

Observation of Rydberg exciton polaritons and their condensate in a perovskite cavity

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The condensation of half-light half-matter exciton polaritons in semiconductor optical cavities is a striking example of macroscopic quantum coherence in a solid-state platform. Quantum coherence is possible only when there are strong interactions between the exciton polaritons provided by their excitonic constituents. Rydberg excitons with high principal value exhibit strong dipole-dipole interactions in cold atoms. However, polaritons with the excitonic constituent that is an excited state, namely Rydberg exciton polaritons (REPs), have not yet been experimentally observed. Here, we observe the formation of REPs in a single crystal CsPbBr₃ perovskite cavity without any external fields. These polaritons exhibit strong nonlinear behavior that leads to a coherent polariton condensate with a prominent blue shift. Furthermore, the REPs in CsPbBr₃ are highly anisotropic and have a large extinction ratio, arising from the perovskite's orthorhombic crystal structure. Our observation not only sheds light on the importance of many-body physics in coherent polariton systems involving higher-order excited states, but also paves the way for exploring these coherent interactions for solid-state quantum optical information processing.

Rydberg exciton | polariton | condensate | perovskite | cavity

Solid-state cavity quantum electrodynamics (CQED) delivers extraordinary control of light-matter interactions in various photonic structures (1). Beyond simply modifying the photonic density of states in the weak coupling regime, CQED also enables the formation of new hybrid light-matter quasiparticles called cavity polaritons (2, 3). In semiconductor microcavities, cavity polaritons are created by strong coupling between excitons and photons when the coupling rate is faster than the dissipation rates of both constituents. These bosonic quasiparticles possess a small effective mass ($\sim 10^{-4}$ electron mass)² from their photonic component and inherit strong interactions from their excitonic component. The combination leads to rich quantum optical phenomena, such as polariton condensation, superfluidity, and quantum vortices, that are similar to those seen in cold atom Bose-Einstein condensation, but at much elevated temperatures (4–8).

Polariton condensation relies on strong nonlinear polariton interactions via their matter constituent, and is characterized by a macroscopic coherent condensate of strongly interacting bosonic particles in a nonequilibrium state (4, 6, 9). The exciton-exciton interactions emerge from underlying Coulomb interactions and strongly depend on the dielectric environment and the exciton radius (4). Currently, the relatively delocalized Wannier-Mott excitons in inorganic semiconductors have formed polariton condensates at densities that are much lower than the exciton's Mott density (10–12), while the tightly bound Frenkel excitons in organic semiconductors have reached polariton condensation at high exciton densities around the exciton saturation density (13, 14). This difference is due to the larger exciton-exciton interaction strength for Wannier-Mott excitons (typical

size, 3 to 10 nm) than for Frenkel excitons (typical size, <1 nm) (15), leading to orders of magnitude stronger interactions in Wannier-Mott excitons. Importantly, there are also excited states of Wannier-Mott excitons that are predicted to provide stronger dipole-dipole interaction strength in the framework of hydrogen-like Rydberg states (16). Polaritons with excitonic constituent of excited states, namely Rydberg exciton polaritons (REPs), have not been naturally observed, due to weak oscillator strength of the excitonic excited states in most optically active semiconductors.

Recently, the emerging lead halide perovskites with Rydberg exciton series (17, 18) have provided a high-quality optoelectronic platform that does not require sophisticated lattice-matched growth (19, 20). The group of lead halide perovskite semiconductors have bulk excitons with exceptional optical properties (21–25), such as a sizeable exciton binding energy, tunable band gaps, high quantum yield, and Rydberg exciton series of strong oscillator strength without applying external magnetic fields (17, 18, 26). They are therefore excellent candidates for investigating exciton-polariton states and polariton condensation, and even for future quantum photonic circuits (7). Encouragingly, polariton lasing based on the ground exciton state has recently been demonstrated in a CsPbCl₃ microcavity

Significance

Rydberg excitons with high principal value exhibit strong dipole-dipole interactions. However, polaritons with an excitonic constituent that is an excited state, namely Rydberg exciton polaritons (REPs), have not yet been experimentally observed. Here, we observe the formation of REPs in a single crystal CsPbBr₃ perovskite cavity without any external fields. These polaritons exhibit strong nonlinear behavior that leads to a coherent polariton condensate with a prominent blue shift. Furthermore, the REPs in CsPbBr₃ are highly anisotropic and have a large extinction ratio, arising from the perovskite's orthorhombic crystal structure.

Author contributions: W.B., X.L., and X.Z. conceived the idea and initiated the project; W.B. and X.L. conducted polariton experiments with assistance from R.T., S.W., Y.X., M.Z., J.K., S.Y., Q.L., Ying Wang, and Yuan Wang under the supervision of X.Z.; F.X. conducted polariton theoretical modeling under the supervision of A.H.M.; F.Z. and L.-W.W. performed calculations of the band structure; W.B., X.L., F.X., A.H.M., and X.Z. analyzed data; and W.B., X.L., and X.Z. wrote the paper with assistance from all authors.

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(27). In this work, we demonstrate the formation of hybrid exciton polaritons in single crystal perovskite CsPbBr_3 , including emerging REPs without external fields. More importantly, we show that Bose–Einstein condensation of polaritons is reached with a prominent blue shift and interesting mode competition that can be explained by our quasiequilibrium mean-field theory. In addition, these polaritons are anisotropic with a large extinction ratio driven by the anisotropy of the potential landscape in the perovskite's orthorhombic phase (28). This precise polarization control is a necessary prerequisite in quantum optical information processing. This work represents a major step in solid-state quantum photonics systems, not only offering a unique platform for new quantum coherent many-body physics (29) but also opening a new door for solid-state quantum photonic applications in communication and computing (30).

The metal halide perovskite CsPbBr_3 is selected as the exciton host. Compared to hybrid organic–inorganic halide perovskites, all-inorganic CsPbBr_3 exhibits superior chemical stability and emission efficiency (31). Fig. 1*D* shows typical absorption spectra of a CsPbBr_3 crystal on mica at 100 K in its thermodynamically stable orthorhombic phase (see additional characterization in *SI Appendix, Materials and Methods* and *Figs. S1–S3*) (28) with clear absorption peaks (E_1 and E_2), consistent with the recent studies (18, 20). According to density functional theory (DFT) in the orthorhombic phase (Fig. 1*E*), there is only one exciton formed in the band edges of CsPbBr_3 , where all other electronic are separated by more than 1 eV. The E_1 and E_2 peaks as the ground and first excited states (Rydberg series $n=1, 2$) of the only

exciton series are identified by the observed energy separation between the 2 absorption peaks, their temperature dependences (*SI Appendix, Fig. S2*), and the reported exciton binding energy (26). The orthorhombic phase also results in the mutually orthogonal birefringence along the a and b crystalline axes (Fig. 1*C* and *E*).

To investigate the strong light–matter interactions in these excitonic states, we embedded the CsPbBr_3 microplate in a Fabry–Perot planar cavity (Fig. 1*A*), where the a and b axes are orthogonal along the in-plane surface. With the full cavity structure, the in-plane wavevector k of the cavity mode is also along this surface (Fig. 1*A, Inset*). As shown in Fig. 1*B*, the microplate transferred onto a distributed Bragg reflector (DBR) substrate has a uniform thickness of 416 nm (*SI Appendix, Fig. S1*) and a typical square shape. Our cavity provides a quality factor in excess of a thousand, derived from the off-resonance cavity linewidth (*SI Appendix, Fig. S6*) as well as polariton linewidth from Fig. 2. This high cavity quality assists the formation of a REP, owing to the sharp interface between the perovskite and the metal mirror (*SI Appendix, Fig. S1*) as well as the reduced metal absorption losses at cryogenic temperature (32).

The coherent coupling of these states and cavity photons is revealed by k -space spectroscopy (*SI Appendix, Fig. S12*, for more details), when the sample is first cooled down to 90 K. The k -space characterization is carried out with selective linear polarization for both photoluminescence (PL) and reflectivity measurements (Fig. 2). The PL measurements are taken using a nonresonant pump laser of 460 nm diagonally polarized between

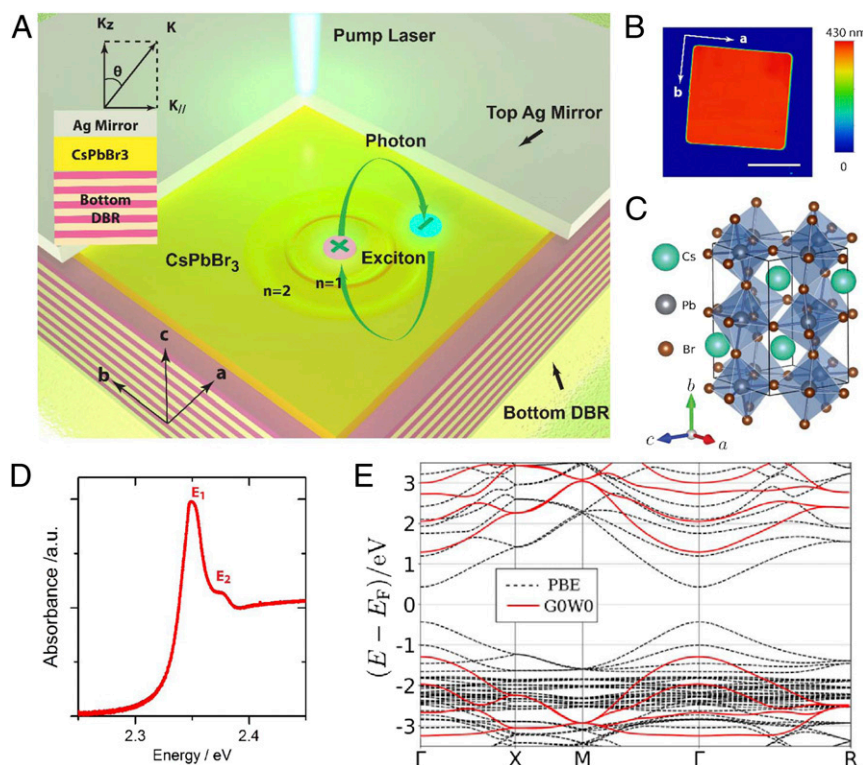


Fig. 1. Schematics of CsPbBr_3 microcavity devices and materials characterization. (A) The CsPbBr_3 microcavity is composed of a 16-pair $\text{SiO}_2/\text{Ta}_2\text{O}_5$ bottom distributed Bragg reflector (DBR), CVD-grown CsPbBr_3 microplates with a thickness of 416 nm, and a 55-nm-thick Ag top mirror. The crystal axes are also indicated. (B) Atomic force microscopy image of the uniform CsPbBr_3 square-shaped single-crystal perovskite used in combination with the bottom DBR mirror in the experiments summarized in Fig. 2. The crystal axes are also labeled. (Scale bar: 10 μm .) (C) The DFT calculated stable crystal structure of orthorhombic CsPbBr_3 , with labeled a , b , and c crystalline axes. This structure results in almost identical refractive indices along the a and c axes, and a distinctly different refractive index along the b axis. (D) The polarization nonselective absorption spectrum of single-crystal CsPbBr_3 film on mica at 100 K. A prominent ground-state E_1 exciton absorption peak is clearly shown along with the excited $n=2$ Rydberg exciton E_2 state. (E) Calculated PBE and G0W0 band structures for orthorhombic CsPbBr_3 . With the inclusion of spin–orbit coupling, the PBE calculated band gap is corrected to 2.5 eV by G0W0, agreeing well with the experiments. Importantly, unlike GaAs, CsPbBr_3 has no degenerate or nearby band states at conduction or valance band edges (Γ point).

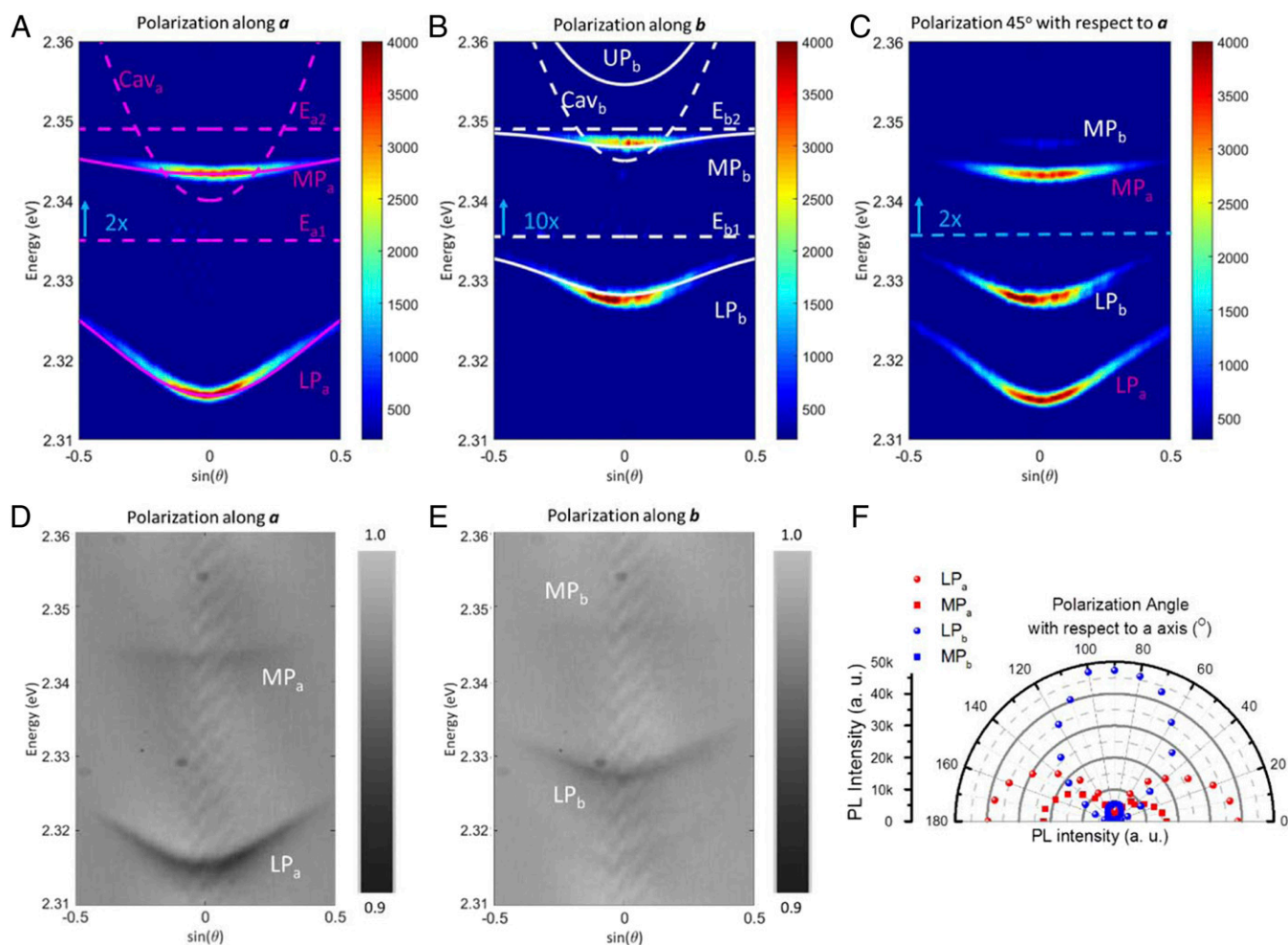


Fig. 2. The k -space angle-resolved PL and white light reflectivity at 90 K. The nonresonantly pumped (460-nm laser) PL map obtained by k -space spectroscopy with detection photon polarization (A) along crystal axis a , (B) along crystal axis b , and (C) 45° in between a and b axes (Fig. 1 A and B). The intensity of the middle branch polariton PL is magnified by 2x, 10x, and 2x in A–C, respectively, due to its weak emission. The horizontal axis represents the sine function of the emission light slant angles θ relative to the z axis (Fig. 1 A, *Inset*), and the vertical axis is the photon energy. Middle branch polariton MP_a and MP_b (better seen in C) are unambiguously formed due to the $n = 2$ exciton state. The polariton dispersion is fit using a coupled oscillator model. The exciton energy and photonic cavity mode (Cav_a and Cav_b) before strong coupling (dashed line) and the fitted polariton dispersion (solid line) are overlaid with the PL map. These fine excitonic states and their polariton structures can only be observed at low temperatures (<150 K). At higher temperatures, the cavity samples transit from only one lower polariton branch to a broad PL peak (similar to bare exciton emission). The corresponding polarization selective white light reflectivity maps of the same sample (D) along crystal axis a and (E) along crystal axis b . The dispersion of k -space reflectivity maps matches the PL dispersion fit very well. (F) The polarization dependence of polariton emission at normal angle ($\theta = 0^\circ$). The extinction ratio of these 2 orthogonal lower branch emission modes is more than 50.

a and b axes (at a 45°, as shown in Fig. 1B), while the reflectivity measurements are carried out using a nonpolarized tungsten halogen white light source. When the detection polarization is set along the a axis, 2 dispersive modes are observed from both PL (Fig. 2A) and reflectivity (Fig. 2D), identified as newly formed polariton states. Although k -space reflectivity has lower color contrast (low reflectivity dips of less than 10%) than PL, they have clear one-to-one correspondence for these polariton states. Both of these polariton modes are distinctively flattened at larger emission angles, indicating that the 2 excitonic states are strongly coupled with the cavity modes. By applying a coupled oscillator model with the 2 excitonic states and a cavity mode at slightly positive detuning ($\Delta_a = E_{cav} - E_{a1} = 5$ meV), the lower and middle branches (labeled as LP_a and MP_a , and overlaid with PL for better visibility) are consistent with the PL and reflectivity dispersion (Fig. 2 A and D; see the model fitting analysis in *SI Appendix*, Supplementary Note S1, additional supporting data in *SI Appendix*, Figs. S4 and S6, and the Hopfield coefficients in *SI Appendix*, Fig. S10). The upper branch (UP_a

and UP_b) dominated by the photon mode component is not visible due to its weak oscillator strength and widely detuned cavity resonance, further aggravated in many cases by fast polariton relaxation (27). These 2 excitonic states corresponding to the $n = 1, 2$ states are confirmed to be in the strong coupling regime based on the temperature-dependent measurements (*SI Appendix*, Fig. S5). The 3-branch polariton dispersion holds up to 150 K, below which the $n = 1, 2$ states can be resolved with large binding energy and oscillator strengths. The polariton dispersion becomes 2-branch above 150 K, beyond which only the ground $n = 1$ state can be resolved. Based on the measurements of a variety of samples, the red shift of the exciton energy levels relative to Fig. 1 is due to sample-to-sample variations, PL Stokes shifts, and temperature-dependent band edge shift (see more details in *SI Appendix*, Figs. S2 and S3 and *Materials and Methods*), consistent with previous reports (13). Thus, this observation implies coherent strong coupling between light and an exciton excited state without an external field for the formation of REP. The extracted energy splittings (37.4 meV between LP_a

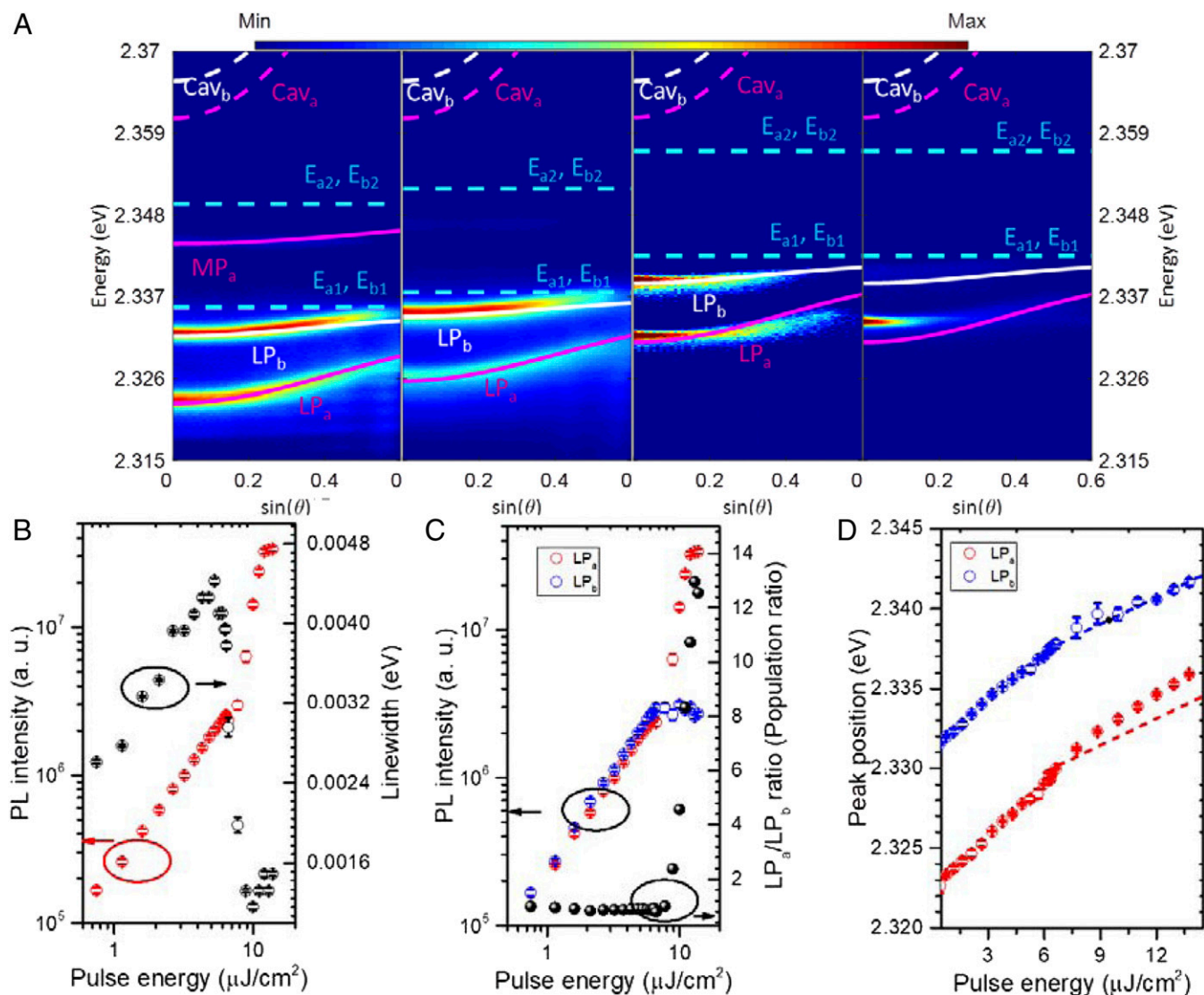


Fig. 3. Anomalous exciton-polariton condensate behavior at 55 K. (A) The k -space power-dependent angle resolved PL map taken at 0.05 P_{th} , 0.4 P_{th} , P_{th} , and 1.4 P_{th} (from Left to Right). The excitation is 460-nm light polarized along the a - b diagonal. The sample is slightly thinner and more positively detuned than in Fig. 2. The 2 sets of orthogonal Rydberg exciton-polariton modes are unambiguously identified and the polariton dispersions are fit using the same coupled oscillator model as in Fig. 2. The uncoupled exciton energy and photonic cavity mode dispersion (dashed line) and polariton dispersion fit (solid line) are overlaid with the PL map. The magenta color represents polarization mode along a axis, while the white color represents the orthogonal polarization mode along b axis (Fig. 1 A and C). The 1.4 P_{th} panel shows the same fit as at P_{th} to emphasize the blue shift above the threshold. The small deviation in the high angle ($\sin\theta$) fitting of the polariton branch LP_a and LP_b at P_{th} and 1.4 P_{th} is due to renormalization of the cavity mode at threshold. The polariton condensate experiences an anomalous condensation process in which the LP_b shows a faster increase than the lower-energy LP_a state between the second and third panels. This is due to a stronger exciton interaction along the b axis. As the pump density gets close to the condensation density, the LP_a finally experiences a superlinear increase with stimulated scattering to the lowest LP_a state, while LP_b shows no further increase. (B) Log-log plot of integrated PL intensity of LP_a mode at $\theta = 0^\circ$ and full width at half-maximum (FWHM) of LP_a mode at $\theta = 0^\circ$ versus pump power. Nonlinearity and linewidth narrowing of the polariton mode is observed as the excitation intensity exceeds the condensation threshold. Fitting error bars from the data processing are shown in B–D. (C) Log-log plot of both LP_a mode (red dot) and LP_b mode (blue dot) at $\theta = 0^\circ$. PL intensity and the ratio of the 2 modes versus pump power. (D) PL peak position of both LP_a mode (red dot) and LP_b mode (blue dot) at $\theta = 0^\circ$ versus pump power. A strong blue shift of polariton modes below the threshold is observed due to the strong exciton interactions and potential system disorder (39). After the threshold of condensation, a prominent blue shift in both LP_a and LP_b mode results from the polariton-polariton interaction and the polariton-reservoir interaction. The theory predicted blue shift contributed from 1s-exciton interaction is plotted in red and blue dot-dot-dash line for guidance. The experimental observed value is larger than the estimation from pure 1s-exciton interaction.

estimate of interaction strength are beyond the scope of the current work. This experimental observation of REP with enhanced interactions promises future explorations of Rydberg interactions in solid-state systems.

In summary, we have surprisingly discovered REPs in a single crystal perovskite cavity, which enables coherent control of these fine quantum states. The intrinsic strong exciton interaction and

optical birefringence in perovskite leads to the observation of polariton-condensation dynamics, which promises a robust macroscopically coherent state for quantum applications. This discovery presents a unique platform to study quantum coherent many-body physics, and potentially enables unprecedented manipulation of these Rydberg states by means such as chemical composition engineering, structural phase control, and external gauge fields.

Controlling the REP and its condensates not only adds flavors on studying polariton lasing, superfluidity, and vortices, but also holds great potential for important applications, such as communication, and quantum simulation.

Materials and Methods

Single-crystal CsPbBr₃ microplates are grown on 150- μ m-thick high-quality muscovite mica substrates via chemical vapor deposition (CVD). The growth surfaces are placed face-down on the top of a quartz crucible that contained a fine power mixture with 20 mg of CsBr (Sigma-Aldrich; 99.999% purity) and 30 mg of PbBr₂ (Sigma-Aldrich; 99.999% purity). Tube pressure is maintained at ambient pressure with an Ar flow rate of 30 sccm. The system is first heated to 400 °C within 24 min, from 400 to 500 °C within 4 min and 30 sccm Ar, held at 500 °C for 20 min, and stopped afterward. When the temperature reaches $\sim$ 200 °C, the furnace is completely opened to achieve rapid cooling. The bottom 16 pairs of SiO₂ and Ta₂O₅ DBR are deposited on top via ion beam sputtering (Veeco IBS) to achieve ultrahigh flatness and >99.95% reflectivity. The as-grown CsPbBr₃ can be directly transferred from the mica substrate to the bottom DBR using a polydimethylsiloxane stamp, which is cured at room temperature and a mixture ratio of 7:1. Before e-beam evaporation of a top Ag mirror, we thermally evaporated 2-nm Al (3 samples, one in Fig. 2 and one in Fig. 3) or 10-nm Sb₂O₃ (one in *SI Appendix*, Fig. S6, and which can preserve PL quantum better) as a seeding layer followed by 5-nm Al₂O₃ atomic layer deposition (Oxford Instruments). Last, the sample is coated with 55-nm 99.99% purity silver using e-beam

evaporation (CHA solution) with a pressure of $<5 \times 10^{-7}$ torr. The evaporation rate is set to be $\sim$ 1 nm/s to avoid oxidation during evaporation. All of the optical *k*-space measurements are carried out in a home-built confocal setup attached with a spectrometer and an electron-multiplying charge-coupled device camera (Andor spectrometer).

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