Supplementary Materials

Polarization superposition of room-temperature polariton condensation

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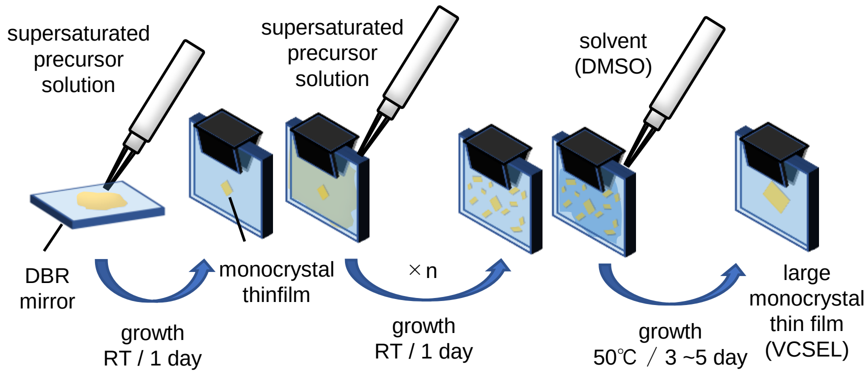

Figure S1 A schematic depiction of microcavity sample using $CsPbBr_3$ crystal. The crystal is grown by a solution-based technique. The distributed Bragg reflectors (DBRs) are fabricated with rf-sputtering deposition of a SiO_2/TiO_2 multilayer.

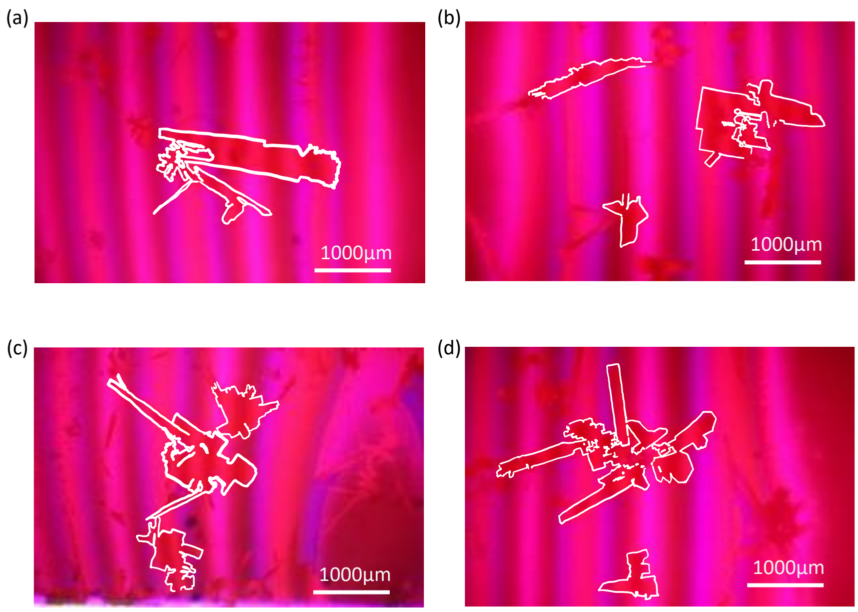

Figure S2 Microscopic images of $CsPbBr_3$ crystals grown in a gap between two DBRs. The areas outlined in white indicate those of flat, high quality crystalline slab.

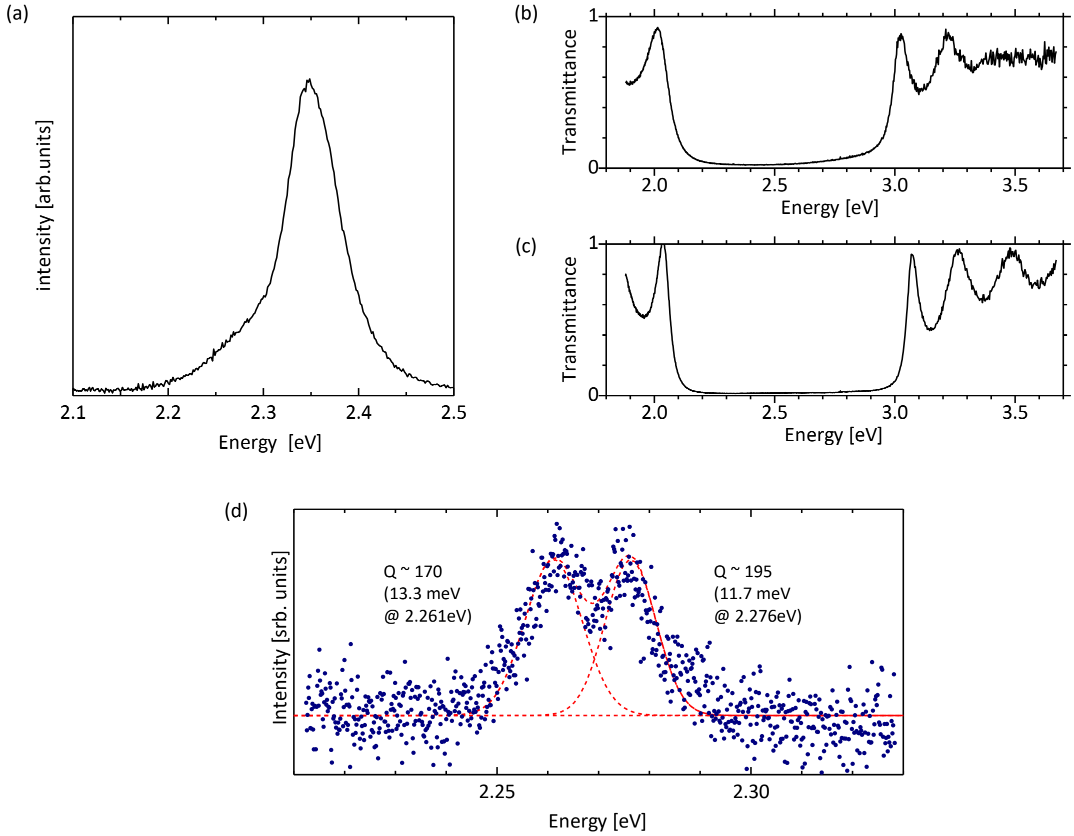

Figure S3 Fundamental evaluations of $CsPbBr_3$ microcavity. (a) Simple photoluminescence (PL) from bare $CsPbBr_3$ crystal without cavity. (b) and (c) Transmittance spectra of DBRs used for bottom and top mirrors, respectively. W can find that the PL signal is well confined in a wavelength range of the reflectivity bands of both mirrors. (d) Unpolarized transmittance spectrum of $CsPbBr_3$ microcavity. The peaks at 2.261 and 2.276 eV corresponds to X- and Y-polarized LPB modes, respectively. The quality factors for these two peaks are estimated as ~170 and ~195.

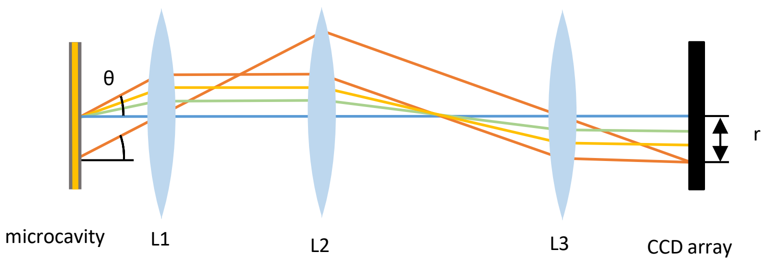

Figure S4 A schematic showing optical setup of Fourier imaging spectroscopy. In the real experiment, we used a Czerny-Turner monochromator equipped with a CCD array (1600 × 400 pixels). The collected light was introduced to an entrance slit of the monochromator and thus the CCD array gathers PL data depending on the wavelength and the angle.

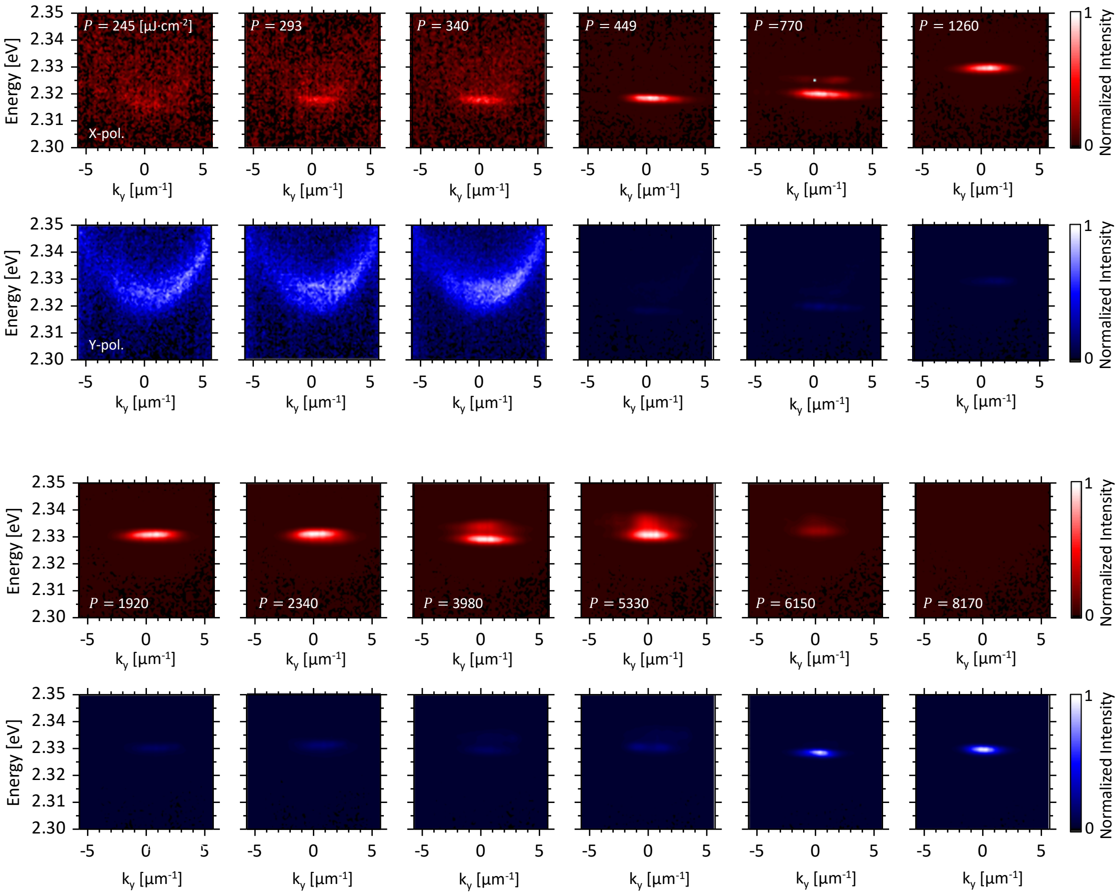

Figure S5 Full data set of angle-resolved PL from $CsPbBr_3$ microcavities. Red and blue colormaps show the data at X- and Y-polarization. The results obtained under pumping fluence of 245 – 8170 μJ/cm^2 are exhibited. At the same pumping fluence, the PL signal intensity is normalized by the stronger of the two polarization directions.

Supplementary text 1

The energy states of exciton polariton can be derived from a eigenvalue problem of a couped oscillator between the energy of original cavity photon (E_{ph}) and the energy of exciton dipole transition (E_{ex}) as

$$\begin{bmatrix} E_{ph} & \hbar\Omega_{Rabi}/2 \\ \hbar\Omega_{Rabi}/2 & E_{ex} \end{bmatrix}\begin{bmatrix} \alpha_{ph} \\ \alpha_{ex} \end{bmatrix} = E_{pol}\begin{bmatrix} \alpha_{ph} \\ \alpha_{ex} \end{bmatrix}. \tag{S1}$$

Here $\lceil \alpha_{ph} \rceil^2$ and $\lceil \alpha_{ex} \rceil^2$ are Hopfield coefficients showing the fractions of photon and exciton, respectively, in the polariton state. E_{pol} and $\hbar\Omega_{Rabi}$ are the energy of polariton state and Rabi splitting energy, respectively. Solving eq. (S1), we get the energies of upper and lower branches of polariton as

$$E_{pol}^{\pm} = \frac{\left(E_{ph} + E_{ex}\right) \pm \sqrt{\left(E_{ph} + E_{ex}\right)^2 + (\hbar\Omega_{Rabi})^2}}{2}, \tag{S2}$$

On the other hand, E_{ph} is shown as a function of wavevector $\boldsymbol{k}$ as

$$E_{ph}(\boldsymbol{k}) = \frac{\hbar n}{c}|\boldsymbol{k}|. \tag{S3}$$

n and c are refractive index and the speed of light, respectively. $\boldsymbol{k}$ is expressed by the in-plane and out-of-plane wavevectors as

$$|\boldsymbol{k}|^2 = |\boldsymbol{k}_\perp|^2 + \left|\boldsymbol{k}_\parallel\right|^2. \tag{S4}$$

Thus, we obtain $E_{ph}(\boldsymbol{k}_\parallel)$ as

$$E_{ph}(\boldsymbol{k}_\parallel) = \frac{\hbar n}{c}\sqrt{\left|\boldsymbol{k}_\parallel\right|^2 + \left(\frac{m\pi}{L}\right)^2}, \tag{S5}$$

where $\boldsymbol{k}_\perp$ is quantized ($m =$ 1, 2, …) in the microcavity with a cavity length L. When $\left|\boldsymbol{k}_\parallel\right|^2 \ll \left(\frac{m\pi}{L}\right)^2$, eq. (S5) is approximated to be

$$E_{ph}(\boldsymbol{k}_\parallel) = E_{ph}(0) + \frac{\hbar^2}{2m^*}\left|\boldsymbol{k}_\parallel\right|^2. \tag{S6}$$

Here $E_{ph}(0)$ is photon energy at $\boldsymbol{k} = 0$ and m^* is noted as the effective mass of polariton mode that is inversely proportional to n.

Figures S6(a) and S6(b) exhibits fitting analyses for the experimental results at the X- and Y-polarization, respectively, which are shown in Fig. 1(d) in the main text. We obtain the $\hbar\Omega_{Rabi}$ of 62 meV and 45 meV for the X-polarized and Y-polarized lower polariton modes, respectively, where E_{ex}= 2.380 eV, n = 2.5, and $E_{ph}(0)$ = 2.349 eV (2.367 eV) for the X- (Y-) polarized mode.

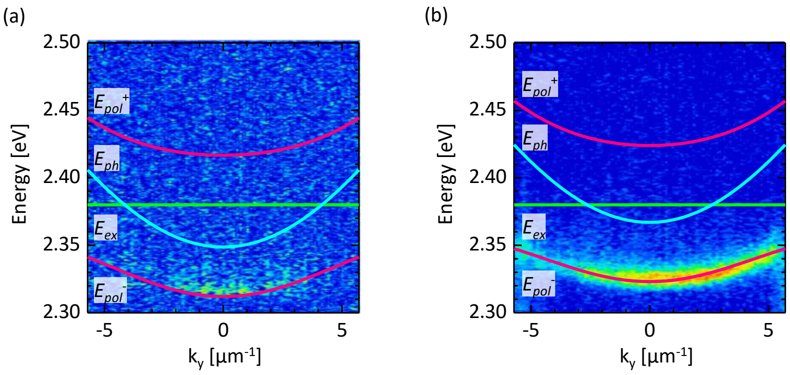

Figure S6 Couped oscillator model analyses of $CsPbBr_3$ microcavity. Lower polariton modes in $CsPbBr_3$ microcavity at (a) X- and (b) Y-polarizations. Angle-resolved photoluminescence results are plotted as colormaps. Blue, green, and purple curves show dispersions of cavity photon, exciton, and polariton modes, respectively.

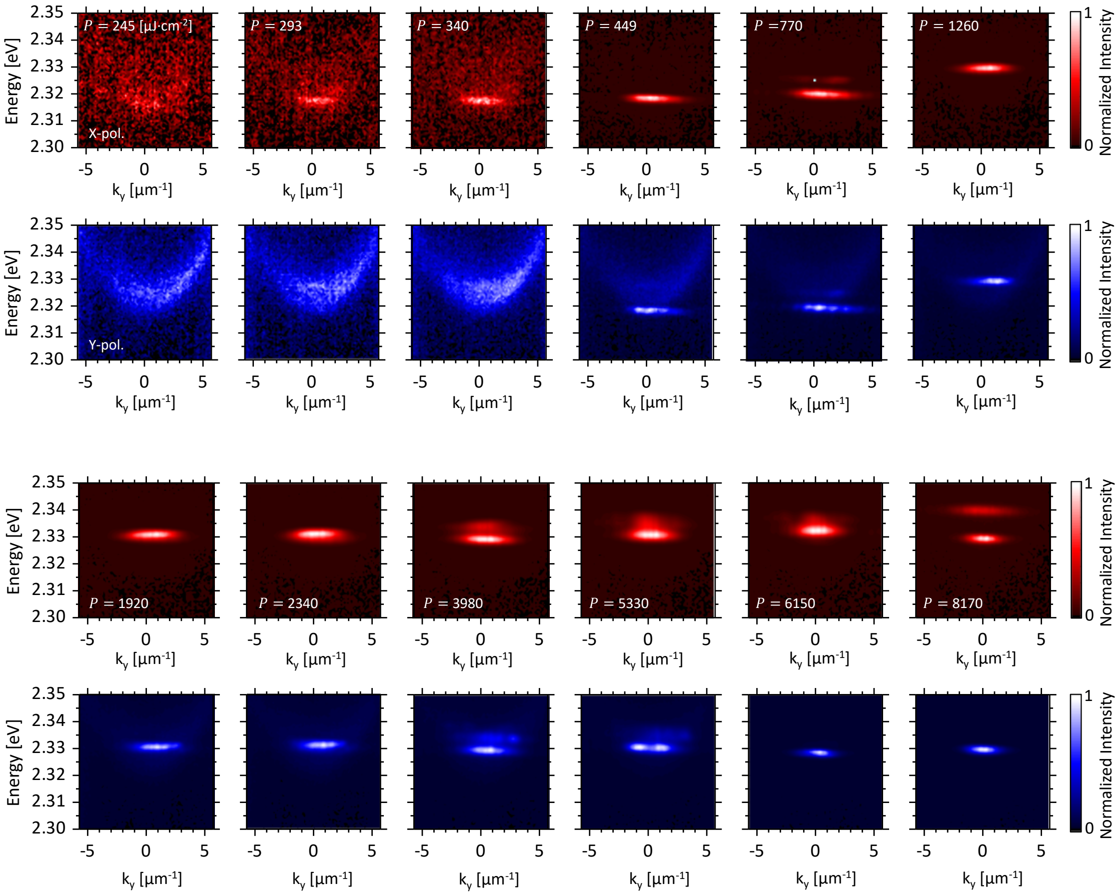

Figure S7 Full data set of angle-resolved PL from $CsPbBr_3$ microcavities. Red and blue colormaps show the data at X- and Y-polarization. The results obtained under pumping fluence of 245 – 8170 μJ/cm^2 are exhibited. The PL signal intensity is normalized within each result.

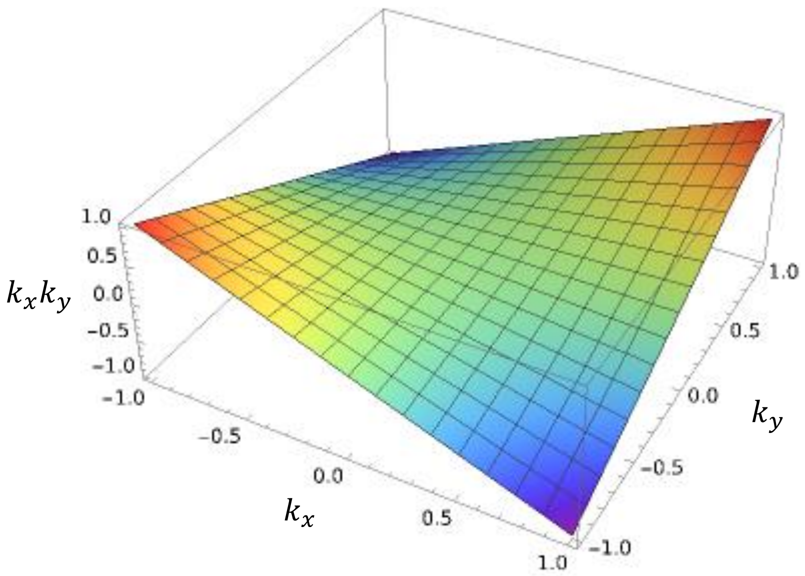

Figure S8 Contour plot of a function of $f(k_x, k_y) = k_x k_y$. This function shows an inversion of sign at the inversion of either one of k_x or k_y, indicating a behavior like an odd function. Along the diagonal directions (i.e. $[k_x,\ k_y]/\sqrt{2}$ or $[k_x, -k_y]/\sqrt{2}$ directions), however, the function reveal a behavior like an even one. As the wavefunction of polariton condensed state is not necessarily isotropically spread in the $k_x k_y$ plane, the off-diagonal terms of equation (4) in the main manuscript are found to be non-zero, showing that the polarization coupling is possible.

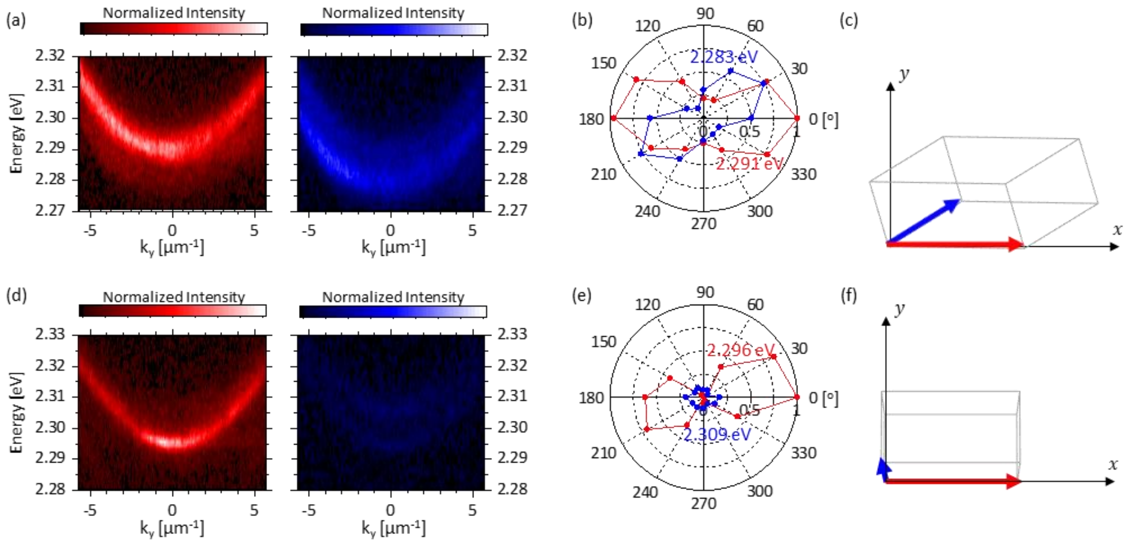

Figure S9 LPB dispersions in $CsPbBr_3$ microcavities with off-axis orientations of perovskite crystal. (a – c) and (d – f) show results of microcavities with cavity thicknesses of ~430 nm and ~440 nm, respectively. (a and d) Colormaps of polarized PL dispersions. (b and e) Polar plots showing PL polarization directions at the Γ point. (c and f) Schematic depictions of off-axis orientations of crystals.

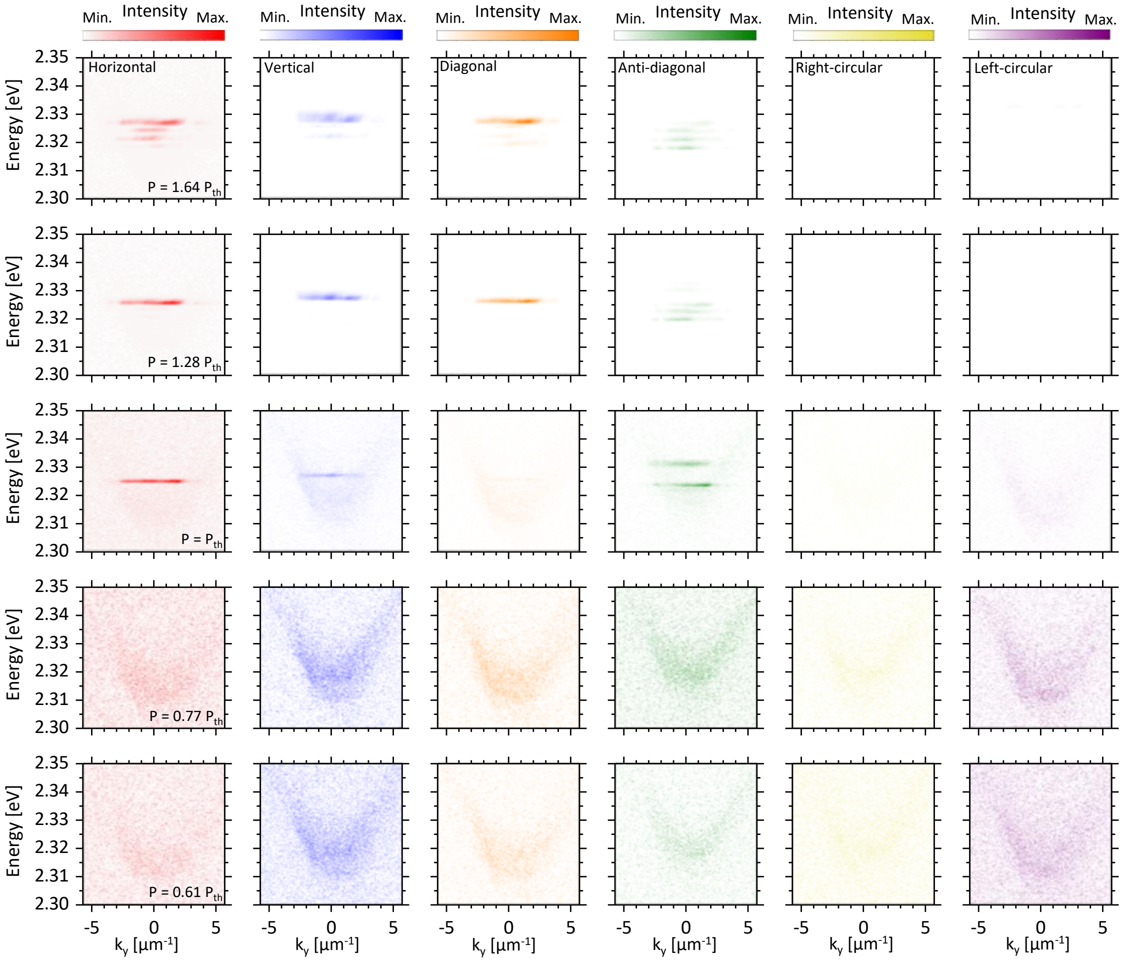

Figure S10 Full data set of angle-resolved PL from $CsPbBr_3$ microcavity around condensation threshold. The results are at pumping fluences are $0.61P_{th}$, $0.77P_{th}$, P_{th}, $1.28P_{th}$, and $1.64P_{th}$, in order from the bottom row. Polarizations are horizontal (red map), vertical (blue map), diagonal (orange map), anti-diagonal (green map), right-circular (yellow map), and left-circular (purple map). The PL intensities are normalized for the data obtained within the same pumping fluence.