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Halide perovskites enable polaritonic *XY* spin Hamiltonian at room temperature

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Exciton polaritons, the part-light and part-matter quasiparticles in semiconductor optical cavities, are promising for exploring Bose–Einstein condensation, non-equilibrium many-body physics and analogue simulation at elevated temperatures. However, a room-temperature polaritonic platform on par with the GaAs quantum wells grown by molecular beam epitaxy at low temperatures remains elusive. The operation of such a platform calls for long-lifetime, strongly interacting excitons in a stringent material system with large yet nanoscale-thin geometry and homogeneous properties. Here, we address this challenge by adopting a method based on the solution synthesis of excitonic halide perovskites grown under nanoconfinement. Such nanoconfinement growth facilitates the synthesis of smooth and homogeneous single-crystalline large crystals enabling the demonstration of *XY* Hamiltonian lattices with sizes up to 10 × 10. With this demonstration, we further establish perovskites as a promising platform for room temperature polaritonic physics and pave the way for the realization of robust mode-disorder-free polaritonic devices at room temperature.

Bose–Einstein condensation (BEC), first proposed in 1924, describes a quantum-statistical phase transition where, below the transition temperature, a substantial fraction of diluted bosonic particles spontaneously occupy the ground state and exhibit macroscopic quantum behaviours. The experimental realization of BEC condensates and their optical lattices in ultracold (approximately microkelvin) atoms enabled the field of analogue simulation, where complex many-body physics problems such as the formation of frustrated spin, superconductivity and superfluidity, to be experimentally tackled in a controllable manner[1–3]. Because of the complexity and extremely low temperature involved in using ultracold atoms, there have been great efforts recently towards building a room-temperature analogue simulator with solid-state BEC systems for practical applications[4–6].

In this context, semiconductor microcavities are an attractive platform. In a semiconductor optical cavity, exciton polaritons, hybrid light–matter quasiparticles, are formed when the coupling rate between excitons and photons is much faster than their respective dissipation rates. These bosonic polaritons have an ultra-small effective mass (~10^{-5} electron mass) inherited from the photon, as well as nonlinear interactions inherited from the exciton Coulombic interactions. Thus, polaritons can undergo BEC at much-elevated temperatures[4,6], ultimately limited by the exciton binding energy. Polariton condensation was first realized in quantum-well microcavities grown with molecular beam epitaxy (MBE)[7,8], where liquid helium temperatures (~4 K) must be maintained to prevent exciton autoionization. Although high exciton binding energy materials, such as organic molecules, show potential for room-temperature polaritonics[9–12], they unfortunately have severe limitations on sample size, homogeneity, polariton lifetime and/or nonlinear interaction strength. Therefore, the large and homogeneous MBE-grown GaAs quantum well remains the only exciton–polariton platform (though at low temperature) where interesting physics, such as soliton formation[13,14], *XY* spin Hamiltonian[15] and topological effects[16,17], have been experimentally explored. This is primarily attributed to two reasons: the maturity of high-quality MBE material growth and patterning, and the strongly interacting excitons of the GaAs quantum wells at liquid helium temperature.

Recently, semiconducting lead halide perovskites have emerged as contenders to GaAs for polaritonics but at room temperature due to their large exciton binding energy[18–20], high photoluminescence (PL) quantum yield[21], tunable bandgap[22] and high room-temperature nonlinear interaction strength[23]. With chemical vapour deposition (CVD), single-crystalline inorganic halide perovskites have shown polariton condensation[24,25]. However, due to the limitations of the current growth methods and the fragile nature of perovskites, only small single crystals can be integrated into the optical cavity (Supplementary Fig. 1). Critically, the small sizes prohibit the studies of the aforementioned large-scale phenomena due to the limited lattice size and the restricted propagation lengths[26].

In this work, we overcome these limitations by direct solution growth of various types of large-area halide perovskite single crystals inside optical nanocavities. Due to the uniform confined environment, our solution growth approach shows uniformity, comparable to the MBE-grown GaAs quantum well, enabling submillimetre-size large single crystals with superb excitonic quality. These crystals with strongly interacting Wannier–Mott excitons allowed us to successfully demonstrate a polaritonic *XY* spin Hamiltonian with all-inorganic perovskite $CsPbBr_3$ at room temperature. Further, we show that a lattice with a large number of coherently coupled condensates up to 10 × 10 can be achieved. This is an important step towards the ultimate goal of realizing a room-temperature polaritonic platform on par with MBE-grown GaAs at low temperatures. In addition, we show that the dispersion of the perovskite system has unique advantages for future studies on synthetic non-Abelian gauge fields and topological physics. Our work establishes a robust

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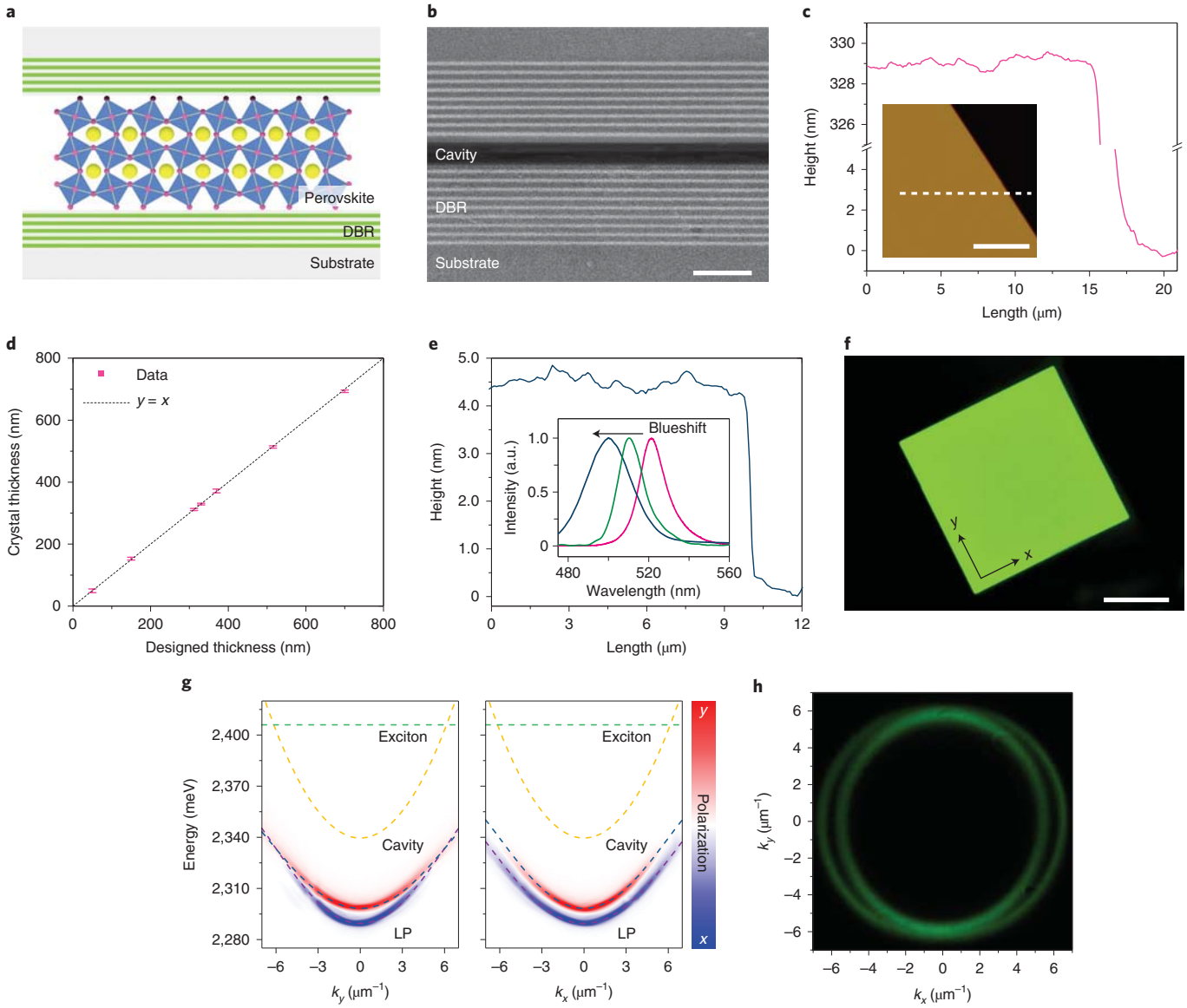

Fig. 1 | Synthesis and characterization of single-crystalline excitonic halide perovskites in DBR nanocavities. a, By using deposited pillar arrays (Supplementary Fig. 2) as spacers and bonders in between two DBR mirrors (nine pairs of SiO_2 and Ta_2O_5 alternating layers), a nanocavity of a designed height for halide perovskite crystals is formed so that the detuning between exciton and photon can be controlled. **b**, The scanning electron microscopy cross-sectional image of a fabricated nanocavity. Scale bar, 1 μm. **c**, The after-growth cavities are opened. The crystal thickness controllability and uniformity is revealed by AFM imaging (inset) and a cross-section profile (the white dashed line in the insert); the thickness of as-synthesized crystal from the nanocavity is ~329 nm (±0.4 nm), which matches the designed height of the nanocavity (~330 nm). Scale bar, 10 μm. **d**, This nanometre-scale-precision thickness can be well controlled in a wide range as measured by AFM. The <1.6% of thickness variance across each chip (2 cm size) is caused by pillar film deposition uniformity. **e**, When shrinking the nanocavity height to only a few nanometres, the $CsPbBr_3$ crystals show a thickness of only 4.45 ± 0.35 nm and strong quantum-confinement-induced blueshifted PL (inset blue curve) down to 499 nm. The green curve in the inset corresponds to a measured thickness of 7.3 ± 0.5 nm (Supplementary Fig. 9b). **f**, A typical $CsPbBr_3$ crystal grown in the nanocavity shows uniform and bright PL across hundreds of micrometres. Scale bar, 100 μm. **g**, The linear-polarized PL of polariton dispersions (energy vs. wavevector k) along the y and x axes (two crystal axes as shown in **f**). The dashed lines are the fittings of the two lower branches (LP; red and blue), exciton energy (green) and cavity (orange) modes.. The two lower polariton branches with polarization along x (blue) and y (red) axis cross at the diabolic points at $k_y = 5.9$ μm^{-1}. **h**, The cross-section of the two polariton branches (PL measurement) in k-space at diabolic points with the energy of 2,332.5 meV, illustrating the Rashba-like spin–orbital coupling.

and excellent platform for room-temperature polaritonics and also paves the way for many other nonlinear polaritonic devices for practical applications.

Device synthesis and characterization

The key that enables our synthesis of high-quality perovskite crystals for large-scale polaritonics and the subsequent realization of the polaritonic *XY* spin Hamiltonian at room temperature is the solution synthesis of halide perovskites directly within prefabricated optical nanocavities (a combination of top-down fabrication and bottom-up synthesis; schematic depiction in Supplementary Fig. 2), inspired by previous efforts in organic polariton growth[27] and recent halide perovskite growth[28]. We first deposited high-quality distributed Bragg reflector (DBR) multilayers on the quartz

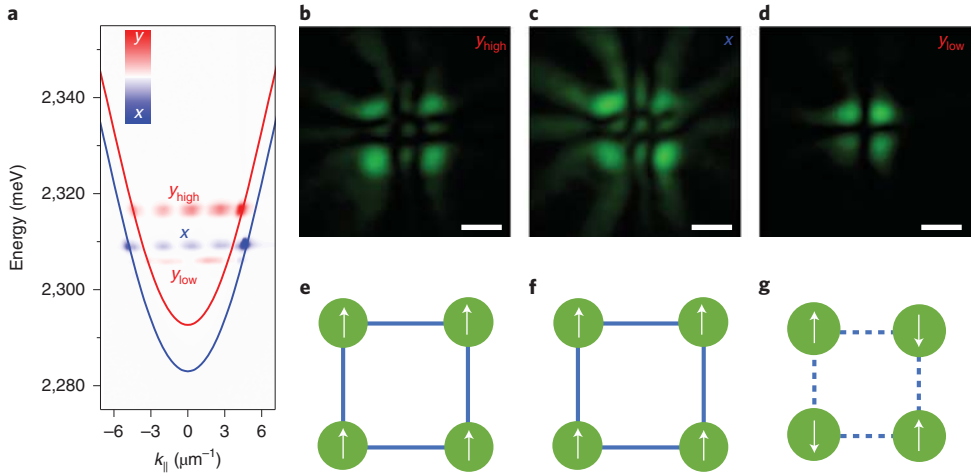

Fig. 2 | Room-temperature demonstration of polariton *XY* Hamiltonian square 2 × 2 lattices. a–d, Time-integrated k-space polariton dispersion before (the solid lines) and after (lines of dots) the condensation (**a**). The polariton condenses simultaneously at two lower branches with perpendicular polarization. At the branch with *x* polarization, the condensation occurs at $k_c = 4.95\,\mu m^{-1}$. The odd number of interference fringes in the corresponding real-space image (**c**) shows an in-phase coupling. At the branch with *y* polarization, the condensation occurs at two different wave vectors (y_{high}, $k_c = 4.3\,\mu m^{-1}$; y_{low}, $k_c = 3.2\,\mu m^{-1}$). Panels **b** and **d** are the time-integrated real-space images of y_{high} and y_{low}, respectively. The intrinsic oscillatory lattice dynamics mechanism reduces the oscillation between the two condensation states with opposite phases and different coupling: in-phase for y_{high} and antiphase for y_{low}. **e–g**, Diagrams of spin configurations of condensations y_{high}, *x* and y_{low}, corresponding to the real-space images of **b**, **c** and **d**, respectively. Panels **e** and **f** show the antiferromagnetic coupling (solid blue lines) with parallel spins, and **g** shows ferromagnetic coupling (dashed blue lines) with antiparallel spins between neighbouring sites. Scale bars in **b**, **c** and **d**, 2 μm.

substrates. Two of these DBR substrates were then bonded together via a thermal compression bonding process with prepatterned gold pillars as the spacers (Methods and Supplementary Fig. 3), creating a symmetric two-dimensional (2D) confined nanocavity (Fig. 1b) to serve as the optical cavity as well as the crystal growth environment. Chemical reaction and crystal growth under nanometre confinement are different than in traditional methods (Methods and Supplementary Fig. 1), and the resulting single-crystalline perovskite crystals are very large (>300 μm; Supplementary Fig. 4) due to a substantially decreased nucleation rate (Supplementary Fig. 6)[29]. In addition, the nanoconfined environment also leads to evaporation only from the nanocavities' exposed edges, resulting in a slow crystal growth rate[30] and thus excellent crystallinity, as evidenced by the sharp X-ray diffraction peaks (Supplementary Fig. 5). To demonstrate its broad applicability, we synthesized three emerging excitonic halide perovskites (Supplementary Fig. 4)—the all-inorganic bromide perovskite $CsPbBr_3$, the 2D iodide layered perovskite $(C_6H_5C_2H_4NH_3)_2PbI_4$ (phenylethylamine lead iodide, PEPI) and the organic–inorganic hybrid chloride perovskite $MAPbCl_3$—all with submillimetre crystal sizes and high crystallinity (Supplementary Fig. 5). Compared with previously reported layer-by-layer stacking of polaritonic devices, our approach produced a laterally large size, yet vertically controllable, nanoscale-thin, single-crystalline halide perovskites free from damage or the contamination from traditional fabrication due to the use of prebonded ultra-flat DBR mirrors (Supplementary Figs. 7 and 8). At the same time, this fabrication design also produces pristine DBR optical reflectivity, which indicates excellent potential for future studies of near thermal equilibrium polariton BEC.

Our work combines large lateral size, nanometre-precision thickness control, homogeneity and excellent excitonic properties as required for large-scale room-temperature polaritonics. The precise control over the crystal thickness here enables accurate tuning of the photonic mode and hence the emission dynamics of the polariton mode. To demonstrate the excellent crystal thickness control and uniformity, we performed atomic force microscopy (AFM) of our $CsPbBr_3$ crystal. The height profile (Fig. 1c) reveals a 329 ± 0.4 nm range (~0.12% variance) across a lateral distance of 20 μm (Fig. 1c inset). This measured height matches well with the designed cavity height (~330 nm; Fig. 1b). By designing cavities of various heights (adjusting the pillar height between DBR substrates), we observed that the crystal grew to fill the cavities completely, with minimal deviation (Fig. 1d). The fully filled cavities in the perovskite chemical synthesis lead to excellent thickness homogeneity, which results in small polariton mode disorders (Supplementary Fig. 14); these cavities are distinct from the partially filled cavities in organics' physical recrystallization, which have substantial polariton mode inhomogeneity[27]. Across 50 to 700 nm, the crystal thickness control shows nanometre-scale precision, with less than 1.6% of the variance across each chip (2 cm size) caused by pillar film deposition uniformity. In addition, we also achieved a cavity thickness down to 4.45 ± 0.35 nm (Fig. 1e and Supplementary Fig. 9), limiting the crystal thickness to a few unit cells. This quantum confinement even led to control over the excitonic energy level.

This method also allows us to fully take advantage of the perovskites' excellent optical properties. PL mapping under ultraviolet excitation showed excellent optical homogeneity (Fig. 1f and Supplementary Figs. 10 and 14); a long PL lifetime (Supplementary Fig. 11), pointing to low non-radiative loss; and a narrow PL emission peak (Supplementary Fig. 12). More importantly, robust excitons were observed at room temperature, supported by the strong excitonic absorption peak (Supplementary Fig. 13). The optical properties, combined with the high reflectivity of the DBR mirrors (cavity quality factor $Q \approx 900$ with two symmetric nine pairs DBR mirrors on both sides of the perovskites), resulted in the strongly coupled exciton polaritons (Fig. 1g). Due to the high optical quality and precise detuning control of the solution-grown $CsPbBr_3$ perovskite sample, a room-temperature polariton BEC at $k_{||} = 0$ ($k_{||}$ is the in-xy-plane wavevector) with non-resonant excitation was readily observed (Supplementary Figs. 16 and 17).

More interestingly, due to the orthorhombic crystal structure of $CsPbBr_3$ at room temperature, optical birefringence breaks

the degeneracy of the two photonics transverse-electric and transverse-magnetic modes at $k_{\parallel}=0$ (ref. [25]; Fig. 1g). The combination of noticeable optical birefringence and spin–orbit coupling that arises from transverse-electric/transverse-magnetic mode splitting in $CsPbBr_3$ creates an interesting k-space dispersion[31] (Fig. 1g) along with the x and y directions (two crystal axes as shown in Fig. 1f). This dispersion effectively creates a synthetic gauge field on the flowing polariton condensate and is predicted to be Rashba-like around the two diabolical points (Fig. 1g,h)[31], where the two polariton branches exactly cross. In contrast to the organics[32–34], the nonlinearity and homogeneity in our perovskites are much superior. Thus, the perovskite microcavities also provide a unique room-temperature playground for future large-scale and nonlinear studies on non-Abelian gauge fields[17,31,35] and topological physics[36].

Construction of *XY* spin Hamiltonian

Given the large and homogeneous samples with interesting polariton dispersion, we were able to explore the potential and limits of our approach by performing the room-temperature polariton graphs of the *XY* Hamiltonian minimizer demonstration with $CsPbBr_3$ perovskites. Many complex optimization problems can be mapped into microscopic spin models with rich macroscopic behaviours such as a distinct Kosterlitz–Thouless[37] phase transition. However, compared with other lithographically patterned polariton lattices[38,39], experimentally implementing *XY* Hamiltonian lattices at room temperature is one of the most challenging tasks due to the stringent sample requirements. Thus, a successful construction has only been reported at 10 K with the best MBE-grown GaAs quantum-well microcavities[15,40], where both excellent sample homogeneity (within a few millielectronvolts potential disorders/variations) and large sample size requirements can be met simultaneously.

Here, taking advantage of nanoconfinement-grown perovskites, we finally construct the polaritonic *XY* spin Hamiltonian at room temperature by non-resonantly exciting polariton lattice patterns with a reflective spatial light modulator (schematics in Supplementary Fig. 15). PL images of the condensate lattice were collected in a transmission configuration to control the excitation and the signal collection separately. Because of the strong polariton–polariton repulsive interaction originating from the Coulombic interaction among excitons, the polaritons under condensation will have a non-zero in-plane momentum and expand ballistically[41,42] with a small excitation laser spot (Supplementary Fig. 18). When these expanding condensates are brought closer, interference and coupling occur, leading to phase synchronization between the condensates[43]. According to the complex Ginzburg–Landau equation model in previous analysis[15], the total amount of polaritons N_{XY} can be approximated as $N_{XY}=N_{iso}+\sum_{ij}J_{ij}\cos\left(\theta_i-\theta_j\right)$, where N_{iso} is the total polariton number with full isolated sites (infinite distance), $\theta_i-\theta_j$ is the phase difference between two condensates and J_{ij} is the nearest neighbour coupling strength. The final macroscopic coherent state will have the highest possible polaritonic occupancy to minimize the polariton losses[44]. Thus, in the ideal cases, the relative phase difference between condensates can be used as pseudo-spins and provides a route to search the global minimum in the *XY* Hamiltonian[15,40,44], $H_{XY}=-\sum_{ij}J_{ij}\cos\left(\theta_i-\theta_j\right)$. When the Josephson coupling is negligible, the coupling strength J_{ij} has an oscillating characteristic as a function of $k_c d$, where k_c is the outflow in-plane momentum and d is the distance between the two adjacent condensates.[15,41] This results in ferromagnetic (positive sign) and antiferromagnetic (negative sign) couplings between sites, as demonstrated in Supplementary Figs. 22 and 23 for a polariton dyad configuration. Thus, by changing the geometry of the polariton lattice via a spatial light modulator, different *XY* spin Hamiltonians can be readily constructed.

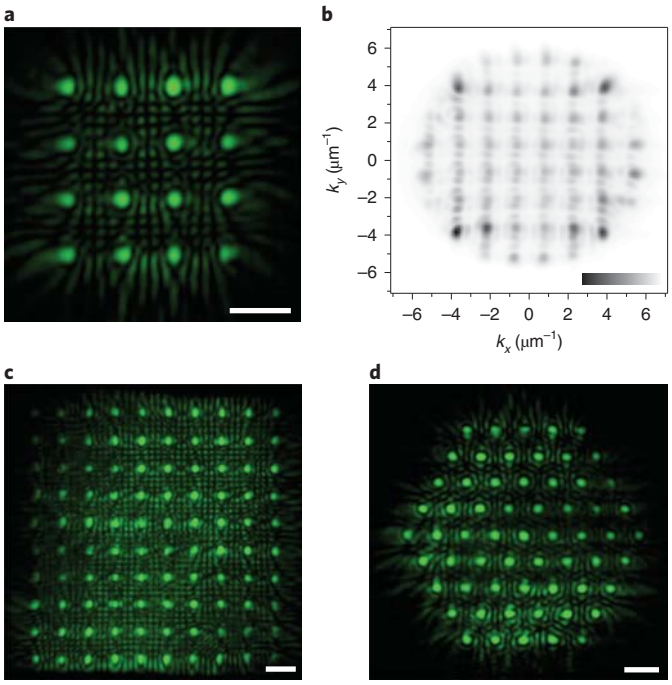

Fig. 3 | Extended polariton lattices. a,**b**, Time-integrated real-space and k-space images of a 4×4 polariton lattice with antiferromagnetic coupling. The even interference fringes can be clearly identified. In the colour bar in **b**, black is high intensity and white is low intensity. **c**, Time-integrated real-space image of a 10×10 polariton lattice with antiferromagnetic coupling. **d**, Time-integrated real-space image of a triangular polariton lattice with ferromagnetic coupling. Scale bars in **a**, **c** and **d**, 5 μm.

When a 2×2 lattice was excited in $CsPbBr_3$ microcavities with a fixed laser spot distance of ~1.5 μm, in contrast with GaAs, the two split lower polariton branches with perpendicular polarizations (Supplementary Fig. 18) condense simultaneously at three slightly different k-space locations (Fig. 2a). The x-polarization branch condenses at $k_c=4.95\,\mu m^{-1}$ with an odd number of interference fringes in k-space dispersion images (line of blue dots in Fig. 2a). The corresponding four real-space condensates (Fig. 2c) also have the same phase, indicating a ferromagnetic coupling as in the spin diagram shown in Fig. 2f. For the y-polarization branch, both ferromagnetic and antiferromagnetic configurations were identified. While the condensates with higher $k_c=4.3\,\mu m^{-1}$ (y_{high}) are synchronized to have the same phase (Fig. 2b), the lower $k_c=3.2\,\mu m^{-1}$ condensates (y_{low}) have a π phase difference between the neighbours (Fig. 2d). The antiphase interference effect also induces the condensates to be closer, consistent with low-temperature GaAs cases[15]. This coexistence of both in-phase and antiphase synchronization of the same polarization branch is due to the intrinsic oscillatory lattice dynamics[45]. In addition, a 90° compass spin model can also be realized with an asymmetrical square configuration (Supplementary Fig. 25).

For the future study of phase transitions in interacting bosonic systems such as the Kosterlitz–Thouless transition with a polariton, it is essential to construct a large-scale, homogeneous and coherently coupled lattice. Towards this goal, we gradually increase the node number of the polariton lattices and tune the lattice geometry simply by dynamically changing the spatial light modulator excitation pattern. High-quality real-space and k-space images of a 4×4 polariton lattice (Fig. 3a,b) are first achieved. The even number of real-space interference fringes between the two neighbouring condensations (Fig. 3a) can be clearly seen, indicating an antiferromagnetic energy minimum state. In addition, the k-space image confirms the antiferromagnetic coupling with the characteristic

pattern, where the slightly lower-intensity centre spots are accompanied by higher-intensity Bragg diffraction peaks in the surrounding corner (Fig. 3b), similar to low-temperature GaAs cases[15]. Next, an 8×8 lattice is also easily constructed (Supplementary Fig. 26). In addition, the square lattice can be further extended to 10×10 coherently coupled condensates with a full lattice size of more than 50 μm. Again, an even number of interference fringes with excellent visibility in between neighbouring condensates suggest an antiferromagnetic coupling. The inhomogeneity of the condensates at the edge region is mainly caused by the polariton outflow (the particle outflow to the sites on the edge happens from fewer sites in comparison with the central sites), as shown in Fig. 3 and Supplementary Fig. 24, instead of the small disorders in our samples (lower polariton-mode spatial root-mean-square variations of only 1.02 meV in Supplementary Fig. 14). These intensity inhomogeneities play an essential role in changing the interactions and the phase difference distribution[44], which can be addressed in the future with feedback excitation schemes recently reported[40]. A real-space PL image of a sizable (40 μm) triangular lattice, one of the most important spin configurations in magnetism research[2,15], is also shown in Fig. 3d. In this case, the *XY* Hamiltonian is minimized when all polariton sites lock in phase and establish a ferromagnetic configuration.

Outlook

We emphasize that, to realize this room-temperature polaritonic *XY* Hamiltonian experiment, strongly interacting polaritons and large homogeneous samples are crucial to generate outflow condensates and to construct a large node lattice without being impacted by the crystal boundaries or disorders, respectively. Thanks to the Wannier–Mott excitons, the polariton–polariton interaction constant g (ref. [23]) in perovskite can be at least two orders of magnitude higher than in the organics with weakly interacting Frenkel excitons[46]. The stringent requirements of homogeneity (one almost needs disorder free), photo-stability and large sample size further hinder other room-temperature systems[10,11,24,25,47] from performing the above spin Hamiltonian experiments. In addition, the experimental room-temperature real-space polariton PL images from our solution-grown samples are comparable to the low-temperature MBE-grown GaAs case[15], which further justifies the high quality of our perovskite platform. The interesting dispersions of perovskite can lead to three sets of *XY* Hamiltonian patterns, due to condensation at three different k-space locations. Last but not least, future innovations on material platforms and optical technology are still highly sought to achieve practical *XY* spin Hamiltonian simulators for optimization problems, where one can precisely control the Hamiltonian at a more quantitative level with a substantially large lattice size.

To conclude, we solution-synthesized various types of halide perovskites using a nanoconfinement growth approach for room-temperature polaritonics. The resulting large-area but nanoscale-thickness crystals with excellent excitonic properties and homogeneity enabled us to demonstrate a polaritonic spin Hamiltonian lattice with a large size at room temperature. Our work establishes a robust room-temperature polaritonic platform for studying non-equilibrium physics. Given the pristine DBR optical reflectivity, the approach is also attractive for near thermal equilibrium polariton BEC at room temperature. The unique combination of intrinsically large optical birefringence and strongly nonlinear interacting polaritons is ideal for studies of quantum fluid physics in topological systems[31] and simulating a lithography-patterned[39] large 2D Hamiltonian with spin–orbital coupling[48]. The strong nonlinearity from Wannier–Mott excitons alone could also lead to many other device applications, such as a 2D topological polaritonic laser[16,49] and potential photonic simulator with synthetic gauge field[31,35], previously inaccessible at room temperature.

Online content

Any methods, additional references, Nature Research reporting summaries, source data, extended data, supplementary information, acknowledgements, peer review information; details of author contributions and competing interests; and statements of data and code availability are available at https://doi.org/10.1038/s41563-022-01276-4.

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