Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics


# Supplementary Information for: Circular Dichroism Nano-Imaging of Two-Dimensional Chiral Metal Nanostructures

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## 1. Contribution of linear dichroism to circular dichroism signals

In circular dichroism (CD) measurements, linear dichroism (LD) signals may potentially be commingled with the CD signals because linear polarization components in a specific direction appear when a modulation detection technique is adopted that involves the use of a photoelastic modulator (PEM). The linear polarization is along the polarization axis of the linear polarizer employed in the system. In the case of the (macroscopic) CD spectrometer, the unfavorable LD contribution is eliminated through rotation of the sample. However, this rotation method is not applicable in our imaging system. Instead, in the present study, we prepared samples with radially placed “S” and mirrored “Ƨ” structures, as shown in Fig. S1(a), to specify the LD contribution. In addition, a symmetric “O” structure was also placed. In an ideal case, no CD signal should be observed for the symmetric “O” structure because it should create no difference in the optical response from irradiation with right and left circularly polarized light. However, in the real experiment, as shown in Fig. S1(b), considerable amounts of positive signals were observed along the direction of the linear polarization (LP), even for the symmetric “O” structure, with the direction of the LP tilted 45 degrees from the vertical direction of the image. These signals are considered to arise from the LD.

With regard to the “S” structures, the measured CD signal distribution also suffered from the LD contribution. The amount and the direction of the false CD signal due to the LD depended on the LP direction relative to the “S” structure. The region apart from the center of each “S” along the LP direction mainly showed positive signals. In contrast, when we carefully observed the central region of each structure, all “S” structures at any orientation exhibited negative signals at the center, whereas, conversely, all “Ƨ” structures exhibited positive signals. These results prove that the CD signal at the center arises from the anti-symmetric features between the "S" and "Ƨ". Similar results were also obtained at other wavelengths of 532 and 633 nm.

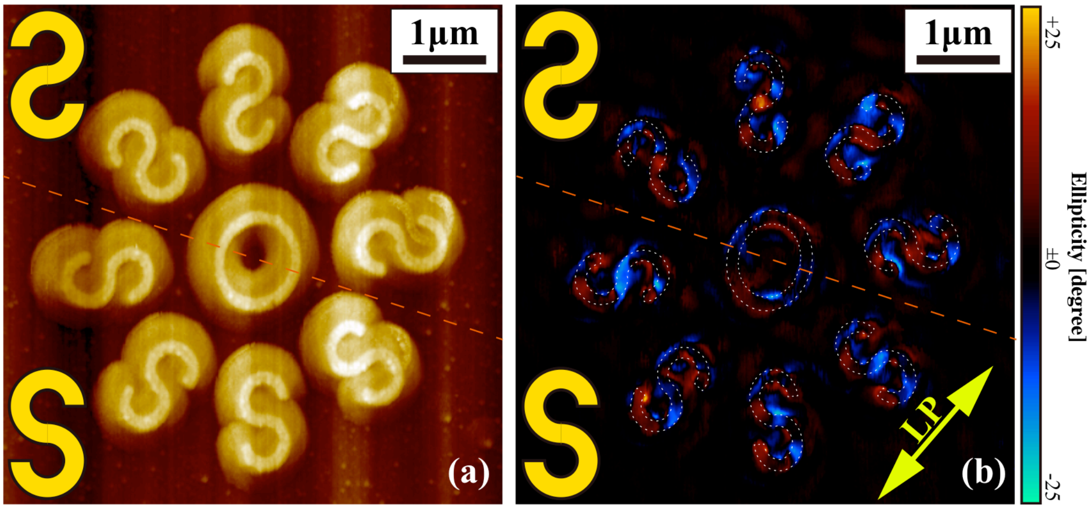


Fig. S1 Simultaneously obtained images of topography (a) and near-field circular dichroism (NF-CD) (b) of asymmetric "S" and "Ƨ" and symmetric "O" structures. To check the contribution from linear dichroism (LD), samples were radially placed. The excitation wavelength was 785 nm. The arrow in (b) represents the LP direction of the modulation optics with the PEM.

## 2. Electromagnetic simulation under circularly polarized light illumination

To evaluate the electromagnetic-field behavior, we performed a three-dimensional electromagnetic simulation based on the finite-difference time-domain (FDTD) method. We used a commercially available FDTD code (Poynting, Fujitsu LTD). As boundary conditions, perfect matching layers were preset for all $x$, $y$, and $z$ directions for a simulated region of 4×4×4 $\mu m^3$. The source of the circularly polarized light (CPL) (plane wave) was placed at the center of the region. The samples of “S” structures were placed 1 μm away from the light source, as shown in Fig. S2. Circularly polarized lights propagating in the upper and lower directions can be regarded as right and left handed CPL, respectively, and the sample “S” structures in each direction undergo opposite rotations of the CPL. The intensity of the electric field it interacted with the sample was evaluated at planes 22 nm away from the sample to mimic the distance between the sample and the near-field probe tip.

Both the upper and lower samples are seen as an “S” from the point of view of the light source. Hence, this configuration provides the electric-field strengths for the “S” structures under the right- and the left-handed CPL illuminations. In contrast, by the reflection symmetry operation in three-dimensional space, the configuration can also be used for the reevaluation of the “Ƨ” structures under left- and the right-handed CPL illumination.

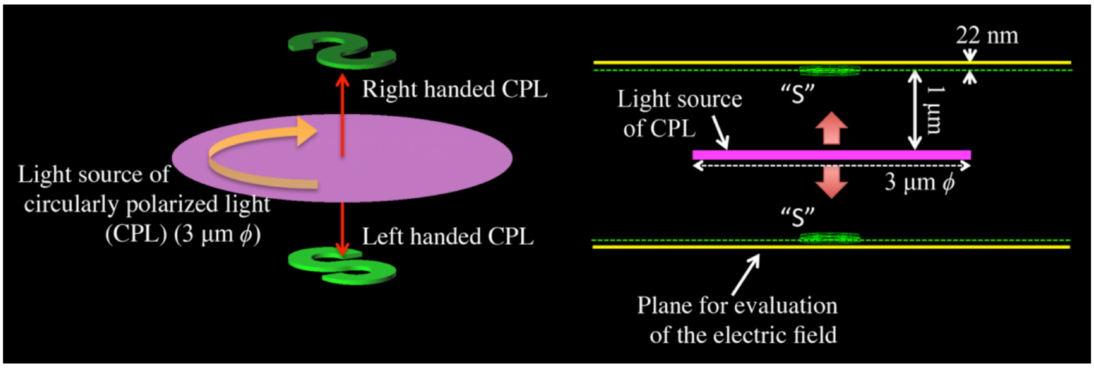


Fig. S2 Configurations of “S” and “Ƨ” samples with the source of the circularly polarized light (CPL). The dimensions of the model “S” structure were 1200 nm × 700 nm, with a linewidth of 200 nm. The thickness of the gold film was 40 nm.