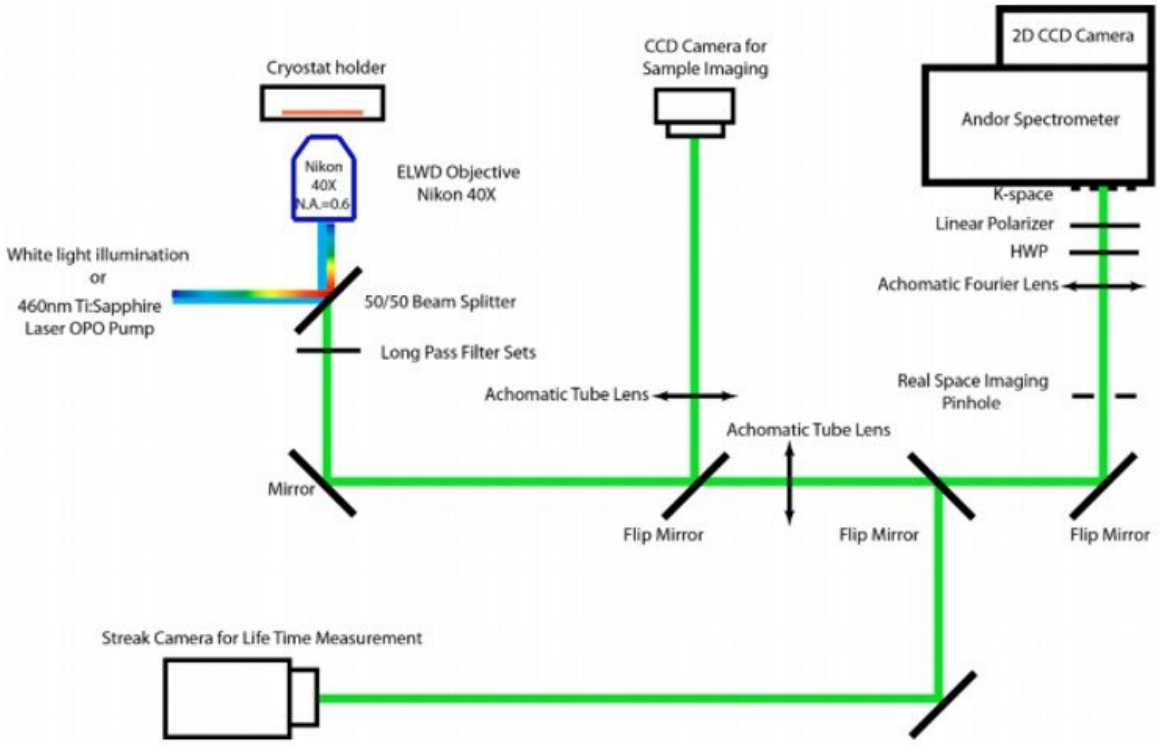
Observations of Rydberg exciton polaritons and their condensate in a perovskite cavity

September 27 2019, by Thamarasee Jeewandara



Schematic of the home-built setup for laser characterization, lifetime measurement, and polarization-dependent k-space photoluminescence (PL) spectroscopy. Credit: Proceedings of the National Academy of Sciences (PNAS), doi: https://doi.org/10.1073/pnas.1909948116

In quantum physics, Rydberg excitons with high principal value can

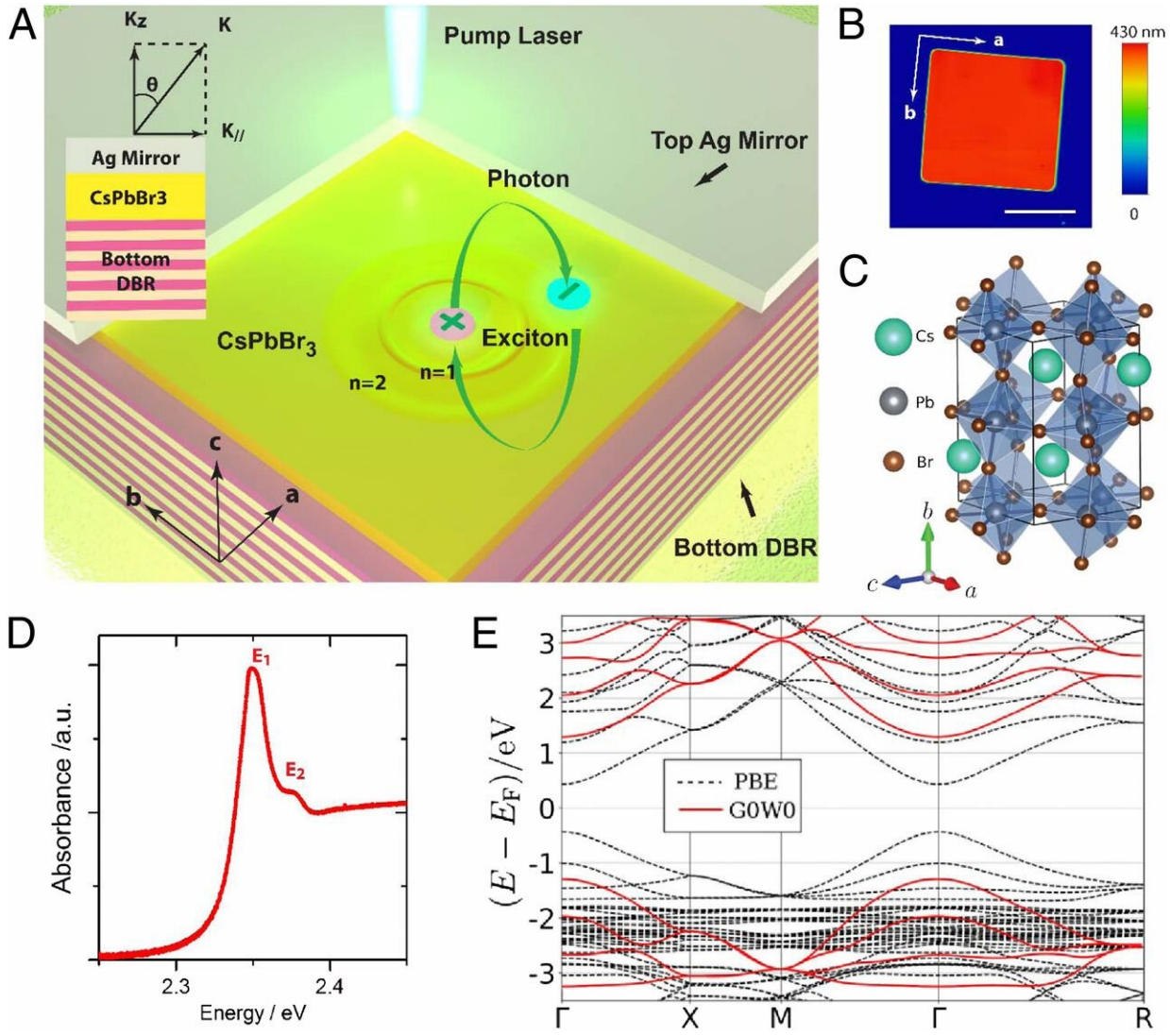
exhibit strong dipole-dipole interactions. However, polaritons (quasiparticles) with an excitonic constituent in an excited state, known as Rydberg exciton polaritons (REPs) remain to be experimentally observed. In a recent study now published on the *Proceedings of the National Academy of Sciences of the United States of America (PNAS)*. Wei Bao and an interdisciplinary research team in the departments of physics, electronics and the National Science Foundation (NSF) Nanoscale Science and Engineering Center in the U.S., observed the formation of REPs in a single crystal $CsPbBr_3$ (cesium lead bromide) perovskite cavity; without any external fields. The researchers noted the polaritons to exhibit strong nonlinear behavior, which lead to a coherent polariton condensate with a prominent blue shift. The REPs in $CsPbBr_3$ cavity were highly anisotropic (showing different properties in different directions) with a large extinction ratio due to the orthorhombic crystal structure of the perovskite.

Quantum coherence is only possible in the presence of strong interactions between exciton polaritons due to their excitonic constituents. The observations by Bao et al. shed light on the importance of many-body physics in coherent polariton systems involving higher-order excited states and paves the way to explore additional coherent interactions. Further investigations will benefit solid-state quantum information processing technologies.

Solid-state cavity quantum electrodynamics (CQED) can deliver extraordinary control of light matter interactions within a variety of photonic structures. Aside from simply modifying the photonic density of states in the weak coupling regime, CQED can also facilitate the formation of new, hybrid light-matter quasiparticles known as cavity polaritons. Cavity polaritons are created in semiconductor microcavities (MC) due to strong coupling between excitons and photons, where the coupling rate can be faster than the dissipation rates of the constituents. The bosonic quasiparticles possessed a small effective mass from their

photonic component to inherit strong interactions from their excitonic component. This combination allowed rich quantum optical phenomena such as polariton condensation, superfluidity and quantum vortices—similar to those observed in cold atom Bose-Einstein condensates (BEC), although at higher temperatures.

Emerging lead halide perovskites with Rydberg exciton series are excellent candidates to investigate exciton-polariton states and polariton condensation for future quantum photonic circuits. Physicists had recently promisingly demonstrated polariton lasing based on the ground exciton state in a $CsPbCl_3$ microcavity. In the present work, Bao et al. showed the formation of hybrid exciton polaritons in a single crystal perovskite $CsPbCl_3$, including emerging REPs without external fields. Importantly, they reached the Bose-Einstein condensation (an exotic quantum phenomenon observed in dilute atomic gases) of polaritons with a prominent blue shift. The polaritons were anisotropic and the observed, precise polarization control was a necessary prerequisite in quantum optical information processing. The work builds a major step forward in solid-state quantum photonic systems and offers a unique platform for new quantum coherent many-body pulses. In parallel, the research also opens a new door to solid-state quantum photonic applications in communication and computing of the quantum internet.

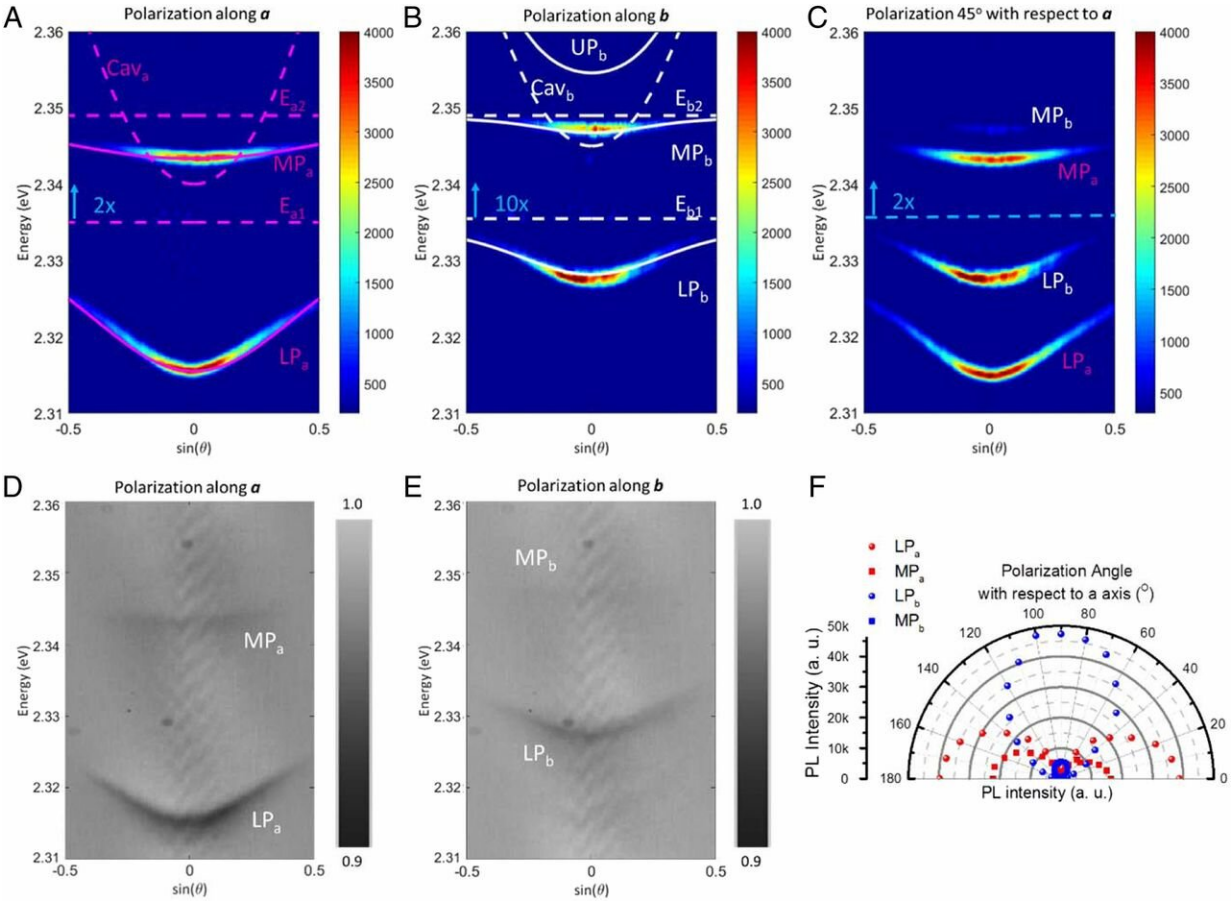


Schematics of CsPbBr3 microcavity devices and materials characterization. (A) The CsPbBr3 microcavity is composed of a 16-pair SiO2/Ta2O5 bottom distributed Bragg reflector (DBR), CVD-grown CsPbBr3 microplates with a thickness of 416 nm, and a 55-nm-thick Ag top mirror. The crystal axes are also indicated. (B) Atomic force microscopy image of the uniform CsPbBr3 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 CsPbBr3, 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 CsPbBr3 film on mica at 100 K. A prominent ground-state E1 exciton absorption peak is clearly shown along with the excited n = 2 Rydberg exciton E2 state. (E) Calculated PBE and G0W0 band structures for orthorhombic CsPbBr3. 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, CsPbBr3 has no degenerate or nearby band states at conduction or valance band edges (Γ point). Credit: PNAS, doi: 10.1073/pnas.1909948116

The research team selected the metal halide perovskite ($CsPbCl_3$) as the exciton host due to its superior chemical stability and emission efficiency compared to organic-inorganic halide perovskites. To investigate the strong light-matter interactions in these excitonic states they embedded the $CsPbCl_3$ microplate in a Fabry-Perot planar cavity (fundamental building block of laser interferometers). This high cavity quality assisted the formation of a REP due to the sharp interface between the perovskite and the metal mirror in the setup, alongside reduced metal absorption losses at cryogenic temperatures.

Bao et al. observed the coherent coupling of these states and cavity photons using k-space spectroscopy after cooling the samples to 90 K. They conducted the k-space characterization using selective linear polarization for photoluminescence (PL) and reflectivity measurements. They measured the PL with a non-resonant pump laser of 460 nm and completed the reflectivity measurements using a non-polarized tungsten halogen white light source. The team obtained two dispersive modes from both PL and reflectivity measurements, which they identified as the newly formed polariton states. The observation implied coherent strong coupling between light and an exciton excited state, without an external field to form the expected REP (Rydberg exciton polariton). The polaritons showed extremely strong polarization anisotropy originating from the perovskite refractive indices.



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. The intensity of the middle branch polariton PL is magnified by 2×, 10×, and 2× 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, and the vertical axis is the photon energy. Middle branch polariton MPa and MPb (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 (Cava and Cavb) 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 (