Electronic Supplementary Material (ESI) for RSC Advances.

Supplementary Information

Dawei Zhang,[1,2] Mengjun Zhu,[1] Linwei Zhu,[2] Qinfeng Xu[3] and Jiannong Chen,[2,*]

[1] *Engineering Research Center of Optical Instrument and System, Ministry of Education, Shanghai Key Lab of Modern Optical System, School of Optical-Electrical and Computer Engineering, University of Shanghai for Science and Technology, 516 Jungong Rd, Shanghai 200093, China*

[2] *School of Physics and Optical-Electrical Engineering, Ludong University, Yantai 264025, China*

[3] *School of physics, Nanjing University, Nanjing, China*

[*] *13220935107@163.com*

1. One lobe linearly polarized illumination source can be created by directing the radially polarized beam onto a half-blocked linear polarizer as shown in Fig.S1. The intensity distribution of one lobe linearly polarized illumination source can be obtained by decomposing the radially oriented electric field vector onto the polarization direction of the linear polarizer as illustrated in Fig.S2. The radially oriented electric vector E_r denoted by red arrow in Fig.S2 at any point in the cross-section of the radially polarized beam can be decomposed into the component E_P parallel to the polarization direction of the linearly polarizer and the component $E_\perp$ perpendicular to it. The blue line segment OL_1 and OL_2 are two different polarization directions of two linear polarizers. The pink line segment Pl_1 and Pl_2 are parallel to OL_1 and OL_2, respectively. We obtain $E_{P1} = E_r \cos(\beta_1 - \alpha)$ and $E_{\perp 1} = E_r \sin(\beta_1 - \alpha)$ or $E_{P2} = E_r \cos(\alpha - \beta_2)$ and $E_{\perp 2} = E_r \sin(\alpha - \beta_2)$. Then we get $E_{x1} = E_{P1} \cos\beta_1$, $E_{y1} = E_{P1} \sin\beta_1$ or $E_{x2} = E_{P2} \cos\beta_2$ $E_{y2} = E_{P2} \sin\beta_2$. In our investigation and calculation, the one dimensional intensity distribution across the center of the beam cross-section $E_r(r)$ is a Gaussian-type which has a full width of half maximum

of 12000nm. It is enough to illuminate the nanostructures with maximum diameters of 6000nm.

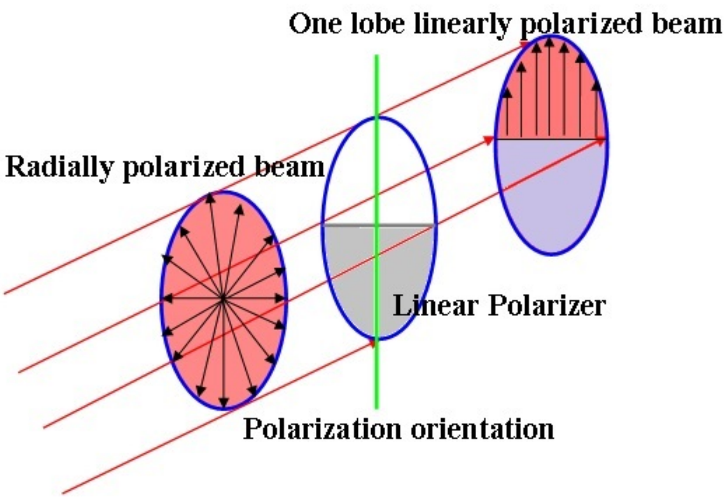

Fig.S1 One lobe linearly polarized illumination source can be created by directing the radially polarized beam onto a half-blocked linear polarizer. The red arrows denote the propagation direction. The green line denotes the polarization orientation of the linear polarizer. The blue elliptical areas denote the cross section of the beam and the linear polarizer. The light impinging on the gray area of the linear polarizer is completely blocked.

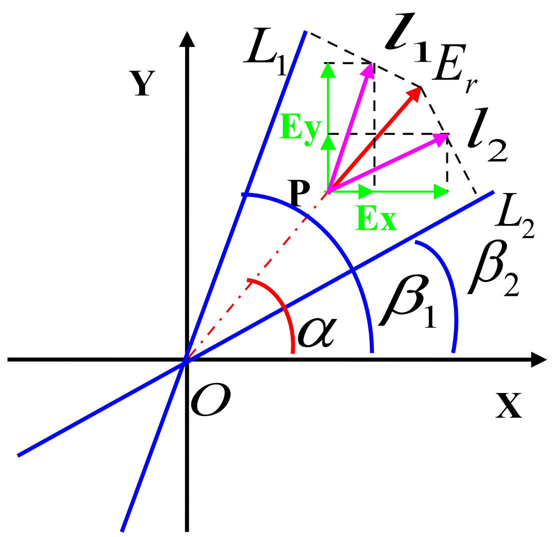

Fig.S2 The schematic diagram of the transformation from a radially oriented electric field vector (red arrow) to two orthogonal components along x-axis and y-axis.

2. The unidirectional propagation property of the second nanostructure (SATPI) when it is illuminated with circularly polarized beams.[1,2] When a multiple pairs of two narrow slits are arranged in a line column as shown

in Fig.S3, the interference of the SPP waves results in time-averaged SPP field intensities propagating to the right (I_R) and the left (I_L) to the column as

$$I_R \propto C[(E_1^2 + E_2^2) - 2E_1E_2 \sin\delta] \tag{1}$$

$$I_L \propto C[(E_1^2 + E_2^2) + 2E_1E_2 \sin\delta] \tag{2}$$

Where I_R and I_L are the SPPs intensity propagating to the right side and the left side, respectively, when the structure is a type of vertical line column. The constant C is proportional to the coupling conversion efficiency. E_1 and E_2 are two components of the incident electric field vector. E_1 and E_2 are perpendicular to two narrow rectangular apertures in an individual pair of the column, respectively. δ is the phase difference between E_1 and E_2. Although these formulae are for a vertical line column composed of multiple pairs of two rectangular apertures perpendicular to each other, it can be reasonably used to explain the inward propagation intensity and outward propagation intensity when the line column is bent to form a circle, especially for a circle with a relatively big diameter. In the case of circularly polarized beam illumination, the phase difference between E_1 and E_2 is 90^0 and we have $E_1 = E_2$. Eq(1) and Eq(2) reduce to:

$$I_{outward} = 0 \tag{3}$$

$$I_{inward} \propto 4CE_1^2 = 4CE_2^2 \tag{4}$$

However, in the case of radially polarized beam illumination, we have $\delta = 0$ and $E_1 = E_2$. Eq(1) and Eq(2) reduce to:

$$I_{outward} = I_{inward} \propto 2CE_1^{\ 2} = 2CE_2^{\ 2} \tag{5}$$

Eq.(3),(4) and (5) shows that the inward energy in the case of circularly polarized beam is two times the inward energy in the case of radially polarized beam. This explains that the enhancement in the case of circularly polarized beam illumination is much better than that in the case of radially polarized beam illumination as illustrated in Fig.5 and Fig.S4

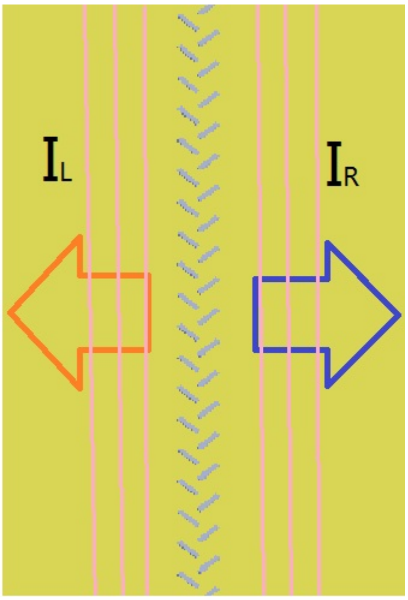

Fig.S3 The SPPs propagating towards the left side and right side when the incident beam is directed onto the nanostructure composed of multiple pairs of two slits perpendicular to each other. The multiple pairs are arranged in a vertical line column.

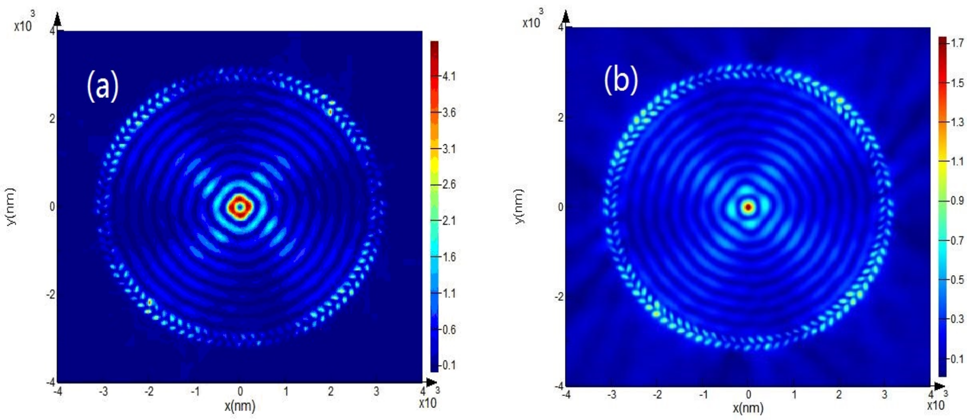

Fig.S4 The intensity distribution of SPPs when the second nanostructure shown in Fig.2 in the main text is illuminated with the circularly polarized beam (a) and the radially polarized beam (b). The size of the monitor is 8000nmx8000nm. The monitor parallel to the surface of the gold film is

10nm away from the surface. The wavelength is 633nm.

3. The enhancement calculation result when a circular gold disk at the center of first nanostructure (Fig.1) illuminated by a radially polarized beam is replaced with a cone-shaped nanoparticle. Fig.S5 shows the cone-shaped nanoparticle (a) and the tip-enhanced intensity distribution (b).

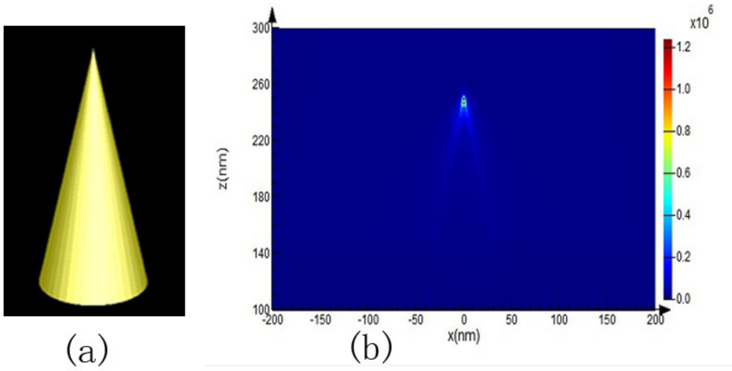

Fig.S5 the cone-shaped nanoparticle (a) and the tip-enhanced intensity distribution (b).

4. The definition and calculation of the overall polarization orientation in a highly confined and enhanced electric field in a nanoscale volume. As illustrated in Fig.S6, the green nanoscale box is the volume in which the overall polarization is defined and calculated. E_{x_volume}, E_{y_volume} and E_{z_volume} can be defined and calculated as

$$E_{x\_volume} = \sum_{i=1}^{M}\sum_{j=1}^{M}\sum_{l=1}^{N} \left|E_x(i,j,l)\right| \frac{\mathrm{Re}[E_x(i,j,l)]}{\left|\mathrm{Re}[E_x(i,j,l)]\right|} \tag{6}$$

$$E_{y\_volume} = \sum_{i=1}^{M}\sum_{j=1}^{M}\sum_{l=1}^{N} \left|E_y(i,j,l)\right| \frac{\mathrm{Re}[E_y(i,j,l)]}{\left|\mathrm{Re}[E_y(i,j,l)]\right|} \tag{7}$$

$$E_{z\_volume} = \sum_{i=1}^{M}\sum_{j=1}^{M}\sum_{l=1}^{N} \left|E_z(i,j,l)\right| \frac{\mathrm{Re}[E_z(i,j,l)]}{\left|\mathrm{Re}[E_z(i,j,l)]\right|} \tag{8}$$

The overall transverse component can be calculated as

$$E_{r\_volume} = \sqrt{E^2_{x\_volume} + E^2_{y\_volume}} \tag{9}$$

The azimuthal angle of the overall polarization is defined as

$$\varphi = \arctan(\frac{E_{y\_volume}}{E_{x\_volume}}) \tag{10}$$

The orientation polar angle with respect to z-axis can be defined as

$$\theta = \frac{\pi}{2} - \arctan(\frac{E_{z\_volume}}{E_{r\_volume}}) \tag{11}$$

i, j and l are the indices of the unit cells in the green nanoscale box. The total number of the unit cells in the box is $M \times M \times N$. Using above-listed formulae, we simulated and calculated the first nanostructure (SPL) shown in Fig.1 and the second nanostructure (SATPI) shown in Fig.2 with azimuthally adjustable and linearly polarized one-lobe illumination as illustrated in Fig.3(c). In the calculation of overall polarization orientation, the unit cells where the field intensity is less than the half of the maximum intensity in the volume are excluded.

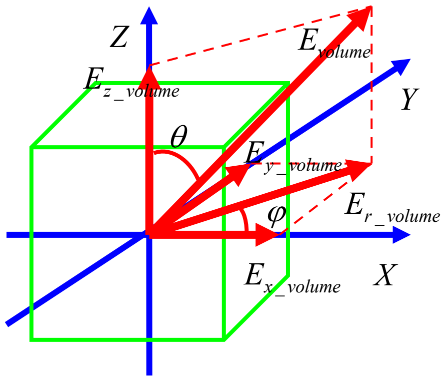

Fig.S6 The schematic diagram for calculating the azimuthal angle φ and orientation polar angle θ with respect to z-axis of the overall polarization orientation of the confined and enhanced electric field in a three-dimensional nanoscale volume.

5. The resonance wavelength of the circular gold disk depends on its parameters. In our investigation, the circular disk is located at the center of surface plasmonic lens which is a circularly symmetrical nanostructure milled into the gold film. The thickness of the gold film is 150nm. The illumination light can not transmit through the film. The localized surface plasmons on the gold disk are not excited directly by external illumination beam but excited by the converging propagating surface plasmons which is excited on the outer circular nanostructure and propagating toward the center[3]. We simulated the enhancement of the first nanostructure (Fig.1) with a gold disk of different diameters in the center. The thickness of the disk is 10nm. The diameter ranges from 20nm to 180nm. The external illumination is the radially polarized beam normally directed onto the nanostructure from the substrate side. We find that the smallest diameter is best for the enhancement in the near field of the disk. The result is shown in the Fig.S7. However, in our investigation, the disk diameter is chosen as the 100nm. We have to take the overall polarization manipulation and potential application into account in choosing the diameter. Here the potential application, just for an example, could be interaction between the enhanced field and nanorods (or a single molecule with dipolar moment). The fluorescent imaging or detection strongly depends on the polarization orientation of exciting fields.

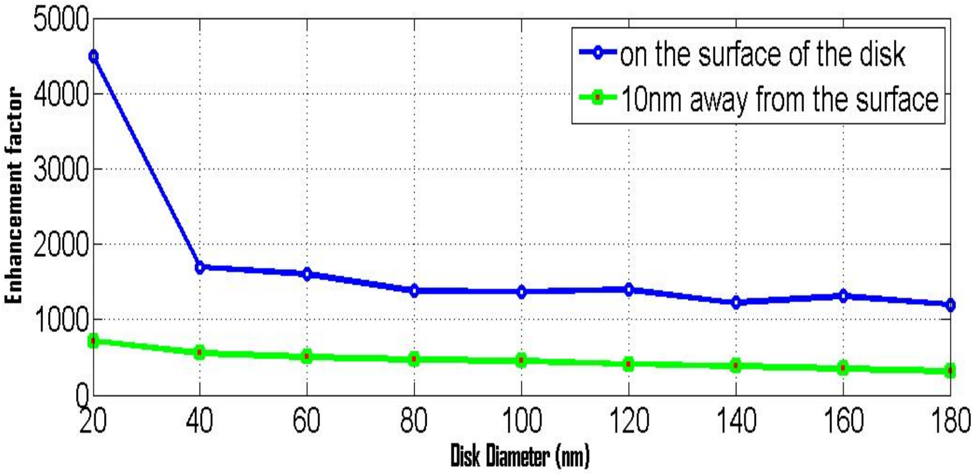

Fig.S7 The enhancement depends on the diameters of the disks. The thickness of all the disks is 10nm. The external illumination wavelength is 633nm.

References

1. Jiao Lin, J. P. Balthasar Mueller, Qian Wang, Guanghui Yuan,3 Nicholas Antoniou, Xiao-Cong Yuan, Federico Capasso, Science, 2013, **340**, 331-334.

2. Takuo Tanemura, Krishna C. Balram, Dany-Sebastien Ly-Gagnon, Pierre Wahl, Justin S. White, Mark L. Brongersma, and David A. B. Miller, Nano.Lett., 2011,**11**,2693-2698.

3. Liang Pan, Yongshik Park, Yi Xiong, Erick Ulin-Avila, Yuan Wang, Li Zeng, Shaomin Xiong, Junsuk Rho, Cheng Sun, David B. Bogy & Xiang Zhang, Scientific Reports, 2011, DOI: 10.1038/srep00175