

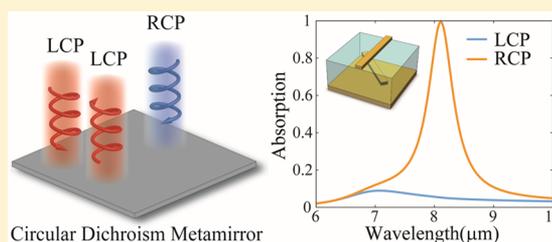
## Circular Dichroism Metamirrors with Near-Perfect Extinction

Zuojia Wang,<sup>†,‡,§</sup> Hui Jia,<sup>†,||</sup> Kan Yao,<sup>⊥</sup> Wenshan Cai,<sup>#</sup> Hongsheng Chen,<sup>\*,‡,§</sup> and Yongmin Liu<sup>\*,†,⊥</sup><sup>†</sup>Department of Mechanical and Industrial Engineering and <sup>⊥</sup>Department of Electrical and Computer Engineering, Northeastern University, Boston, Massachusetts 02115, United States<sup>‡</sup>Department of Information Science and Electronic Engineering, Zhejiang University, Hangzhou 310027, China<sup>§</sup>The Electromagnetics Academy at Zhejiang University, Hangzhou 310027, China<sup>||</sup>College of Science, National University of Defense Technology, Changsha 410073, China<sup>#</sup>School of Electrical and Computer Engineering, Georgia Institute of Technology, Atlanta, Georgia 30332, United States

## Supporting Information

**ABSTRACT:** In nature, the beetle *Chrysin gloriosa* derives its iridescence by selectively reflecting left-handed circularly polarized light only. Here, an optical analogue is suggested based on an ultrathin metamaterial, which is termed circular dichroism metamirror. A general method to design the circular dichroism metamirror is presented under the framework of Jones calculus. It is analytically shown that the building block of such a metamirror needs to simultaneously break the  $n$ -fold rotational ( $n > 2$ ) symmetry and mirror symmetry. By combining two layers of anisotropic metamaterial structures, a circular dichroism metamirror is designed in the mid-infrared region, which shows perfect reflectance for left-handed circularly polarized light without reversing its handedness, while it almost completely absorbs right-handed circularly polarized light. These findings offer a new methodology to implement novel photonic devices for a variety of applications, including polarimetric imaging, molecular spectroscopy, and quantum information processing.

**KEYWORDS:** metamaterials, symmetry, polarimetry, circular dichroism, plasmonics



In addition to the intensity, wavelength, and phase, the polarization state of light plays a critical role in light–matter interactions. Biological species show remarkable examples in this regard. For instance, the vision sensitivity to linearly polarized light helps squids to detect transparent prey,<sup>1</sup> while *Gonodactylus smithii* (mantis shrimps) can unambiguously distinguish left-handed circularly polarized (LCP) and right-handed circularly polarized (RCP) light.<sup>2</sup> *Chrysin gloriosa* (jeweled beetles) under LCP illumination appear more brilliant than those under RCP illumination.<sup>3</sup> In condensed matter physics and quantum information technology, manipulating the spin of electrons with circularly polarized light is also of critical importance.<sup>4–7</sup> Therefore, the efficient analysis and engineering of the polarization state is imperative in diverse disciplines, including biology, physics, materials science, and quantum optics.

Because of the unprecedented ability to create novel material properties and control the flow of light in a prescribed manner, metamaterials have gained tremendous interest over the past decades.<sup>8–10</sup> A number of metamaterial-based polarization converters have been proposed and demonstrated.<sup>11–14</sup> In particular, metamaterials provide a powerful strategy to achieve enormous chiral responses.<sup>15</sup> For instance, three-dimensional metallic helices, rotating rods, or DNA-assembled nanoparticles exhibit strong intrinsic chirality,<sup>16–22</sup> which can further induce negative refractive index or negative reflection.<sup>23–26</sup> Metamaterials also demonstrate giant gyrotropy and strong circular

dichroism<sup>27–29</sup> that are far beyond the reach of natural media.<sup>30</sup> Very interestingly, diode-like asymmetric transmission has been demonstrated through the manipulation of the anisotropy and dissipation in planar chiral structures.<sup>31–33</sup> However, most of these works focus on the performance of circular dichroism in transmission, while much less attention has been paid to reflective chiral structures, which are equally important in optical engineering.

The recently developed ultrathin, planar metamaterials, known as metasurfaces,<sup>34,35</sup> promise simple and low-loss designs in comparison with the counterpart of bulk metamaterials. They allow us to tailor the polarization,<sup>36–38</sup> effectively preserve the handedness in reflection,<sup>39</sup> and manipulate the ray trajectory of circularly polarized light.<sup>40–42</sup> Quarter-wave plates<sup>43</sup> and optical activity<sup>44</sup> with nonchiral structures can be achieved by controlling the phase discontinuities. With these new approaches, metasurfaces have also been employed for unidirectional coupling of surface plasmon polaritons,<sup>45–47</sup> which could be very useful in on-chip nanophotonics. In addition, integrating a ground metal layer can dramatically enhance the light–matter interaction in metasurfaces and leads to high-efficiency generation of circularly polarized light.<sup>42,48</sup> Nevertheless, these demonstrated

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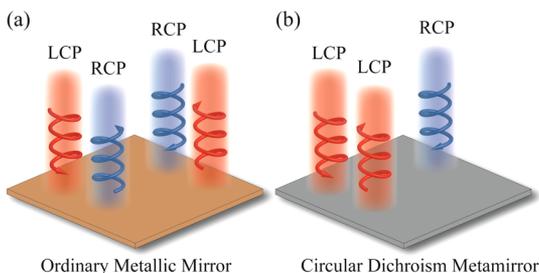
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metasurfaces so far modulate the amplitude and/or phase for both LCP and RCP light with equal efficiency. It is thus important to exploit a novel approach to selectively control the states of circular polarization for distinctly different responses, especially for reflected light.

In this Letter, we propose and demonstrate the concept of circular dichroism (CD) metamirrors, which enables selective, near-perfect reflection of designated circularly polarized light without reversing its handedness, yet complete absorption of the other polarization state. Such a metamirror can be considered as the optical analog of *Chrysin gloriosa* in nature, while exhibits nearly maximal efficiency. To achieve this functionality, Jones calculus is first employed to predict the required reflection matrix. The subsequent analyses in structural symmetry show that the necessary condition is to break both the  $n$ -fold rotational ( $n > 2$ ) and mirror symmetries. On the basis of the transfer matrix method (TMM), a proof-of-concept CD metamirror utilizing an ultrathin, bilayer metamaterial is then designed and optimized in the mid-infrared region. Simulation results show 94.7% reflectance and 99.3% absorption for LCP and RCP waves, respectively. Hence we can achieve selective perfect absorption for circularly polarized light, which is different from previous metamaterial-based absorber work.<sup>49–52</sup> In addition, the device performance is insensitive to the incident angle and the offset between the two layers of metamaterials. Such an extremely high extinction ratio as well as the tolerance in geometry and illumination manifests promising applications in polarimetric detection, CD spectroscopy, and quantum optical information processing. Furthermore, the planar metamirror is only quarter-wavelength thin, superior to chiral sculptured thin films<sup>53</sup> with thickness larger than wavelength, and hence it is highly desirable for on-chip device integration. It should be noted that Zheludev's group recently presented a similar concept in the microwave region,<sup>54</sup> and Valentine's group demonstrated circularly polarized light detection at the near-infrared wavelength.<sup>55</sup> However, our current work, as shown in the following, provides the general requirements of structural symmetries, which are more stringent than the 2D chirality as previously discussed.<sup>54,55</sup> These analyses clearly unravel the underlying physical mechanism of the proposed device.

## RESULTS

The concept of CD metamirrors is illustrated in Figure 1. For an ordinary metallic mirror, reversal of handedness occurs when LCP or RCP light is reflected off its surface at the normal



**Figure 1.** Schematic of the optical response of an ordinary metallic mirror and a CD metamirror. (a) An ordinary mirror reverses the handedness of both LCP and RCP waves in reflection. (b) A CD metamirror reflects only LCP waves and preserves the handedness, while it totally absorbs RCP waves.

incidence. Here RCP (LCP) is defined if the electric vector rotates clockwise (counterclockwise) when an observer looks along the wave propagation direction. For oblique incidence, the reflected light is elliptically polarized in general. This phenomenon dramatically complicates the optical setup and characterization when circularly polarized light is involved. In contrast, the function of the proposed CD metamirror is to selectively reflect a certain circularly polarized wave and preserve its handedness over a relatively wide range of incident angles. For simplicity, selective reflection of an LCP wave is considered in the following. This means incident LCP light is expected to be perfectly reflected by the metamirror while the RCP wave is completely absorbed. Such a property is distinctly different from the previous metasurface designs, which act as a half-wave plate to convert the handedness of the reflected wave regardless if it is LCP or RCP.<sup>39</sup>

The functionality of the CD metamirror is urgently needed in optical engineering. Circularly polarized light is usually generated by the combination of a linear polarizer and a quarter-wave plate, which are often bulky optical elements. Multiple reflections from a series of metallic mirrors are often unavoidable in an optical setup. Consequently, optical signals can hardly maintain their original handedness after many reflecting surfaces. The proposed metamirror may be an ideal means to address these issues. As we know, linearly polarized light can be decomposed into two circularly polarized light with opposite handedness. When it illuminates on the CD metamirror, only the LCP component can be reflected with preserved handedness and the RCP component will be completely absorbed, thus generating circularly polarized light. Furthermore, the preservation of the handedness after reflection can suppress the mode conversion between two circular polarization states and reduce the complexities in optical instruments.

In the following, we first analyze the necessary condition for selective reflection for one designed state of circular polarization based on Jones calculus. Consider two half-spaces separated by a metamirror at  $z = 0$ . The fields in the two regions can be related via Jones calculus:<sup>56</sup>

$$\begin{pmatrix} E_r^x \\ E_r^y \end{pmatrix} = \begin{pmatrix} r_{xx} & r_{xy} \\ r_{yx} & r_{yy} \end{pmatrix} \begin{pmatrix} E_i^x \\ E_i^y \end{pmatrix} = \mathbf{R} \begin{pmatrix} E_i^x \\ E_i^y \end{pmatrix} \quad (1)$$

$$\begin{pmatrix} E_t^x \\ E_t^y \end{pmatrix} = \begin{pmatrix} t_{xx} & t_{xy} \\ t_{yx} & t_{yy} \end{pmatrix} \begin{pmatrix} E_i^x \\ E_i^y \end{pmatrix} = \mathbf{T} \begin{pmatrix} E_i^x \\ E_i^y \end{pmatrix} \quad (2)$$

where  $\mathbf{R}$  and  $\mathbf{T}$  are the reflection and transmission matrices of the metamirror, while  $E_i^x$ ,  $E_r^x$ , and  $E_t^x$  are the incident, reflected, and transmitted electric fields polarized in the  $x$  direction, respectively. The similar notations with a superscript of  $y$  represent the field polarized in the  $y$  direction. Accordingly, RCP and LCP waves can be represented by  $(1, i)^T$  and  $(1, -i)^T$ , respectively. The transmission matrix must equal zero ( $\mathbf{T} = 0$ ) for an ideal, reflecting mirror. For the reflection matrix  $\mathbf{R}$ , we can readily prove that complete reflection of LCP waves and total absorption of RCP waves require (see Supporting Information)

$$\mathbf{R} = \begin{pmatrix} r_{xx} & r_{xy} \\ r_{yx} & r_{yy} \end{pmatrix} = \frac{e^{i\alpha}}{2} \begin{pmatrix} 1 & i \\ i & -1 \end{pmatrix} \quad (3)$$

where  $\alpha$  is an arbitrary phase shift through the metamirror and a time-harmonic propagation of  $e^{-i\omega t}$  is assumed. To characterize the optical behavior, we can analyze the eigenstates of the polarization, which are uniquely related to the symmetry of the structure. By solving the eigenvalue problem of the reflection matrix  $\mathbf{R}$  given by eq 3, we obtain the eigenvalue  $\kappa = 0$  with the eigenvector  $(1, i)^T$ . This implies that no light is reflected at all if the metamirror is illuminated by RCP light. In contrast, LCP incident light is completely reflected, and its handedness is preserved.

To better understand the physical meaning behind the desired reflection matrix in eq 3, now we perform symmetry analysis and identify the nature of structural symmetry required for the CD metamirror. Mathematically, rotating the structure by an arbitrary angle with respect to the  $z$ -axis can be accomplished by applying the following matrix operation:<sup>56</sup>

$$\mathbf{R}_{\text{new}} = \mathbf{D}_{\varphi}^{-1} \mathbf{R} \mathbf{D}_{\varphi}, \text{ with } \mathbf{D}_{\varphi} = \begin{pmatrix} \cos(\varphi) & \sin(\varphi) \\ -\sin(\varphi) & \cos(\varphi) \end{pmatrix} \quad (4)$$

where  $\varphi$  is the rotation angle and  $\mathbf{R}_{\text{new}}$  is the new reflection matrix of the structure after rotation. If the structure has a certain rotational symmetry, the invariance of the reflection matrix under transformation requires  $\mathbf{R}_{\text{new}} = \mathbf{R}$ . The general condition of such rotational symmetry can then be obtained, given by

$$\sin(\varphi) \begin{pmatrix} r_{xy} + r_{yx} & r_{yy} - r_{xx} \\ r_{yy} - r_{xx} & -r_{xy} - r_{yx} \end{pmatrix} = 0 \quad (5)$$

To satisfy both the desired reflection matrix in eq 3 and the rotational symmetry condition in eq 5, the only solution is  $\varphi = m\pi$ ,  $m = 0, \pm 1, \dots$ . Consequently, to achieve an ideal CD metamirror, only the  $C_2$ -symmetric group is allowed.

Other than the rotational symmetry, another basic characteristic of artificial structures is the mirror symmetry. Similarly, we can employ the method of matrix transformation to predict the requirement of the mirror symmetry for an ideal CD metamirror. If the metamirror is mirror-symmetric with respect to a plane that is rotated from the  $xz$ -plane by an angle  $\varphi$  with respect to the  $z$ -axis, the reflection matrix for the structure reflected at that plane is identical to the original one. Therefore, we have

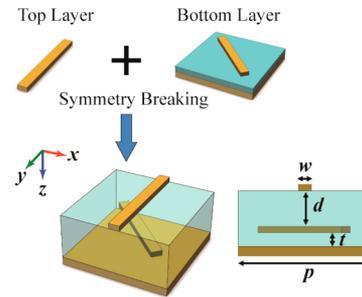
$$\mathbf{R}_{\text{new}} = \mathbf{A}_x^{-1} \mathbf{D}_{-\varphi}^{-1} \mathbf{R} \mathbf{D}_{-\varphi} \mathbf{A}_x \quad (6)$$

where  $\mathbf{A}_x = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}$  is the mirror matrix with respect to the  $x$ -axis and  $\mathbf{D}_{-\varphi}$  is given by changing  $\varphi$  to  $-\varphi$  in  $\mathbf{D}_{\varphi}$ . Again by applying the symmetry condition  $\mathbf{R}_{\text{new}} = \mathbf{R}$ , we obtain

$$\sin(2\varphi)(r_{xx} - r_{yy}) + 2 \cos(2\varphi)r_{xy} = 0 \quad (7)$$

According to eq 3, the ratio of  $(r_{xx} - r_{yy})$  to  $r_{xy}$  is a pure imaginary number, so that the mirror-symmetric condition described by eq 7 can never be met. Hence, selective reflection of circular polarization does not exist in structures that contain mirror symmetry with respect to the incidence plane.

The proceeding analyses conclude the general yet necessary condition to realize CD metamirrors: simultaneous breaking of the  $n$ -fold rotational ( $n > 2$ ) and mirror symmetries. A straightforward design to satisfy the symmetry condition is to combine two anisotropic metamaterial interfaces with a relative twisting angle, as schematically illustrated in Figure 2. Here, a continuous metallic wire array, probably the simplest structure



**Figure 2.** Schematic of the design procedure for CD metamirrors. Bilayer metamaterials with a relative twisting angle are employed to simultaneously break the  $n$ -fold rotational ( $n > 2$ ) and mirror symmetries. One unit cell is shown in the figure.

for anisotropy, is adopted as the top layer, and the bottom layer consists of a metallic rod array placed above an optically thick metallic reflector. The total transmission is hence zero. A dielectric material with refractive index  $n_d$  is used as the spacer between the top metallic wires and the bottom metallic reflector.

We apply the transfer matrix method in the CD metamirror design and optimization. In general, for a stratified planar structure between two media  $a$  and  $b$ , a  $4 \times 4$  matrix  $\mathbf{M}$  can be used to relate the forward to the backward propagating fields:<sup>12</sup>

$$(E_{bx}^{(f)}, E_{by}^{(f)}, E_{bx}^{(b)}, E_{by}^{(b)})^T = \mathbf{M} (E_{ax}^{(f)}, E_{ay}^{(f)}, E_{ax}^{(b)}, E_{ay}^{(b)})^T \quad (8)$$

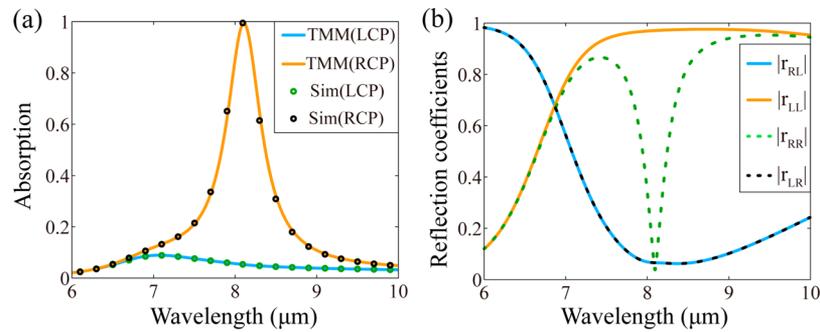
Here the superscripts (f) and (b) denote the forward and backward propagating light, respectively. The overall transfer matrix  $\mathbf{M}$  can be expressed as multiplication of the transfer matrix of each layer (see Supporting Information). Once the transfer matrix is fixed, the reflection matrix for the entire system is then given by

$$\mathbf{R} = - \begin{pmatrix} m_{33} & m_{34} \\ m_{43} & m_{44} \end{pmatrix}^{-1} \begin{pmatrix} m_{31} & m_{32} \\ m_{41} & m_{42} \end{pmatrix} \quad (9)$$

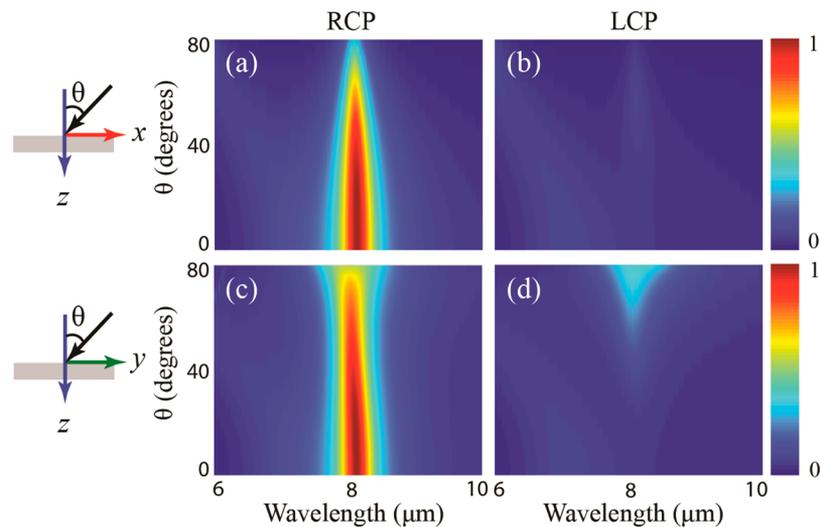
where  $m_{nl}$  is the  $n$ th row and  $l$ th column component of the overall  $4 \times 4$  transfer matrix  $\mathbf{M}$ . By combining the transfer matrix method with the appropriate optimization algorithm, we can determine the desired thickness of the dielectric spacing layer.

The proof-of-concept CD metamirror is numerically demonstrated in the mid-infrared frequency region. The width of both the top metallic wire and the bottom metallic rod is  $w = 0.2 \mu\text{m}$  and the length of the latter is  $l = 2 \mu\text{m}$ , with the period of  $p = 2.5 \mu\text{m}$ . The metallic rod is rotated by  $\pi/4$  with respect to the  $z$ -axis to eliminate the  $n$ -fold rotational ( $n > 2$ ) and mirror symmetries. A dielectric material with refractive index  $n_d = 1.51$  is used to separate different metallic layers with the thickness  $d = 1.55 \mu\text{m}$  and  $t = 0.09 \mu\text{m}$ . Gold is taken as the metal, and the thickness of the metallic wire and rod is  $0.1 \mu\text{m}$ . The permittivity of gold is described by the Drude model after fitting the experimental data.<sup>57</sup>

We validate the function of the CD metamirror by performing full-wave numerical simulations (CST Microwave Studio). As shown in Figure 3a, the absorption of the structure is as high as 99.3% for RCP light, while only 5.3% for LCP light at the wavelength of  $8.1 \mu\text{m}$ . The simulation results are in perfect agreement with the analytical calculation based on the transfer matrix method, because of the weak coupling between the top and bottom layers. If the thickness of the middle



**Figure 3.** (a) Numerically simulated absorption spectra of the CD metamirror by CST (data dots) and calculated results based on the transfer matrix method (solid lines). (b) Simulated reflection coefficients of the CD metamirror for circularly polarization incidences.



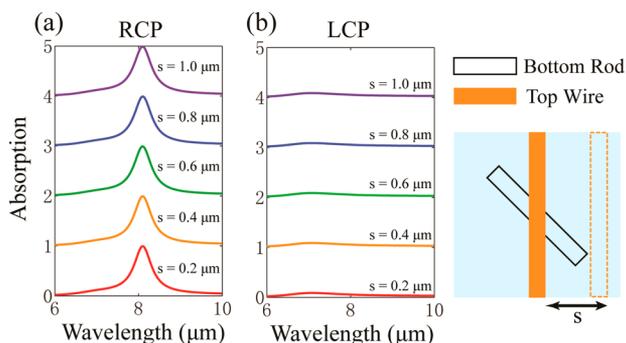
**Figure 4.** Simulated absorption spectra of (a, c) RCP and (b, d) LCP states as a function of incident angle  $\theta$ , when the incident light propagates in the  $xz$ -plane or  $yz$ -plane, respectively.

dielectric is reduced to deep subwavelength scales, the interaction between the top and bottom gold structures gradually dominates and leads to a slight discrepancy (see [Supporting Information](#)). Moreover, from the reflection spectrum presented in [Figure 3b](#), we find that for LCP incidence the magnitude ratio of the reflected LCP ( $r_{LL}$ ) to the reflected RCP light ( $r_{RL}$ ) can reach up to 14.8. Therefore, not only can the proposed metamirror selectively reflect LCP waves with high efficiency, but also the handedness is preserved. This is distinctly different from previous metasurface work,<sup>39,42</sup> which maintains the handedness of the reflected wave for both LCP and RCP light without any extinction.

The underlying physical mechanism of selective reflection originates from the interference of multiple reflections. The LCP and RCP waves can be considered as mirror images of each other with respect to the  $yz$ -plane. The mirror symmetry in the top layer of continuous metallic wires guarantees the identical optical behavior for the two states of circular polarization. However, the lack of  $yz$ -plane mirror symmetry in the bottom layer of metallic rods enforces different phase shifts for the two polarization states, which can be understood in the context of the Pancharatnam–Berry phase.<sup>40</sup> The orientation of the bottom metallic rod is rotated 45 degrees from the  $y$ -axis, giving rise to 90-degree phase accumulation in opposite signs for the LCP and RCP light, respectively. The total phase difference between the reflected circularly polarized light with the opposite handedness is therefore 180 degrees.

When constructive interference and thus perfect reflection occur for the LCP incidence, the RCP light is totally absorbed, arising from the destructive interference.

Another remarkable feature of our CD metamirror is its good performance over a wide range of incident angles. The angular sensitivity of the metamirror is investigated for two different cases for the wave vector confined in the  $xz$ -plane and  $yz$ -plane, respectively. One can clearly see from [Figure 4](#) that the operating wavelength does not significantly change under oblique incidences. Moreover, the absorption of two polarization states still maintains a high contrast (>80%) for an incident angle range up to 40 degrees. The main reason for this feature is that the bottom metamaterial structure exhibits insensitivity of resonance frequency to the incident angle (see [Supporting Information](#)). Such an omnidirectional performance of the CD metamirror is highly beneficial in its application as a single-mode mirror for circularly polarized light. We further investigate the influence of the offset between the two layers of metamaterials, which may occur in practical fabrications. As shown in [Figure 5](#), the performance of the metamirror is very robust even when the top wire has a lateral offset of up to 1  $\mu\text{m}$  with respect to the bottom rod. Such a property is understandable, because the current structure works in the weak-coupling regime. The top layer can be always considered as a homogeneous anisotropic metasurface regardless of the offset.



**Figure 5.** Dependence of absorption on the offset between the top and bottom metamaterial structures. (a) and (b) are for RCP and LCP illuminations, respectively. The neighboring absorption spectra are offset by unity along the vertical axis for clarity.

## DISCUSSION

In summary, we have proposed a circular dichroism metamirror to mimic the iridescent attributes of beetles in nature, yet showing near-perfect extinction. The breaking of both the rotational and mirror symmetries is theoretically proven as the necessary condition in the structural design. By employing the transfer matrix method, we design and optimize a metamirror comprising two layers of anisotropic metamaterials in the mid-infrared region, which in principle can be scaled to other wavelengths. It is shown that such a metamirror can almost perfectly reflect all the LCP light with preserved handedness, while totally absorbing the RCP incidences. The prominent tolerance in the incident angle and geometry offset strongly implies the promising applications of the CD metamirror in the detection and manipulation of circularly polarized light. Furthermore, similar to the recently demonstrated helicity-dependent near-field optics, intriguing phenomena may arise when we combine the proposed structure with plasmonics.<sup>45,58,59</sup> Finally, we think that the method developed here is universal, opening a new gateway to achieving advanced single-mode devices for circularly polarized light that are beneficial for both classical and quantum topics.

## ASSOCIATED CONTENT

### Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acsp Photonics.6b00533.

Derivation of the reflection matrix for circular dichroism metamirrors, discussion of the requirements of structural symmetry for different cases, details of the structural optimization based on the transfer matrix method, discussion of the influence of interaction between two metamaterial interfaces, an explanation of why the performance of the designed metamirror is insensitive to the incident angle, and discussion of the dependence of the metamirror performance on the twisted angle of the two layers (PDF)

## AUTHOR INFORMATION

### Corresponding Authors

\*E-mail: y.liu@northeastern.edu.

\*E-mail: hansomchen@zju.edu.cn.

### Notes

The authors declare no competing financial interest.

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