Supporting Information

Plasmonic nanoantenna arrays as efficient etendue reducers for optical detection

Shaojun Wang[†*], Quynh Le Van[†], Thibault Peyronel[‡], Mohammad Ramezani[†], Niels Van Hoof[†], Tobias G. Tiecke[‡], and Jaime Gomez Rivas[†,§*]

[†]Dutch Institute for Fundamental Energy Research, P.O. Box 6336, 5600 HH Eindhoven, The Netherlands

[‡]Facebook Inc., Connectivity Lab, 1 Hacker Way, Menlo Park, California 94025, USA

[§]Department of Applied Physics, Eindhoven University of Technology, P.O. Box 513, 5600 MB Eindhoven, The Netherlands

[*]E-mail: s.wang@differ.nl

[*]E-mail: j.gomez.rivas@tue.nl

1. OPTICAL METHODS AND EXTINCTION SPECTRA

The variable angle extinction spectrum of the particle array with dye layer on top (shown in Figures S1a and S1b) was measured using a collimated white light beam from a halogen lamp. The zero-order transmission of the white light from the sample under different angles of incidence ($T(\theta)$) was normalized by the transmission through the bare dye layer on the quartz substrate. The extinction was recorded as 1- $T(\theta)$. The photoluminescence enhancement (PLE) is 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 a flat substrate. The PLE was measured by exciting the dye molecules with a continuous-wave 532 nm laser at a low power to avoid any nonlinear or bleaching effect. The excitation beam size on the sample was 1 mm^2.

To correlate the features in the extinction spectra with the lattice resonances supported by the particle array, we calculate the Rayleigh anomaly conditions (RAs), i.e., the dispersion of the diffracted orders grazing to the plane of the array, using the grating equation,

$$\boldsymbol{k_{//d}} = \boldsymbol{k_{//i}} \pm \boldsymbol{G}\,, \tag{1}$$

where $\boldsymbol{k_{//d}} = (2\pi/\lambda)n_{eff}\cdot\mathbf{u_d}$ and $\boldsymbol{k_{//i}} = (2\pi/\lambda)\sin(\theta)\cdot\mathbf{u_i}$ are the parallel to the surface components of the diffracted and incident wave vectors respectively, $\mathbf{u_d}$ and $\mathbf{u_i}$ are the unitary vectors along the diffracted and incident directions projected on the plane of the array, $\boldsymbol{G}=(2\pi m/a, 2\pi n/a)$ is the reciprocal lattice vector of the square array with (m, n) defining the diffraction order, a is the lattice constant of the array, θ is the angle between the wave vector of the incident beam and the direction normal to the plane of the array, and n_{eff} is the effective index defining the phase velocity of the in-plane diffracted wave (RAs). For simplicity, we assume an average effective refractive index n_{eff} of 1.53 considering the inhomogeneous surrounding of the particle array. The dispersions of the (±1, 0) and (0, ±1) RAs are plotted in Fig. S1(a) with dashed blue lines. Along with the RAs, we calculate the dispersion of the quasi-guided modes in the polymer which is sandwiched between air and the substrate. The upper red (TM) and green (TE) dashed lines correspond to the zeroth-order quasi-guided modes coupled into free space through the (±1, 0) and (0, ±1) diffracted orders. The first-order quasi-guided modes are also shown in the same figure by the lower red and green dashed lines. Note that the coupling between modes is neglected in the calculations.[1,2]

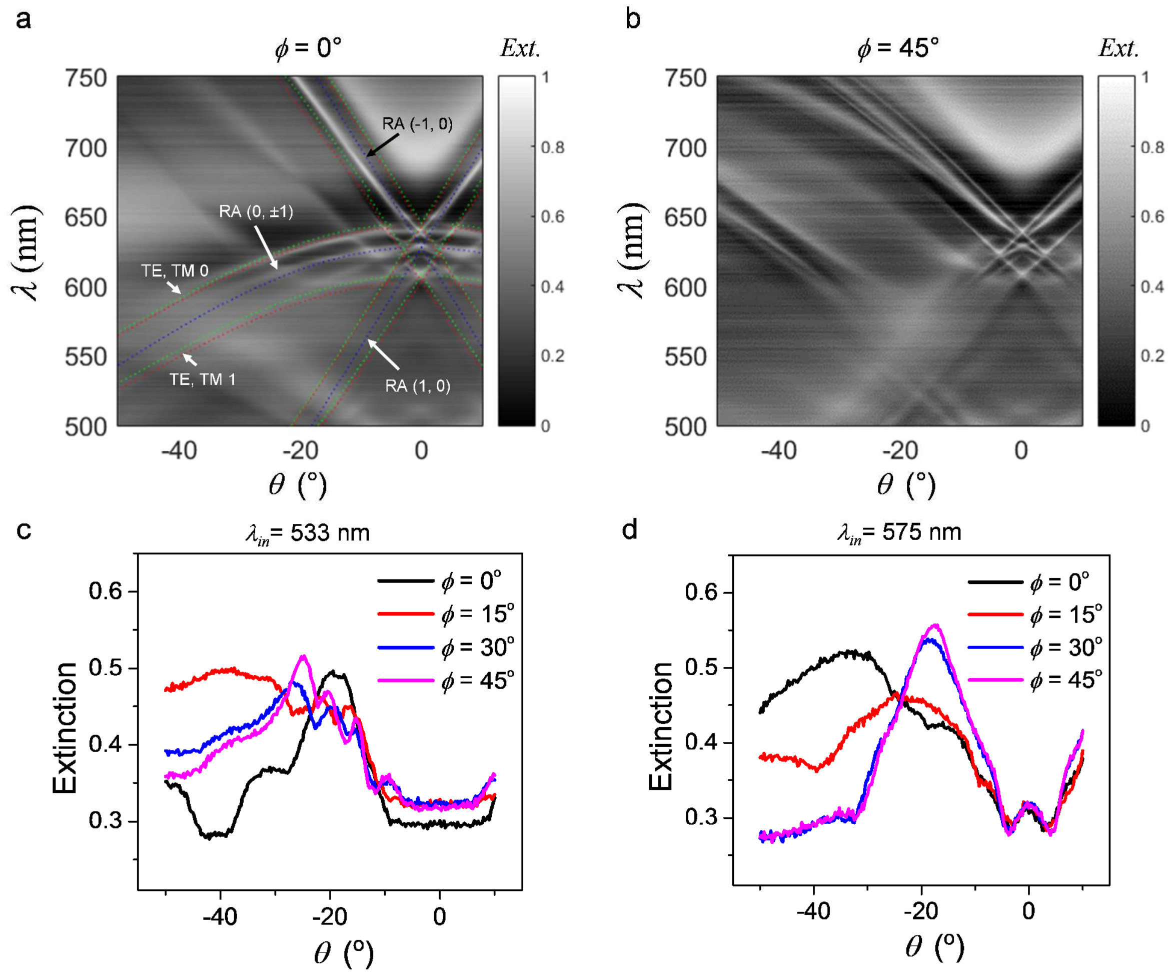

Figure S1. Angle-resolved extinction spectra of the particle array with 8.5 wt% dye layer on top at $\phi = 0°$ (a) and $\phi = 45°$ (b). Extinction as a function of the angle of incidence of the incident beam at the wavelength of λ_{in} = 533 nm (c) and λ_{in} = 575 nm (d) for different azimuthal angles, $\phi = 0°$ (black curve), 15° (red curve), 30° (blue curve) and 45° (magenta curve).

2. PHOTOLUMINESCENCE LIFETIME

The photoluminescence lifetime of the excited dye was measured using time-correlated single photon counting under picosecond pulsed excitation (150 ps pulse duration, 20 MHz repetition rate at λ_{in}=375 nm). Figure S2 displays the measurements for the bare dye layer and the dye layer on top of the nanoparticle array.

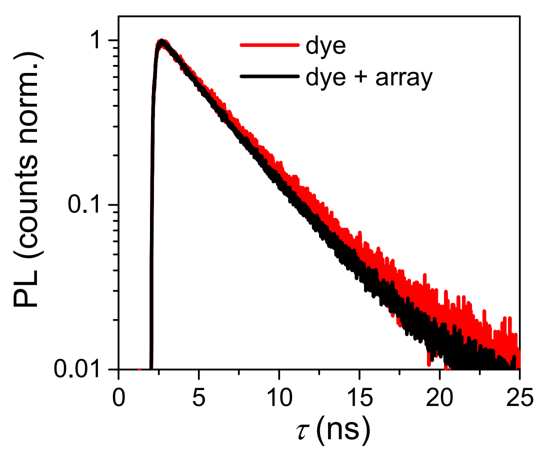

Figure S2. Time-correlated single photon counting measurements showing the PL decay of the dye layer on the particle array (black curve) and the same dye layer on the flat quartz substrate (red curve).

3. CONFOCAL FOURIER SETUP

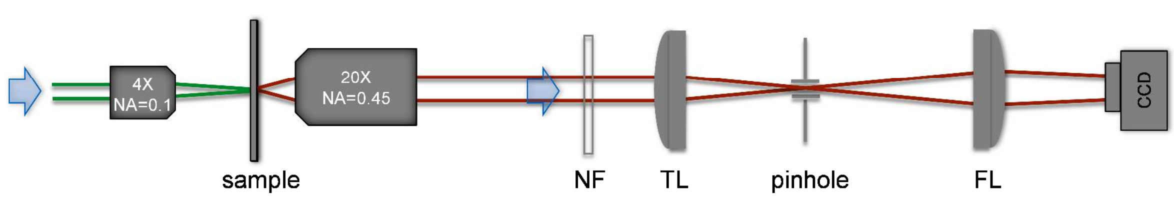

Figure S3. Confocal Fourier setup used for the angle-resolved emission pattern measurements. The sample is excited with a focused green laser beam (λ = 532 nm). The emitted red light from the sample is collected by a 20x objective lens (NA = 0.45), passed through a 532-nm notch filter (NF), tube lens (TL), pinhole, Fourier lens (FL) and is detected by the CCD camera.

The PL intensity of the sample as a function of the emission angle was imaged with a confocal Fourier setup (see schematic representation in Figure S3). The red emitting dye is excited with a focused λ = 532nm, cw laser beam. The emitted light from the sample is collected by an objective lens (NA = 0.45) and passed through a notch filter (532 nm, FWHM = 18 nm). A pinhole with the diameter of 400 μm placed at the image plane of the tube lens allows to spatially filter out the non-conjugated points of the two objective lenses. The PL intensity from the pinhole as a function of angle is imaged on the CCD at the back focal plane of a Fourier lens.

4. COMPARISON OF THE ENHANCEMENT FACTORS

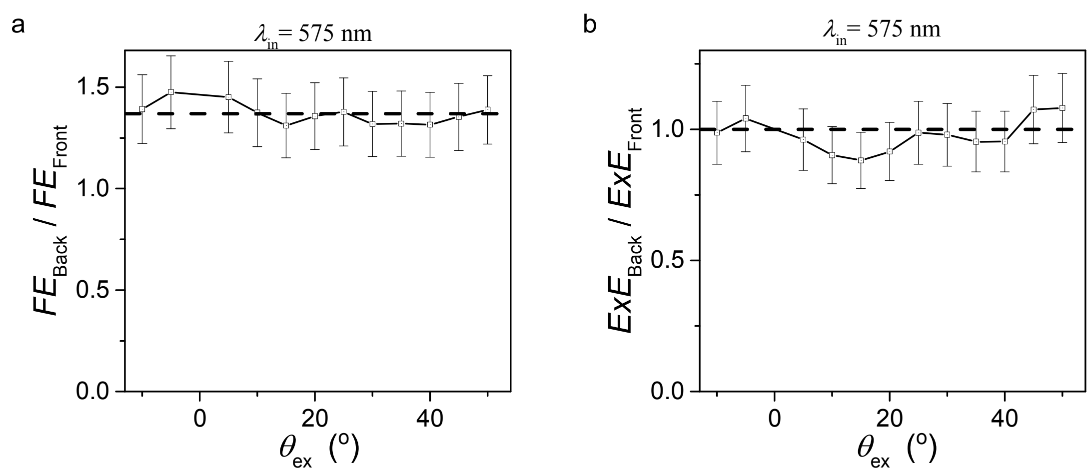

Figure S4. (a) Ratio of the figure of merit enhancement (*FE*) and the excitation enhancement (*ExE*) (b) measured from the two sides of the particle array sample as a function of the angle of incidence of the pump laser beam at the wavelength λ_{in} = 575 nm.

REFERENCES

(1) Guo, K.; Lozano, G.; Verschuuren, M. A.; Gómez Rivas, J. Control of the External Photoluminescent Quantum Yield of Emitters Coupled to Nanoantenna Phased Arrays. *J. Appl. Phys.* **2015**, *118*, 73103.

(2) Ramezani, M.; Lozano, G.; Verschuuren, M. A.; Gómez Rivas, J. Modified Emission of Extended Light Emitting Layers by Selective Coupling to Collective Lattice Resonances. *Phys. Rev. B* **2016**, *94*, 125406.