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Induced Optical Chirality and Circularly Polarized Emission from Achiral CdSe/ZnS Quantum Dots via Resonantly Coupling with Plasmonic Chiral Metasurfaces

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Abstract

Functionalization of quantum dots to induce circular dichroism as well as the direct emission of circularly polarized light has been the subject of intensive studies for the obvious importance not only in practical applications but also in gaining a deeper understanding of quantum semiconductor heterostructures. In this work, we report remarkable chiral features in achiral CdSe/ZnS colloidal quantum dots (CQDs) induced by plasmonic chiral metasurfaces. We observe not only giant circular dichroism but also a high degree of circular polarization in the light emitted by the CQDs hybridized with the chiral metasurfaces. By probing the response of CQDs combined with tunable chiral metasurfaces, we show that the induced chiral features are closely linked to the correlation between the absorption bands of CQDs and the chiral bands of the metasurfaces. Our findings indicate that plasmonic chiral metasurfaces is an effective medium in reshaping CQDs with useful chiral properties.

Keywords: Quantum dots, chiral metasurfaces, induced chirality, circularly polarized

light

1. Introduction

Chiral materials are a pivot in modern chemical and medical industry, and have been studied intensely in many frontier research fields ranging from ultrasensitive chiral recognition^[1,2] in life sciences to being standard active materials in quantum computation and communications^[3,4]. Compared with the chiral materials in nature, artificial chiral quantum nanocrystals^[5-8] (NCs) have overwhelming advantages in showing chiral behaviors, such as highly enhanced optical circular dichroism (CD) and direct emission of circularly polarized electromagnetic waves, which are traditionally obtained by converting the linearly polarized waves from the light sources by inserting retardation optical elements which are expensive and bulky.

Because of these promising applications, various kinds of chiral quantum NCs have been proposed. Among them, colloidal quantum dots (CQDs) have long been recognized as appealing optoelectronic materials by the virtue of the solution processability, large absorption coefficient, size-controllable emission wavelength and high photoluminescence quantum yields^[9-14]. Realizing directly circularly polarized emission (CPE) and induced chirality from CQDs will enable a lot of new possibilities for integrated optical systems. In an attempt to achieve chiral CQDs, the most commonly used method to date is to deliberately connect chiral ligands or molecules to various CQDs, such as CdSe^[15-18], CdTe^[16,19] and CdS^[20,21]. The CPE and induced CD can be realized from the hybridization of CQDs molecular orbitals with those of the chiral ligands. However, by this way, the induced chiral properties are normally very weak, rendering these chiral NCs far more from being practical usage. Therefore, it is extremely important and desired to establish an efficient platform to reshape the CQDs with highly enhanced chiral features.

Recently, plasmonic metamaterials^[22-27] with engineered electromagnetic response have been used to interact with active materials, which has been proved to be an effective approach to enhance the light matter interaction and control the photonic properties of the active

materials^[28-30]. As a crucial branch, chiral metasurfaces^[19,31-39] have been reported in manipulating circular polarization with giant CD values^[40-42] and strong chirality of the evanescent field in the vicinity of the chiral metasurfaces^[43-44], which offers not only a chiral platform for coupling various potential chiral systems but also the possibility to break the time-reversal symmetry of achiral objects for highly efficient chiral light-matter interactions, hence providing a way to functionalize CQDs possessing significant chiral properties.

In this work, we demonstrate a hybrid system in which controllable chiral metasurfaces are coupled with achiral semiconductor quantum dots. In order to explicitly reveal the interactions between the quantum emitters and plasmonic metasurfaces, we introduce tunable chiral responses from chiral metasurfaces to probe the genuine achiral CQDs in both the absorbing and emitting processes. The clear chiral features from both absorption and emission processes clearly suggest a coupling mechanism between the chiral metasurfaces and CQDs. Firstly, circular dichroism spectroscopic measurements indicate that CQDs on chiral metasurfaces absorb preferentially for the circular polarization associated to the metasurfaces' chirality. More importantly, the induced CD shifts according to the tuned chiral responses of the metasurfaces, verifying that the achiral CQDs are chirally functionalized by the metasurface. Secondly, a preferential spin state was further observed in the photoluminescence, leading to a high degree of circular polarization of 17%, which represent a new record for CQDs based direct CPE in the visible range. Our work, as the first demonstration of hybrid CQDs/chiral metasurface system, shows that the achiral CQDs can be reshaped by certain chiral metasurfaces to turn into both an efficient circular polarized light (CPL) emitter and absorber, which holds great potentials for optimization and miniaturization of optical systems such as bio-sensing, bio-imaging, and quantum communications.

2. Results and Discussion

The chiral metasurface adopted to couple with CQDs is schematically illustrated in the Figure 1a. The unit cells of this square lattice array are composed of 4 identical nanoslits fabricated by focused ion beam (FIB) milling in gold film of 100nm in thickness. The scanning electron microscope (SEM) image of the chiral unit cell (left handedness) is shown in figure 1b with nanoslits of length 400nm, width 100nm and lattice period P . The neighboring nanoslits are perpendicular to each other and separated by a distance of 50nm. We used the canonical CdSe/ZnS core/shell QDs as the active materials. The photoluminescence spectrum of CdSe/ZnS QDs on a glass substrate peaks at 623nm as shown in figure 1c and the morphology of the individual quantum dot can be seen in the transmission electron microscope (TEM) image of figure 1d. The CD response of the chiral metasurface is tuned by altering the lattice period, as previously reported^[36, 37], which facilitates the resonance matching between the CQDs and the chiral metasurface. More importantly, the interaction between the chiral metasurfaces and achiral CQDs is hereto not limited to employing the localized mode. The propagating mode of circular polarization dependent surface plasmon polaritons (SPPs) that propagate between unit cells on the whole surface enables a larger area of interaction between the random spin-coated CQDs and metasurface, contributing to a more intense coupling.

In order to reveal the interaction between the plasmonic chiral metasurface and CQDs, CD spectroscopy technique is used to measure the transmission difference between the right and left CPL of the hybrid system. First, the chirality of the pure CQDs was determined by measuring CD spectrum of the CQDs spin-coated on sapphire substrate. It should be noted that the spin-coating conditions were kept identical across all reported results on both sapphire and metasurfaces. The results shown in the figure 2a obviously indicate that the CQDs, as expected^[17,18], show no preference for either left or right CPL. A chiral metasurface was then fabricated with lattice period $P=1000\text{nm}$ and total square array area of $50\mu\text{m}\times 50\mu\text{m}$ (see

details in Figure S1). Its CD spectrum is depicted in the black curve of figure 2b showing strong chiral responses in the visible range with a peak at 650nm. Notably, the wavelength of the CD peak is shorter than the lattice period, which could introduce the ordinary diffraction in this regime. However, these ordinary diffractive channels are from the interference with a non-resonant contribution, while the chiral properties in our case originate from the plasmonic resonance which is reflected in the zero order. Therefore we don't consider the influence from these diffractive channels as many pervious reports [39, 43, 45] do. When the metasurface is hybridized with CQDs, the CD spectral response of the hybrid system (red curve in figure 2b) shows a dramatic rise around the 617nm wavelength when compared to the bare chiral metasurface (black curve in figure 2b). The rise can be attributed to the fact that, at this wavelength, the hybrid metasurface/CQDs system absorbs more right CPL than the bare chiral metasurface, while the pure CQDs show no preference, as mentioned earlier. This confirms that there exists coupling between the CQDs and chiral metasurface, inducing new chiral features. Full wave simulations (figure 2c) of the chiral metasurface show good spectral agreement to the experimental data (figure 2b). A clear dependence of the spectral features on the excitation spin states can be seen in the electric field maps for LCP and RCP light at the CD peak of 650nm (figure 2d and 2e). Figure 2f illustrates the experimental setup used to measure the CD, where a quarter waveplate is inserted in the optical path of a microspectrometer to obtain the left and right CPL to illuminate the sample (see details in supporting information).

Furthermore, to verify the chiral nature of the hybrid system, both left and right handed chiral metasurfaces are prepared (see details in Figure S1). The chiral responses of bare left and right handed chiral metasurfaces are presented in Figure 3a, in which a very typical mirror image is seen in the CD spectra due to the intrinsic chirality of the chiral pattern. Then, we measured the CD spectra of the two hybrid systems respectively formed by the left and right handed metasurfaces combined with the identical achiral CQDs. The experimental

results shown in Figure 3b illustrate another almost perfect mirror image as well as the fact that the whole hybrid system possesses a chiral nature.

In order to explore the origin of the chiral features in the hybrid system, the CQDs are combined with metasurfaces of varying lattice periods (900nm, 950nm and 1000nm, see details in Figure S1), and the CD spectra of the bare metasurface and hybrid CQDs/metasurface systems are compared on the background of the absorption spectrum of the bare CQDs. Figure 4a reports the CD spectra for a metasurface with period $P=1000\text{nm}$. The CD spectra of both the bare chiral metasurface (black curve) and the hybrid system (red curve) have two peaks (dashed arrows serve as guides for the eye), with the peak at shorter wavelength showing a significant increase in magnitude in the case of the hybrid metasurface/CQDs. By comparing the CD spectra with the absorption of the CQDs (blue dashed line), we find that the positions of the CD increase well coincides with the peak position in the CQDs absorption spectrum while there is no obvious change detected (black dashed arrow) at longer wavelengths where the CQDs absorption is low. This confirms that the achiral CQDs can be chirally activated when their absorption bands overlap with the chiral responses of the metasurface. To verify this interpretation, we studied the hybrid systems of CQDs combined with chiral metasurfaces having identical unit cell but different lattice periods, which provide controllable shifting chiral responses. It should be emphasized here that the ultra-fine control of chirality is achieved by modifying the propagating mode of the chiral metasurfaces, which has been well studied and confirmed in *ref 36 and 37*. Figure 4b shows a similar analysis for a chiral metasurface with period of 950nm, whose CD spectral peaks are $\sim 20\text{nm}$ blue-shifted with respect to the metasurface of 1000nm period (black curve in figure 4b). This implies that the overlap of the CD spectra with the CQDs absorption spectra is also shifted and the effect is reflected in the CD spectrum of the hybrid system (red curve in figure 4b). The peak at longer wavelengths, which has now a better overlap with the CQD absorption, shows a rise while a drop can be observed for the peak at shorter wavelength

because its chiral response is shifting away from the absorbing peak of the CQDs. A third metasurface, with period of 900nm and more blue-shifted chiral response has confirmed this trend (figure 4c). In this case the longer wavelength peak of the CD spectrum (black curve in figure 4c) fully overlaps with the absorption peak of the CQDs (blue dashed line in figure 4c), resulting in a remarkable lift of the corresponding CD peak for the hybrid system (red curve in figure 4c), while the CD peak at short wavelengths, now overlapping with a dip in the CQD absorption, almost disappears. The described differences in the behavior between the hybrid CQDs/metasurface system and the bare metasurface clearly suggest a preferential absorption of the right CPL for the CQDs when combined with an appropriately designed chiral metasurface. This indicates that metasurfaces can be applied to induce chiral response into otherwise achiral CQDs.

We also studied the chiral response from the perspective of light emission from the CQDs and analyzed the luminescent circular polarizations of a hybrid CQDs/chiral metasurface (left-handed) system with a period of $P=1000\text{nm}$. The experimental setup is shown in figure 5a. The sample was excited by a continuous wave He-Cd laser at optical wavelength of 442 nm with an angle of $\sim 45^\circ$ to surface normal. The photoluminescence (PL) signal was collected in the forward direction and analyzed by a silicon-based charge-coupled device (CCD) and a monochromator after passing through a zero-order quarter-wave plate, a linear polarizer and an achromatic half-wave plate (see details in the Methods). We measured and compared the RCP and LCP emission intensity of the CQDs on the chiral metasurface and that of CQDs outside the chiral patterns. The results are displayed in figures 5b and 5c. It can be clearly seen that for the CQDs on the chiral metasurface, there is a very strong asymmetry in the RCP and LCP PL intensity due to the coupling of the PL from CQDs to the left-handed chiral metasurfaces (figure 5b). The degree of circularly polarized luminescence (g) was determined to be as high as 0.17 based on the formula: $g = (I_R - I_L) / (I_R + I_L)$, where I_R , I_L represent the intensity of left and right circularly polarized luminescence, respectively.

1 Additionally, we also measure the chiral emission from the enantiomeric system based by
2 CQDs in a right-handed chiral metasurface, which displays a dissymmetric degree around -
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4 0.13 (see details in Figure S2). The result further confirms the induced chiral nature of the
5
6 CQDs. It is understandable that the small difference between the absolute value of the two
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8 dissymmetric degrees is caused by fabrication imperfections and uneven distribution of CQDs.
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11 To account for the high degree of CPL, we refer to the induced CD of CQDs which is
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13 displayed in the figure 5d. This CD spectrum is obtained by using the CD of hybrid system
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15 minus the CD of metasurface. It is found that the CD spectrum shows a strong CD band from
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17 600nm to 650nm, which approximately corresponds to the PL emission band of CQDs. Due
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19 to the resonant coupling between the metasurface and CQDs, the up transition rates are
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21 different for spin-up and spin-down electrons at the valence band maximum to spin-down and
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23 spin-up states respectively at the conduction band minimum under the excitations of CPL. In
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25 further, this different up transition rates(absorption rates of CPL) lead to different down
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27 transition rates(emission rates of CPL) based on Einstein coefficients^[46], which thus enables
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29 the selective CPL emission. As for the case without coupling with metasurface, these
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31 transition rates are kept equal, as a result, there is no CD and CPL emission detected from the
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33 sole CQDs, which is also consistent with previous reports ^[17,18] and further proves the
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35 resonant coupling between the CQDs and chiral metasurface. Notably, the remarkable CPL
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37 emission directly from the CQDs combined with the chiral metasurfaces is in free space and
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39 room temperature, which may enable practical CPL emitters and benefit various applications,
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41 such as bio-sensing.
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54 3. Conclusion

55 In conclusion, we have demonstrated a hybrid system that uses chiral metasurfaces to
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57 induce chiral properties into achiral CQDs. A significant CD can be induced by the chiral
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metasurfaces on the achiral CQDs as evidenced by the CD and absorption spectra. By utilizing the identical CQDs but different chiral metasurfaces with tunable chiral responses through altering their lattice period, it is shown that enhanced chiral responses of the hybrid system happen only if the chiral response of the chiral metasurface overlap the CQD absorption bands. A direct circular polarized emission from the hybrid system has been achieved with a degree of circular polarization approaching 0.17, which further demonstrates the resonant coupling between the CQDs and chiral metasurfaces. This is the first demonstration of chirally driving CQDs by chiral metasurfaces. Our proposed system and findings may provide a new path to modulate CQDs response with chirality and hold great potentials in various applications, including CQD spintronics and optoelectronics, quantum communications and bio-sensing.

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Supporting Information

Supporting Information is available from the Wiley Online Library.

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Figures

Figure 1. CQDs combined with chiral metasurfaces. (a) Schematic illustration of the hybrid system in which the red dots represent CQDs and yellow film with slits is the chiral metasurface. (b) Scanning electron microscope (SEM) image of the left-handed chiral unit cell. (c) PL spectra of the CQDs without chiral metasurface. (d) Transmission electron microscope (TEM) image of the CQDs.

Figure 2. Circular dichroism difference caused by introducing the achiral CQDs on chiral metasurface. (a) CD spectrum of pure CQDs on sapphire substrate. (b) CD spectrum of the chiral metasurface (black curve) and CD spectrum of the CQDs combined chiral metasurface (red curve). (c) Stimulated CD spectrum of the chiral metasurface. (d) Electric field distribution of the chiral metasurface excited by LCP at the wavelength of 650nm. (e) Electric field distribution of the chiral metasurface excited by RCP at the wavelength of 650nm. (f) The setup for measuring the CD spectra by microspectrophotometer.

Figure 3. The experimental demonstration of the chiral nature in the CQDs combined system. (a) CD spectra of the left and right handed chiral metasurfaces. (b) CD spectra of the hybrid systems with opposite handedness.

Figure 4. CQDs coupled with the chiral metasurfaces with various lattice periods. CD spectra of the chiral metasurfaces (black curve) and the CQDs combined system (red curve) with the chiral metasurfaces' period of (a) 1000nm, (b) 950nm and (c) 900nm.

Figure 5. CPE detected from the photoluminescence of the CQDs combined system. (a) The setup adopted for measuring the luminescent circular polarizations from the CQDs' emitting light. The experimental circular polarized PL intensity of the CQDs (b) on the chiral metasurfaces and (c) out the chiral metasurface. (d) The CD spectrum of CQDs which is measured by using the CD of hybrid system minus the CD of metasurface.

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