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Chirality Switching Via Rotation of Bilayer Fourfold Meta-Structure

Yew Li Hor[1] · **Wee Kee Phua**[1] · **Eng Huat Khoo**[1]

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Abstract Chiral effects have been observed from the interaction of chiral plasmonics nanostructures with light. Such nanostructures enhance the chiral response of molecules and provide an ideal platform for biological and chemical sensing. Here, we investigate the chiral switching effects of an array of subwavelength nanostructures with a unit cell composed of four double-layered nanostrips arranged to be rotationally symmetric. We observe chiral switching leading to a change in circular dichroism (CD) signature when the mutual angle between the first and second layer increases from 0° to 90° with respect to each other. This mutual angle can be manipulated to switch the handedness of the nanostructure and cause a change in the outgoing light. We also investigated the field distribution of each mode when circularly polarized light is normally incident into the structure. These modes can be categorized into longitudinal and transverse modes depending on the orientation of their dipole moments. The mode studies clearly show the nature of each plasmonics mode.

Keywords Chirality · Plasmonics · Optical activity · Switching · Dipole

Introduction

Chirality in molecules has fascinated scientists ever since the phenomenon was first observed. The enantiomers of chiral molecules greatly affect the biological and pharmacological properties of drugs. Even though these enantiomers have similar molecular composition, they may have significantly different effects in both pharmacokinetics and pharmacodynamics. In 1980s, enantioselective drugs became important when it was discovered that half of the drug molecules have no effects on the human body. Enantioselective differences of drugs drew awareness and became widely discussed [1–3].

Circular dichronism (CD) is one of the preferred methods in chiral measurements to identify the left/right enantiomer of the chiral molecule because of its ease of measurement and its ability to detect large varieties of the chiral molecule or compound [4, 5]. In CD detection, the absorption difference due to the left-handed circular polarized (LCP) and right-handed circular polarized (RCP) is obtained. However, the signal arising from the absorption difference in both enantiomers [6] only yield structural information of relatively low resolution compared to other methods [7, 8].

Using metallic nanoparticles with localized field resonance [9–11] is a common and successful technique in biological and chemical sensing. For sensor designs involving nanostructures, the characteristics, properties, and the configuration of the particular molecule have to be well studied so that the sensor can react with such molecules [12–14]. In supramolecules, we can make use of the coupling between building blocks to achieve a chiral effect. Translating this concept to nanostructures, we propose a structure consisting of four double-layered coupled symmetric chiral blocks as a good candidate for supramolecular detection. Our proposed structure enhances the localized electromagnetic field [15] since its dimensions are significantly smaller than the

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✉ Yew Li Hor
Horyl@ihpc.a-star.edu.sg

✉ Eng Huat Khoo
khooeh@ihpc.a-star.edu.sg

[1] A*STAR Institute of High Performance Computing, 1 Fusionopolis Way, #16-16 Connexis, Singapore City 138632, Singapore

wavelength of the incident light. The electromagnetic field enhancements are resultant of the conduction band electrons that oscillate with the incident light. Depending on the size and geometry of the nanostructures, a strong resonance can be achieved for a given wavelength of light, allowing for strong absorption and scattering of light. When circularly polarized light is incident upon the nanostructure, we can excite plasmons on the surface of the nanostructure. The polarization state of the circularly incident light can either enhance or suppress the surface plasmons on the chiral nanostructure.

In this paper, a chiral subwavelength nanostructure made up of four double-layered rotationally symmetric nanostrips will be discussed. The double layers designate the coupling chiral effect while the four rotationally symmetric nanostrips designate the helical chiral occurrence. The subwavelength nanostrips made up of noble metal cause local electromagnetic field enhancements when light is normally incident on the nanostructure. In addition to the coupling effect, the double-layered structure has the ability to manipulate CD upon rotation of the two layers.

Bilayer Fourfold Meta-Structure

Our designed nanostructure consists of two layers of gold nanostructure arrays with a sandwiched layer of thin silicon nitrate. Gold is the most stable noble metal while silicon nitride is a solid that is relatively chemically inert and has a high melting point. Both materials are suitable for use in medical applications, especially orthopedic applications for silicon nitrate [16, 17]. The pitch of the unit cell in Fig. 1a is 250 nm. Gold (t_{Au}) layer is 20 nm thick and Si_3N_4 (t_{sub}) is 30 nm thick. Each gold layer consists of four nanostrips arranged in a windmill arrangement, with each nanostrip having a length (L) of 60 nm, separation (d) of 30 nm, and width (w) of 30 nm.

We simulated the nanostructure in a liquid environment of refractive index of 1.33 with LCP or RCP light incident onto the system. Upon interaction of the incident light with the nanostructure, localized surface plasmons are excited which enhances the field amplitude on the structure. The second layer is rotated from $\theta = 0°$ to 90° relative to the first layer. At $\theta = 0°$ and 90°, the nanostructure in the second layer is aligned coherently with the first layer. For other angles, angular deviation increases the chiral effect of the whole nanostructure.

To model the system, we consider a unit cell with boundary conditions of Floquet mode implementation to allow the consideration of an infinite nanostructure expanding in x and y direction. The boundary condition for z direction uses perfectly matched layers. We used finite different time domain (FDTD) method with a mesh size of 4 nm in all directions. Circularly polarized light introduced along the z-axis is incident normally to the xy-plane of the nanostructure (see Fig. 1a). Monitors are set up to measure the transmittance and reflectance during the simulation with time of 10^6 fs. We investigated the interaction of the bilayer fourfold rotationally symmetric nanostructure with RCP and LCP light by varying the rotation angle, θ of the second layer. The absorption, CD spectrum, and field distribution of this nanostructure were studied. CD is given by;

$$CD = A_L - A_R$$

where A_L and A_R represent the absorption when LCP and RCP are applied, respectively.

CD Manipulation and Switching Effect

The absorption profile of the nanostructure when the angle rotate from $\theta = 0°$ to 90° is extracted from the simulation data. Figure 2 shows the orientation of the nanostructure upon rotation and the absorption profile as a function of wavelength. We observed a reversal of RCP and LCP absorbance peak when θ rotated from 0° to 90° with $\theta = 45°$ as the turning point. Taking $\theta = 15°$ and $\theta = 75°$ as an example, the arrows indicate a reversal of the absorbance peak at different wavelengths. At 980 nm, RCP absorbance is dominant for $\theta = 15°$ while for $\theta = 75°$ the LCP absorbance is dominant. The reversal is due to the change in handedness of the nanostructure. From $\theta = 0°$ to 30° the nanostructure is left-handed but from $\theta = 45°$ to 75° the nanostructure became right handed.

A positive or negative circular dichroism signal depends on the extent by which LCP light is absorbed more (CD signal positive) or less (CD signal negative) than RCP light. Figure 3 shows the CD profiles with CD switches between positive and negative as a function of wavelength. The CD profile at each rotation may exhibit both positive and negative peaks/dips in different wavelength region. The observed CD peaks/dips fall around ~630 , ~690 , ~900, and ~980 nm. These CD peaks can be denoted as modes A, B, C, and D. Mode A and mode D exhibit negative CD for $\theta = 15°$ and 30°, and positive CD for $\theta = 60°$ and 75° while in $\theta = 0°$, 45°, and 90°, the sign of the CD is undefined. The CD signature for mode C is opposite to mode A and mode D. Mode C and mode D exhibits opposite chiral effects and a bisignate CD appearance where the spectra consist of peaks next to dips (or vice versa). The CD sign flipped, indicating a switching behavior due to the change in handedness of the nanostructure.

When the second layer rotates, a change in CD from positive to negative magnitude is observed. This is due to the nanostructure acting as a left-handed nanostructure for certain angles, and as a right-handed structure for other angles. Figure 4 shows the switching effect of the nanostructure for the three modes with change in rotation angle from 0° to 180°. Mode D provided the most significant switching effect among the three

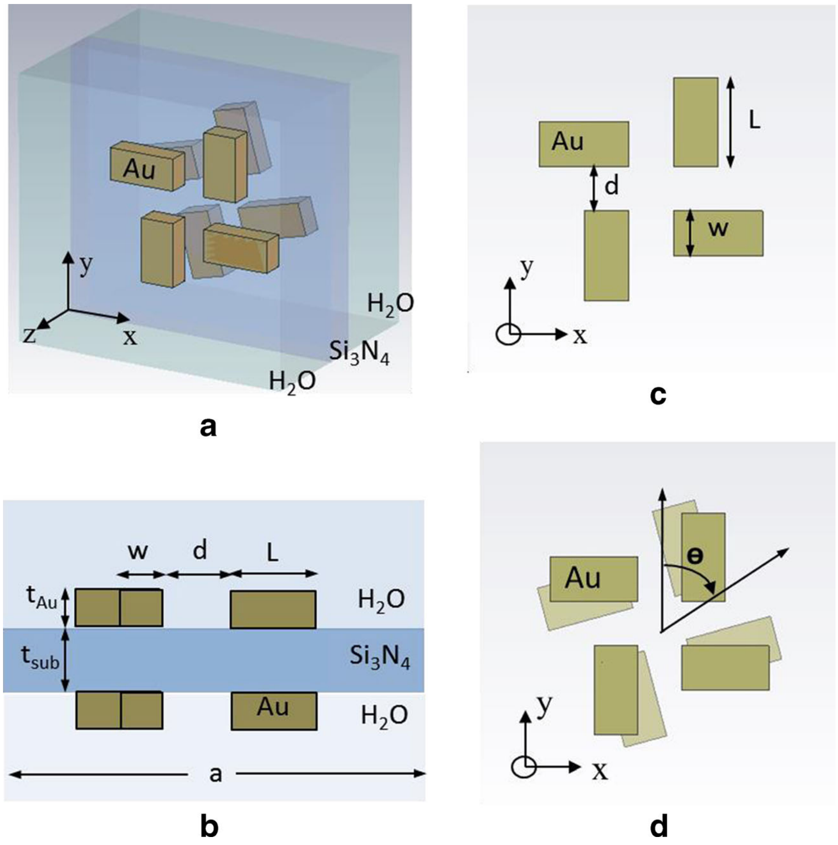

Fig. 1 3D schematic of unit cell with pitch of 250 nm (**a**); side view (**b**); top view of single layer nanostructure layout (**c**); top view of double layer with the second layer rotated θ degrees with respect to the first layer (**d**)

modes. The strong switching effect is observed in this bilayer fourfold symmetry nanostructure when one of the layers rotates upon the other layer, thus indicating that chirality can be manipulated.

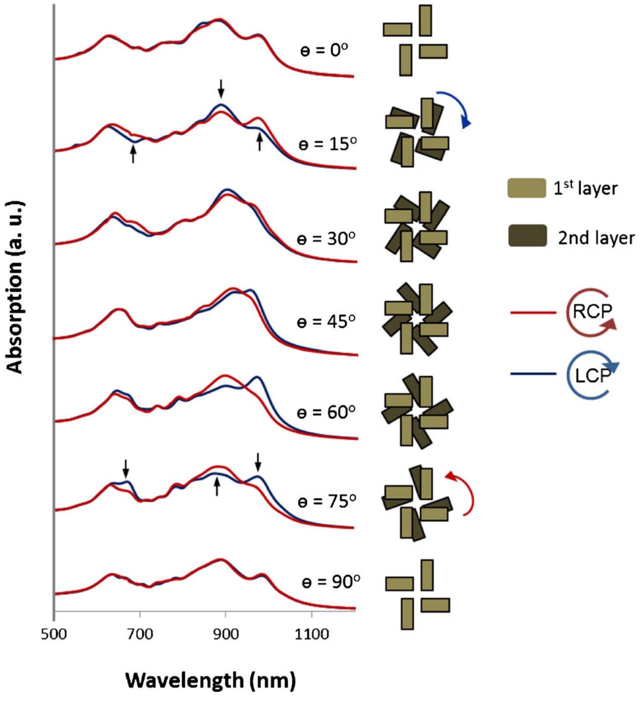

Fig. 2 (*left*) Absorption vs. wavelength and (*right*) the nanostructure with rotated second layer. *Black arrows* indicate a switch of the absorbance peak in RCP and LCP (use $\theta = 15°$ and $\theta = 75°$ as examples)

For metal nanoparticles with small diameters relative to the incident wavelength, the excitation of surface plasmons produces oscillating dipole fields [18]. We begin by considering the modes of nanostructures when circularly polarized light is normally incident onto the nanostructure. The modes are categorized into two categories, depending on the orientation of the dipole moments of the individual nanostrip, which are

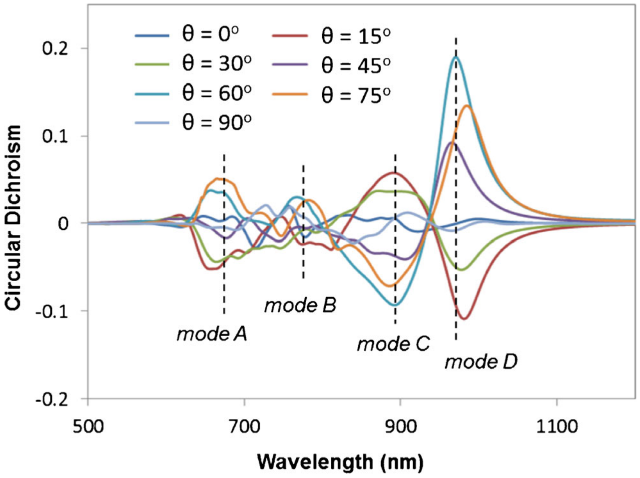

Fig. 3 CD spectrum of the bilayer fourfold rotationally symmetric nanostrips

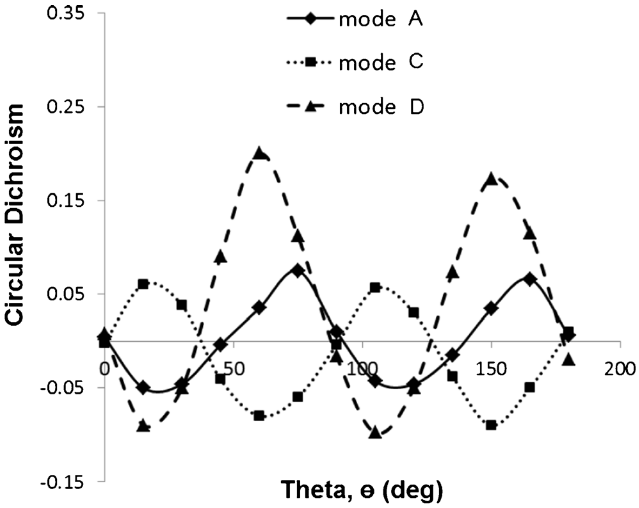

Fig. 4 The switching effect of CD for mode A, mode C, and mode D when the second layer rotates from 0° to 180° with respect to the first layer. Mode B is not included since it is of less significance

longitudinal and transverse modes. In the longitudinal mode, the dipole moments oriented parallel along the direction strips while the transverse modes have their dipole moments oriented perpendicular to the strip. Figure 5 shows the LCP and RCP E-field distribution for first and second layers of the designed structures for modes A, B, C, and D at the transmittance profile. The field responses of LCP and RCP in the nanostructures are identical except for the switch between positive and negative behavior in the effective region. The detail plot of the '+' and '−' charge distribution illustrated schematically in Fig. 5, clearly shows the nature of each plasmonics mode. The field distribution plots clearly show that modes A and B are transverse modes while C and D are longitudinal modes. For mode A with wavelength of 633 nm, the '+' and '−' charge are distributed along the transverse direction and the charge are accumulated along the edges. Therefore, we have designated them as transverse edge modes. Similarly for mode B with wavelength of 690 nm, the charges are also transversely separated and accumulated at the corners, so we designated this as a transverse corner mode [19]. Modes C and D with wavelength of 904 nm and 978 nm, respectively, are longitudinal modes with different magnitude of the x- and y-component of the RCP and LCP [20]. Mode C has only purely x- or y-components of LCP/RCP while mode D has hybridized x- and y-components of RCP/LCP. Therefore, for mode C, we only observed the field distribution in two nanostrips while for mode D, the field distribution is approximately identical in all four nanostrips. The role of the second layer of the nanostructure is to identify the handedness of the nanostructure. For example, in mode D, the E-field distribution of the second layer with RCP is stronger than E-field distribution with LCP; this indicates that the right-handed characteristic of the nanostructure has dominated in this mode.

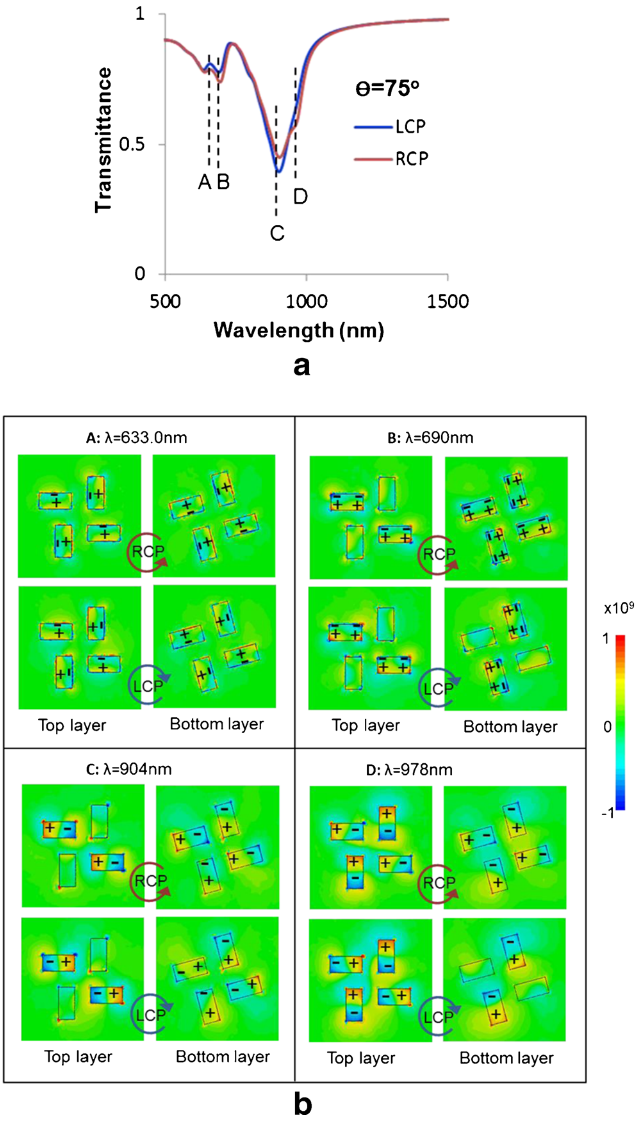

Fig. 5 **a** The transmittance profile indicate the location of modes A, B, C, and D. **b** E-field response of individual layer under RCP and LCP light incident at resonance peaks indicate on the transmittance profile above. The *red arrow* indicates the polarization of the dipole

Conclusion

The proposed fourfold symmetric bilayer nanostructure attains its CD switching effect upon rotating one of the layers of the nanostructure, with each layer consisting of a unit cell of four nanostrips arranged in windmill orientation. For chiral molecules detection, this allows fine tuning of the spectrum to obtain the maximum CD as well as the desired sign of the CD, and consequently, the chirality of the molecular filaments can be detected. Depending on the circularly polarized states and also the angle between the planar axis of the first and second layer, CD may change from negative to positive or vice versa, indicating a switching phenomenon in the nanostructure. From 0° to 90°, the CD switching effect completes a cycle due to the fourfold symmetric structure. Thus, by controlling the rotation angle from 0° to 90°, a switch in the handedness of the nanostructure will cause a change in the outgoing light. The subwavelength nanostrips give rise to interesting optical phenomena such as localized surface plasmon polaritons. The field study of each mode shows clear pictures of the field resonance of each individual layer of the nanostructure explaining the CD switching phenomenon.

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