Room-Temperature $CsPbBr_3$ Mixed Polariton States

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Abstract: Light-matter interactions are known to lead to the formation of polariton states through what is called strong coupling, allowing the formation of two hybrid states usually tagged as Upper and Lower Polaritons. Here, we consider a similar interaction between excitons and photons in the realm of strong interactions, with the difference that it enables us to obtain a mixed-polariton state. In this case, the energy of this mixed state is found between the energies of the exciton state and the cavity mode, resulting in an imaginary coupling coefficient related to a specific class of singular points. These mixed states are often considered unobservable, although they are predicted well when the dressed states of a two-level atom are considered. However, intense light confinement can be obtained by using a Bound State in the Continuum, reducing the damping rates, and enabling the observation of mixed states resulting from the correct kind of exceptional point giving place to strong coupling. In this study, using the Transfer Matrix Method, we simulated cavities made of porous silicon coupled with $CsPbBr_3$ perovskite quantum dots to numerically observe the mixed states as well as experimentally, by fabricating appropriate samples. The dispersion relation of the mixed states is fitted using the same equation as that used for strong coupling but considering a complex coupling coefficient, which can be directly related to the appropriate type of exceptional point.

Supplementary Information

TEM pictures of perovskites QDs.

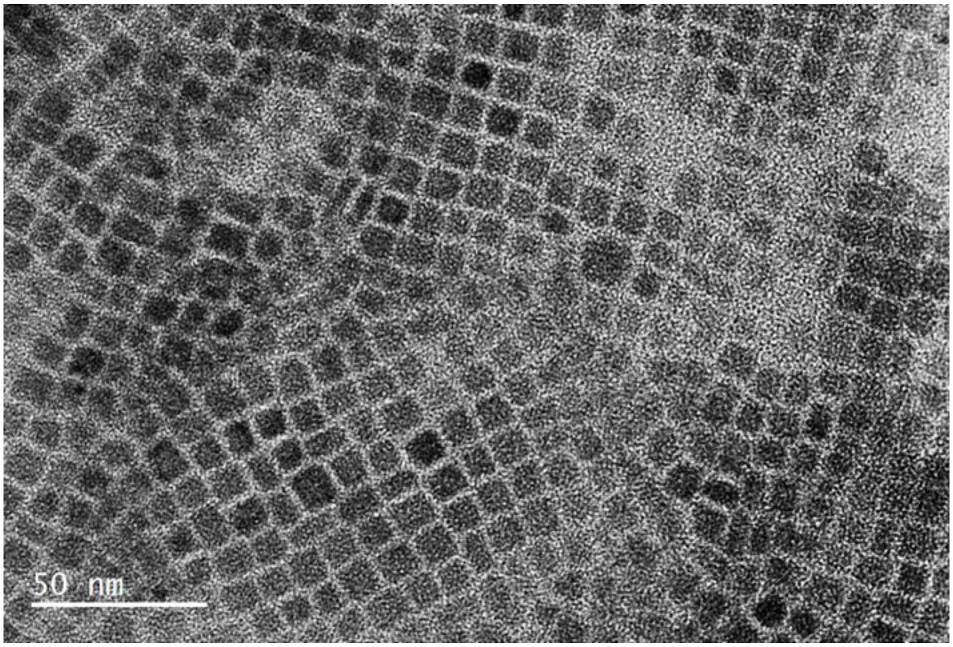


Fig. S1. TEM micrograph of $CsPbBr_3$ perovskite QDs before deposition on the p-Si microcavities. An average size of 8-10 nm could be calculated from the corresponding statistical analysis.

Evolution of the pol-BIC dispersion relation for real and imaginary $\tilde{g}$ values, and positive and negative detuning.

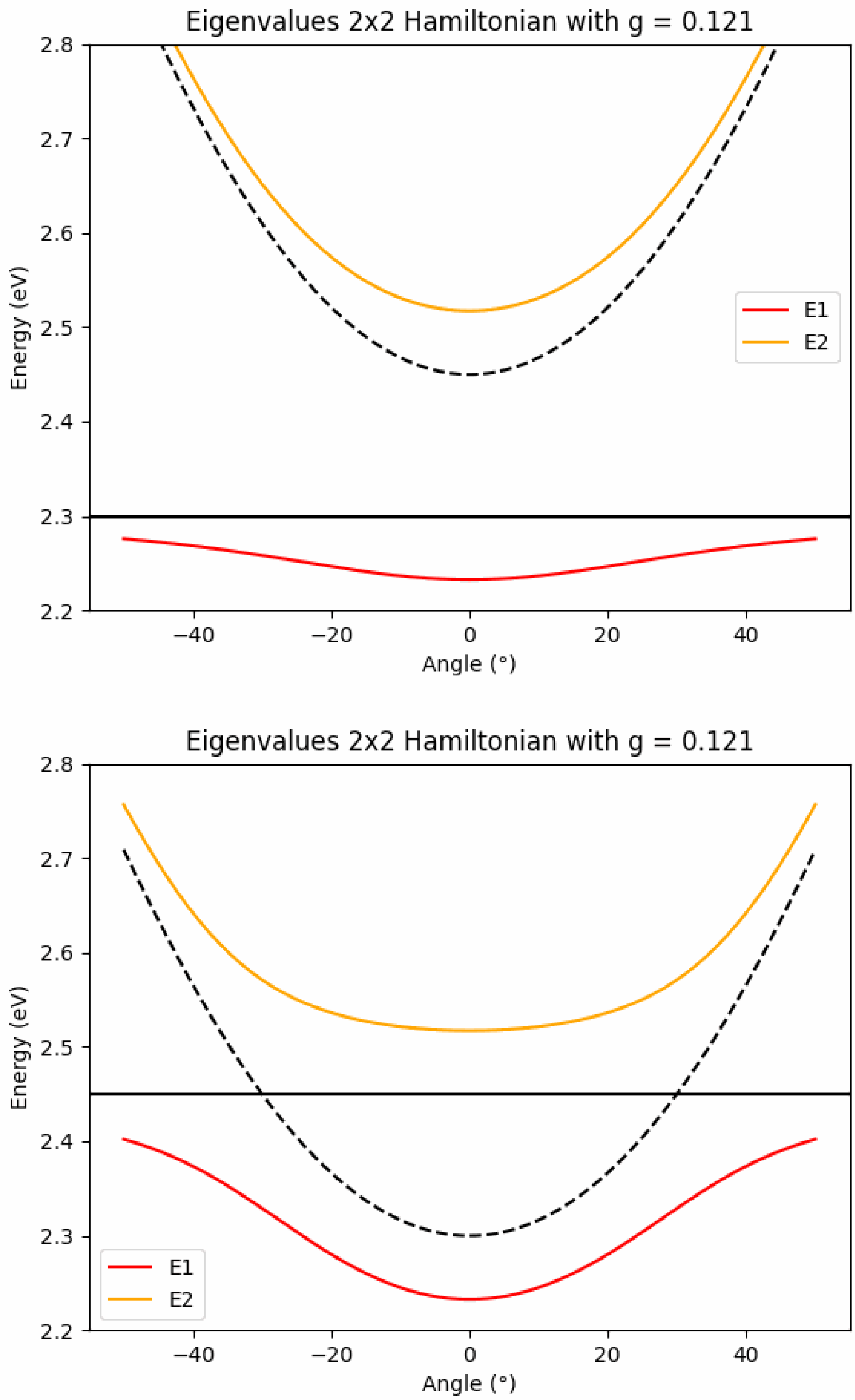


Fig. S2. GIF images showing the evolution of the pol-BIC dispersion relation for real and imaginary $\tilde{g}$. Positive (top) and negative (bottom) cavity-detuning are considered.

Parameter space showing normal polaritons (yellow zones) and mixed polaritons (red zones). These last occur at periodic regions defined by ${}^{\lambda_{DBR}}/_{2}$ integral periods.

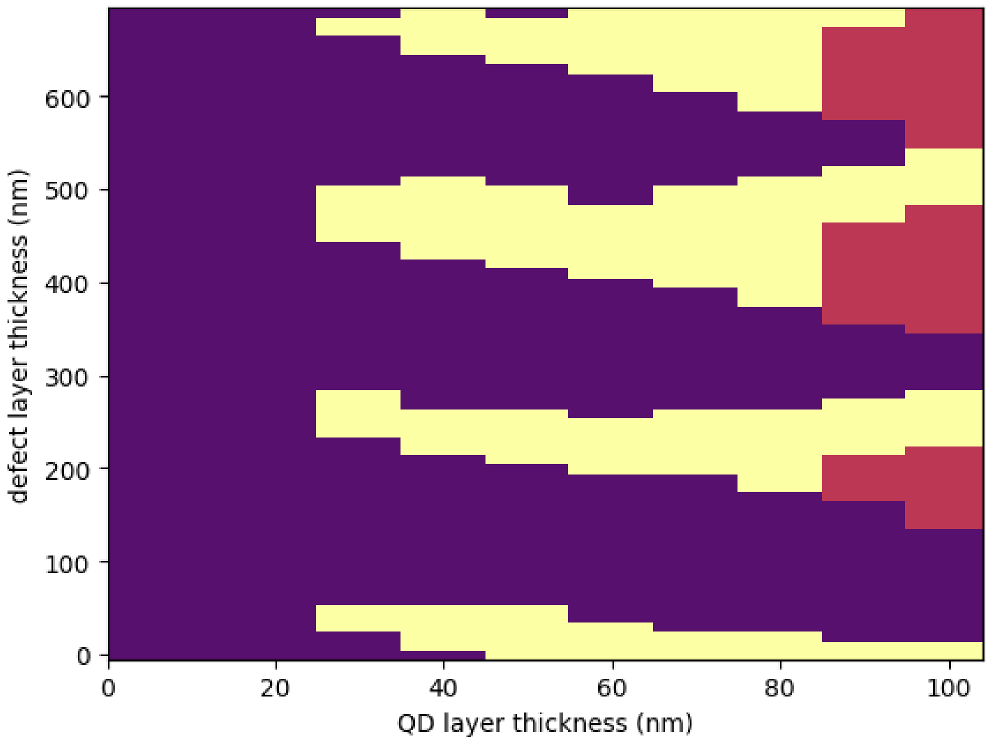
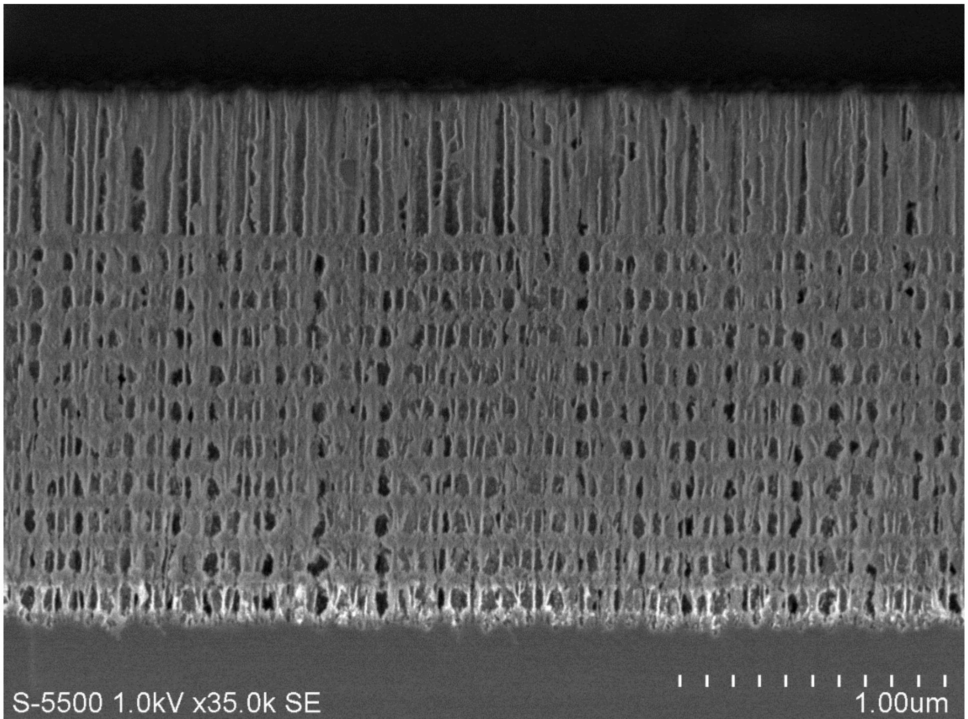
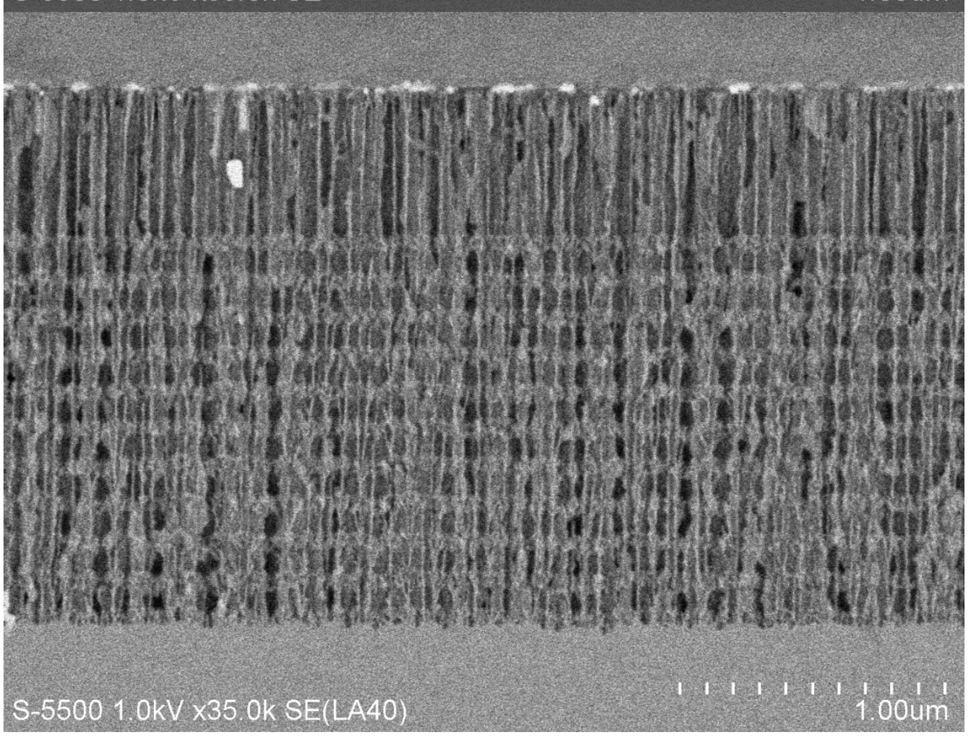


Fig. S3. Cartography of different types of pol-BICs: yellow, normal LP and UP polaritons; red, mixed polaritons. Violet spots correspond to uniform intensities, that is where no coupling is observed.

SEM pictures from the surface and the cross-section of the porous Si system and the perovskite deposit on the surface.


S-5500 1.0kV x35.0k SE
1.00um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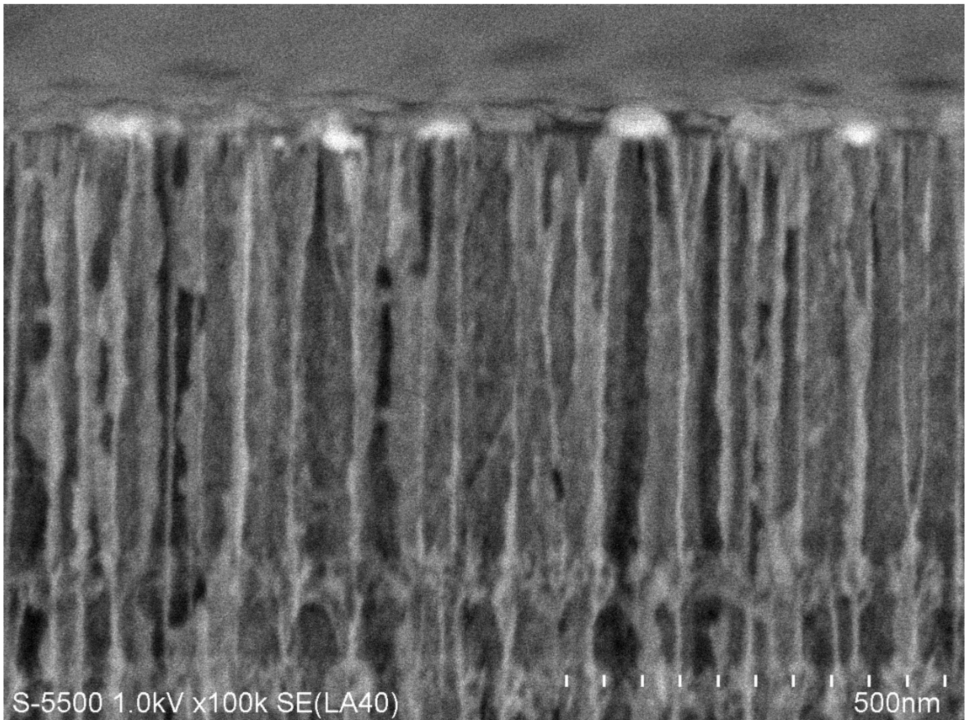
1.00um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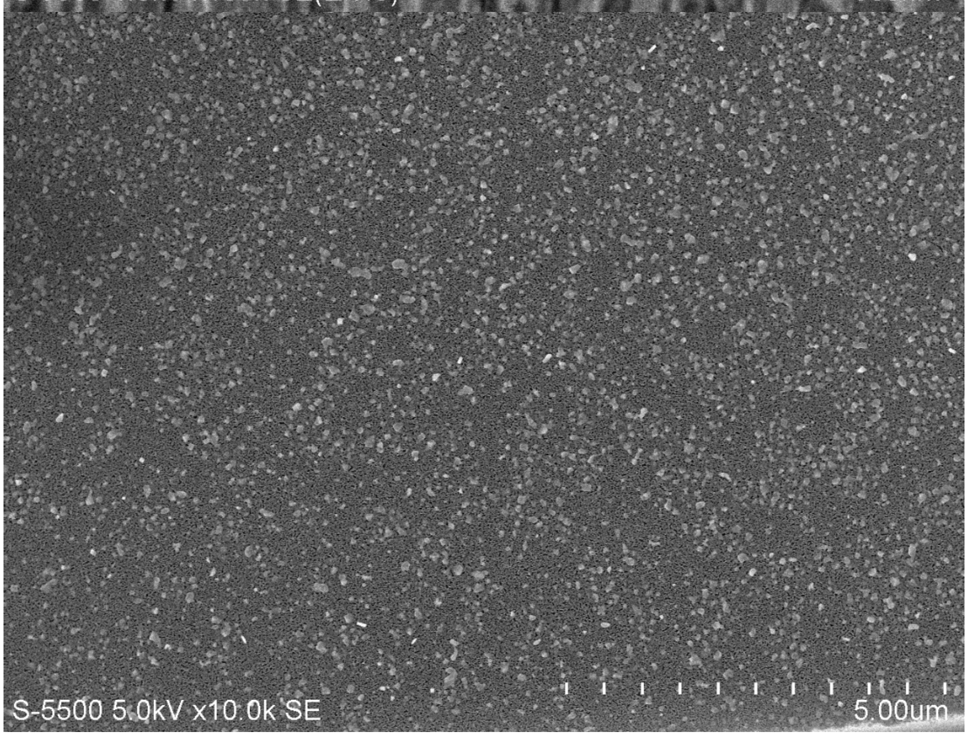
500nm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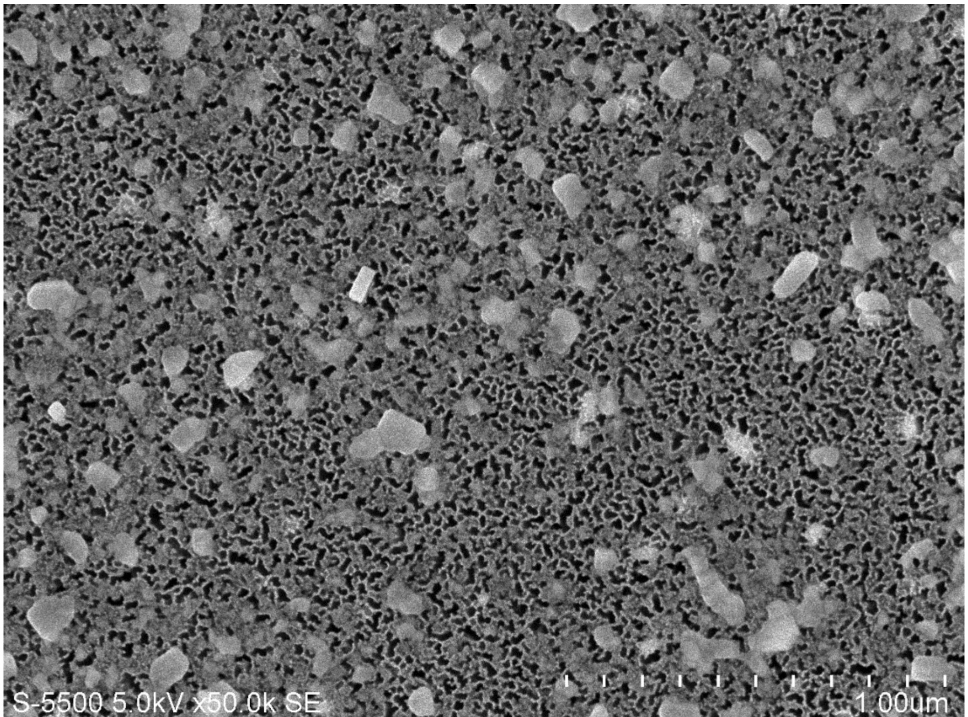
5.00um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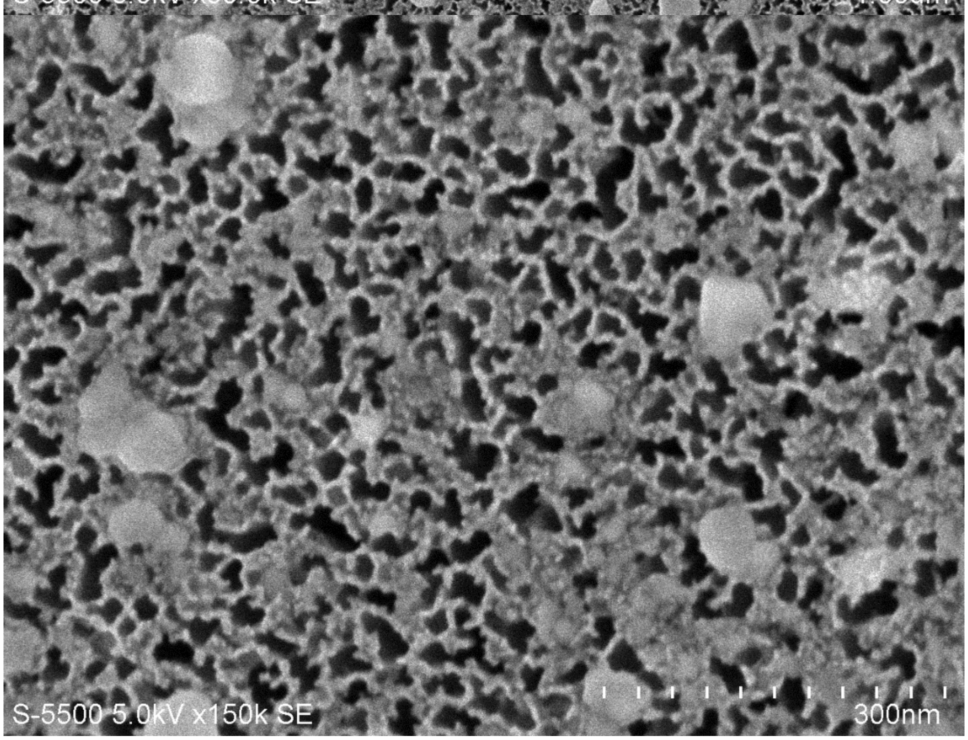
S-5500 5.0kV x150k SE
300nm

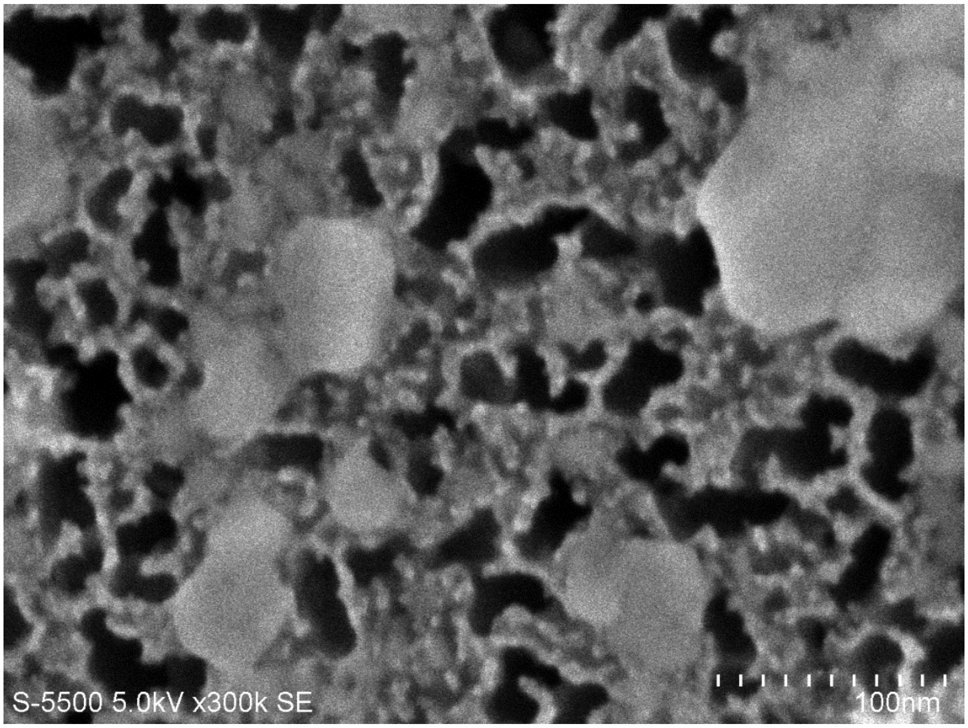

Fig. S4. SEM micrographs from the cross section and surface of the coupled system, showing the perovskite islands formed from the deposited film because of the action of the electron beam.

Evidence of no PL shift for the perovskite deposition on a single p-Si monolayer. That is, there is not strong coupling between the perovskite emitters and the single silicon monolayer.

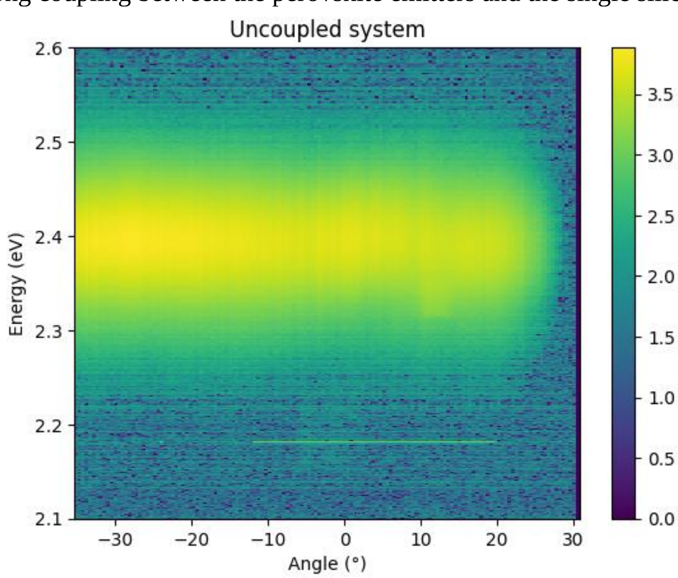

Fig. S5. Angle-resolved PL measurements obtained for $CsPbBr_3$ QDs deposited on a single p-Si monolayer. In the absence of strong coupling between the QD excitons and the cavity photons, the emission peak is angle-independent, and its energy corresponds to that of the QDs in solution.

A set of three pairs of polaritons can be used to fit the simulated and experimental data, with one pair always being a normal polariton and a real coupling corresponding to a Rabi splitting of 28 meV. The other two pairs can be fitted either using normal polaritons, with a real coupling corresponding to a Rabi splitting of 16 meV for both (Fig. S6(a)), or mixed polaritons, with an imaginary coupling corresponding to a negative Rabi splitting of -16 meV (Fig. S6(b)).

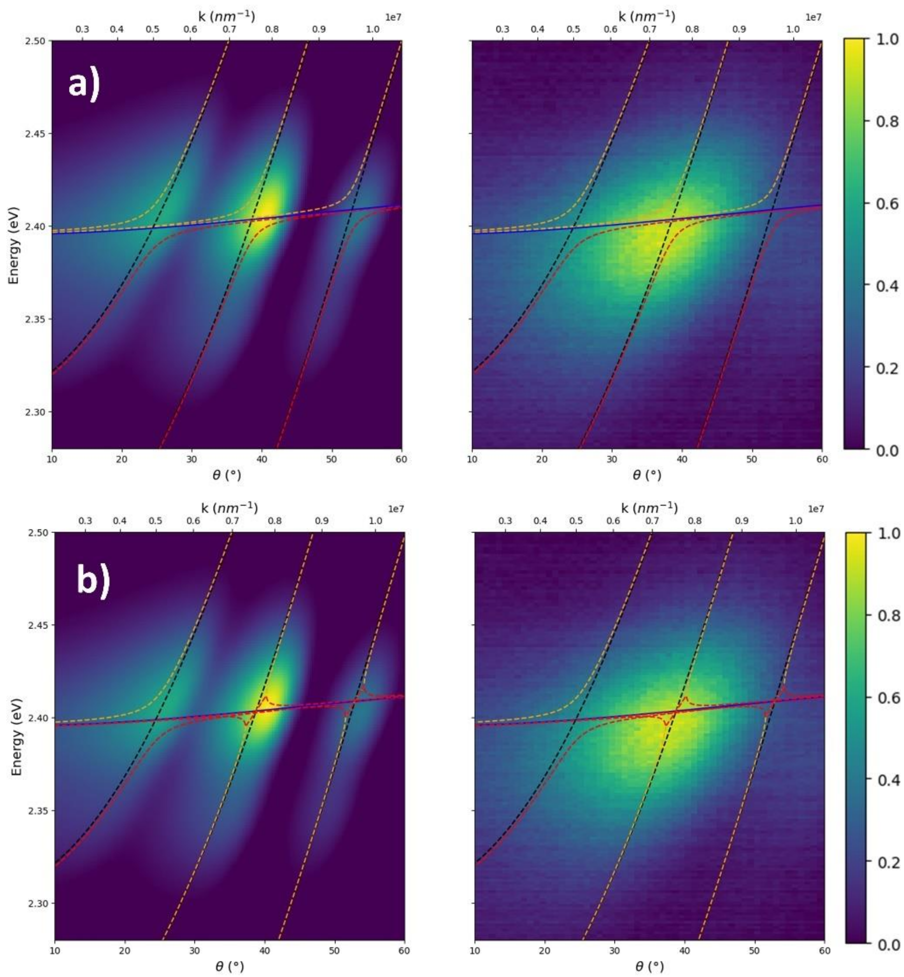

Fig. S6. ARPL best-fit simulation for $d_{QD} = 32$ nm (left) and experimental results for M1 sample (right). Dispersion relationships using (a) normal (real g) and (b) mixed (imaginary g) polaritons. Blue dashed line represents the exciton dispersion, black dashed lines the cavity modes, and orange (red) dashed lines represent the upper (lower) polaritons.