Supplementary Materials for

Spin-orbit–coupled exciton-polariton condensates in lead halide perovskites

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The PDF file includes:

Supplementary Text
Figs. S1 to S20
Legends for movies S1 and S2

Other Supplementary Material for this manuscript includes the following:

Movies S1 and S2
Supplementary text 1: Justification of Hamiltonian

The Hamiltonian is written using the formalism of a spin ½ particle in the presence of a magnetic field. This is possible due to the mathematical equivalence of the Poincaré Sphere and the Bloch Sphere, where the Poincaré Sphere represents the possible spin states of a free photon, whereas the Bloch Sphere represents possible spin states of an electron. To see why this is helpful, first consider relevant bases either in the presence of just the crystalline birefringence or just the usual TE-TM splitting that occurs in any DBR cavity.

Let’s consider first consider crystalline anisotropy. For the purposes of this discussion, we will consider only the in-plane components of the dielectric tensor, as this simplification is sufficient to capture the features observed in this report, although in principle at increasingly large angles of incidence, an incoming TM-polarized wave will be affected by this out of plane component of the dielectric function. Within this approximation, we can consider the in-plane birefringence to be that of a uniaxial crystal, where the orientation of the fast and slow axes with respect to the coordinate frame is determined by the angle the crystal is oriented. For example, if the a = (1,0,0) axis has the lowest refractive index, and is aligned along the x axis, then the y axis will correspond
to \(b = (0,1,0)\), and would be the slow axis. In this case, an incident wave, oriented normal to the cavity, polarized along the y axis would experience an effective cavity length larger than one polarized along the x axis. Therefore, we will see two optical modes; the photon spin degeneracy is broken, in favor of modes polarized along either \(\mathbf{\hat{x}}\) or \(\mathbf{\hat{y}}\). The principle holds in general – any incident waves will be decomposed into the ordinary and extraordinary rays as a consequence of this in-plane birefringence, as the eigenmodes are polarized strictly along the crystallographic axes, which may be rotated as a consequence of crystal rotation.

TE-TM splitting, a more familiar consequence of DBR cavities, may also be incorporated. The physical origin of this effect is derived from the interfacial field matching conditions prescribed by Maxwell’s Equations which distinguish between fields oscillating along or perpendicular to the plane of an interface. The TE wave will always be oscillating in the plane of the interface, whereas for increasingly large incidence angles the TM wave will oscillate perpendicular to the plane of the interface, thus giving it a different reflectivity. Near normal incidence, the polarization direction of a TM wave will also be in the plane, but will be polarized perpendicular to the TE polarization. It is also important to note that because the TE polarization direction is always perpendicular to the plane of incidence, the TE polarization direction will rotate \(4\pi\) for a \(2\pi\) rotation of polar angles. This effect is well documented (e.g. Panzarini et. al., 1999, Physics of the Solid State), and leads to a TE-TM splitting the grows like \(k_\parallel^2\). Because the energy difference is with respect to the TE and TM polarizations, these polarizations remain the basis of the eigenmodes in a cavity where this is the dominant effect breaking spin symmetry.

Within the Poincarè Sphere formalism, the splittings discussed above may be easily incorporated as an effective field which leads to eigenfunctions that correspond to the proper basis. Importantly, it also allows for efficient calculation of a situation, such as in this report, where multiple effective fields are present, leading to optical modes which are not well described either in the basis of X,Y polarized modes or TE-TM polarized modes. In the same way that a magnetic field’s direction can cause the electron spin to be aligned with that direction, we introduce an effective field which will cause the photon spin to be polarized along that given direction. For the fields considered here, that corresponds to all possible linearly polarizations, which lie on the equator of the sphere.
Supplementary text 2: SOC polariton modeling

When the SOC cavity photons are strongly coupled to an exciton resonance with Rabi splitting $\Omega$, we write the SOC polariton Hamiltonian, excluding the photonic and excitonic damping rates, as,

$$
H_{\text{pol}} = \begin{pmatrix}
E_o + \frac{\hbar^2 k_{\parallel}^2}{2m} & \Omega/2 & -\alpha e^{-i\phi} + \beta k_{\parallel}^2 e^{-2i\phi} & 0 \\
\Omega/2 & E_{xc} & 0 & 0 \\
-\alpha e^{i\phi} + \beta k_{\parallel}^2 e^{2i\phi} & 0 & E_o + \frac{\hbar^2 k_{\parallel}^2}{2m} & \Omega/2 \\
0 & 0 & \Omega/2 & E_{xc}
\end{pmatrix}
$$

This Hamiltonian is written with the assumption that the exciton resonance is itself isotropic, or at least negligible compared with the linewidth of the polariton modes. This Hamiltonian suggests four polariton bands: two lower polariton branches, and two upper polariton branches, where the two modes in a given branch correspond to one of two polarization states. To highlight the role of the effective magnetic field in the strong coupling regime, we use the eigenvectors of the traditional $2 \times 2$ coupled oscillator Hamiltonian to partially resolve the effects of strong coupling, and identify how strong coupling effectively modulates the strength of the effective magnetic field within the cavity. To achieve this, we perform a matrix transformation as demonstrated below:

$$
H'_{\text{pol}} = M^{-1} H_{\text{pol}} M, \quad M = 1 \otimes V
$$

where $V$ is written by placing the eigenvectors of the coupled oscillator Hamiltonian as column vectors, i.e., we perform a unitary transformation into the eigenbasis of the isotropic polariton modes which have been introduced into the spin space via a tensor product. These eigenvectors are written below for completeness. The final matrix result (below) has also had the indices rearranged to highlight the re-appearance of the effective magnetic field, $G$, in this form of the matrix.

$$
|v_1\rangle = X_{up}|Exc\rangle + P_{up}|Ph\rangle, \quad |v_2\rangle = X_{up}|Exc\rangle + P_{up}|Ph\rangle,
$$

$$
V = (v_1, v_2) = \begin{pmatrix} X_{up} & X_{lp} \\ P_{up} & P_{lp} \end{pmatrix}
$$
Here we have written the effective magnetic field terms as $G = -\alpha e^{-i\phi_0} + \beta k_\parallel e^{-2i\phi}$. The $X, P$ are the Hopfield Coefficients, which are given by: $X_{UP} = \frac{1}{\sqrt{2}} \left( 1 + \frac{\Delta E}{\Delta E^2 + \Omega^2} \right)^{\frac{1}{2}}, P_{UP} = \frac{1}{\sqrt{2}} \left( 1 - \frac{\Delta E}{\Delta E^2 + \Omega^2} \right)^{\frac{1}{2}} \Delta E = E_{ex} - E_{ph} - \frac{\hbar^2 k_\parallel^2}{2m}$. The polariton branch energies are given by: $E_{UP,LP} = \frac{1}{2} \left( \Delta E \pm \sqrt{\Delta E^2 + \Omega^2} \right)$. Note that the Hopfield coefficients switch when considering a different polariton branch, i.e., $P_{UP} = -X_{LP}, X_{UP} = P_{LP}$. We maintain the distinction of the upper and lower branch terms to help clarify the role of the photonic component in the effective magnetic field. These terms quantify the projection of a given eigenvector onto the basis of the Hamiltonian – a pure photon or exciton state, and therefore their squares represent the photon or exciton fraction of a given eigenfunction. Their values can range anywhere from zero to one, and in the case of strong coupling at the anti-crossing point, they are both equal to 1/\sqrt{2}.

The first important property to note about this transformed 4×4 Hamiltonian is that it can be well approximated as being two independent blocks, with a negligible coupling between the blocks. These coupling terms are given by $XPG$, and will couple a given polarization within the upper/lower branch to either polarization of the opposite polariton branch. The subscripts are dropped here because either subscript denotation would be equivalent. Because the coupling terms within a branch refer to a splitting amongst otherwise degenerate modes, they are much more consequential than coupling terms to another mode which is energetically well separated. The closest the upper and lower polariton branches can be is at the anti-crossing point, where they are separated by the Rabi splitting. In our system there is approximately 30 meV of separation. The effective magnetic field’s energetic coupling in this same range is about an order of magnitude smaller, leading to a negligible coupling outside of a given polariton branch. As an example calculation, using typical values found within this report ($\Omega = 30 \text{ meV}, k_\parallel = 5 \mu \text{m}^{-1}, \alpha = 5 \text{ meV}, \beta = 0.5 \text{ meV} \left( \mu \text{m}^{-1} \right)^{-2}$), we identify that a typical effective magnetic field (restricted to the approximate form shown below) leads to approximately a 5 meV energy correction at the anti-
crossing point, and that the addition of the full field correction, changes the energies less than 1 meV. For this reason, we further discuss the approximate form of the Hamiltonian;

\[
H'_{pol} \approx \begin{pmatrix}
E_{UP} & P^2_{up}G & 0 & 0 \\
0 & 0 & E_{LP} & P^2_{LP}G \\
0 & 0 & P^2_{LP}G^* & E_{LP} \\
0 & 0 & 0 & E_{LP}
\end{pmatrix}
\]

In this format, is now clear that the phase of the field field is approximately preserved, noting that there is some small polarization mixing we neglect in this form, although its total strength is strongly modulated by the appearance of strong coupling. To be precise, the strength of the effective magnetic field in a polariton branch is simply the photonic fraction at a given momentum times the effective field strength that corresponds to a purely photonic mode. Example simulations (e.g. Fig. S10) demonstrate that the anisotropy splitting is only important when the polariton mode exhibits an appreciable photonic component, which is why the lower polariton spin branches collapse onto one another as they approach the exciton resonance, whereas the upper polariton spin branches converge onto the purely photonic modes at high momenta. Fig. S10 further demonstrates the preservation of the effective magnetic field, where panels C,D show simulated spin texture at the energy corresponding to the diabolical point, showing that the major effect of strong coupling is to raise the effective mass of the mode.

For the modeling, we estimated the exciton resonance based on polariton dispersions with a series of cavity detunings and the reflectance spectra of the crystals. The wedged thickness of our sample allows us to continuously tune the cavity mode across the exciton resonance. By examining the polariton dispersions with various detunings, the exciton resonance can be estimated based on the flatten curvature at high \( k \) momentum. For example, the exciton resonance at 77K is about 2.3525 eV, which is slightly larger than the PL emission peak. We have further confirmed that this value matched well to the absorption peak measured in ref. Nat. Communs. 2019, 10, 1175, i.e., 2.3525 eV at 80 K. Since our crystals have thickness about several micrometers, Fig. S3, the optical properties are expected to be same as bulk crystals measured in the reference. The reflectance spectra of our samples are also provided in Fig. S4. The excitonic feature at room temperature is much less noticeable than at low temperature. The various dispersions are provided in Fig. S13 for room temperature and Fig. S16 for 77 K.
Supplementary text 3: Rashba-Dresselhaus Hamiltonian around the diabolical points

In this section we provide a mathematical justification towards representing the Hamiltonian (Eq.1) as a modified Rashba-Dresselhaus Hamiltonian in the vicinity of the diabolical points. That is, we will expand this Hamiltonian for momentum vectors, \( k \), close to the of a diabolical point, \( k_{db} \), i.e., small \( q = k_\parallel - k_{db} \). We find it useful to decompose this small vector \( q \) into the components parallel and perpendicular to the diabolical point vectors – see figure S19 for visual aid. Further, for sufficiently small vectors \( q \) the projection onto \( k_\parallel \) is identical to the projection \( k_{db} \). By use of a small angle approximation and noting that \( q_\perp \) is always perpendicular to the diabolical point vector \( k_{db} = \pm \frac{\alpha}{\beta} \left( \cos \left( \frac{\phi_o}{2} \right), \sin \left( \frac{\phi_o}{2} \right) \right)^T \), we can write an equation that relates the change in the polar angle for small excursions about \( \phi = \frac{\phi_o}{2} \): \( \delta = \frac{\sin \left( \frac{\phi_o}{2} \right) q_x - \cos \left( \frac{\phi_o}{2} \right) q_y}{k_{db}} = \frac{q_\perp}{k_{db}} \).

We will prefer to keep things written in terms of \( q_\perp, q_\parallel \) for convenience, although to appreciate the connection to the Rashba-Dresselhaus Hamiltonian, they will be decomposed into \( q_x \cdot q_y \) later. We also note that the quantity, \( k_{db}^2 \), which appears repeatedly in Eq.1, will now be represented in terms of the diabolical point and the small vector \( q: k_{db}^2 = k_{\parallel db}^2 + q^2 + 2k_{db} \cdot q = k_{\parallel db}^2 + q^2 + 2k_{db}q_\parallel \).

Using small angle approximation, we write the exponential (Eq.1) as follows:

\[
e^{-2i\phi} = e^{-2i\left(\frac{\phi_o}{2}+\delta\right)} \approx \cos(\phi_o) + \frac{2q_\perp}{k_{db}}\sin(\phi_o) - i \left(\sin(\phi_o) - \frac{2q_\perp}{k_{db}}\cos(\phi_o)\right)
\]

Using this expression, we can express the upper right hand side of the Hamiltonian as:

\[
H_{0,1} = -\alpha(\cos(\phi_o) - i\sin(\phi_o)) + (\alpha + \beta q^2 + 2\sqrt{\alpha\beta}q_\parallel)\left(\cos(\phi_o) + \frac{2q_\perp}{k_{db}}\sin(\phi_o) - i \left(\sin(\phi_o) - \frac{2q_\perp}{k_{db}}\cos(\phi_o)\right)\right)
\]

After cancelling certain terms, we are left with the following expression:

\[
H_{0,1} = \beta q^2(\cos(\phi_o) - isin(\phi_o)) + \alpha \frac{2q_\perp}{k_{db}}(\sin(\phi_o) + i\cos(\phi_o)) + 2\sqrt{\alpha\beta}q_\parallel(\cos(\phi_o) - i\sin(\phi_o))
\]

\[
H_{0,1} = \left(\beta q^2 + i\alpha \frac{2q_\perp}{k_{db}} + 2\sqrt{\alpha\beta}q_\parallel\right)e^{-i\phi_o} = \left(\beta q^2 + 2\sqrt{\alpha\beta}(q_\parallel + iq_\perp)\right)e^{-i\phi_o}
\]

By similar calculation, the Hamiltonian can be written in the following form:
\[
H = \left( E_o + \frac{\hbar^2}{2m} \left( k_{db}^2 + q^2 + 2k_{db}q_\parallel \right) \right) \left( \beta q^2 + 2\sqrt{\alpha\beta} (q_\parallel + iq_\perp) \right) e^{-i\phi_o} \\
= \left( E_o + \frac{\hbar^2}{2m} \left( k_{db}^2 + q^2 + 2k_{db}q_\parallel \right) \right) \left( \beta q^2 + 2\sqrt{\alpha\beta} (q_\parallel + iq_\perp) \right) e^{-i\phi_o}
\]

\[
H = \left( E_o + \frac{\hbar^2 k_\parallel}{2m} \right) I + \begin{pmatrix}
0 & \beta q^2 + 2\sqrt{\alpha\beta} (q_\parallel + iq_\perp) \\
\beta q^2 + 2\sqrt{\alpha\beta} (q_\parallel + iq_\perp) & 0
\end{pmatrix} = H_o + H_I
\]

In the second line, we have identified a distinction between the diagonal terms and the off-diagonal terms, which correspond to the field, that dictate the phase of the polariton branches at a given wavevector. We have neglected at this point the exponential phase terms, which correspond to a rotation about the z-axis on the Poincare sphere, i.e., \( H' = R_z(\phi_o)H_I R_z(-\phi_o) \). This corresponds to a universal shift of the emission angle. It is not physically observable, but in principle is derived from the rotation of the crystal by an angle \( \phi_o \).

At this point we switch to representing the small wavevector in terms of its x and y components.

\[
q_\parallel = q \cdot \frac{k_\parallel}{|k_\parallel|} \approx \cos \left( \frac{\phi_o}{2} \right) q_x + \sin \left( \frac{\phi_o}{2} \right) q_y; \quad q_\perp = q \cdot \frac{-k_{ix}}{|k_\parallel|} \approx \sin \left( \frac{\phi_o}{2} \right) q_x - \cos \left( \frac{\phi_o}{2} \right) q_y
\]

\[
H_I = \beta q^2 \sigma_x + \begin{pmatrix}
0 & \kappa_x q_x + \kappa_y q_y \\
\kappa_x q_x + \kappa_y q_y & 0
\end{pmatrix}
\]

Here we have used \( \kappa_x = 2\sqrt{\alpha\beta} \cos \left( \frac{\phi_o}{2} \right), \kappa_y = 2\sqrt{\alpha\beta} \cos \left( \frac{\phi_o}{2} \right) \) for notational simplicity. In this way we finally write

\[
H = \left( E_o + \frac{\hbar^2}{2m} \left( k_{db}^2 + q^2 + k_{db}q_\parallel \right) \right) I + \beta q^2 \sigma_x + \kappa_x \sigma \cdot q + \kappa_y \sigma \times q
\]

This is the form of the Hamiltonian about the diabolical points, in the limit of very small \( q \). Here when \( \sigma \) is the vector of Pauli matrices. Because of the cavity the momentum in the z-direction is quantized, we consider only the x,y directions for the momenta and Pauli matrices. Similarly, we consider only the z-component of \( \sigma \times q \). We can observe now that the Hamiltonian takes on the form of a modified Rashba-Dresselhaus Hamiltonian, with the addition of a term that goes like \( \beta q^2 \). Importantly, note that the weighting of the Rashba and Dresselhaus terms correspond to the crystal rotational angle, i.e., that for certain crystal orientations a purely Rashba- or Dresselhaus-like Hamiltonian can be achieved.
We now will highlight the connection of this interaction Hamiltonian with the graphene tight-binding Hamiltonian similarly expanded about the Dirac cone region. We can write the Hamiltonian in the following way, representing the phase terms as a complex exponential, rather than the $\kappa_x, \kappa_y$ terms:

$$H_l \approx \beta q^2 \sigma_x + 2\sqrt{\alpha \beta} \begin{pmatrix} 0 & (q_x - i q_y)e^{i\phi_0/2} \\ (q_x + i q_y)e^{-i\phi_0/2} & 0 \end{pmatrix}$$

This is the same Hamiltonian, but now written in a way that more intuitively relates to the crystal rotation angle. We can now clearly see how the rotation of the crystal rotates the momentum vector providing the coupling between the previously spin-degenerate branches. In addition, we note that this is very similar to the Hamiltonian in graphene (to first order in $q$, see Neto, A.C., Guinea, F., Peres, N.M., Novoselov, K.S. and Geim, A.K., 2009. Reviews of Modern Physics, 81(1), p.109), with the addition of a ‘twist’ which can turn the spin texture from a divergence to a curl structure. These properties are displayed in figure S.20, which shows the evolution of the spin texture upon crystal rotation.

We will now turn our attention to the eigenvalue within this same region. Based on the form of our Hamiltonian, we should expect to observe the traditional spin-split parabolas characteristic of a Rashba-Dresselhaus type Hamiltonian. We employ the same type of expansion of the eigenvalue equation to arrive at the following expression:

$$E_\pm(q \approx 0) - \left(E_o + \frac{\hbar^2 k_{db}^2}{2m}\right) = \frac{\hbar^2}{2m}(q^2 + 2k_{db} \cdot q) \pm 2\sqrt{\alpha \beta} q$$

If we write the eigenvalue strictly along the direction $q_\perp$, we then have the familiar Rashba-Dresselhaus eigenvalue equation:

$$E_\pm(q \approx 0) - \left(E_o + \frac{\hbar^2 k_{db}^2}{2m}\right) = \frac{\hbar^2}{2m}q_\perp^2 \pm 2\sqrt{\alpha \beta} q_\perp$$

**Supplementary text 4: Stokes Vectors and Connecting Hamiltonian To Polarization**
Stokes Vector Experimental Characterization: Stokes Vector is a 4-dimensional vector, which characterizes the full polarization state of an optical state, projecting the polarization onto 3 different bases, with a fourth component describing total intensity/ degree of polarization.

\[ \mathbf{S} = \begin{pmatrix} S_0 \\ S_1 \\ S_2 \\ S_3 \end{pmatrix} \]

Here, \( S_0 \) is the total intensity, and the degree of polarization is given by:

\[ p = \frac{\sqrt{s_1^2 + s_2^2 + s_3^2}}{s_0} \]

\( S_1, S_2, S_3 \) measure the projection onto the basis of horizontal/vertical polarizations, diagonal/anti-diagonal polarizations, and circular polarizations, respectively. So-called Mueller Matrices, \( \mathbf{M} \) can be constructed which allow computation of an output stokes vector, given an input stokes vector and an optical element;

\[ \mathbf{S'} = \mathbf{MS} \]

For the elements of interest here (quarter wave plate at some angle \( \theta \), and linear polarizer fixed to horizontal polarization transmission) we have the Mueller Matrices:

\[ \mathbf{M}_{\text{hbp}} = \frac{1}{2} \begin{pmatrix} 1 & -1 & 0 & 0 \\ -1 & 1 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix} \]

\[ \mathbf{M}_{\text{qwp}} = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & \cos^2(2\theta) & \cos(2\theta)\sin(2\theta) & \sin(2\theta) \\ 0 & \cos(2\theta)\sin(2\theta) & \sin^2(2\theta) & -\cos(2\theta) \\ 0 & -\sin(\theta) & \cos(2\theta) & 0 \end{pmatrix} \]

From these we can compute the \( S'_0 \) measured by the camera, after passing through both optical elements, in terms of the original stokes vector emitted from the microcavity:

\[ S'_0 = \frac{1}{2} \left( S_0 + S_1 \cos^2(2\theta) + S_2 \cos(2\theta)\sin(2\theta) + S_3 \sin(2\theta) \right) \]
To determine the Stokes vector, we measure the energy-, momentum-dependent dispersion for many choices of $\theta$, and then perform a discrete Fourier transform on this data to extract the Stokes vector parameters. Often the data is presented showing the angle of the linear polarization, which corresponds to the angle along the equator of the Poincaré Sphere, which may be computed as $\phi_{\text{stokes}} = \tan^{-1}\left(\frac{S_2}{S_1}\right)$. Note that this angle is not the same as the geometric angle expressed in the Hamiltonian. Rather, it is the same angle as those labelled in the scale bar of the phase plots, either experimental (Figs. 1G, 1H & 1I) or theoretical (Figs. 1G3, 1H3 & 1I3).

**Connecting Hamiltonian To Polarization:** The Hamiltonian under consideration here is written within the basis of circular polarizations, and its eigenvectors are complex-valued superpositions of left- and right-handed circular polarization states:

$$|\psi\rangle = c_1|+\rangle + c_2|-\rangle$$

From this (normalized) state, one can calculate the Stokes vector parameters, based on the transformation between the basis states;

$$\mathbf{S} = \begin{pmatrix}
|c_1|^2 + |c_2|^2 \\
2 \Re(c_2^*c_1) \\
-2 \Im(c_2^*c_1) \\
|c_1|^2 - |c_2|^2
\end{pmatrix}$$

where $\Re$, $\Im$ denote taking only the real or imaginary part of an expression. As noted before, this allows for the computation of the in-plane angle of the Stokes vector, i.e., the orientation of the polarization: $\phi_{\text{stokes}} = \tan^{-1}\left(\frac{S_2}{S_1}\right) = \tan^{-1}\left(-\frac{\Im(c_2^*c_1)}{\Re(c_2^*c_1)}\right)$. Results of these calculations are shown in Fig.S10, showing the spin texture of an entire mode, and are used to generate the theoretical comparison in Figs. 1G3, 1H3 & 1I3 and Figs S10, S20 by displaying the hue from the phase calculation and in the case of figures S10, the saturation is derived from using a Lorentzian-convolution of eigenvalue dispersion and then taking a cut of that convolution at a given energy.
Supplementary movies

Movie S1. Wavelength resolved Fourier space imaging of PL from an isotropic MAPbBr3 microcavity at room temperature.

Movie S2. Wavelength resolved Fourier space imaging of PL from an anisotropic CsPbBr3 microcavity at room temperature.
Supplementary figures

Fig. S1. Optical image of representative MAPbBr$_3$ (A) and CsPbBr$_3$ (B) single crystals in microcavity. Based on the crystal shape and the crystal growth behavior, we identify the $<100>$ axis of the (pseudo)cubic perovskite structure (indicated by the red arrows). At room temperature, the MAPbBr$_3$ is cubic phase (space group $Pm\bar{3}m$, $a = 5.948$ Å), whereas the CsPbBr$_3$ is orthorhombic phase (space group $Pbnm$, $a = 8.202$ Å, $b = 8.244$ Å, $c = 11.748$ Å). The orthorhombic phase can be considered as a pseudo-cubic phase, in which the unit cell parameter $a_c$ can be related to that of the orthorhombic cell, by $a_c = a/\sqrt{2} \approx b/\sqrt{2} \approx b/2$. Note that the spot-like area in B is the epoxy used to bond the DBRs.

Fig. S2. PXRD of the as-grown CsPbBr$_3$, in comparison with the standard PXRD pattern for CsPbBr$_3$ perovskite phase at room temperature. The perovskite crystals are highly oriented along $<110>$ direction in the $z$ direction.
Fig. S3. Atomic force microscopy of a representative CsPbBr$_3$ crystal, showing a thickness of ~2 µm.

Fig. S4. Reflectance spectra of the CsPbBr$_3$ crystal measured at room temperature and 77 K.
Fig. S5. Optical setup of Fourier space imaging with polarization-resolved capability. The pump laser is focused near the back focal plane of the objective lens to obtain a large uniform excitation spot on the sample. The Fourier plane is imaged through the entrance slit of the spectrometer, where the grating disperses the wavelengths perpendicular to the orientation of the entrance slit, onto a 2D camera array. The fully energy-momentum resolved Fourier space imaging can be constructed by (i) rotating the perovskite microcavity, i.e., equivalent to rotating the Fourier plane; or (ii) scanning the lens, i.e., equivalent to moving the Fourier plane across the entrance slit of the spectrometer. The full Stokes vectors can be measured through the use of a rotating quarter waveplate and a linear polarizer positioned before the entrance slit. The emission polarization is measured by positioning a half waveplate and a linear polarizer in front of the entrance slit.
Fig. S6. Polarization-dependent energy dispersions of an isotropic MAPbBr₃ microcavity, with the <100> axis of the perovskite crystals set to be parallel to the entrance slit of the spectrometer. The sample is at room temperature. The linear polarizer is oriented (i) vertical, i.e., parallel to the direction of the entrance slit; (ii) horizontal, i.e., perpendicular to the entrance slit; (iii) diagonal, i.e., oriented 45º with respect to the entrance slit. One can see that the TE and TM modes are degenerate at $k_\parallel = 0$. However, energy splitting between the two modes becomes evident at high $k_\parallel$, as shown by the arrows in c pointing two pairs of TE-TM modes. In general, the TE-TM splitting occurs when the frequency of the cavity is not equal to the center frequency of the stop bands in the DBRs, and the magnitude of the splitting scales with $k_\parallel^2$. 
Fig. S7. Tuning the synthetic SOC Hamiltonian by way of structural phase transition in a MAPbBr₃ microcavity: (A) cubic phase; (B) tetragonal phase; (C) orthorhombic phase. Top panels: energy dispersions of the three phases. The cubic phase exhibits single mode dispersions at $k_\parallel = 0$. By comparison, the tetragonal phase exhibits a small mode splitting at $k_\parallel = 0$, due to small in-plane anisotropy. As the structure enters the orthorhombic phase, the mode splitting at $k_\parallel = 0$ becomes obvious, due to the increased in-plane anisotropy. Middle panels: polarization-resolved PL spectra at $k_\parallel = 0$ of the three phases. The cubic phase exhibits no polarization dependence at $k_\parallel = 0$, as expected in the absence of crystalline anisotropy. Note that the intensity gradually decreases due to sample degradation. By comparison, the tetragonal and orthorhombic phases show the two split modes are orthogonal to each other. Bottom panels: polarization-resolved PL emissions of the peaks at $k_\parallel = 0$. One can observe the polarization pattern changes as the structure undergoes phase transition. Note that the discontinuity at 0° is due to slight photobleaching of the sample during the time course of the measurement.
Fig. S8. PL emission of the polariton at k = 0 from representative microcavities measured at room temperature and 77 K.

Fig. S9. Polarization-resolve photoluminescence spectra of a CsPbBr$_3$ single crystal at room temperature. One of the <100> axis of the (pseudo)cubic crystal is aligned with the entrance slit in front of the spectrometer. The vertical and horizontal polarizations are relative to the entrance slit.
Fig. S10. Phase-Resolved Properties of Anisotropic Polariton Hamiltonian (a,b). The hue represents the stokes vector orientation along the equator of the Poincarè Sphere, and the vector represents the effective field orientation and magnitude at a given momentum vector. Note that for eigenvector (a) the phase is aligned with the field, whereas for the other eigenvector (b) the phase is opposite to the effective field. (c,d) Figure c represents the phase-resolved simulated emission intensity about the diabolical point energy, in the absence of strong coupling. Figure d represents the phase-resolved simulated emission intensity about the diabolical point energy, including the strong coupling, showing the effective increase in the emission linewidth as a function of the decreased slope. (e,f) Black lines show the result of the full 4x4 Hamiltonian. The dotted red line shows the exciton energy, and the dotted blue lines show the results of the anisotropy Hamiltonian, before accounting for strong coupling. (g,h) Simulated phase-resolved momentum cuts depict a switch in the emission polarization of the polariton mode along the x direction (h), and showing the convergence of the two different polarized polariton modes as the exciton resonance is approached, along the y direction (g). All calculations presented here use the band parameters: $E_o = 2.1415 \text{eV}$, $m = 2.4 \times 10^{-4} m_e$, $\alpha = 5 \times 10^{-3} \text{eV}$, $\beta = 2.5 \times 10^{-4} \text{eV} \cdot \mu m^{-2}$, $E_{xc} = 2.165 \text{eV}$
Fig. S11. Polarization-resolved energy dispersion along $k_y$ in CsPbBr$_3$ microcavity measured at room temperature. (A) The energy dispersion along $k_y$. The two pairs of anisotropic modes of interest here are labeled as LP1a, LP1b, LP2a, and LP2b. (B, D, E) Polarization-resolved PL emission at $k_y = 0$, showing the two modes in a given pair are mutually orthogonal, and linearly polarized, in agreement with the theoretical predictions shown in Fig. S5. (C, F) Polarization-resolved PL emission of the LP2a at $k_y \sim 5.6$ $\mu$m$^{-1}$, above the diabolical point, showing linear polarization orthogonal to that of at $k_y = 0$. This is consistent with the pseudospin singularity at the diabolical point shown in Fig. S5. At high momentum, the emission of LP2b mode severely mix with the emission from the next, energetically lower mode or the nearby Bragg mode of the cavity, causing deviations from the theoretical predictions. Note that the discontinuity at 0° is due to slight photobleaching of the sample during the time course of the measurement.
Fig. S12. Polarization-resolved energy dispersion along $k_x$ in an anisotropic CsPbBr$_3$ microcavity, measured at room temperature. (A) The energy dispersion along $k_x$. (B, D, E) Polarization-resolved PL emission at $k_x = 0$, showing the two modes in a given pair are mutually orthogonal linear polarized, in agreement with the theoretical predictions shown in Fig. S5. (C, F) Polarization-resolved PL emission at $k_x \sim 5.6 \mu$m$^{-1}$, showing roughly orthogonal linear polarizations for the two modes. The polarizations of the two modes maintain the same along $k_x$. Note that the discontinuity at 0º is due to slight photobleaching of the sample during the time course of the measurement.
Fig. S13. Details of polariton dispersions near the diabolic points for MAPbBr$_3$ in the orthorhombic phase. (A) Constant energy cross section from an orthorhombic MAPbBr$_3$ microcavity showing the diabolical points. (B) The dispersion in the direction perpendicular to $k_\ast$, and intersecting the diabolical points shows two offset probolas.
**Fig. S14.** Dispersions collected on a series of crystals grown at different positions at room temperature. These dispersions show deviations from parabolic dispersions at high $k$ and avoided crossings with the exciton resonance for the higher energy polariton modes.
Fig. S15. Temperature-dependent energy dispersion measured at a selected spot on CsPbBr₃ microcavity. The evolution of the dispersion again confirms strong exciton-photon coupling presence at low temperature. It is also shown that that the strength of coupling gradually decreases as the temperature increases, due to the broadening of excitonic linewidth (as revealed by the temperature-dependent PL and absorption spectra in a previous study). Note that the energies of both cavity modes and excitons change when the temperature changes.
Fig. S16. Dispersions collected on a series of crystals grown at different positions at 77 K, along with the corresponding models with the coupled oscillator, showing positive, resonant, and negative detunings for the lower polariton branches. The white and magenta dashed lines are the dispersion of two SOC optical cavity modes. The solid lines are the modeled lower polariton branches using the coupled oscillator. The corresponding modeling gives $\Omega \sim 20$-25 meV.
Fig. S17. Pump power dependences showing the two lasing thresholds. (A) Integrated PL intensity (left axis) as a function of $P$ in a log-log scale of the main lasing peaks (blue solid triangles, 530-534 nm; red solid circles, 544-551 nm), showing the two-threshold behavior for the $\delta = +1$ meV cavity. Also shown are the FWHMs (right axis) of the lasing peaks at ~533 nm (blue open triangles) and ~547 nm (red open circles). (B) Total PL intensity (525-555 nm) (grey squares, left axis) and lasing peak positions (open circles, right axis) as a function of $P$ for the $\delta = +1$ meV cavity. (C) $P$-dependences of integrated PL intensities (left axis) of the two lasing peaks from (blue solid triangles, 534-538 nm; green solid circles, 538-541 nm) and corresponding peak FWHMs (right axis; blue open triangles ~537 nm, green open circles, ~540 nm) for the $\delta = -40$ meV cavity. (D) Total PL intensity (525-555 nm) (grey squares, left axis) and lasing peak positions (open circles, right axis) as a function of $P$ for the $\delta = -40$ meV cavity.
Fig. S18. Integrated total PL intensity as a function of $P$ in a log-log scale for a microcavity measured at room temperature (black curve), showing only one nonlinear threshold. The low temperature at 77 K is also shown for comparison (red curve).

Fig. S19. Demonstrating the expansion of the Hamiltonian about the Diabolical Point, in orders of a small vector $q$. The left panel shows the two rings associated with the energy corresponding to the diabolical point, at an arbitrary crystal angle. The right panel shows the vectors associated with the expansion about this point. Note that the vector to the diabolical point makes an angle $\phi_0/2$ with the x-axis, and then the total wave vector makes the same angle with the x-axis, with a small deviation $\delta$. The right panel also demonstrates how, under the small angle approximation, the tangential component of the small wave vector $q$ may be used to express the small angle.
Fig. S20. Demonstrating the non-Abelian character of the effective magnetic field/spin texture, and the role of crystal orientation on the spin texture about the diabolical point. Panels A-C depict the field (vector field) as a function of the orientation of the anisotropy field, showing the phase of the lowest energy eigenvector (color) as a function of this orientation. We observe the divergence-like field in the case of the anisotropy field aligned along the x axis (A), and a curl-like field in the case aligned against the x axis (C). We observe an intermediate case in panel B, where the anisotropy field is aligned along the y axis.