Plasmonic Nanoantenna Arrays as Efficient Etendue Reducers for Optical Detection

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ABSTRACT: Optical detectors require the efficient collection of incident light onto a photodetector. Refractive or reflective optics are commonly used to increase the collected power. However, in the absence of losses, such optics conserve etendue and therefore pose a limit on the field of view and the active area of the detector. A promising method to overcome this limitation is to use an intermediate layer of fluorescent material that omnidirectionally absorbs the incident light and preferentially emits toward the photodetector. We demonstrate here that plasmonic nanoantenna phased arrays are a promising platform to improve the emission efficiency of thin luminescent layers and provide an efficient method to reduce optical etendue. In particular, we show an almost constant optical absorption of the luminescent layer on top of the array with the angle of incidence and a strong beamed emission in small solid angles in the forward direction. These results pave the way for novel optical communication detectors incorporating nanofabricated plasmonic materials as optical etendue reducers.

KEYWORDS: free-space optical communication, plasmonics, surface lattice resonances, directional emission, fluorescence materials

Optical communication applications often require the concentration of light onto a photodetector to magnify the received signal. Using refractive or reflective optics allows for the concentration of light, however, it typically comes at the expense of a reduced field of view because the etendue, that is, the product of the area and field of view, is conserved in the absence of losses. Luminescent materials can act as etendue reducers to concentrate energy onto a photodetector and have been used for solar concentrators,† high-energy particle detection,‡ and recently for free-space optical communications (FSOC).§

Desirable characteristics of a photodetector for FSOC include a large bandwidth, a large area and a large field of view to allow for high data rate in the presence of multimode beams with large spatial extent. However, only two of the above characteristics can be matched simultaneously with traditional solid-state detectors. Large area detectors have a slow response time and focusing optics minimize the field of view by conservation of etendue. The etendue can be effectively reduced by using a fluorescent material as an intermediate light conversion layer before detection.† In this process the laser beam carrying the information is absorbed with near unity efficiency over a wide angle of incidence and is preferentially emitted in the direction of the photodetector. In this approach, the spontaneous decay rate of the photoluminescent material should be as fast as possible for large bandwidth optical communication and the conversion of the incident light and collection efficiency of the emission as high as possible.‡

In this work, we demonstrate a novel geometry where luminescent materials, comprised of fluorescent dye molecules near metallic nanoparticle arrays, achieve light conversion from an incident laser beam over a large field of view and an emission in a small solid angle toward a semiconductor photodetector.¶ This process reduces the etendue of the light field and, when combined with a conventional lens focusing the directionally emitted light, results in a photodetector with an effective large area and a response time set by the decay rate of the fluorescent dye, which can be significantly faster than a semiconductor detector of the same size. Metallic nanoparticles supporting localized surface plasmon resonances (LSPRs), that is, coherent oscillations of the free electrons in the nanoparticle driven by an electromagnetic field, have emerged in recent years as interesting structures to control the emission properties of quantum emitters in their close proximity.¶,† This characteristic has led to the description of these structures as optical nanoantennas.¶ Antenna phased arrays are designed to beam electromagnetic waves in certain directions by controlling the

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Figure 1. (a) Schematic representation of the sample. From bottom to top: fused silica substrate, Al nanoparticle array and polymer layer containing dye molecules. (b) Side view scanning electron microscope image of the Al nanoparticle array. The scale bar in the inset represents 400 nm. (c) Absorbance (black curve, left axis) and normalized photoluminescence (red curve, right axis) spectrum of a 700 nm thick, 8.5 wt % Lumogen red dye layer.
layer where the dye molecules are isotropically dispersed in the polymer matrix and the emission is Lambertian, the absorption of the excitation beam can be maximized by increasing the concentration of the molecules or the layer thickness. However, in order to increase the emission in the forward direction with the nanoparticle array, as shown later, the layer thickness needs to be kept below 1 μm. Increasing the molecular concentration also leads to concentration quenching of the emission and the reduction of the QY. This reduces the efficiency of the luminescence process quantified by $F(4\pi)$, which is given by the product of the absolute absorption and the quantum yield (QY) of the bare dye film. Therefore, $F$ for thin molecular layers are typically much smaller than one.

Figure 2 displays measurements of the absorptance ($A$), QY, and integrated $F$ for various dye layers with the same thickness of 700 nm, containing different molecular concentrations and for different excitation wavelengths ($\lambda_{ex} = 450, 533,$ and 575 nm). The absorptance was determined from the conservation of energy, that is, $A = 1 - T - R$, where $T$ is the transmittance of the normal incident beam through the dye film and $R$ is the...
The absorption of the dye molecules at each wavelength increases linearly as a function of the dye concentration. At $\lambda_{\text{ex}} = 575$ nm, the dye molecules present the highest absorbance as shown in Figure 2a. The QY of the same layers was measured with an integrating sphere and it is shown in Figure 2b. The QY of the dye is reduced by increasing concentration, which is mainly associated with the aforementioned emission quenching due to the increase of the nonradiative energy transfer between neighboring dye molecules. Consequently, a maximum value of the integrated $F$ of 0.2 for the bare dye film is obtained for a concentration of $\sim$8.5 wt% and by exciting the layer at $\lambda_{\text{ex}} = 575$ nm (see Figure 2c). In what follows, we show that the plasmonic nanoparticle array can improve $F$ by enhancing the directional emission of the dye layer.

### DIRECTIONAL EMISSION OF NANOANTENNA ARRAYS

The emission directivity of the dye layer with and without the nanoparticle array was characterized with a confocal Fourier setup schematically represented in Figure S3 and described in the Supporting Information. The emission from the sample was collected by a 20x microscope objective with a numerical aperture (NA) of 0.45. The emission intensity of the sample as a function of the angle was recorded in the back focal plane of the objective with a Fourier lens. Figure 3a-c show the Fourier images of the emission from the bare dye layer, the emission of the dye layer on top of the nanoparticle array detected from the front side, that is, the particle array side facing the objective lens, and the back detected emission of the dye layer on top of the nanoparticle array, that is, the substrate side of the sample facing the objective lens, respectively. Note that these measurements were performed under the same conditions and normalized to the same value to facilitate a direct comparison. The emission from the bare dye film displays a nearly isotropic distribution within the maximum angle measured ($\theta_{\text{max}} = 26^\circ$), corresponding to its Lambertian properties. The emission from the dye layer on top of the nanoparticle array is predominantly confined in a small solid angle. This pronounced beaming effect is due to the emission decay into the SLRs and quasi-guided modes supported by the dye layer on the particle array and the subsequent outcoupling of the emission to free space in a defined direction.

Interestingly, the beaming effect detected from the back side of the particle array is more pronounced than the one observed from the front side. As we will show later, this is mainly due to the different fractional density of optical states (FLDOS) in the two directions because of the tapered shape of the nanoparticle array.

The photoluminescence enhancement (PLE) spectra, defined as the PL of the dye layer on top of the particle array normalized by the PL of the same layer on top of the flat substrate and recorded in the normal direction ($\theta_{\text{max}} = 0^\circ$) are presented in Figure 3d. To distinguish the different peaks in the spectra, we have determined the angular dispersion of the particle array sample by measuring the extinction (see Figure S1). The dispersion curves are well described by the grating and planar waveguide phase matching equations and neglecting the coupling between the different modes. Based on these extinction measurements, we can assign the three main peaks of the PLE spectra in Figure 3d to the zeroth-order quasi-guided modes (TE−TM$_0$) coupled into free space through the lattice diffraction orders, the Rayleigh anomalies (RAs), and the first-order quasi-guided modes (TE−TM$_1$). Due to the small energy differences between TE and TM modes, a single peak, instead of two, is observed in the PLE spectra. The solid black curve in Figure 3d corresponds to the emission detected from the backside of the particle array sample. This emission shows a 14-fold, 10-fold, and 5.8-fold PLE at the TE−TM$_0$, RAs, and TE−TM$_1$ frequencies, respectively. The dash-dotted red curve in the same figure corresponds to the emission detected from the front side of sample and shows an overall lower enhancement. The integrated directional enhancement over the emission spectra detected from the back side is 1.2X higher than the one measured from the front side.

To gain more physical insight in the asymmetry of the detected emission intensity, we have simulated the electric field ($E$) intensity profile at the position of the dye layer when illuminated by a plane wave from both sides using a commercial finite-difference in time-domain (FDTD) solver. Bloch periodic boundary conditions were used on the boundaries of the unit cell of the array and perfectly matched absorbing layers were used on the vertical boundaries of the simulation volume. The optical constants of the Al used in the simulation was obtained from the literature and fitted using the Drude model. The incident wave in the simulations was a broadband pulse incident along the normal direction. These simulations represent the FLDOS of the system and are related to the experiments by reciprocity, that is, a local source will emit preferentially in a given direction when the local field at the position of the source for a plane wave incident from this direction is the highest. As an example for the TE−TM$_0$ mode, Figure 3e shows the $E$-field intensity in the x0z plane crossing the center of a nanoparticle in a unit cell of the array when illuminated by a plane wave incident from the front side of the sample along the normal direction. The color scale represents the field intensity normalized by the incident field intensity. This simulation can be compared to the emission shown in Figure 3b. Similarly, in Figure 3f the $E$-field intensity is calculated for illumination by a plane wave incident from the back side of the sample to compare with the emission shown in Figure 3c. The results of Figures 3e,f show that the $E$-field intensity for the illumination from the back and front side have different enhancement and slightly different shape, which leads to the detected asymmetry in the emission intensity from the array.

After the optimization of the dye concentration and the description of the asymmetric beaming effect of the nanoparticle array, we set out to measure the $F(\Omega_0)$ of the nanoparticle array. Experimentally, the emitted power was measured in two steps due to the broad emission spectrum of the dye layer (from 560 to 780 nm): First, the integrated power was measured in two steps due to the broad emission spectrum of the dye layer (from 560 to 780 nm): First, the integrated power was measured by a plane wave incident from the back side of sample to compare with the emission shown in Figure 3c. The results of Figures 3e,f show that the $E$-field intensity for the illumination from the back and front side have different enhancement and slightly different shape, which leads to the detected asymmetry in the emission intensity from the array.

The power of the emitted photon flux passing through a 620 nm band-pass filter with a bandwidth of 10 nm was measured with a calibrated power meter, $P_{\text{out}}(620)$. Second, the emission spectrum over the same $\Omega_0$ was recorded with a fiber-coupled spectrometer with and without the band-pass filter. To ensure that the spectrum was measured correctly, we have calibrated its photon counts response in the wavelength range from 400 to 800 nm with a certified halogen lamp. Here, we call the ratio of the photon counts measured by the spectrometer with and without band-pass filter as $R(620)$.

The power of the emitted photon flux without the band-pass filter is given by

$$P_{\text{out}} = P_{\text{out}}(620) \cdot R(620)$$

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Figure 4. (a) Schematic representation of the optical setup used for the characterization of the figure of merit of luminescent nanoparticle arrays. A laser beam excites the sample at an angle $\theta_{in}$ with respect to the sample normal. The emitted photon flux passes through a diaphragm, an achromatic (AC) lens, a band-pass filter (BF) and is detected by a silicon power meter or fiber coupled spectrometer. The distance between the sample and the diaphragm is fixed to 50 mm. (b) Ratio of the measured emitted power ($P_{out}$) and the excitation laser beam power ($P_{in}$) as a function of the collection solid angle (or the elevation angle $\theta_{em}$) in the case of bare dye layer (red squares) and the nanoparticle array with the dye layer on top (black circles). (c) Emission power enhancement of the dye layer due to the nanoparticle array as a function of the collection solid angle.

The maximum $P_{out}/P_{in}$ enhancement factor in the normal direction is $\sim 3.4$. The enhancement factor decreases for larger collection solid angles because of the strong beaming in small solid angles provided by the nanoparticle array (shown in Figure 3c). In addition, and as expected from eq 1, the $F$ of the bare dye layer is enhanced by the particle array by a factor of $\sim 3$ for $\Omega_0 = 0.015 \text{ sr}$.

Figure 5a,b show the results of $F(\Omega_{0,\text{max}})$ measured with the excitation laser beam at $\lambda_{in} = 533$ and 575 nm, respectively, and $\Omega_{0,\text{max}} = 6.3 \times 10^{-3} \text{ sr}$, that is, the solid angle of maximum enhancement of the emission on the forward direction. Note that in such regime of small solid angles, $F$ is independent of $\Omega_0$ as shown by the linear behavior on Figure 4b. With these measurements, we have also investigated the directional excitation dependence of the sample by varying the elevation of the incident angle of the laser beam with respect to surface normal and recording $F$ at the direction perpendicular to the surface for the two sample sides. Comparing Figure 5a,b to Figure 3c). In addition, and as expected from eq 1, the solid angles provided by the nanoparticle array (shown in Figure 4b). The significantly lower $F$ presented in Figure 4b. The significantly lower $F$ compared to $P_{in}$ is mainly due to the small collection angle in these experiments and not to losses in the array of metallic nanoparticles. For instance, the power reduction is 50 dB when the emitted light is collected within $\Omega_0 = 0.01 \text{ sr}$. However, the value of $P_{out}/P_{in}$ increases by enlarging the collection solid angle (see Figure 4b). The ratio of $P_{out}/P_{in}$ measured with the particle array to the one measured onto the bare dye layer is displayed in Figure 4c as $P_{out}/P_{in}$ enhancement.
Consequently, the enhancement of $F$ obtained from (b) by dividing the $F$ of dye layer on the particle array by the $F$ of the bare dye layer. The $F$ as a function of $\theta$, normalized by the averaged value of $F$ measured in the range $-5^\circ \leq \theta \leq 5^\circ$ (indicated by the vertical dashed lines in (c)), result in the excitation enhancement ($ExE$) shown in (d). In (c) and (d), the solid black circles and green triangles denote the results for the front and back detection configuration, respectively.

To understand the enhancement of $F$ by the nanoparticle array, we have analyzed its optical resonances at $\lambda_{in} = 533$ and $575$ nm by measuring the extinction spectra of the sample as a function of the angle of incidence ($\theta$, $\phi$), where $\theta$ is the elevation angle and $\phi$ azimuthal axis measured from one of the principle axis of the square lattice. Figure S1c,d in the Supporting Information show these measurements as a function of $\theta$ and for $\phi = 0^\circ$, $15^\circ$, $30^\circ$, and $45^\circ$. The extinction as a function of $\theta$ at $\lambda_{in} = 575$ nm shows broad peaks that can be partially attributed to LSPRs in the individual nanoparticles. In the angle range $-5^\circ \leq \theta \leq 5^\circ$ there are no scattering resonances excited, which means that the excitation enhancement ($ExE$) for these incident angles can be neglected. Consequently, the enhancement of $F$, which we call FE, at other angles of incidence can be normalized by the averaged results in this range (indicated by the vertical dashed lines in Figure S2), leading to the $ExE$ assisted by the LSPRs at the excitation wavelength of 575 nm. This $ExE$ is displayed in Figure Sd, where it can be appreciated that the $ExE$ fluctuates around 1. Additionally, the $ExE$ is insensitive to the detection configuration as illustrated by the triangles and circles shown in Figure S4b. This is an expected result due to the small mode volume of LSPRs, which only efficiently enhances the absorption of the small fraction of dye molecules distributed at the vicinity of the nanoparticles. The FE could also have its origin in the emission enhancement due to an enhanced radiative decay rate of the dye molecules assisted by the higher local density of optical states. However, the fluorescence lifetime of the dye layer on the particle array is only slightly reduced (see Figure S2). Therefore, we conclude that the substantial enhancement of $F$ is mainly due to the asymmetric beaming effect of the tapered nanoparticle.

**Conclusion**

In summary, we have demonstrated the potential application of arrays of metallic nanoparticles to act as etendue reducers and thereby enhance the response of optical detectors for free-space optical communication. In particular, we have shown that a luminescent layer on top of a periodic array of Al nanoparticles has a strong beamed emission in a solid angle in the forward direction. This beamed emission can be used to improve the detected signal by small field of view photodetectors. Using this...
system, we achieve an emission figure of merit close to unity using a thin layer of nonperfectly absorbing dye molecules, which is equivalent to the figure of merit of a perfectly absorbing layer with a quantum efficiency of 100%. In addition, this figure of merit is almost independent of the angle of incidence, which will enable to suppress complex pointing and tracking systems for optical communication. Further improvement of the beam emitted can be achieved by replacing the layer of dye by a material with a higher absorption efficiency and high quantum yield. Potential candidates are atomic monolayers of 2D semiconductors, inorganic perovskites, and aggregation induced emission crystals. A thinner layer of these materials with higher absorption efficiency will improve the beaming of the emission and, hence, the figure of merit can reach values higher than the maximum value of 1 for bare, perfect emitting layers. This improved beam can be achieved by positioning the thin layer at the height on the sample of maximum electric field amplitude as has been recently shown in ref. Alternative to metallic nanonatennas, also arrays of dielectric Mie resonators could be used, suppressing the losses in the metal. However, the size of the resonators should be larger than the nanoparticles to achieve comparable polarizabilities and scattering efficiencies. In this work, we have not addressed the band-limiting effects induced by the dyes, which can limit the performance if data rates higher than ~2 Gbps are required. It has been recently demonstrated that using plasmonic enhancement the radiative rate of the luminescent materials can be boosted over 2 orders of magnitude by leveraging the Purcell effect and, therefore, can increase the bandwidth of the optical detectors. In conclusion, plasmonic enhancement provides a promising method to improve the performance of luminescent detectors.

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