contribution stems from nanocrystal emission which is not coupled to the NW as presented for nanocrystals in the absence of a NW (Figure 2 (a)). A single nanocrystal contains between $10^2 - 10^4$ dipolar emission centers without preferred orientation, thus the resulting BFP pattern can be treated as the sum of patterns from randomly oriented dipoles. This contribution is modeled using the theoretical description of dipolar emission on an interface from *Lieb et al.*²⁹ Due to the symmetry of the resulting functions the random orientation of dipoles can be represented by two orthogonal in-plane dipoles and an additional dipole perpendicular to the surface, also indicated in Figure 4 (a) by black arrows. Thus the intensity in the the BFP can be described by the sum of the individual patterns of the single dipoles:

$$I_{\text{NC}}(\vec{k}_{\text{BFP}}) = I_{\text{dipole},x} + I_{\text{dipole},y} + I_{\text{dipole},z} \quad (1)$$

as depicted in Figure 2 (b) and Figure 4 (a).

The second contribution originates from SPPs launched by nearby nanocrystals propagating along the NW surface which are detected as leakage radiation.^{20,22} A characteristic signature of this radiation channel is the fringe pattern in the BFP image that results from the finite length of the NW in the μm range emitting *via* leakage radiation. We describe this channel by a leaky 1D antenna resonator model, following *Taminiau et al.*³² The configuration is defined by the finite NW length L , the wire orientation given by the in-plane angle Ψ and the input excitation at the position a (Figure 4 (b)). Plasmon propagation depends on k_{SPP} supported by the NW and a complex reflection coefficient at its end including the phase change ϕ taken up with each reflection:

$$r = |r| \cdot e^{i\phi}; \quad 0 \leq |r| \leq 1 \quad (2)$$

In order to include losses in the metal the plasmon wavevector k_{SPP} is defined as a complex value in direct correlation with the propagation length L_D :¹

$$k_{\text{SPP}} = k'_{\text{SPP}} + ik''_{\text{SPP}} = k'_{\text{SPP}} + i\frac{1}{2L_D} \quad (3)$$

Here we also consider the air/glass interface on which the NW is deposited by multiplying the antenna BFP intensity distribution taken from³² with the BFP pattern of an in-plane dipole oriented parallel to the wire axis I_{dipole}^Ψ :

$$I_{\text{NW}}(\vec{k}_{\text{BFP}}) = I_{\text{dipole}}^\Psi \cdot \left| \frac{i}{1-r^2e^{2ik_{\text{SPP}}L}} \left(A \left[\frac{re^{ik_{\text{SPP}}L}e^{-i(k_{||}-k_{\text{SPP}})z}}{k_{||}-k_{\text{SPP}}} - \frac{e^{-i(k_{||}+k_{\text{SPP}})z}}{k_{||}+k_{\text{SPP}}} \right]^a - \frac{L}{2} \right) + B \left[\frac{e^{-i(k_{||}-k_{\text{SPP}})z}}{k_{||}-k_{\text{SPP}}} - \frac{re^{ik_{\text{SPP}}L}e^{-i(k_{||}+k_{\text{SPP}})z}}{k_{||}+k_{\text{SPP}}} \right]^a \right| \quad (4)$$

with

$$\begin{aligned} A &= e^{ik_{\text{SPP}}a} + re^{ik_{\text{SPP}}L}e^{-ik_{\text{SPP}}a} \\ B &= re^{ik_{\text{SPP}}L}e^{ik_{\text{SPP}}a} + e^{-ik_{\text{SPP}}a} \end{aligned} \quad (5)$$

and

$$\begin{aligned} k_{||} &= \vec{k}_{\text{BFP}} \cdot \vec{e}_{\text{NW}} \\ \vec{e}_{\text{NW}} &= \begin{pmatrix} \cos(\Psi) \\ \sin(\Psi) \end{pmatrix} \end{aligned} \quad (6)$$

Figure 4 (b) shows a representative NW BFP pattern I_{NW} calculated for a wire with a length of $L = 4.0 \mu\text{m}$, a diameter of 200 nm and a plasmon wave vector of k'_{SPP} predicted for SPPs in nanowires surrounded by glass excited at the nanocrystal emission at 670 nm.^{24,33} The modulus of the reflection coefficient $|r|$ was taken to be 0.3, the phase shift $\varphi = 100^\circ$ ³⁴ and the excitation position $a = 200 \text{ nm}$ away from the wire end. Together with the clearly visible fringes resulting from the finite wire length the schematic Figure 4 (b) illustrates the formation of the BFP pattern which explains the asymmetry in the intensity distribution that is also seen in the experimental patterns after exciting the wire at the ends (Figure 3 (b), (d)). If the NW is excited at the left end, SPPs can travel only to the right. In Fourier space the wave vector components are sorted by their propagation direction. Therefore it is evident that the excitation in the middle of the NW results in a symmetric BFP pattern with similar intensities on both sides (Figure 3 (c),(f)). The influence of the excitation position a on the BFP pattern is shown in detail in the Supporting Info, Figure 3. The present model treats the several μm long NWs as quasi-1D structures. For shorter NWs with considerably smaller aspect ratio the contribution of sub- and super-radiant modes becomes more significant.^{32,35,36}

We fitted all experimental BFP patterns with a combination of the NW pattern I_{NW} and the pattern consisting of randomly oriented dipoles I_{NC} :

$$I_{\text{BFP,fit}}(D, W, k'_{\text{SPP}}, L, L_D, a) = D \cdot I_{\text{NC}} + W \cdot I_{\text{NW}}(k'_{\text{SPP}}, L, L_D, a) \quad (7)$$

After adjusting the orientation angle Ψ with respect to the NW axis the free fit parameters were the amplitudes of the different channel contributions D and W , the real part of the plasmon wavevector k'_{SPP} together with the propagation length L_D , the wire length L and the NC position a . Representative fits of experimental BFP patterns are shown in Figure 3 (e)-(f) and in the Supporting Info, Figure 2. The fitted patterns agree very well with the experimental results supporting our simplified model describing the emission by the two distinct channels. Moreover, the optically derived NW length L that is the source of the characteristic fringe patterns is in general agreement with the values determined by AFM (see Supporting Information Figure 4).

Besides the two emission channels used in our model, forward scattering at the nanowire end leading to a radiation cone as reported by *Li et al.*³⁷ could occur. From the good agreement between the experimental data and the fitting results we conclude that the relative contribution of such an additional channel cannot be substantial in the present system.

Since the theoretical BFP patterns used in the fit for the NC and the NW were normalized to the intensity radiated into all possible angles below and above the air/dielectric interface:

$$\int_0^\pi \int_0^{2\pi} I_{\text{NC}} d\varphi d\theta = 1 \quad \int_0^\pi \int_0^{2\pi} I_{\text{NW}} d\varphi d\theta = 1 \quad (8)$$

the amplitudes D and W can be seen as the number of photons emitted *via* each channel. The branching ratio F between the two radiation channels, that reflects the coupling strength

between NC and NW can be defined as $F = \frac{W}{D}$. For the 49 studied NW positions this branching ratio was found to be 0.7 ± 0.3 on average. No correlation between NW diameter and branching ratio was observed. The spread in F probably reflects different NC to NW distances also caused by different thicknesses of the polymer layer covering the nanowires.

We note, the finite length L of the NW over which it emits *via* leakage radiation leads to a fringe pattern in the BFP enabling the determination of k'_{SPP} even if k'_{SPP} lies outside the detectable wave vector range of the microscope objective (Figure 5). In order to compare the values for k'_{SPP} determined by fitting the BFP patterns for the different NWs with theoretical calculations we used the model of thin metal cylindrical waveguides.³³ This model was already employed for similar silver NWs to calculate k'_{SPP} for a given wire thickness.²⁴ As shown in Figure 5 (b) for larger NW diameters the experimental values k'_{SPP} are situated in between the theoretical k' values for the SPPs at the metal glass and metal air interface. This is a strong indication for a hybridized plasmon as described by *Shegai et al.*²⁴ For

decreasing wire thickness the values approach the larger and more bound $k'_{\text{SPP, glass}}$. While the fitting results for the propagation length L_D do not show a clear correlation with the nanowire diameter most likely due to varying surface scattering losses and reflectivities r we observe a weak trend of increasing L_D for larger diameters. This trend agrees with recent theoretical predictions by *Li and Qiu*³⁸ indicating that longer propagation lengths in thicker nanowires result from reduced ohmic losses due to a decreasing fraction of energy within the silver.

CONCLUSIONS

By recording the radiation patterns of a coupled system of rare earth nanocrystals and silver nanowires in the back focal plane we found that the emission in the vicinity of a nanowire can be approximately described by two emission channels that can be calculated analytically: Dipolar emission, also observed in the absence of the nanowire, and leakage radiation from the nanowire. The latter can be calculated using an antenna-resonator model that considers the air-dielectric interface on which the nanowire is deposited and the position of excitation along the nanowire. Fitting of the experimentally observed patterns provides estimates of the ratio F of photons emitted *via* the nanowire mediated and dipolar channels with mean value of $F = 0.7$. Finally, the fit of the back focal plane patterns gives access to the wave vector k_{SPP} of the SPP supported by the nanowire. Our results are important for plasmonic applications of nanowires since the understanding of emission characteristics, coupling efficiency and SPP properties supported by such nanowires, plays a key role in device integration.

METHODS

Experimental setup

A conventional inverted confocal microscope with an oil immersion objective (NA = 1.4) and a laser source operating at 980 nm was used. Detection of scanning confocal signals and back focal plane (BFP) patterns followed the setup described in .³¹ A detailed description is given in the supporting information.

Subsequently the same sample area was mapped *via* atomic force microscopy (AFM) (Nanoink DPN System, tapping mode, constant amplitude) to measure the topographic height of each investigated nanowires.

Materials

The used sample consisted of silver nanowires with diameters between 100 and 250 nm and lengths up to several microns,³⁹ spin coated on a microscope glass coverslide. In a second step, erbium and ytterbium doped NaYF₄ nanocrystals⁴⁰ were spin coated on top of the nanowire sample. Based on a dopant concentration of 2%(wt.) Er and the cubic lattice structure of NaYF₄ we can estimate the number of emitting centers for a NC with a diameter of 20 (50) nm to be $\approx$ 600 (9200).

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

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Figure 1.

(a) Confocal PL map of a representative sample area together with the corresponding topography data in (b). In the PL map the NWs show up as elongated structures with higher intensity at the NW ends. Several single PL spots are located around the NWs which can be attributed to single or small clusters of rare earth nanocrystals which can also be seen in the AFM data. The scalebar represents 2.0 μm in both images.

Figure 2.

(a) Back-focal plane PL emission pattern detected for a nanocrystal on glass. The detected angular range is limited by the maximum collection angle of the microscope objective that is given by its $NA = 1.4$. (b) Theoretical emission pattern for an isotropic emitter represented by two orthogonal in-plane and one out-of-plane point dipoles on glass. Calculated according to eq. 1

Figure 3.

(a) Confocal PL map of a wire decorated with nanocrystals. The scalebar represents $2.0\ \mu\text{m}$. Upon excitation at different positions (b-d) the corresponding BFP patterns (b)-(d) were recorded. The detected angular range is limited by the maximum collection angle of the microscope objective that is given by its $\text{NA} = 1.4$. The lower row (e)-(g) shows the corresponding theoretical patterns obtained from a superposition of plasmon leakage radiation from the NW and direct dipolar emission calculated according to eq. 4. Each BFP pattern is scaled to its maximum intensity, respectively.

Figure 4.

(a) The BFP pattern for randomly oriented dipoles represented by two orthogonal inplane and one out-of-plane dipole, indicated by black arrows. Also indicated are the projections of the critical angle $\theta_{crit.}$, the maximum detectable angle $\theta_{max,NA}$ and k vector $k'_{max, NA}$ determined by the objective NA. (b) depicts the resulting BFP pattern for the leaky antenna resonator as described in eq. 4. Indicated by the arrows are the wave vectors for the right (red $k'_{SPP, right}$) and the left (green $k'_{SPP, left}$) direction and their projection onto the Fourier plane after leaking into the glass substrate at the resonance angle θ_{SPP} given by $k'_{SPP} = k_0 n_{obj.} \sin \theta_{SPP}$.

Figure 5.

The fitted k'_{SPP} is plotted against the thickness for each corresponding wire together with the uncertainty of the fit indicated by the gray shaded area. The black solid and the red dashed line represent the theoretical k values for the SPP at the metal glass interface $k'_{\text{SPP, glass}}$ and at the metal air interface $k'_{\text{SPP, air}}$ based on.³³ Indicated by the green dashed dotted line is the maximum detectable $k'_{\text{max, NA}}$ with the used microscope objective (NA = 1.4).