Room temperature coherently coupled exciton-polaritons in two-dimensional organic-inorganic perovskite

Wang, Jun; Su, Rui; Xing, Jun; Bao, Di; Diederichs, Carole; Liu, Sheng; Liew, Timothy Chi Hin;
Chen, Zhanghai; Xiong, Qihua

2018

Wang, J., Su, R., Xing, J., Bao, D., Diederichs, C., Liu, S., . . . Xiong, Q. (2018). Room temperature coherently coupled exciton-polaritons in two-dimensional organic-inorganic perovskite. *ACS Nano*, 12(8), 8382-8389. doi:10.1021/acsnano.8b03737

<https://hdl.handle.net/10356/141657>

<https://doi.org/10.1021/acsnano.8b03737>

This document is the Accepted Manuscript version of a Published Work that appeared in final form in *ACS Nano*, copyright © American Chemical Society after peer review and technical editing by the publisher. To access the final edited and published work see <https://doi.org/10.1021/acsnano.8b03737>

Downloaded on 26 Apr 2023 22:22:11 SGT

Room Temperature Coherently Coupled Exciton Polaritons in Two-Dimensional Organic-Inorganic Perovskite

Jun Wang,^{1,2,⊥} Rui Su^{1,⊥} Jun Xing,¹ Di Bao,¹ Carole Diederichs,^{3,4} Sheng Liu,¹ Timothy C.H. Liew,¹ Zhanghai Chen,^{2,} Qihua Xiong,^{1,3,5,*}*

¹Division of Physics and Applied Physics, School of Physical and Mathematical Sciences, Nanyang Technological University, Singapore 637371, Singapore.

²State Key Laboratory of Surface Physics, Key Laboratory of Micro and Nano Photonic Structures (Ministry of Education), Department of Physics, Collaborative Innovation Center of Advanced Microstructures, Fudan University, Shanghai 200433, P. R. China.

³MajuLab, CNRS-UCA-SU-NUS-NTU International Joint Research Unit, UMI 3654, Singapore 639798, Singapore.

⁴Laboratoire Pierre Aigrain, Département de physique de l'ENS, Ecole normale supérieure, PSL Research University, Université Paris Diderot, Sorbonne Paris Cité, Sorbonne Universités, UPMC Univ. Paris 06, CNRS, Paris 75005, France.

⁵NOVITAS, Nanoelectronics Center of Excellence, School of Electrical and Electronic Engineering, Nanyang Technological University, Singapore 639798, Singapore.

*To whom correspondence should be addressed. E-mail address: Qihua@ntu.edu.sg and zhanghai@fudan.edu.cn.

[⊥]These authors contributed equally to this work.

ABSTRACT

Two-dimensional (2D) organic-inorganic perovskite semiconductors with natural multi-quantum well structures and confined 2D excitons are intriguing for the study of strong exciton-photon coupling, due to their large exciton binding energy and oscillation strength. This strong coupling leads to a formation of the half-light half-matter bosonic quasiparticle called exciton-polariton, consisting of a linear superposition state between photonic and excitonic states. Here, we demonstrate room temperature strong coupling in exfoliated wavelength-tunable 2D organic-inorganic perovskite semiconductors embedded into a planar microcavity, exhibiting large energetic splitting-to-linewidth ratios (>34.2). Angular dependent spectroscopy measurements reveal that hybridized polariton states act as an ultrafast and reversible energy oscillation, involving 2D perovskite exciton, cavity modes (CM) and Bragg modes (BM) of the distributed Bragg reflector (DBR). Meanwhile, sizeable hybrid particles dominantly couple to the measured optical field through the CMs. Our findings advocate a considerable promise of 2D organic-inorganic perovskite to explore fundamental quantum phenomena such as Bose-Einstein condensation, superfluidity and exciton-polariton networks.

KEYWORDS: *two-dimensional perovskite, exciton-polariton, light-matter strong coupling, Fabry-Pérot cavity, Bragg mode*

Exciton-polaritons are half-light half-matter quasiparticles formed by strong coupling of excitons and photons in a semiconductor microcavity.¹⁻³ This coupling is a result of the reversible exchange of energy between the excitonic state and the confined cavity photon mode, when the coupling strength (Rabi splitting) is larger than the average dissipation rates of uncoupled excitons and cavity photons. Exciton-polaritons have a bosonic nature, following Bose-Einstein statistics, with an extremely small effective mass and long coherence length, enabling Bose-Einstein condensation,⁴⁻⁷ superfluidity,⁸ quantum vortices,⁹ the optical spin Hall effect,¹⁰⁻¹² *etc.* However, most implementations of exciton-polaritons have focused on III-V¹³⁻¹⁷ and CdTe^{4, 18, 19} quantum wells (QWs) with Wannier-Mott excitons embedded in lattice-matched, monolithic, crystalline Fabry-Pérot (FP) cavities, which are limited to cryogenic operating temperatures due to the small exciton binding energy and oscillator strength. On the contrary, III-nitride^{20, 21} and ZnO-based semiconductors²² can support polaritons at room temperature, but they are hindered by broad exciton linewidth and sophisticated epitaxial techniques are needed for fabricating high-quality microcavities.

Organic-inorganic perovskite semiconductors have recently become a rejuvenated family of optoelectronic materials owing to their remarkable properties and applications in photovoltaics,²³ high optical gain photonics²⁴⁻²⁷ and optoelectronics.²⁸⁻³¹ Particularly, 2D organic-inorganic perovskite semiconductors with the natural multiple QW crystal structure have exhibited fantastic properties,³²⁻³⁶ *i.e.*, large exciton binding energy (> 0.23 eV at room temperature), huge oscillator strength, high fluorescence yield and low nonradiative recombination loss, due to a small excitonic Bohr radius and strong Coulomb interaction between an electron and a hole in the confined exciton. As a van der Waals material, exfoliated atomically flat 2D perovskites are conveniently transferred to any substrates for fabricating devices including microcavity structures without considering the lattice match. Considerable overlaps of excitons and CMs can promote the maximum light-matter coupling, as a result of multiple molecularly thin active QWs in the strongest amplitude of electric field in the microcavity. Motivated by these exceptional properties, 2D organic-inorganic perovskites are ideal candidates for

realization of the strong light-matter coupling at room temperature and applications in quantum information, communication and computing.

In this work, we have demonstrated that 2D organic-inorganic perovskite excitons are strongly coupled to a single CM or multiple hybrid CMs, based on the system consisting of a FP microcavity and an exfoliated wavelength-tunable 2D perovskite crystal ((PEA)₂PbI₄, (PEA)₂PbBr₄). Here coherently coupled exciton-polaritons have presented: (i) a large Rabi splitting (~ 242 meV) and a large energetic splitting-to-linewidth ratio (SLR) (~ 34.2), *i.e.*, the average number of energetic oscillations between excitons and cavity photons during the polariton lifetime, which is fundamental to maintain the strong coupling and support nonlinear polariton interactions;³⁷ and (ii) a large energetic splitting in polaritons with transverse electric (TE) and transverse magnetic (TM) linear polarizations (> 15 meV, see Supplementary Materials), which can provide an effective magnetic field in k -space to act on the pseudospin of polaritons for demonstrating the optical spin Hall effect. Moreover, angle-resolved reflectivity and photoluminescence (PL) spectra reveal that angular dependent hybrid compositions of polariton dispersions are modulated by tuning the thickness of exfoliated 2D perovskite. Interestingly, in the dispersions of multi-mode polaritons with high quality factor ($Q \sim 2200$), the coherent strong couplings involving 2D perovskite excitons, multi-mode CMs and BMs of DBRs, have resulted in the unambiguous anti-crossings and splitting of energy at resonance, indicating a reversible energetic oscillation among these three states. A significant hybridization for these three states is clearly observed though the mixing coefficients of hybrid Bragg-mode polariton (HBP) states, along with sizeable hybridized particles dominantly coupling to the measured optical field through the CMs. Our findings provide an ideal platform to modulate the coherent light-matter coupling and hybridization, and to explore fundamental quantum phenomena in 2D perovskite at room temperature.

RESULTS AND DISCUSSION

The microcavity structure is schematically shown in Figure 1a, consisting of a single-

crystal 2D organic-inorganic perovskite sandwiched between the bottom and top DBR mirrors. 2D organic-inorganic perovskite bulk crystals were grown by a supersaturation precipitation method,³⁸ then the 2D perovskite sample was transferred from bulk crystal to the bottom DBR by an exfoliating process. Finally, a top DBR was deposited onto the 2D perovskite sample without an additional spacer, because numerous natural molecularly thin QW layers of the 2D perovskite near the antinode of electric field in the microcavity can support maximum coupling. The thin (PEA)₂PbI₄ flake was characterized by absorption and PL spectroscopy at room temperature. Figure 1c shows that the 2D (PEA)₂PbI₄ flake has a single strong excitonic absorption peak at 516 nm (2.4 eV) with narrow full width at half maximum (FWHM) of 16 nm (74 meV). Due to the excitonic absorption peak far from the band edge absorption, 2D perovskites exhibit a large exciton binding energy of about 0.23 eV at room temperature, larger than in GaN and ZnO.²² The PL emission spectrum shows a sharp and narrow peak at about 524 nm (2.366 eV), with a FWHM of 11 nm (49 meV) and a red-shift corresponding to the absorption peak. The schematic energy-band structure of 2D perovskite crystal is displayed in Figure 1a. Excitons with the lowest energy can be well confined in the inorganic layers, due to the lower bandgap inorganic QW layers sandwiched by the higher bandgap organic barrier layers. Because of the high contrast in dielectric constants between the inorganic and organic layers, the Coulomb interaction in the QWs is hardly screened by the barriers. Consequently, 2D organic-inorganic perovskites present large exciton binding energy, high fluorescence yield and huge oscillator strengths at room temperature. When the exfoliated thick 2D perovskite crystal of several micrometers was transferred onto a silicon substrate, its PL emission showed multiple peaks with unequal energy difference, as shown in Figure 1d; a signature of the formation of a FP microcavity by its atomically flat surface and own QW nature.

By angle-resolved reflectivity (ARR) and angle-resolved PL (ARPL) measurements, we investigated the behavior of polaritons in the 2D perovskite crystal with FP microcavity at room temperature. More details of the setup for the *k*-space

characterization are shown in the Supplementary Materials,²⁷ and all data shown here are for the TE polarization unless otherwise noted. Figures 2a and 2b show the ARR spectra for k -space mappings of polaritons with negative (-31.7 meV) and positive (61 meV) detuning ($\Delta = E_{cav} - E_{ex}$) in (PEA)₂PbI₄, respectively. Figures 2d and 2e are the corresponding ARPL spectra. Figures 2c and 2f show the ARR and ARPL spectrum for k -space mapping of the polariton with positive detuning (10.8 meV) in (PEA)₂PbBr₄, respectively. All dispersions obtained in ARPL mappings agree well with corresponding ARR measurements. The 2D perovskite exciton energy (E_{ex}) and the cavity photon dispersion ($E_{cav}(\theta) = E_{ph}/\sqrt{1 - [\sin(\theta)/n_{eff}]^2}$) are shown as the yellow lines and curves, respectively. Figures 2d-2f show that the FWHM of the lower polariton (LP) branch is 0.77 nm (3 meV), 1.6 nm (6 meV) and 1.2 nm (8 meV), the Rabi splitting energy ($\hbar\Omega$) is about 205 meV, 221 meV and 242 meV, and the SLR is 34.2, 18.4 and 15.1 (which is greater than in 2D transition-metal dichalcogenides),³⁷ respectively. These large SLRs indicate sufficient reversible energetic oscillations between excitonic states and confined photon states in the strong coupling regime. In Figures 2d-2f, dispersion curvatures of LP branches are unambiguously flattened at larger angles and display an onset of the anti-crossing behaviors between the FP cavity dispersion and the flat perovskite exciton energy at resonance. These phenomena are the distinct signature of the strong light-matter coupling. In the perovskites inserted in DBRs cavity, the upper polariton (UP) whose Rabi splitting energy is comparable to (even larger than) the exciton binding energy, is hardly observed, due to the band edge absorption, the thermal relaxation, and the high reflectivity index of the top DBR mirror (Supplementary Materials Figure S3). However, in the thick perovskite exfoliated on the SiO₂ substrate, the UP branches can be clearly observed, and their dispersions are flattened at small angles when approaching the exciton dispersion (Supplementary Materials Figure S2), which is a typical signature of exciton-polariton dispersions with UP-LP branches. Note here another parabolic-like dispersion in Figure 2f is attributed to the bare microcavity without any embedded perovskites (Supplementary Materials Figure S4), owing to the size of the 2D (PEA)₂PbBr₄ perovskite sample being smaller

than the collection area of the objective.

To further confirm the strong coupling regime, we have extracted the FWHM of the 2D perovskite excitons from the PL spectrum, which is $\hbar\Gamma_{ex} = 49$ meV. It is proved that the light-matter strong coupling condition is satisfied with $\hbar\Omega > \sqrt{(\hbar\Gamma_{ex})^2 + (\hbar\Gamma_{cav})^2}/2$ in our systems, where $\hbar\Gamma_{cav}$ is the FWHM of the CM. As shown as the white dashed curves in Figure 2, we have fitted the polariton dispersion and obtained the Rabi splitting energy by the coupled oscillator model⁵ expressed as

$$\begin{pmatrix} E_{ex} & V \\ V & E_{cav}(\theta) \end{pmatrix} \begin{pmatrix} \alpha \\ \beta \end{pmatrix} = E(\theta) \begin{pmatrix} \alpha \\ \beta \end{pmatrix} \quad (1)$$

where V is the coupling strength between exciton and CM. $E_{UP}(\theta)$ and $E_{LP}(\theta)$ are two polariton branches with higher and lower eigenenergy given by

$$E_{UP,LP}(\theta) = \frac{1}{2} [E_{ex} + E_{cav}(\theta) \pm \sqrt{4V^2 + (E_{ex} - E_{cav}(\theta))^2}] \quad (2)$$

and $|\alpha|^2$, $|\beta|^2$ represent the exciton and photon component of the corresponding polariton branch, respectively, which are referred to the Hopfield coefficients.³⁹ The Hopfield coefficients for the LP branches are plotted in Figures 3a-3c, corresponding to Figures 2d-2f. As the cavity-exciton detuning changes from negative to positive through tuning cavity lengths, the Hopfield coefficients reveal that the LPs at small angles can be tuned from a more photon-like state, to a photon-exciton-mixed state where photons and excitons could not be distinguished, then to a more exciton-like state. On the one hand, at negative detuning (Figure 3a), the LP is a more photon-like state with smaller effective mass and faster decay rate at small angles, while turning into a more exciton-like state at large angles. On the other hand, at slight positive detuning (Figure 3c), approximate hybrid fractions of the exciton and the photon at small angles can exhibit the polariton state is a half-light half-matter superposition state. Moreover, as shown in Figure 3d, we have obtained the relationship of Rabi splitting and the

cavity-exciton detuning at normal incidence in the samples with different cavity lengths, which displays an anti-crossing and minimum separation between LP and UP energy levels when tuning the CM across the exciton energy.⁵

In the thick 2D perovskite sample embedded in a microcavity, we have investigated the dispersions for a number of polaritons hybridized with BMs at room temperature. Figures 4a and 4c show both the ARR (left panel) and ARPL (right panel) spectra for the dispersions of thick 2D perovskite microcavities (high Q-factor ~ 2200), respectively, and we call them sample A in Figure 4a and sample B in Figure 4c, with different cavity length. Note here slight dispersions of the TM linear polarization mode exist in the ARR spectrum of Figure 4c. In Figures 4a and 4c, the presence of several energetic anti-crossings for adjacent HBP dispersions are clearly seen at resonance between BMs and exciton-cavity coupling states. This phenomenon is the distinct signature of the coherent hybridization between the cavity components of polariton states and BMs of DBRs (Supplementary Materials Figure S3), due to the overlap of wave functions with each other. To confirm the coherent strong coupling, some parameters are extracted from the ARPL spectrum. The dissipation of the cavity photon components ($\hbar\Gamma_{cav}$) and the BM components ($\hbar\Gamma_{BM}$) for HBPs is 1 meV and 9 meV, respectively. At the anti-crossing position, the energetic splitting of the hybridization of photonic components for sample A and B is 22 meV and 9.4 meV, respectively, which can completely withstand the dissipation of CMs and BMs to support the coherent hybridization. We have fitted the experimental HBP dispersions by coupled oscillator models, as shown as figures 4b and 4d. More fitting details are shown in the Supplementary Materials. Various color curves are fitting dispersions of HBP modes labeled by HBP_n , where n takes values from 1 to 4 and from 1 to 14, respectively.

HBPs are forming by the strong coupling between the exciton and hybrid CMs with BMs. As we discuss below, we can observe a strong angular variation of mixing CM, exciton and BM components into the HBP branches, through the analysis of mixing coefficients for each part in the HBP. Figures 5a and 5b show the mixing coefficients of the 3rd HBP branch (HBP_3) for sample A and the mixing coefficients of the 11th HBP

branch (HBP_{11}) for sample B, respectively. These two HBP branches are dominantly attributed to adjacent CM, BM and exciton components. For figure 5a as example, the HBP_3 branch of sample A is mostly composed of the 3rd CM (cav_3) at small angles, then the BM component becomes dominant ranging from 45° to 60°, finally this HBP branch is mostly composed of the 2nd CM (cav_2) at large angles. When the HBP_3 branch approaches the BM around 56°, the BM component reaches a maximum value of 85%. Compared to Figure 4b, it can be observed that the dispersion of the HBP_3 branch steepens as it approaches the BM, where it presents more BM-like. The similar process still exists in the other branches of the sample. In figure 5b, at positions of predominance of CM components, the HBP_{11} branch of sample B, mostly composed of CM and exciton state, exhibits a more photon-like state. When the HBP_{11} branch approaches the BM_1 and the BM_2 , there are three approximate mixing coefficients among CMs, exciton and the BMs, indicating hybrid polaritons are part-light part-matter and these three modes are indistinguishable. Meanwhile, the dispersion of the HBP_{11} branch steepens twice and has maximum mixing coefficients of BMs, when it approaches the BM_1 and the BM_2 , corresponding to the red curve of HBP_{11} in Figure 4d. Thus, the HBP states have effectively hybridized the 2D perovskite exciton, CMs and BMs.

The angular variation of relative photonic population in the different HBP branches is determined by dividing the ARPL emission intensity over the total photonic component of the HBP branches ($P_{ph} \propto I_{PL} / \sum_{cav,BM} |\alpha_n|^2$), and is plotted in figures 5c and 5d. Figure 5d shows the relative populations of photonic components for different HBP branches (HBP_8 to HBP_{12}) in sample B. It can be observed that there are two dips in the angle dependence relative populations of photonic components for the different HBP branches. These traces of two dips comply with the dispersions of BMs. We can observe a majority of photonic populations exist close to the CMs. Due to the large SLR and the low dissipation rate of CMs which is smaller than that of BMs ($\hbar\Gamma_{cav} < \hbar\Gamma_{BM}$), most hybridized particles, originating from the strong coupling of these three modes, will tend to be populated in CMs for coupling to the measured

optical field. This angle dependence distribution of the relative photonic population agrees well with the ARPL emission intensity (figure 4c). Furthermore, as shown in figures 4c and 5f, the mode dependence of the relative photonic population of different HBP branches presents a redistribution, that is, the relative photonic population of some HBP states are greater than the other states, which is dependent on the occupancy of the corresponding polariton states. The microcavities studied here have a relatively high Q-factor (2200). Consequently, a crucial mode dependent redistribution for the occupancy of different polariton states is determined by the dynamic equilibrium between the relaxation along each polariton state and the radiative recombination of polaritons. This similar mechanism still exists in sample A. This effective hybridization can enable polaritons with extremely smaller effective mass and longer lifetime to realize low-threshold polariton condensations.

CONCLUSIONS

In summary, we have realized unambiguous strong light-matter couplings with large Rabi splitting (242 meV) and SLR (> 34.2) in the exfoliated wavelength-tunable 2D organic-inorganic perovskite FP microcavity at room temperature. A significant coherent strong coupling involving exciton state, CMs and BMs, has been demonstrated, enabling a reversible energetic oscillation in these three states. Furthermore, the mixing coefficients and the relative photonic populations of HBPs reveal that sizable hybrid particles coupled to the measured field, through the adjacent CMs with the low photon dissipation and high Q-factor. Our findings provide an ideal platform to realize high-performance polariton devices and to explore fundamental quantum phenomena such as Bose-Einstein condensation, superfluidity and exciton-polariton networks.

MATERIALS AND METHODS

Microcavity Fabrication. The bottom DBR mirror was deposited on a silicon chip by the E-beam evaporation, consisting of 20.5 $\lambda/4$ pairs of tantalum oxide (thickness of 60.2 nm, refraction index of 2.159) and silicon dioxide (thickness of 87.5 nm, refraction index of 1.456). As an optical gain, 2D van der Waals organic-inorganic perovskite single crystal was exfoliated and transferred from the bulk crystal to the bottom DBR mirror using the scotch tape method. The detailed procedure of the synthesis for the 2D perovskite bulk crystal is the same as that described in our previous works.³⁸ The top DBR mirror was finally deposited on the top of 2D perovskite sample using the E-beam evaporation, consisting of 7.5 $\lambda/4$ pairs of tantalum oxide and silicon dioxide.

Optical Characterizations. The fluorescence image of the 2D perovskite was obtained through an Olympus microscope, where the 2D perovskite was illuminated by an Olympus U-HGLGPS lamp. The absorption spectrum of thin 2D perovskite flakes was measured by the commercial micro-transmission/absorption spectrometer (Craic 20/20). The steady-state PL spectra were measured by a confocal microscopic spectrometer (LabRAM HR800, Horiba) at room temperature using a helium cadmium laser (325 nm) as a pump source. ARR and APL spectroscopy were measured by a home-built micro-PL setup within the Fourier imaging configuration.⁴⁰ For the white light illumination and continuous wave laser of 473 nm off-resonant pumping, ARR and ARPL were measured through a high numerical aperture ($NA = 0.9$) 100 $\times$ microscope objective, covering an angular range of $\pm 64^\circ$. The light emission from the 2D perovskite microcavity system was collected and sent to a 550 mm focal length spectrometer (Horiba iHR550) with a 256×1024 pixels liquid nitrogen cooled charge-coupled device (CCD) and a 600 lines/mm grating.

ACKNOWLEDGMENTS

Q.X. acknowledges the strong support from the Singapore National Research Foundation through the NRF Investigatorship Award (NRF-NRFI2015-03) and Competitive Research Programme (NRF-CRP14-2014-03), and the Singapore Ministry of Education *via* AcRF Tier 2 grants (MOE2015-T2-1-047 and MOE2017-T2-1-040) and Tier 1 grants (RG 103/15, RG 113/16, RG 194 /17). Z.C. acknowledges National Natural Science Foundation of China (No.11674069) and Program of Shanghai Subject Chief Scientist (14XD1400200), and National Key R&D Program of China (No.2018YFA0306304). J.W. acknowledges the support of China Scholarship Council for studying at Nanyang Technological University (No.201706100056).

Supporting Information Available: The Q-factor and fitting of HBPs, atomic force microscope of samples, the reflectivity of DBRs, ARPL spectra of polaritonic dispersions (TM mode), and the TE-TM energetic splitting for the HBPs. This material is available free of charge *via* the Internet at <http://pubs.acs.org>.

REFERENCES

1. Weisbuch, C.; Nishioka, M.; Ishikawa, A.; Arakawa, Y. Observation of The Coupled Exciton-Photon Mode Splitting in A Semiconductor Quantum Microcavity. *Phys. Rev. Lett.* **1992**, *69*, 3314.
2. Houdré, R.; Weisbuch, C.; Stanley, R.; Oesterle, U.; Pellandini, P.; Ilegems, M. Measurement of Cavity-Polariton Dispersion Curve from Angle-Resolved Photoluminescence Experiments. *Phys. Rev. Lett.* **1994**, *73*, 2043.
3. Tredicucci, A.; Chen, Y.; Pellegrini, V.; Börger, M.; Sorba, L.; Beltram, F.; Bassani, F. Controlled Exciton-Photon Interaction in Semiconductor Bulk Microcavities. *Phys. Rev. Lett.* **1995**, *75*, 3906.
4. Kasprzak, J.; Richard, M.; Kundermann, S.; Baas, A.; Jeambrun, P.; Keeling, J.; Marchetti, F.; Szymańska, M.; Andre, R.; Staehli, J. Bose-Einstein Condensation of Exciton Polaritons. *Nature* **2006**, *443*, 409.
5. Deng, H.; Haug, H.; Yamamoto, Y. Exciton-Polariton Bose-Einstein Condensation. *Rev. Mod. Phys.* **2010**, *82*, 1489.
6. Schneider, C.; Rahimi-Iman, A.; Kim, N. Y.; Fischer, J.; Savenko, I. G.; Amthor, M.; Lerner, M.; Wolf, A.; Worschech, L.; Kulakovskii, V. D. An Electrically Pumped Polariton Laser. *Nature* **2013**, *497*, 348.
7. Byrnes, T.; Kim, N. Y.; Yamamoto, Y. Exciton-Polariton Condensates. *Nat. Phys.* **2014**, *10*, 803.
8. Amo, A.; Lefrère, J.; Pigeon, S.; Adrados, C.; Ciuti, C.; Carusotto, I.; Houdré, R.; Giacobino, E.; Bramati, A. Superfluidity of Polaritons in Semiconductor Microcavities. *Nat. Phys.* **2009**, *5*, 805.
9. Lagoudakis, K. G.; Wouters, M.; Richard, M.; Baas, A.; Carusotto, I.; André, R.; Dang, L. S.; Deveaud-Plédran, B. Quantized Vortices in An Exciton-Polariton Condensate. *Nat. Phys.* **2008**, *4*, 706-710.
10. Kavokin, A.; Malpuech, G.; Glazov, M. Optical Spin Hall Effect. *Phys. Rev. Lett.* **2005**, *95*, 136601.
11. Leyder, C.; Romanelli, M.; Karr, J. P.; Giacobino, E.; Liew, T. C.; Glazov, M. M.; Kavokin, A. V.; Malpuech, G.; Bramati, A. Observation of The Optical Spin Hall Effect. *Nat. Phys.* **2007**, *3*, 628.
12. Liew, T.; Leyder, C.; Kavokin, A.; Amo, A.; Lefrère, J.; Giacobino, E.; Bramati, A. Interplay Between Weak Localization of Exciton-Polaritons and The Optical Spin Hall Effect. *Phys. Rev. B* **2009**, *79*, 125314.
13. Lai, C.; Kim, N.; Utsunomiya, S.; Roumpos, G.; Deng, H.; Fraser, M.; Byrnes, T.; Recher, P.; Kumada, N.; Fujisawa, T. Coherent Zero-State and π -State in An Exciton-Polariton Condensate Array. *Nature* **2007**, *450*, 529.
14. Deng, H.; Solomon, G. S.; Hey, R.; Ploog, K. H.; Yamamoto, Y. Spatial Coherence of A Polariton Condensate. *Phys. Rev. Lett.* **2007**, *99*, 126403.
15. Bajoni, D.; Senellart, P.; Wertz, E.; Sagnes, I.; Miard, A.; Lemaître, A.; Bloch, J. Polariton Laser Using Single Micropillar GaAs-GaAlAs Semiconductor Cavities. *Phys. Rev. Lett.* **2008**, *100*, 047401.

16. Azzini, S.; Gerace, D.; Galli, M.; Sagnes, I.; Braive, R.; Lemaître, A.; Bloch, J.; Bajoni, D. Ultra-Low Threshold Polariton Lasing in Photonic Crystal Cavities. *Appl. Phys. Lett.* **2011**, *99*, 111106.
17. Bhattacharya, P.; Xiao, B.; Das, A.; Bhowmick, S.; Heo, J. Solid State Electrically Injected Exciton-Polariton Laser. *Phys. Rev. Lett.* **2013**, *110*, 206403.
18. Richard, M.; Kasprzak, J.; Romestain, R.; André, R.; Dang, L. S. Spontaneous Coherent Phase Transition of Polaritons in CdTe Microcavities. *Phys. Rev. Lett.* **2005**, *94*, 187401.
19. Kasprzak, J.; Richard, M.; Baas, A.; Deveaud, B.; André, R.; Poizat, J. P.; Dang, L. S. Second-Order Time Correlations within A Polariton Bose-Einstein Condensate in A CdTe Microcavity. *Phys. Rev. Lett.* **2008**, *100*, 067402.
20. Christopoulos, S.; Von Högersthal, G. B. H.; Grundy, A.; Lagoudakis, P.; Kavokin, A.; Baumberg, J.; Christmann, G.; Butté, R.; Feltin, E.; Carlin, J. F. Room-Temperature Polariton Lasing in Semiconductor Microcavities. *Phys. Rev. Lett.* **2007**, *98*, 126405.
21. Das, A.; Heo, J.; Jankowski, M.; Guo, W.; Zhang, L.; Deng, H.; Bhattacharya, P. Room Temperature Ultralow Threshold GaN Nanowire Polariton Laser. *Phys. Rev. Lett.* **2011**, *107*, 066405.
22. Sun, L.; Chen, Z.; Ren, Q.; Yu, K.; Bai, L.; Zhou, W.; Xiong, H.; Zhu, Z.; Shen, X. Direct Observation of Whispering Gallery Mode Polaritons and Their Dispersion in a ZnO Tapered Microcavity. *Phys. Rev. Lett.* **2008**, *100*, 156403.
23. Liu, M.; Johnston, M. B.; Snaith, H. J. Efficient Planar Heterojunction Perovskite Solar Cells by Vapour Deposition. *Nature* **2013**, *501*, 395.
24. Zhang, Q.; Ha, S. T.; Liu, X.; Sum, T. C.; Xiong, Q. H. Room-Temperature Near-Infrared High-Q Perovskite Whispering-Gallery Planar Nanolasers. *Nano Lett.* **2014**, *14*, 5995.
25. Ha, S. T.; Shen, C.; Zhang, J.; Xiong, Q. H. Laser Cooling of Organic-Inorganic Lead Halide Perovskites. *Nat. Photonics* **2016**, *10*, 115.
26. Wang, J.; Cao, R.; Da, P.; Wang, Y.; Hu, T.; Wu, L.; Lu, J.; Shen, X.; Xu, F.; Zheng, G. Purcell Effect in An Organic-Inorganic Halide Perovskite Semiconductor Microcavity System. *Appl. Phys. Lett.* **2016**, *108*, 022103.
27. Su, R.; Diederichs, C.; Wang, J.; Liew, T. C.; Zhao, J.; Liu, S.; Xu, W.; Chen, Z.; Xiong, Q. H. Room-Temperature Polariton Lasing in All-Inorganic Perovskite Nanoplatelets. *Nano Lett.* **2017**, *17*, 3982.
28. Xing, J.; Liu, X. F.; Zhang, Q.; Ha, S. T.; Yuan, Y. W.; Shen, C.; Sum, T. C.; Xiong, Q. H. Vapor Phase Synthesis of Organometal Halide Perovskite Nanowires for Tunable Room-Temperature Nanolasers. *Nano Lett.* **2015**, *15*, 4571.
29. Xing, J.; Yan, F.; Zhao, Y.; Chen, S.; Yu, H.; Zhang, Q.; Zeng, R.; Demir, H. V.; Sun, X.; Huan, A.; Xiong, Q. H. High-Efficiency Light-Emitting Diodes of Organometal Halide Perovskite Amorphous Nanoparticles. *ACS Nano* **2016**, *10*, 6623.
30. Zhang, Q.; Su, R.; Liu, X.; Xing, J.; Sum, T. C.; Xiong, Q. H. High-Quality Whispering-Gallery-Mode Lasing From Cesium Lead Halide Perovskite Nanoplatelets. *Adv. Funct. Mater.* **2016**, *26*, 6238.
31. Ha, S. T.; Liu, X.; Zhang, Q.; Giovanni, D.; Sum, T. C.; Xiong, Q. H. Synthesis of

- Organic-Inorganic Lead Halide Perovskite Nanoplatelets: Towards High Performance Perovskite Solar Cells and Optoelectronic Devices. *Adv. Opt. Mater.* **2014**, *2*, 838.
32. Lanty, G.; Jemli, K.; Wei, Y.; Leymarie, J.; Even, J.; Lauret, J.-S.; Deleporte, E. Room-Temperature Optical Tunability and Anhomogeneous Broadening in 2D-Layered Organic-Inorganic Perovskite Pseudobinary Alloys. *J. Phys. Chem. Lett.* **2014**, *5*, 3958.
 33. Tsai, H.; Nie, W.; Blancon, J.-C.; Stoumpos, C. C.; Asadpour, R.; Harutyunyan, B.; Neukirch, A. J.; Verduzco, R.; Crochet, J. J.; Tretiak, S. High-Efficiency Two-Dimensional Ruddlesden-Popper Perovskite Solar Cells. *Nature* **2016**, *536*, 312.
 34. Yuan, M.; Quan, L. N.; Comin, R.; Walters, G.; Sabatini, R.; Voznyy, O.; Hoogland, S.; Zhao, Y.; Beauregard, E. M.; Kanjanaboos, P. Perovskite Energy Funnels for Efficient Light-Emitting Diodes. *Nat. Nanotechnol.* **2016**, *11*, 872.
 35. Byun, J.; Cho, H.; Wolf, C.; Jang, M.; Sadhanala, A.; Friend, R. H.; Yang, H.; Lee, T. W. Efficient Visible Quasi-2D Perovskite Light-Emitting Diodes. *Adv. Mater.* **2016**, *28*, 7515.
 36. Huo, C.; Cai, B.; Yuan, Z.; Ma, B.; Zeng, H. Two-Dimensional Metal Halide Perovskites: Theory, Synthesis, and Optoelectronics. *Small Methods* **2017**, *1*, 1600018.
 37. Liu, X.; Bao, W.; Li, Q.; Ropp, C.; Wang, Y.; Zhang, X. Control of Coherently Coupled Exciton Polaritons in Monolayer Tungsten Disulphide. *Phys. Rev. Lett.* **2017**, *119*, 027403.
 38. Liu, W.; Xing, J.; Zhao, J.; Wen, X.; Wang, K.; Lu, P.; Xiong, Q. Giant Two-Photon Absorption and Its Saturation in 2D Organic-Inorganic Perovskite. *Adv. Opt. Mater.* **2017**, *5*, 1601045.
 39. Hopfield, J. Theory of The Contribution of Excitons to The Complex Dielectric Constant of Crystals. *Phys. Rev.* **1958**, *112*, 1555.
 40. Wang, J.; Da, P.; Zhang, Z.; Luo, S.; Liao, L.; Sun, Z.; Shen, X.; Wu, S.; Zheng, G.; Chen, Z. Lasing From Lead Halide Perovskite Semiconductor Microcavity System. *Nanoscale* **2018**, *10*, 10371.

Figures and Captions

Figure 1. Microcavity structure and emission. (a) Schematic of energy-band structure of 2D organic-inorganic perovskite (left) and a 2D perovskite microcavity (right), consisting of a 2D perovskite crystal inserted in a planar microcavity with bottom and top DBRs. (b) The atomic force microscope image of a typical exfoliated 2D (PEA)₂PbI₄ perovskite crystal with a width of 25 μm and a height of 254 nm after complete microcavity fabrication. (c) Room temperature absorption (red curve) and PL emission spectrum (blue curve) of the thin 2D (PEA)₂PbI₄ perovskite flake, the polariton emission (olive curve) of 2D perovskite embedded into the microcavity. (d) Room temperature PL spectrum of thick 2D (PEA)₂PbI₄ perovskite crystal on silicon substrate. The inset represents the corresponding fluorescence microscopy image, showing strong and uniform green emission. The scale bar is 10 μm in length.

Figure 2. Angle-resolved reflectivity and PL spectra of 2D organic-inorganic perovskite microcavity system with single CM. (a-c) Angle-resolved reflectivity spectra of 2D (PEA)₂PbI₄ perovskite with negative detuning (-31.7 meV) and positive detuning (61 meV), and (PEA)₂PbBr₄ perovskite with positive detuning (10.8 meV), respectively. Dashed white lines show the theoretical dispersion of the upper (UP) and lower (LP) polariton states. Solid yellow lines display the dispersion of uncoupled 2D perovskite exciton (Ex) and cavity photon modes (CM). The angles represent the detecting angles. (d-f) Angle-resolved PL spectra of 2D perovskite microcavity corresponding to (a-c). Dashed white lines show the theoretical dispersions of the UP and LP states. Solid yellow lines display the dispersions of Ex and CM.

Figure 3. Hopfield coefficients of lower polariton (LP) states and the relationship between Rabi splitting and the cavity-exciton detuning. (a)-(c) The components of exciton and photon in LP states at cavity-exciton detuning of -31.7 meV, 61 meV and 10.8 meV, respectively, corresponding to Figure 2. Red and blue curves represent the excitonic and photonic components, respectively. (d) The relationship of Rabi splitting (Ω) and the cavity-exciton detuning (Δ) at normal incidence in the samples with different cavity lengths. Red squares and black dots are the fitting positions of UP states and CMs, respectively. Red and blue curve represent the simulation of UP and LP states position.

Figure 4. Angle-resolved reflectivity and PL spectra of 2D (PEA)₂PbI₄ perovskite microcavity system with multi-mode dispersions. (a, c) The left panel and right panel are angle-resolved reflectivity and PL spectra of samples A and B ($Q \sim 2200$), respectively. Energetic anti-crossings of dispersions occur around 45° indicated by the white dashed box. (b, d) The fitting dispersions of hybrid Bragg-mode polaritons (HBP) for samples A and B. The black dashed line represents the exciton state. Color curves are dispersions of HBPs labeled by HBP_n , where n takes values from 1 to 4 and from 1 to 14 for samples A and B, respectively. The traces of anti-crossing gap comply with the dispersions of bare BMs.

Figure 5. Mixing coefficients and relative populations of HBPs. (a, b) Mixing coefficients of the 3rd HBP branches and the 11th HBP branches for samples A and B, respectively. The solid lines, the dashed lines and the dot dashed lines represent the components of CMs, BMs and the exciton, respectively. (c, d) Angle dependence of the relative photonic populations of the HBP branches for samples A and B, respectively. The photonic populations are the number of photons which couple to the measured optical field. (e, f) Mode dependence of the relative photonic populations of the HBP branches for samples A and B, respectively.

TOC graphic