The ability to control the spin-orbit interaction (SOI) of light in optical microresonators is of fundamental importance for future photonics. Organic microcrystals, due to their giant optical anisotropy, play a crucial role in spin-optics and topological photonics. Here, the controllable and wavelength-dependent Rashba–Dresselhaus (RD) SOI is realized that is attributed to the anisotropic excitonic response in an optical microcavity filled with an organic microcrystalline. This work investigates the transition of the spin-splitting from twice winding caused by the splitting of the transverse-electric and transverse-magnetic modes to once winding caused by the RD effect. The interplay of the two allows engineer the SOI of light in organic microcavities, which besides its fundamental interest promises applications in spin-controlled on-chip integrated nanophotonic elements, toward exploiting nonmagnetic and low-cost spin-photonic devices.

1. Introduction

The spin degree of freedom of electrons has been extensively exploited in condensed matter physics and is at the heart of many protocols for efficient data transfer, quantum-information processing, and storage. The fundamental physics of their potential applications relies on the splitting of the electronic energy bands induced by spin-orbit interaction (SOI), whose essential feature is that a spin-polarized electron moving in a static electric field experiences an effective magnetic field. The SOI of electrons in solid-state systems was originally realized in structure-inversion-asymmetric materials and bulk-inversion-asymmetric materials by Rashba and Dresselhaus, respectively. Light possesses the intrinsic polarization degree of freedom, where the polarization of the photons is akin to the spin characters of electrons. In optics, the SOI of light has opened remarkable new opportunities to manipulate the pseudospin of photons, which has inspired the study of the optical spin Hall effect (OSHE) and optical topological insulators. In these areas, the synthetic gauge fields created by the SOI play a central role.

It is known that the SOI of light in optical microcavities can be caused by the transverse electric-transverse magnetic (TE-TM) mode splitting, which can be modeled as an effective magnetic field and by constructing artificial gauge fields. Besides the TE-TM splitting, the SOI in optical microcavities can also be induced by other effects, such as the chirality of the materials as well as the so-called Rashba–Dresselhaus (RD) Hamiltonian, which can also contribute to the construction of the synthetic gauge fields to further manipulate the spin states of photons. It is known that the effective magnetic field caused by the TE-TM splitting winds twice in reciprocal space, while the effective magnetic field caused by the RD effect winds only once. It remains a great challenge but is highly desirable to integrate multiple of these different types of SOI into the same optical microcavity. This would not only facilitate the tuning of the spin splitting between different types but also provide an interesting platform for the study of a novel type of topological photonics to efficiently control the pseudospin of light.

In the study of the SOI of light, organic crystalline materials have received significant attention because their large optical anisotropy brings out significant H (horizontal)-V (vertical) splitting. Recently, the controllable spin splitting and observation of different types of SOI have been reported in optical microcavities filled with liquid crystals (LC) and α-perylene single-microcrystals. In the α-perylene single-microcrystal
microcavities, the SOI arises because of the H-V splitting, but it is difficult to realize the RD effect due to the nearly constant energy splitting of the linearly polarized modes of opposite parity. This energy splitting results mainly from the nonresonant linear birefringence of the anisotropic organic crystals, with a much smaller contribution from the excitonic response that is not explicitly used or discussed in detail. Tuning the optical modes more into resonance with the excitonic excitations, however, introduces a pronounced exciton-mediated contribution to the system’s SOI with fundamental changes to the topology of the overall optical response: i) Due to the resonant nature the energy splitting of the linearly polarized modes becomes wavelength-dependent instead of a constant. In other words, the excitons only strongly influence those cavity photon modes that are near the resonance. As the cavity modes move away from the excitonic resonance, the influence of the excitons reduces gradually. ii) With the pronounced anisotropy of the dipole coupling to the excitonic excitations, coupling only occurs to one of the two perpendicular cavity modes, leaving the other one unaffected. With these fundamental observations, involving the excitonic excitations, coupling only occurs to one of the two perpendicular cavity modes, leaving the other one unaffected. Further, the moderate crystalline anisotropy, in the vicinity of the exciton resonance, the curvatures of the V modes become much smaller, while the H modes are not substantially influenced. This leads to the possibility for one H mode to simultaneously cross with multiple V modes and allows the introduction of RD and chirality SOIs in the same microcavity. The interplay of the spin splittings induced by different types of SOI is investigated in detail. With this we introduce an approach not relying on strong external magnetic/electric fields that by using purely optical means allows the SOI control in organic microcrystalline resonators, which gives access to a unique toolbox for advanced quantum control strategies in on-chip integrated photonics.

2. Result and Discussion

2.1. Principles of Spin-Orbit Interaction in Optical Microcavities

To realize complex SOIs in one microcavity, we consider an anisotropic cavity medium which can possess TE-TM splitting, linear birefringence, as well as resonant exciton-photon interaction. In a simplified picture the system’s effective Hamiltonian \( H = H_{\text{VX}} + H_{\text{TEM}} + H_{\text{RD}} \) in the circular polarization basis can be written in a 2 × 2 matrix form. The TE-TM splitting appears as off-diagonal terms in the dispersive part of the Hamiltonian, making use of the Pauli spin matrices \( \{ \sigma_x, \sigma_y, \sigma_z \} \) and introducing a polariton pseudospin vector that experiences an effective magnetic field as a result of finite TE-TM splitting. The contribution of the linear birefringence can lead to the energy splitting at \( k = 0 \), giving rise to the so-called YX (or H-V) splitting part \( H_{\text{VX}} = \hat{\sigma}_x \). Due to the coupling to anisotropic excitons, the energy splitting that occurs at \( k = 0 \) becomes wavelength-dependent and only one of the two orthogonally polarized cavity modes is significantly influenced. As a result, the strongly affected cavity modes start to move away from their counterparts of the same parity and toward their counterparts of opposite parity. When two perpendicular cavity modes of opposite parity meet each other at \( k = 0 \), the RD SOI is induced, accompanied by a spin splitting, i.e., \( H_{\text{RD}} = -2a \hat{\sigma}_y k_x \). The total Hamiltonian takes the explicit form:

\[
H (k) = \begin{pmatrix}
\frac{\hbar^2 k_x^2}{2 m_x} + \frac{\hbar^2 k_y^2}{2 m_y} - 2a k_y & \beta_0 + \beta_1 k_x e^{2i\varphi} \\
\beta_0 + \beta_1 k_x e^{-2i\varphi} & \frac{\hbar^2 k_x^2}{2 m_x} + \frac{\hbar^2 k_y^2}{2 m_y} + 2a k_y
\end{pmatrix}
\]

where \( m_x (m_y) \) is the effective mass of cavity photons along \( x (y) \) direction, which are typically different in anisotropic media. \( \alpha \) represents the strength of the linear splitting or RD effect when the Rashba and Dresselhaus coupling strengths are equal to each other, contributing to only \( k_x \) direction in the in-plane momentum space. \( \beta_0 \) represents the strength of the TE-TM splitting, and \( \varphi (\varphi \in [0, 2\pi]) \) is the polar angle. \( \beta_1 \) denotes the energy splitting of linearly polarized modes of opposite parity at \( k = 0 \) (here, we define it as \( \beta_1 = E_{V_x} - E_{H_x} \), where \( E_{V_x} \) and \( E_{H_x} \) are the ground state energies of V and H modes of opposite parity). Different from the \( \alpha \)-perylen crystal, the value of \( \beta_1 \) in this case is determined by two factors, i.e., the linear birefringence and the anisotropic exciton-photon interaction. The contribution from the linear birefringence is linked to the crystallographic axes and thus remains constant due to the immutability of organic crystals. However, due to the wavelength-dependent anisotropic exciton-photon interaction, the value of \( \beta_1 \) varies gradually with the wavelength of the cavity modes. Although the influence of the excitons is not evidently given in the Hamiltonian (1), the mediation of them is reflected in the changing of the effective mass of optical modes. Without the participation of the excitons, the effective mass of the optical modes is almost wavelength-independent, while the excitons can strongly alter the effective mass of the near resonant optical modes.

We first consider the case of \( \beta_1 < 0 \). Two eigenstates of the Hamiltonian (1) are shown in Figure 1a,d,g,j with \( \alpha \neq 0, \beta_1 \neq 0 \), and \( m_x = 1.5 m_y \). Here, the two perpendicular cavity modes have opposite parity. The anisotropic effective mass and strong H-V splitting result in that the eigenmodes in reciprocal space are strongly squeezed (not circles anymore) along \( k_x \) and \( k_y \) directions (Figure 1d, where two perpendicular ovals are formed), respectively. The lower eigenmode in Figure 1g is flatter compared with the upper one, while the situation reverses in Figure 1j where the two modes cross at finite momentum. These kinds of crossing points cannot be opened in Figure 1j, because the linear splitting, which is the determining factor to open them, lies only along \( k_y \) direction, without contribution to \( k_x \) direction.
When $\beta_0 = 0$, the two cavity modes are in resonance at $k = 0$, leading to the spin splitting (i.e., the RD effect) along $k_y$ direction as shown in Figure 1h. When $\beta_0 > 0$, the linear splitting results in the anti-crossing of the two modes of opposite parity (Figure 1i), which was observed in our previous report.\cite{21} In Figure 1f, one can see that the two perpendicular ovals are rotated by 90° compared with the ones in Figure 1d. According to the above analysis, the strong anisotropy of the material and the mediation of the excitons may give rise to the intersecting of an $H$ (V) mode with multiple V (H) modes (c.f. Figure 1m). This configuration requires more cavity photon modes with small energy difference between them, which can be achieved in a relatively thicker microcavity. In this case, the anti-crossing at finite momentum and the RD effect close to zero momentum may occur simultaneously as predicted in Figure 1n (note that the dispersion relation presented in Figure 1n is a schematic diagram, not calculated directly from a $4 \times 4$ Hamiltonian).

2.2. Experimental Realization

To experimentally realize the above theoretical prediction, we fabricate an optical microcavity, in which $\beta$-phase single-crystalline perylene nanosheets are sandwiched between two-layer metallic films as sketched in Figure 2a. Perylenes, due to their excellent optical properties,\cite{26,27} special polaritonic bands, and birefringent crystals,\cite{19} provide a wonderful platform for the study of topological photonics.\cite{21} More importantly, perylene polymorphs provide an effective approach that affects the physico-chemical properties of crystals and avoid the difficulty in time-consuming molecular synthesis.\cite{28,29} The microscope image clearly shows that these nanosheets have uniform rhombus morphology and smooth surface (Figure 2b). The X-ray diffraction (XRD) analysis indicates that these rhombus sheets possess the $\beta$-phase structure (Figure S1, Supporting Information), i.e., their unit cell comprises two molecules and forms a standard herringbone stacking pattern (Figure 2c). The characteristic photon stopband in the transmission spectrum has also confirmed this assignment (Figure S2, Supporting Information).

We perform angle-resolved reflectivity (ARR) measurements of the microcavity with 3.2-µm $\beta$-phase perylene (Figure S3, Supporting Information) at room temperature. The thicker microcavity gives rise to the relatively small energy difference between neighboring cavity modes, which is essential to observe the crossing of multiple cavity modes. Figure 2d shows the corresponding two-dimensional (2D) reflectivity dispersion at $k_y = 0$ (i.e., along $Y$-direction as indicated in Figure 2c) and two sets of modes with different curvatures can be clearly seen. One set of the modes marked by the red dotted lines has larger curvatures, which is almost wavelength-independent and evenly distributed in the spectral range between 400 and 660 nm. These modes are consistent with the fitted individual H-polarized cavity modes which
Figure 2. Organic single-crystalline microcavity and its angle-resolved reflectivity (ARR). a) Schematic diagram of the microcavity structure. PS and SiO$_2$ represent polystyrene film and silicon dioxide, respectively. b) Microscopy image of the as-prepared $\beta$-phase perylene microcrystals. c) Molecular orientation in the $\beta$-phase perylene crystal. Its unit cell comprises two molecules, forming a standard herringbone stacking pattern. The two testing $X$- and $Y$-directions are denoted by the red and blue arrows, respectively. d) ARR of the microcavity with the organic-layer thickness of 3.2 $\mu$m. The red and yellow dashed lines represent the simulation of the H and V cavity modes, respectively. The blue dashed line represents the exciton line. The numbers are the corresponding indices.

are calculated by using the coupled harmonic oscillator (CHO) model. The other set of the modes (V-polarized) marked by the yellow dotted lines exhibits wavelength-dependent curvatures, i.e., they become smaller as approaching the exciton resonance of 469 nm (the blue dashed line in Figure 2d). Note that the experimentally measured V modes agree well with the calculated ones (considering the participation of excitons) at larger momenta, but they bifurcate at smaller momenta because of the RD effect (see the detailed discussion below). We further measured the polarization-dependent ARR of the cavity modes by adding a linear polarizer to the detection optical path (Figure S4, Supporting Information). It is found that these two sets of the cavity modes are perfectly orthogonally polarized, i.e., H modes are polarized along $X$-direction, whereas V modes along $Y$-direction. Obviously, the wavelength-dependent curvatures of V modes are a typical feature given by the exciton-photon interaction. The influence of the excitons on H modes, however, can be neglected. The anisotropic light-matter interaction enables the intersection of an H mode with multiple exciton-mediated V modes, toward the realization of the theoretical hypothesis in Figure 1n.

2.3. Interaction of Multiple Cavity Modes

Figure 3a shows the magnified ARR spectrum (Figure 2d). Clearly, the energy splitting $\beta'_n$ (defined as $\beta'_n = E'_n - E'_{n-1}$, where the superscript $n$ represents the mode index) reduces gradually and finally approaches 0 around 575 nm (see also Figure 4a). The nonconstant splitting $\beta'_n$ results in complicated and rich mode interaction in such a microcavity. First, the adjacent H and V modes of the same parity cross at $|k| > 0$, for example, clear crossing points appear at 509 nm (black dashed circle). Second, H and V modes of opposite parity anti-cross at around $k_y = \pm 10.15$ $\mu$m$^{-1}$ (white dashed circles), similar to the phenomenon observed in the $\alpha$-phase-perylene microcavity. Such an anti-crossing produces a nonzero local Berry curvature, although the global Berry curvature remains zero. The anti-crossing section is validated by the 2D tomography measurements (Figure 3b, more details can be found in Figure S5, Supporting Information). It is worth mentioning that the anti-crossings occur for the V$_n$ mode and the H$_{n+1}$ mode, hence it corresponds to the case of $\beta_0 > 0$ as shown in Figure 1i (also compare with Figures 1f and 3b). Importantly, at longer wavelengths, H and V modes of opposite parity are close to each other and eventually present energy degeneracy at $k = 0$ (blue dashed circle). Comparing the experimental and simulated (yellow lines in Figure 2d) results of the V modes, one can see their obvious deviation at $k_y = 0$. At the same time, different spin components split along $k_y$ directions (see Figure 3k and Figure S6, Supporting Information), evidencing the RD effect. If focusing on the cavity H$_4$ mode in Figure 3a, it is clear that it anticrosses with the V$_4$ mode at $k = 0$, which triggers the RD spin splitting (Figure 3j). This behavior is consistent with our theoretical expectation in Figure 1n.
2.4. Spin Splitting and Transition

We performed three-dimensional (3D) tomography and measured the Stokes vector components\(^\text{(31)}\) in energy-momentum space to analyze the spin splitting. From the 2D wavevector map of the tomography in Figure 3b, we can clearly see two mutually perpendicular and independent ovals that represent the \(H_2\) (the inner most horizontal oval marked by the dashed green line) and \(V_3\) (the vertical oval marked by the dashed red line) cavity modes, respectively. Additionally, the \(V_1\) mode crosses with the \(H_3\) mode (the mode outside of the \(H_2\) mode) and anti-crosses with the \(H_4\) mode (the mode outside of the \(H_3\) mode). Around the anti-crossing point at larger momentum (\(k_y = 10.15 \, \mu m^{-1}\)), one can see the twice winding of the circular polarizations (the region marked by the dashed circle in \(S_3\) component of the Stokes vector in Figure 3g). We note that at this wavelength the RD SOI cannot be clearly recognized. As the wavelength increases, \(V\) modes shrink faster than \(H\) modes in reciprocal space. At some point (e.g., white dashed line in Figure 3a), one can see that the \(V_4\) mode becomes tangent to the outer \(H_4\) mode along \(k_y\) direction and simultaneously is tangent to the inner \(H_3\) mode along \(k_x\) direction (Figure 3c). Here, the near resonant \(H_3\) and \(V_4\) modes enhance the RD effect, leading to the partial spin splitting (i.e., the spin components of the two innermost circles are not completely split) as shown in Figure 3h. At the wavelength 532 nm (blue dashed line in Figure 3a), although the \(H_1\) mode disappears, the \(V_4\) mode is still visible (Figure 3d) and the spin components strongly split as can be seen in Figure 3i. Remarkably, in Figure 3i, the twice winding of the circular polarizations of the \(H_4\) mode (the mode outside of the innermost one) is still visible although the contrast becomes weaker in comparison with the \(H_4\) mode (red dashed circle) in Figure 3g. However,
as the wavelength increases further to 542 nm, corresponding to the cyan dashed line in Figure 3a, the spin splitting and once winding of the $H_4$ mode can be clearly seen (innermost ring in Figure 3i). The detailed transition process between the states in Figures 3i,j can be found in Figure S7 (Supporting Information). From Figure 3e, one can see that there is no exchange between the $H_4$ mode and the outer $V_5$ mode, indicating that the RD effect is not dominating here. The spin splitting caused by the RD SOI can be distinctly observed at round 550 nm (Figure 3f,k). The dependence of the RD splitting value ($\alpha = 2\beta_0'k_0$)\(^{[12]}\) on the mode numbers is shown in Figure 4a. For an even longer wavelength (≈600 nm), two orthogonally polarized modes of opposite parity ($H_6$ and $V_7$) are brought into resonance with $\beta_0' = 0$ (Figure 4a) and a very clear splitting of the parabolic dispersions along $k_y$ direction can be seen in Figure 3a (spin-dependent dispersions can be found in Figure S6, Supporting Information).

Figure 4b–d presents six polarization components, collected at $k_y = 3 \mu m^{-1}$ along the vertical black dashed line in Figure 3a, depending on the mode indices (or the wavelength). The horizontally and vertically polarized components ($S_1$ in Figure 4b) become weaker at larger indices (longer wavelength), while the trend of the circularly polarized components ($S_3$ in Figure 4d) is opposite, demonstrating that the RD SOI dominates at longer wavelength where $\beta_0' \rightarrow 0$. Interestingly, the diagonally and anti-diagonally polarized $S_2$ components swap sign around the transition region of different windings of the circular polarizations (Figure 4c).

### 3. Conclusions

In conclusion, we have realized different SOIs in a $\beta$-phase single-crystalline perylene (organic) microcavity, mediated by anisotropic and frequency dependent exciton-photon interaction. In such microcavity, the exciton dipoles oriented along $Y$-direction affect only the cavity $V$ modes, which lead to the dispersion of the $V$ modes to have much smaller curvature. Consequently, we observed simultaneously the spin splitting carrying a twice winding at larger momenta and the spin splitting carrying once winding, induced by the RD SOI, at smaller momenta. Different spin splittings can even appear on the same cavity mode. Hence this system provides a platform for the study of the transition of different SOIs. Our work also paves the way for studying the influence of the RD SOI on the strong light-matter coupling states, e.g., exciton-polaritons, in semiconductor microcavities and manufacturing low-cost spin photonic devices.

### Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Data Availability Statement

Research data are not shared.

Conflict of Interest

The authors declare no conflict of interest.

Keywords

anisotropic excitonic response, organic microcrystalline cavities, Rashba–Dresselhaus effect, spin-orbit interactions, TE–TM splitting

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