Supporting information

Chiral Assembly of Gold Nanorods with Collective Plasmonic Circular Dichroism Response

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1. Au nanorods: materials, methods, and electron micrograph

Sodium borohydride (NaBH4), hydrogen tetrachloroaurate(III) trihydrate ($HAuCl_4$. $3H_2O$), CTAB, silver nitrate ($AgNO_3$), and L-ascorbic acid (AA) were purchased from Alfa and used as received. Milli-Q water (18 MΩ cm, Millipore-Q) was used for all solution preparations. All glassware used in the synthesis procedures was cleaned in a bath of piranha solution (70% H_2SO_4/30% H_2O_2 7:3 v/v) and boiled for 30 min.

The Au NRs were prepared by the seed-mediated growth method. Briefly, 7.5 mL of 0.1 M CTAB solution was mixed with 250 μL of 10 mM HAuCl4. Then, 0.6 mL of ice-cold 0.01 M NaBH4 solution was quickly added, resulting in the formation of a light-brown solution. The seed solution was vigorously stirred for 3 min and then kept at room temperature. This seed solution was used within 2-5 h. The synthesis procedure of Au NRs was as follows. The growth solution of Au NRs consisted of 100 mL of 0.1 M CTAB, 5 mL of 0.01 M HAuCl4, 2 mL of 0.5 M H_2SO_4, 0.45 mL of 10 mM $AgNO_3$, and 800 μL of 0.1 M AA. After the color of the solution changed from orange to colorless, 240 μL of the seed solution was added to the growth solution. The resulting mixture was left undisturbed and aged overnight at room temperature. Au

NRs were separated from the growth solution by centrifugation (9 000 rpm for 7 min). The precipitate was collected and redispersed in 20 mL deionized water. The concentration of the Au NRs was 2.0 nM.

The high-resolution TEM (HRTEM) images were obtained using a JEOL model JEM-2010 microscope. (100 nanorods have been measured to obtain the average length and diameter of the Au NRs.) The mean diameter and length of the Au NRs are 16.0±2.4 nm and 43.5±7.4 nm (averaged from 100 nanorods), respectively.

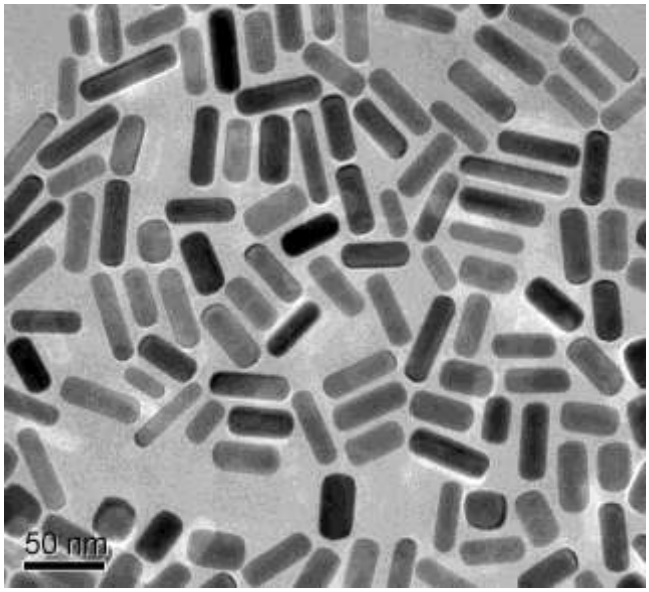

Fig. S1 Typical TEM image of Au nanorods

2. Chiral template of CTAB/Lipid: Materials, methods, and electron micrograph and CD spectrum

In control experiments, a dried lipid film consisting of phospholipids 1,2-Dimyristoyl-sn-glycero-3-phosphatidylcholine (DMPC, 3.5 mg, Avanti Polar Lipid) and small quantities of fluorescent-labeled lipid, rhodamine B 1,2-dihexadecanoyl-sn-glycero-3-phosphoethanolamine, triethylammonium salt

(rhodamine DHPE, 0.02 mg, Invitrogen) was added with CTAB aqueous solution (7.4 mM, 1.5 mL) and phosphate buffer saline (PBS, 2.5 mL, pH~7.4). The molar ration of CTAB: DMPC: Rhodamine DHPE is 67.93%: 32.0%: 0.07%. This mixture was then hydrated at 65 °C for 2 h. After hydration, the sample solution was naturally cooled down to room temperature. The SEM image was obtained from a vacuum-dried sample by electron microscope (JEOL, JEM-2010).

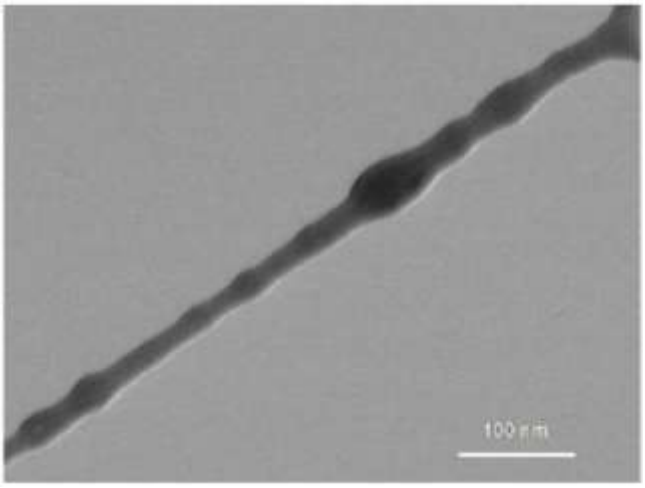

Fig. S2 SEM image of CTAB/Lipid nanotubes with oscillatory perturbation in the diameter. The scale bar is 100 nm.

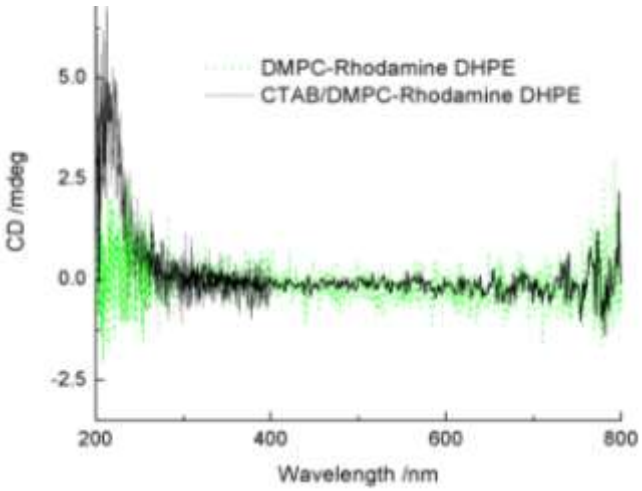

Fig. S3**.** CD spectra from CTAB/DMPC- Rhodamine DHPE solution (black solid line), and from DMPC-Rhodamine DHPE solution (green dotted lines).

3. Chiral hybrid superstructure of Au NRs/CTAB/Lipid: electron and optical micrographs, and CD spectra.

For Cryo-TEM micrographs, a small droplet of the Au NRs/CTAB/Lipid sample solution (~2 μL) was placed on the surface of a pretreated Cu grid covered with a perforated cellulose acetate butyrate film. Through blotting and freezing, a very thin film of specimen was obtained at freezing temperature of liquid ethane. Then the sample was transferred to by FEI Tecnai T12 TEM and examined the microstructure at 120kV. The temperature of the specimen was kept below -165 ^{o}C during both transfer and examination of samples. For SEM micrographs, air-dried sample films were prepared on Si substrates, and characterized by electron microscope (JOEL, JSM-7500F).

Optical images were taken by a confocal laser scanning microscope (CLSM, Zeiss LSM 410, Jena Germany) equipped with a laser source and a 100W Hg lamp, a 100× oil immersion objective, and a digital camera (software Fluoview FV500). 5 μL of the naturally sediment solutions was placed between two glass coverslides. For optical observation, two optical paths were used here: (1) Laser excitation at 543 nm, with a LP 560 nm filter; (2) Hg light excitation at 450-490 nm, with a LP 515nm filter.

CD spectra were measured by using Jasco J-810 spectropolarimeter. For the spectra in 200 ~ 265 nm region, the Quartz cell with 1 mm path length was used, and scans were taken at a rate of 20 nm/min with the sampling interval of 0.2 nm and the response

time of 2 s. For the region of 265 ~ 850 nm, 2 or 1 mm path length were used, and scans were taken at a rate of 100 nm/min with the sampling interval of 1 nm and the response time of 1s. All scans are taken with PBS as background.

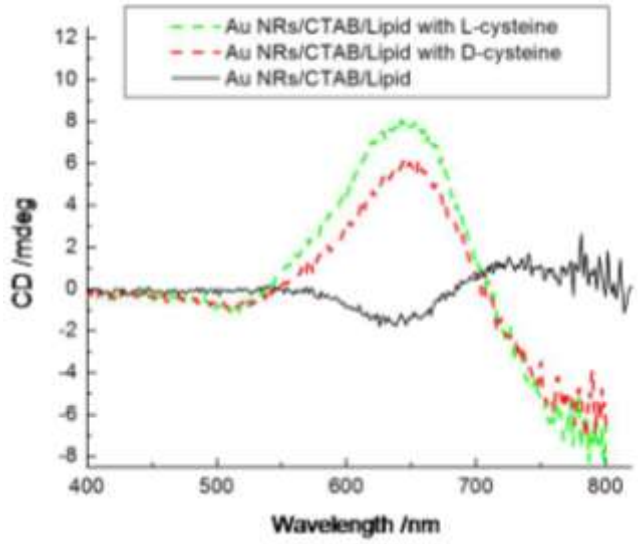

Fig. S4 Cysteine-induced spectral change of plasmonic CD of Au NRs/CTAB/Lipid hybrid superstructure. (1) for D-cysteine additives (4.8 vol%, 1.3mM, red dotted line). (2) for L-cysteine (1.8 vol%, 0.5mM, blue solid line).

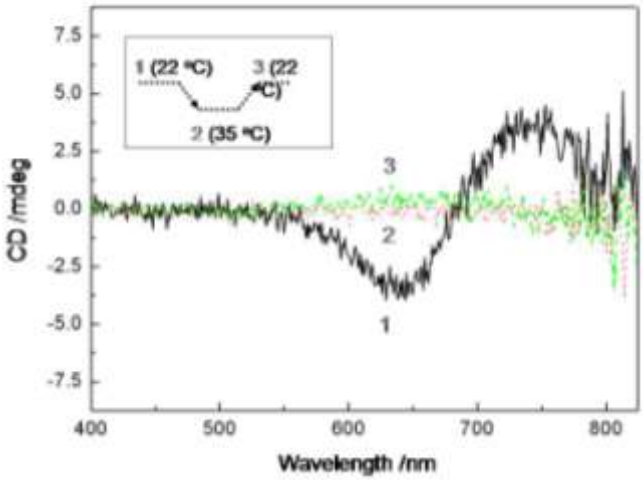

Fig. S5 Temperature dependence of plasmonic CD response of the Au NRs/CTAB/lipid hybrid superstructure.

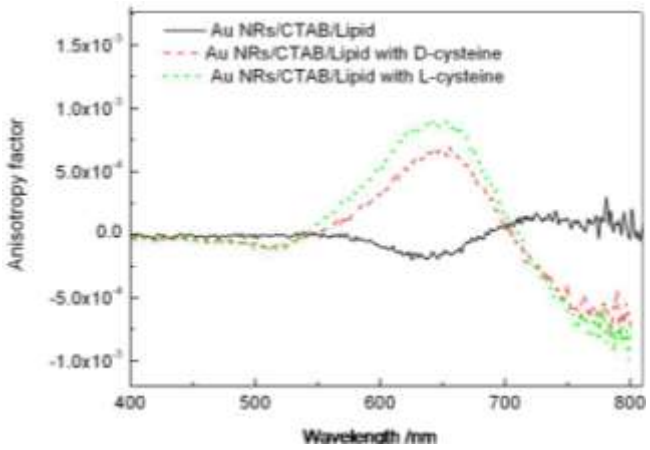

Fig. S6 Anisotropy factors (g factor) for the plasmonic CD of Au NRs/CTAB/Lipid chial hybrid superstructure with and without cysteine additives, which is derived from the spectra shown in Figure S3.