Electronic Supplementary Material (ESI) for Nanoscale.

Supporting Information

Plasmonic Nanobar-on-Mirror Antenna with Giant Local Chirality: a New Platform for Ultrafast Chiral Single-Photon Emission

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S1. Comparison of a 60 nm NCOM antenna

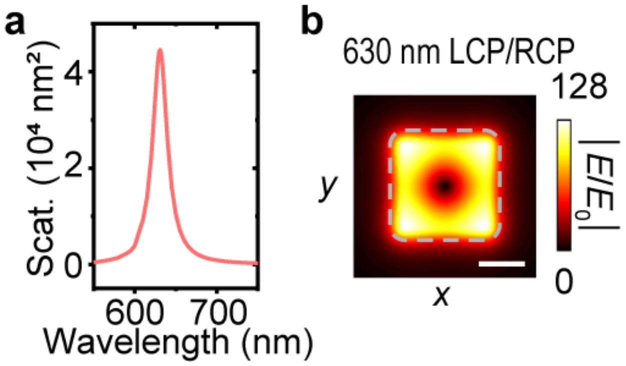

Figure S1. (a) Scattering cross-section of a 60 nm Ag nanocube-on-mirror (NCOM) antenna. (b) Degenerated left-circularly polarized (LCP) and right-circularly polarized (RCP) field enhancement of a Ag NCOM.

As shown by the scattering spectra in Fig. S1a, NCOM (red line) has a degenerated magnetic resonance at 630 nm. Both RCP and LCP show the same electric field enhancement results, revealing no intrinsic chirality of such two geometries. The quality factor $Q = 29$, mode volume $V_{\mathrm{eff}} = 5.6\times10^{-5}(\lambda/\mathrm{n})^3$.

S2. Determination of the size of nanobar

We chose the nanocuboids to construct the antenna for two specific reasons: (1), the Nanocuboid-on-mirror system inherits the outstanding radiation performance from the NCOM (nanocube-on-mirror) configurations due to the same magnetic mode. It provides outstanding radiation decay and antenna efficiency (> 50%), which are crucial for ultrafast bright single-photon sources. (2), synthesized nanocuboids have a controllable size and aspect ratio[1,2] whose unequal lengths support nondegenerated plasmonic resonances, which could result in considerable circular polarization (chiral effect, also see ref.[3]). This effect contributes to the high degree of circular polarization (DCP) of the chiral quantum sources. It should be noted that works[3] from Zu et.al is a bare nanofabricated rectangular nanoplate on SiO2/Si substrates (instead of the metal mirror), whose radiative performance from the nanoparticle's dipole emission is much worse than the magnetic mode in NCOM which has a thin nanogap (compared and discussed in detail in our former work[4] SI Section 13).

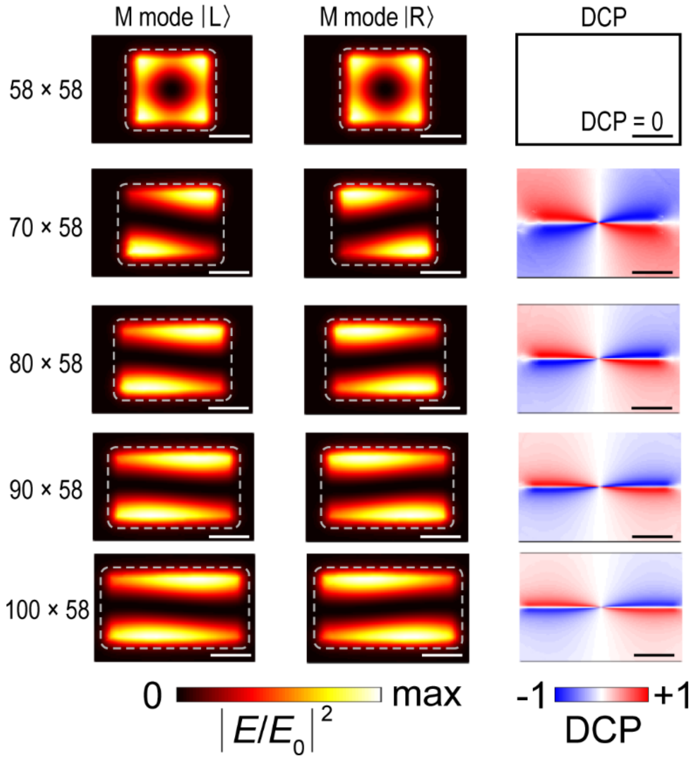

Figure S2. Mode distribution (intensity enhancement) of left-circularly polarized (first column), right-circularly polarized (second column) M modes of NBOM with different sizes. Third column: the DCP of the NBOM with different sizes. The short length of the nanobar was set as 58 nm, and the longer side had a step of roughly 10 nm (i.e. 58 nm, 70 nm, 80 nm, 90 nm, and 100 nm). The scale bars represent 40 nm. The grey dashed lines show the edge of the nanobars.

The specific particle sizes used in the main text are only for proof-of-principle demonstration, and one could design their dimensions by just matching the plasmonic resonances (M mode supported by the shorter length) with the emission of the emitter (e.g. 630 nm QD assumed in our work). The reason for choosing M mode to enhance the emission rather than the M' mode is due to the larger DCP possessed by M mode than the M' mode (see Fig. 3a). Therefore, the shorter sides of the cuboid are set as 58 nm (in Figs. 1, 2) and 80 nm (in Figs. 3, 4) in order to guarantee a plasmonic M resonance around the QD emission peak ~ 630 nm. On the other hand, the longer side has a much flexible selection range. However, the aspect ratio of the cuboid would influence the DCP. As shown in Fig. S2, when the short axis is fixed, the larger aspect ratio (larger long axis) may slightly decrease the DCP. Here, we chose roughly 10 nm as a step (i.e., 58 nm, 70 nm, 80 nm, 90 nm, 100 nm) and calculated the mode distribution and the DCP. We could find that the larger the aspect ratio is, the less chiral the mode would be. Because the chiral modes come from the superposition of two linear modes supported on each side,[3] when the aspect ratio is too large (M and M' modes have significant energy differences), the LCP/RCP input could not efficiently excite both components, and the DCP would decrease. Therefore, the selection of the longer side may not be significantly larger than the short side.

S3. Energy proportion of each component

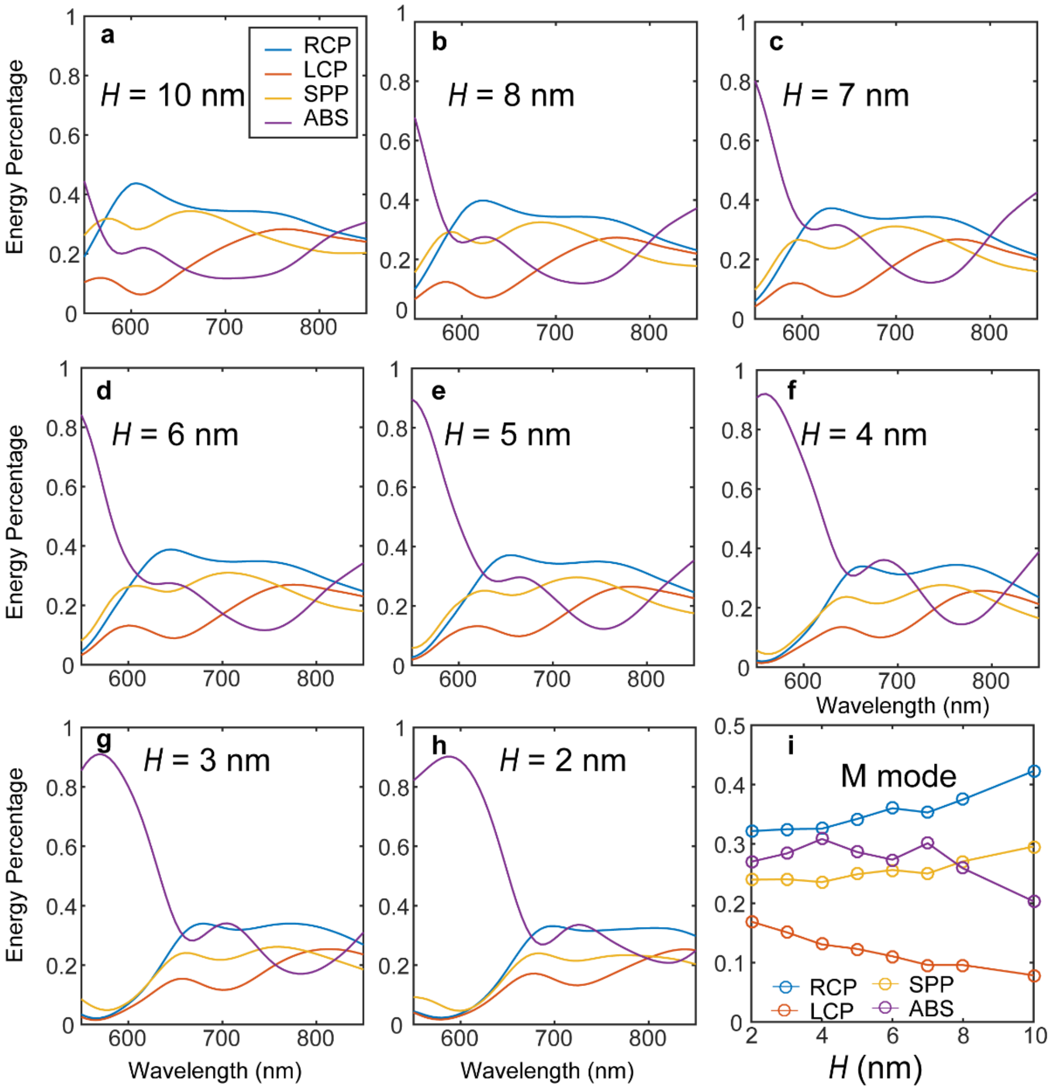

Figure S3. (a-h) Energy proportion of RCP (blue), LCP (red), SPP (yellow), absorption (purple) in the NBOM system with *H* ranging from 10 nm to 2 nm, where the energy proportions on M resonance dependent on *H* are represented in (i). *H* is the diameter of the quantum emitter.

When putting a QE in the nanogap of the NBOM, the energy will decay through multiple channels: (i) radiative channels into left- and right-circularly polarized light (LCP/RCP), (ii) non-radiative channels into surface plasmon polaritons (SPP, which is lossy and would end up to be absorbed) and absorption (heat in metal). Here we calculate each part of the energy in the NBOM with *H* ranging from 10 nm to 2 nm, and summarize their energy proportion on resonance (M mode) against the quantum emitters' diameter *H*. As shown in Fig. S3i, as the *H* is increasing, the RCP component is increasing while the LCP component is decreasing, which means a higher chirality and degree of circular polarization (DCP). The absorption and SPP components both roughly vary around a fraction of 25%. From the result, we could summarize that the total radiative decay (RCP + LCP) occupies a proportion of roughly 50%, and the absorption and SPP each account for 25%.

Reference

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