Controlling matter–light interactions has mostly been implemented either in microscopic systems (individual quantum particles) or in macroscopic ensembles of free particles that can often be modelled classically. Bridging the two limits to realize collective coupling among arrays of quantum particles ushers new paradigms in quantum many-body physics and quantum simulation research, such as those pursued using atomic optical lattices and cavities. In solid-state systems, however, such a pursuit has been exceedingly difficult, because nonlinearity is often too weak, owing to screened Coulomb interactions, and inhomogeneity is too large. Here, we show that moiré lattices formed in hetero-bilayer (hBL) transition metal dichalcogenide (TMDC) crystals may provide a platform that overcomes these limitations of conventional solids.

When two monolayer (ML) crystals are placed together, a moiré lattice can form owing to a slight mismatch in the lattice constants or crystal orientations of the MLs. Its period is tunable by the twist angle between the ML crystals from a few to a few tens of nanometres—comparable to the range of Coulomb interactions in semiconductors. The natural formation of the moiré lattice promises the prospect of uniformity among different moiré cells across the lattice, thereby potentially enabling a new solid-state platform for cooperative phenomena between light and arrays of nonlinear quantum particles. Although exciton states induced by the moiré lattice have been reported, the fundamental question remains of whether a sufficiently uniform lattice of quantized excitations can be formed in a moiré system. Exciton–light interactions have remained in the perturbative regime, and cooperative phenomena have not been reported.

**Moiré polaritons in WS₂–MoSe₂ hBLs**

To enable cooperative coupling between moiré excitons and photons, we use WS₂–MoSe₂ hBLs enclosed in a microcavity (Fig. 1a). The hBL is encapsulated in hexagonal boron nitride (hBN) with a twist angle measured to be $56.5° \pm 0.9°$ (the uncertainty corresponds to the 95% confidence interval in fitting the polarization-dependent second-harmonic-generation spectra) (Extended Data Fig. 1). The WS₂–MoSe₂ hBL is unique among frequently studied TMDCs hBLs in that the two lowest-energy moiré exciton modes have large oscillator strengths, inherited from the ML MoSe₂ A-exciton. This not only allows ready identification of moiré excitons via the absorption spectra, but also suggests that both of the moiré excitons may strongly couple with light and form stable moiré polaritons.

We first identify the existence of a moiré lattice and moiré excitons before enclosing the hBL in a cavity. As shown by the comparison of the hBL and ML reflection spectra in Fig. 1c, the ML MoSe₂ A-exciton is split into two moiré excitons in the hBL, $X_1$ and $X_2$, separated by about...
**Fig. 1** Moiré polariton system and the constituent moiré excitons and microcavity. a, Schematic of the moiré polariton system, formed by excitons confined in a moiré lattice and coupled with a planar cavity. b, Moiré band structure for hBL excitons along paths connecting the centre (y) and the corners (k, k′, k″) of the moiré Brillouin zone, where \( X_1 \) and \( X_2 \) are two bright moiré exciton states. See Methods section ‘Theory of moiré excitons’ for the details of the calculation. c, Top, theoretical optical absorption spectrum calculated from the moiré exciton band structure. Bottom, reflection contrast spectra near the MoSe\(_2\) A-exciton resonance, from the WS\(_2\)-MoSe\(_2\) hBL (red), MoSe\(_2\)-ML (blue) and WS-ML (green). The spectra are displaced vertically for clarity. The MoSe\(_2\) ML A-exciton (\( X_1 \)) splits into two well resolved moiré excitons (\( X_1 \) and \( X_2 \)) in the hBL. a.u., arbitrary units. d, Angle-resolved white-light reflection spectrum of the bare cavity, measured in a region where there is no hBL, showing a single cavity dispersion with a half-line width of \( \gamma = 2.7 \) meV. \( k_i \) is the in-plane wavevector.

40 meV, both with pronounced absorption, consistent with calculations (Fig. 1b, c) and the temperature dependence of the hBL (Extended Data Fig. 2).

When the hBL is placed at the anti-node of a \( \lambda/2 \) microcavity, where \( \lambda \) is the wavelength of the cavity resonance (Fig. 1d; see Extended Data Fig. 3 for more details of the cavity), in place of the moiré excitons or the bare cavity, three dispersive modes are observed up to 70 K, as shown in Fig. 2a, b. The modes anti-cross at the two moiré exciton resonances, showing clearly strong coupling of both moiré excitons (\( X_1 \) and \( X_2 \)) with the cavity photon. The emission temperature dependence of the reflectance spectra and time-resolved studies are provided in Extended Data Figs. 4–6.

The measured dispersions (Fig. 2d, e) are described very well by calculations based on a three-coupled oscillator model with a Hamiltonian:

\[
H = \begin{bmatrix}
E_{X1} & 0 & \Omega_1 \\
0 & E_{X2} & \Omega_2 \\
\Omega_1 & \Omega_2 & E_c
\end{bmatrix},
\]

(1)

Here, \( E_i \) is the energy of the cavity mode, \( E_{X1,2} \) are the energies of the two moiré excitons and \( \Omega_{1,2} \) are their coupling strengths with the cavity photon. The three distinct dispersions measured correspond to the three new light–matter hybrid eigenmodes of the Hamiltonian: the upper (UP), middle (MP) and lower (LP) polaritons. The fitted \( E_{X1,2} \) values agree with the independently measured moiré excitons energies (Fig. 1c) within 5 meV, and this difference is usually observed as a result of strain due to the deposition of the top mirror. The fitting gives \( \Omega_1 = 10.1 \pm 0.3 \) meV and \( \Omega_2 = 8.5 \pm 0.3 \) meV at 4 K, which change slightly to \( \Omega_1 = 9.6 \pm 0.4 \) meV and \( \Omega_2 = 8.7 \pm 0.4 \) meV at 70 K. From independent measurements, the exciton half linewidths increase from \( \gamma_{1,2} = 7.5 \) meV and \( \gamma_{1,2} = 8.4 \) meV at 4 K to \( \gamma_1 = 8.8 \) meV and \( \gamma_2 = 8.9 \) meV at 70 K (Extended Data Fig. 5), which are mainly due to inhomogeneous broadening. The oscillator strengths of the moiré and ML excitons correspond to radiative linewidths of hundreds of microelectrons volts and radiative decay rates of hundreds of femtoseconds. Therefore, the measured linewidth of both types of exciton is still dominated by inhomogeneous broadening. The measured photoluminescence decay times are a few picoseconds for both moiré and ML excitons (Extended Data Fig. 6), which are consistent with the expected very short radiative lifetime and are lengthened owing to the energy relaxation dynamics. The moiré excitons have broader linewidths (half-width of 7–8 meV) than the ML excitons (about 2 meV), probably due to inhomogeneity in lattice alignment or strain distribution introduced during transfer and stacking of the two MLs.). The strong-coupling condition \( \Omega_{1,2} > (\gamma_1 + \gamma_{X1,2})/2 \) is satisfied up to 70 K, where \( \gamma_1 = 2.7 \) meV is the half-width of the cavity mode.

In comparison, a similar cavity enclosing an ML MoSe\(_2\) also exhibits clearly strong coupling of the ML MoSe\(_2\) A-exciton and the cavity photon (Fig. 2c, f). Fitting the dispersions of the polaritons yields the coupling strength of the A-exciton, \( \Omega_{X1} = 17.1 \pm 0.1 \) meV, which is comparable to, but slightly larger than, \( \sqrt{\gamma_1^2 + \gamma_{X1}^2} = 13.2 \) meV. This confirms that most of the oscillator strength of the ML MoSe\(_2\) A-exciton is distributed to the moiré states \( X_1 \) and \( X_2 \). The reduction may be due to additional, higher-energy moiré states or additional disorder introduced into the bilayer during stacking of the MLs.

### Zero-dimensionality of moiré excitons

Using the robust polariton modes formed with both moiré hBL and ML excitons, we study the effect of the underlying moiré lattice via their nonlinear response to the excitation density \( n \). We focus on \( n < n_{\text{Mott}} = 1/\alpha_0^2 = 10^5 \) \( \mu \text{m}^{-2} \), where \( n_{\text{Mott}} \) is the Mott density and \( \alpha_0 = 1 \) nm the Bohr radius, and vary \( n \) from 10 \( \mu \text{m}^{-2} \) up to 3 \( \times 10^4 \) \( \mu \text{m}^{-2} \) using a resonant 150-fs pulsed laser (see Methods for details).

A few representative reflectance spectra at different excitation densities are shown in Fig. 3a, b for the hBL and ML cavities, respectively. By fitting the absorption dips in the spectra with Lorentzian functions, we obtain the polarization energies plotted in Fig. 3c, d.

With increasing excitation densities, the moiré LPs and MPs shift symmetrically towards the moiré exciton resonance \( E_{X1,2} \) while their linewidths remain the same (Fig. 3a, c). These results suggest reduced exciton–photon coupling strength, yet constant exciton energy or dephasing, which are typical properties of zero-dimensional (0D) excitons. In sharp contrast, the ML LPs and MPs shift together to higher energies, accompanied by considerable linewidth broadening. These observations suggest a much weaker saturation of the exciton–photon coupling strength but pronounced many-body effects, as expected of two-dimensional (2D) excitons.

To analyse the density dependence quantitatively, we use the coupled-oscillator model to extract from the polariton spectra the three basic properties of excitons: the exciton energy \( E_X(n) \), linewidth \( \gamma_X(n) \) and photon coupling strength \( \Omega(n) \) (see Methods for details). The results can be compared with well established models for free 2D excitons:

\[
E_X(n) = E_X(0) + \beta n - \beta n^2,
\]

(2)

\[
\gamma_X(n) = \gamma_X(0) + \alpha n.
\]

(3)

\[
\Omega(n) = \Omega(0) \sqrt{1 + \frac{n}{n_s}}.
\]

(4)
These equations describe the energy shift due to two- and three-particle exchange interactions with coefficients $\beta_{1,2}$, the linewidth broadening due to exciton-induced dephasing with coefficient $\alpha$, and oscillator strength saturation due to Pauli blocking with a saturation density of $n_s$ (refs. 21,27).

Pronounced differences between moiré and ML excitons are clearly seen in all three properties (Fig. 4a–c). For the ML exciton, all three properties are described very well by equations (1)–(3) for 2D excitons (blue diamonds and lines in Fig. 4a–c). The exciton energy blueshifts by 1.5 meV owing to exchange interactions and the linewidth broadens considerably by 3 meV owing to exciton-induced dephasing. The fitted coefficients, $\beta_{1,ML} = (1.2 \pm 0.1) \times 10^{-1} \mu eV \mu m^2$, $\beta_{2,ML} = (2.9 \pm 0.4) \times 10^{-6} \mu eV \mu m^4$, and $\alpha = 0.11 \pm 0.01 \mu eV \mu m^2$, all agree well with reported values. The coupling strength decreases slightly by up to 5%; the corresponding polarization energies versus carrier density (logarithmic scale) obtained from a and b, respectively. The shaded areas show the range in the linear scale, plotted over the same ranges of the horizontal and vertical axes as the main figure. Solid lines are calculations using parameters obtained from fitting the data in Fig. 4a–c. The error bars on the energy data correspond to the 95% confidence interval of the Lorentzian fit.
Fig. 4 | Enhanced nonlinearity by moiré lattice confinement. a–c. Shift of exciton energy \( \Delta \), half linewidth \( \gamma \), and normalized coupling strength \( 2\Omega /\gamma \) of the hBL LPs (red) and ML LPs (blue) as a function of carrier density, obtained from the data in Fig. 3. The hBL \( \Delta_{\text{exc}} \), and \( \gamma_{\text{exc}} \) (red circles in a, b) are approximately constant. The ML \( \Delta_{\text{exc}} \), and \( \gamma_{\text{exc}} \) (blue diamonds in a, b) are fitted by a second-order polynomial and a linear fit, respectively (black solid lines in a, b). In c, the black solid lines are fits with equation (4) with a constant \( n_0 \) for the moiré excitons at \( n > 1,000 \mu m^{-2} \) and for ML excitons. The black dashed line is a fit with a density-dependent effective saturation density \( n_{\text{sat,eff}} \), and is used to calculate the polariton energies in Fig. 3c. d. Real-space distribution of the interlayer component of the X state with a linear colour scale. The white lines mark a moiré unit cell. The scale bar is 1 nm. e. Measured (symbols) and fitted (lines) nonlinear coefficient, \( g \), versus carrier density for the moiré hBL LPs (red) and ML LPs (blue). The magenta dashed line and blue solid line are the calculations using the fitted polariton energies in Fig. 3c. d. The hBL LP is at zero detuning; the red solid line and magenta dashed lines correspond to fitted \( \Omega_{\text{sat}} \) using a constant and effective saturation density, \( n_0 \) and \( n_{\text{sat,eff}} \) respectively, in equation (3). For the ML LP, the blue solid and dashed lines correspond to the measured detuning and zero detuning, respectively. The error bars in a–c and density in e are explained in Methods. The error bars of \( g \) correspond to the 95% confidence interval of the fit using \( g(n) = |dE(n)/dn| \).

\[ n_{\text{ML}} = (2.8 \pm 0.4) \times 10^4 \mu m^{-2} \] is comparable to \( 1/\alpha_0^2 = 10^5 \mu m^{-2} \). These results confirm that the ML excitons behave as 2D particles; they also provide a consistency check of our density calibration.

In contrast to the ML excitons, the moiré excitons show no measurable energy shift, a much smaller line-broadening of <1 meV and a much stronger saturation of the coupling strength by up to 20% (Fig. 4a–c, red circles). These cannot be explained with the 2D exciton picture, but are expected of OD excitons, as we discuss next.

In a moiré lattice, the exciton centre-of-mass wavefunction is no longer a plane wave, but becomes localized near the potential minimum of each moiré cell, with a localization length \( l = 1/\alpha_0 \) (where \( \alpha_0 \) is the moiré period), as illustrated in Fig. 4d. Evaluating \( l \) for our device of \( \alpha_0 = 4.2 \text{ nm} \) yields \( l = 1.2 \text{ nm} \). Therefore, the confinement leads to an increase of the effective local density \( n_{\text{eff}} \) by (\( \alpha_0^2/2 \)) \( = 3 \) at the potential minima, and thus to enhanced exchange and dipole–dipole interactions. If the moiré excitons are 2D-like band excitons, then the enhanced interactions should lead to a correspondingly enhanced energy shift and enhanced exciton-induced dephasing. By contrast, we observe no energy shift and a much smaller line-broadening. Therefore, our observations show that the moiré excitons in our device are no longer 2D band excitons.

On the other hand, the suppressed energy shift and linewidth broadening are characteristic of quantum dots with strong exciton blockade. Exciton blockade takes place when the interaction energy between two excitons in the same cell becomes greater than the exciton linewidth, so that multiple excitations correspond to distinct, quantized energy levels. In our hBL, the on-site exchange interaction is \( U_{\text{exc}} = \frac{1}{2} \langle \frac{\alpha_0}{\omega} \rangle E_0^2 = 40 \text{ meV} \) for a binding energy of \( E_0 = 250 \text{ meV} \), and the exciton wavefunction extension is \( \omega = 1 \text{ nm} \). The dipole interaction due to the interlayer component is \( U_{\text{intra}} = \frac{1}{2} \Delta_{\text{exc}} E_0 = 30 \text{ meV} \) for a dipole length of \( d = 0.7 \text{ nm} \) (ref. 2). These values agree with a more detailed calculation (Extended Data Fig. 9) and are much larger than the exciton full linewidth of \( 2\gamma_{\text{exc}} = 15 \text{ meV} \). Therefore, we expect exciton blockade in a moiré cell. At the same time, because both the exchange and dipole–dipole interactions decrease quickly with distance, they are both suppressed for excitons in different moiré cells (Extended Data Fig. 9). The intra-cell exciton blockade, together with suppressed inter-cell interactions, leads to suppression of many-body effects for the single-exciton resonance, which manifests as an absence of an energy shift or exciton-induced dephasing, in agreement with our observations.

Consistent with exciton blockade, exciton–phonon coupling saturates at one exciton per moiré cell, \( n_{\text{sat}} = \gamma_{\text{exc}}^{-1}/\alpha_0^2 = \gamma_{\text{exc}}^{-1} \approx 0.8 \times 10^4 \mu m^{-2} \) for \( n > 1,000 \mu m^{-2} \) (bottom solid line in Fig. 4c). By contrast, if the moiré excitons are 2D band excitons, \( n_0 \) would have remained the same as \( n_{\text{sat}} \), because the total oscillator strength is conserved in the band across the lattice. We note that the fit does not explain the anomalously strong saturation at very low densities of \( n < 1,000 \mu m^{-2} \), which we will discuss later.

Moiré lattice-induced nonlinearity of moiré polaritons

The distinctly different density dependence of the moiré polaritons from that of ML exciton polaritons, as discussed above, suggests the possibility of achieving a much higher polariton nonlinearity, even for polariton modes that are stable at high temperatures. Figure 4e shows the measured nonlinear coefficient \( g(n) = |dE(n)/dn| \) for both the moiré and ML LPs (symbols).

Whereas the nonlinearity increases with decreasing density for both moiré and ML LPs, the moiré LPs show a surprisingly larger nonlinearity. The nonlinearity of ML LPs originates primarily from exciton-exchange interactions and \( g_{\text{ML-IP}} \) Saturates below \( n < 1,000 \mu m^{-2} \) to 0.02 \text{ eV} \text{ nm}^2 \text{ (0.04 eV \text{ m}^2) at the measured (zero) detuning, in agreement with reported values}^{21,25,26}. The nonlinearity of moiré LPs results primarily from exciton blockade. On the basis of the data at \( n > 1,000 \mu m^{-2} \), we obtain a \( g_{\text{ML-IP}} \) value about four times higher than \( g_{\text{ML-IP}} \).

At very low densities of \( n < 1,000 \mu m^{-2} \), whereas the ML polaritons or excitons show no measurable shift, the moiré polaritons show clearly saturation-induced shifts down to \( n = 10 \mu m^{-2} \) (Fig. 3c), corresponding to a stronger exciton saturation than that expected from exciton blockade, exciton interactions, or effects of trions and defect states (Fig. 4c). (The strong saturation observed at very low excitation densities cannot be explained by trions or states in deeper trapping potentials. These states would have a small initial oscillator strength and lower resonance energy, so the increase in saturation density would have been accompanied by an increase of the coupling strength and exciton energy, contradicting our observations.) However, this abnormally large nonlinearity is repeatable over multiple measurements in multiple devices (see Extended Data Fig. 7 for another example), suggesting hidden mechanisms for large polariton nonlinearity in moiré lattices. Phenomenologically, \( \Delta_{\text{exc}}(n) \) can be described very well over the full density range by equation (3) if \( n_0 \) is replaced by a density-dependent effective saturation density of \( n_{\text{sat,eff}} = \langle n_{\text{sat}}(1 - A n - B n^2) \rangle \) for fitted \( A = 4.4 \pm 0.1 \times 10^{-3} \mu m^{-2} \), \( B = 0.98 \pm 0.04 \) and \( B = (7.4 \pm 0.5) \mu m^{-2} \) (Fig. 4c).

The polariton nonlinearity is a key figure of merit that distinguishes polariton systems from pure photon systems. Together with the robust coherence enforced by the photon component, it gives rise to a wide range of novel nonlinear many-body and quantum phenomena.\textsuperscript{29–34}
For polaritons made of 2D excitons, however, a larger $g$ is obtained only at the expense of $\Omega$ or of the stability of polariton modes. This is because the exciton-exchange interaction, $g_{\text{exc}}$, and the exciton–photon coupling strength saturation, $g_{\text{sat}}$, are both reduced with the reduced mass of the exciton, $\mu g_{\text{exc}} = E_a q^2 = 1/\mu$ and $g_{\text{sat}} = \alpha_0^2 = 1/\mu^2$. However, the exciton–photon coupling strength is $\Omega \propto \sqrt{f} \approx 1/\Omega_0 \propto \mu$. The highest polariton nonlinearity is achieved in single- or few-quantum-well GaAs microcavities$^{22,33}$, with $g \approx 3 \mu eV \mu m^2$ and $\Omega$ of only ~3 meV. Wide-bandgap semiconductors, organic crystals and ML TMDs all feature greater $\Omega$ and high-temperature polaritons, but a much weaker polariton nonlinearity$^{22,25,35,36}$. Higher-order excitations in TMDs, such as trions with strong band-filling effect$^{23,28}$ and 2$s$ excitons with a larger Bohr radius$^{34}$, have shown enhanced nonlinearity, but again at the compromise of stability.

The moiré polariton system uniquely combines strong nonlinearity, due to quantum confinement of excitons within each moiré cell, and a large total photon coupling strength, due to collective coupling among cells. It thereby provides a new route to achieving strong nonlinearities simultaneously with robust exciton–photon coupling.

In summary, we have demonstrated a polariton system formed via collective coupling of a 2D lattice of 0D excitons with light in a microcavity. The system introduces quantum dot-like nonlinearity into a cooperatively coupled solid-state system, opening a door to novel quantum many-body physics and polariton devices$^{26}$. Polariton blockade and strongly correlated polariton gases may become possible with reduced inhomogeneous broadening of moiré excitons, improved cavity quality factors and a better understanding of the abnormal enhancement of the polariton nonlinearity at very low excitation densities. Electrical gating and electrical field tuning of the hBL can be implemented to further control the nonlinearity and many-body phenomena in the moiré polariton system$^{35}$.

### 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/s41586-021-03228-5.

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**Article**

**Methods**

**Sample fabrication**
The ML MoSe₂, WS₂, and few-layer hBN flakes are obtained by mechanical exfoliation from bulk crystals. A polyethylene terephthalate (PET) stamp is used to pick up the top hBN, WS₂, ML, MoSe₂, ML, and the bottom hBN under a microscope. The WS₂ and MoSe₂ MLs are rotationally aligned to about 0° in the hBL. The bottom half of the cavity consists of a distributed Bragg reflector (DBR; 16 pairs of SiO₂/TiO₂) and a 1/4 SiO₂ layer. The heterostructure on the PET stamp is stamped onto the bottom half of the cavity and the PET is dissolved in dichloromethane for 6 h at room temperature. Then, a 67 nm polyethylene methylacrylate (PMMA) film is spin-coated on the top of hBL to form the second half of the h/2 cavity. Subsequently, a 40 nm thick silver film is deposited using electron-beam evaporation as the top mirror of the cavity.

**Optical measurements**
For low-temperature measurements, the sample was kept in a 4-K cryostat (Montana Instrument). The excitation and collection were carried out with a home-built confocal microscope with an objective lens with a numerical aperture of NA = 0.42. To characterize the dispersion of the polariton device, we performed angle-resolved reflection measurements using white light from a tungsten halogen lamp. The average power of the laser is P. The laser profile can be fitted with the Gaussian function

\[
G(E) = \frac{A_{\text{laser}} \cdot e^{-\frac{2(\frac{E - E_{\text{laser}}}{\gamma_{\text{laser}}})^2}{\nu_{\text{FWHM}}^2}}}{\nu_{\text{FWHM}}},
\]

where \(A_{\text{laser}}\) is the area of the Gaussian function and \(\nu_{\text{FWHM}}\) is the linewidth. The power absorbed by the LP and MP, \(P_{LP,MP}\), can be calculated using the convolution

\[
P_{LP,MP} = P \int G(E) \text{Absor}_{LP,MP}(E) dE.
\]

The total carrier density, including both LPs and MPs, created per pulse and its error \(\delta n\) can be calculated by

\[
n = \frac{P_{LP} + P_{MP}}{A_{\text{beam}}},
\]

\[
\delta n = \sqrt{\left(\frac{\delta P_{LP}}{A_{\text{beam}}}\right)^2 + \left(\frac{\delta P_{MP}}{A_{\text{beam}}}\right)^2},
\]

where \(A_{\text{beam}} = 1.5 \mu m\) is the beam area and \(\delta P_{LP,MP} = P_{LP,MP} \times \delta n\).

**Theory of moiré excitons**
The Hamiltonian for the moiré excitons is

\[
H = \frac{\hbar^2 k^2}{2M_X} \left(\omega_1 e^{i(b_{-x} r - e^{-i b_{-x} r})} + \omega_2 e^{i(b_{-y} r + e^{-i b_{-y} r})} \right) + \frac{\hbar^2 (k - k_0)^2}{2M_X} \delta_0 + \hbar^2 \delta \omega_0 \left[\frac{1}{2} \frac{3 \omega_0}{\sqrt{\delta_0}} \left(\frac{3}{2} \omega_0 \right)\right].
\]

where \(E_c\) is an energy constant, \(M_X\) and \(M_\delta\) are the effective masses of intralayer and interlayer excitations, respectively, \(x = (0, 4\pi/(3a_{WS})), \alpha = (a_{MoSe2} + a_{WS2})/2 = 3.26 \AA, \delta = \left|a_{MoSe2} - a_{WS2}\right| / a_0 = 0.82\text{ meV}, \text{ and } M_\delta = 0.71 m_e\text{ where } m_e\text{ is the bare electron mass. The energy spectrum of the moiré excitons is obtained by diagonalizing the moiré Hamiltonian }H\text{ using plane-wave expansion, as shown in Fig. 1b. For the lowest-energy exciton }X_0, \text{ the interlayer component is plotted in Fig. 4d, which shows spatial localization.}

**Polariton density calibration**
In this section, we use the bilayer device data as an example; the same procedure applies to the monolayer device. To extract the polariton density, first, we fit the reflection spectra in Fig. 3 using

\[
R = 1 - \text{Absor}_{LP} - \text{Absor}_{MP}.
\]

where \(\text{Absor}_{LP}\) and \(\text{Absor}_{MP}\) represent absorption by the LP and MP in the bilayer, respectively, which are described by the Lorentzian functions

\[
\text{Absor}_{LP,MP}(E) = \frac{A_{LP,MP}}{(E - E_{LP,MP})^2 + \gamma_{LP,MP}^2}.
\]

The resonance energy \(E_{LP,MP}\), linewidth (half-width at half-maximum) \(\gamma_{LP,MP}\) and absorption amplitude \(A_{LP,MP}\) are fitting parameters with uncertainties \(\delta E_{LP,MP}, \delta \gamma_{LP,MP}, \delta A_{LP,MP}\), respectively, corresponding to 95% confidence intervals.

We used pulsed laser with a pulse duration of 150 fs and repetition rate of \(f = 80 \text{ MHz}\). We calculate the polariton density \(n\) injected per pulse. The average power of the laser is \(P\). The laser profile can be fitted with the Gaussian function

\[
G(E) = \frac{A_{\text{laser}} \cdot e^{-\frac{2(\frac{E - E_{\text{laser}}}{\gamma_{\text{laser}}})^2}{\nu_{\text{FWHM}}^2}}}{\nu_{\text{FWHM}}},
\]

where \(A_{\text{laser}}\) is the area of the Gaussian function and \(\nu_{\text{FWHM}}\) is the linewidth. The power absorbed by the LP and MP, \(P_{LP,MP}\), can be calculated using the convolution

\[
P_{LP,MP} = P \int G(E) \text{Absor}_{LP,MP}(E) dE.
\]

The total carrier density, including both LPs and MPs, created per pulse and its error \(\delta n\) can be calculated by

\[
n = \frac{P_{LP} + P_{MP}}{A_{\text{beam}}},
\]

\[
\delta n = \sqrt{\left(\frac{\delta P_{LP}}{A_{\text{beam}}}\right)^2 + \left(\frac{\delta P_{MP}}{A_{\text{beam}}}\right)^2},
\]

where \(A_{\text{beam}} = 1.5 \mu m\) is the beam area and \(\delta P_{LP,MP} = P_{LP,MP} \times \delta n\).

**Extraction of exciton energy, linewidth and coupling strength at different densities**
To analyse the results quantitatively, we extract the density dependence of the exciton properties from the measured polariton spectra. In the hBL cavity, we focus on the MP and LP modes and neglect changes caused by \(X_2\), given that \(X_2\) is at a much higher energy and its change affects MPs and LPs only negligibly (see Extended Data Fig. 10 for details). The cavity resonance energy \(E_c\) and linewidth \(\gamma\) are assumed to change negligibly with exciton density. Using the two-coupled oscillator mode, the energies of the MP and LPs of the hBL cavity, as well as the LPs and MPs of the ML cavity, can be extracted. In the following, we use the bilayer device data as an example; the same procedure applies to the monolayer device.

\[
E_{LP,MP}(n) = \frac{1}{2} \left[\left|X_0(n)\right| + \sqrt{\left|X_0(n)\right|^2 + \frac{1}{4} \left(\left|E_{\text{hBN}}(n) - E_{\text{hBN}}(n)\right| + \left|\gamma_\phi(n) - \gamma_\phi(n)\right|\right)^2}\right].
\]

Here the cavity resonance energy \(E_{\text{hBN}}\), and cavity half-linewidth, \(\gamma_\phi\), do not change with carrier density. Therefore, from the measured polariton energies and half-linewidth, \(E_{LP,MP}\), we obtain the density dependence of the exciton energy, \(E_X\), half-linewidth, \(\gamma_X\), and exciton–photon coupling strength, \(\Omega\).
The exciton energy $E_x$ and its uncertainty $\delta E_x$ are

$$E_x(n) = E_{LP}(n) + E_{MB}(n) - E_c,$$

$$\delta E_x = \sqrt{(\delta E_{LP})^2 + (\delta E_{MB})^2 + (\delta E_c)^2},$$

where $E_c$ and $\delta E_x$ are the cavity resonance energy and its uncertainty, respectively, obtained by fitting the angle-resolved reflection spectrum in Fig. 3, which do not change with polariton density.

The exciton linewidth $\gamma_x$ and its uncertainty $\delta \gamma_x$ are

$$\gamma_x(n) = \gamma_{LP}(n) + \gamma_{MB}(n) - \gamma_c,$$

$$\delta \gamma_x = \sqrt{(\delta \gamma_{LP})^2 + (\delta \gamma_{MB})^2 + (\delta \gamma_c)^2},$$

where $\gamma_c$ and $\delta \gamma_x$ are the cavity linewidth and its uncertainty, respectively, obtained by fitting the reflection spectrum of the bare cavity in Fig. 1d, which do not change with polariton density.

The coupling strength $\Omega$ is

$$\Omega(n) = \frac{1}{2} \times$$

$$\sqrt{[E_{LP}(n) - E_{MB}(n) + i|\gamma_{LP}(n) - \gamma_{MB}(n)|]^2 - [E_x(n) - E_c + i|\gamma_x(n) - \gamma_c|]^2.}$$

### Dipole–dipole interaction

The interlayer component of moiré excitons contributes to dipole–dipole interactions. The wave function for the interlayer component of a moiré exciton localized near the potential minimum around the origin can be described as

$$W(r_1, r_2) = x_{ix} \Phi \left(\frac{r_1 + r_2}{2}\right) \phi(r_1 - r_2).$$

where $x_{ix}$ is the interlayer component weight and $\Phi$ and $\phi$ are the centre-of-mass and relative-mass wave functions, respectively. We approximate $\Phi$ and $\phi$ by the Gaussians $\Phi(R) = \frac{1}{\sqrt{\pi}} e^{-R^2/2}$ and $\phi(r) = \frac{1}{\sqrt{\pi} a} e^{-2r/a}$, where $R = \frac{r_1 + r_2}{2}$, $r = r_1 - r_2$ and the localization length $l$ can be estimated using the exciton state shown in Fig. 4d.

The dipole–dipole interaction between two excitons localized at different moiré sites can be approximated as

$$U_{dd}(L) = \int dr_1 \int dr_2 \int dr_3 \int dr_4 |W(r_1, r_2)W(r_3, r_4)|^2 [V_r(r_1 - r_3) + V_L(r_2 - r_4) - V_r(r_1 - r_2) - V_L(r_3 - r_4)]$$

$$= 2\chi^2 \int \frac{d^2 q}{(2\pi)^2} \frac{e^{-q^2/2}}{(1 + \frac{1}{4\pi^2 q^2})} e^{-i q \cdot L},$$

where $L$ is a moiré lattice vector, $V_r(r) = e^2/(2r^2)$ is the intralayer Coulomb interaction, $V_L(r) = e^2/(2(r^2 + d^2))$ is the interlayer Coulomb interaction with $d$ the interlayer distance, and $V'(q) = \frac{2e^2}{\pi q^2}(1 - e^{-q^2})$. To calculate $U_{dd}(L)$, we take $\chi = 1/\sqrt{2}$, $d = 0.65$ nm, $c = 5$ and $a_0 = 1$ nm.

Extended Data Fig. 9 shows the onsite and nearest-neighbour dipole–dipole interactions as a function of twist angle. The onsite repulsion is sizable (~24 meV for a twist angle of 56.3°), whereas the offsite repulsion is negligible.

### Data availability

The data that support the findings of this study are available from the corresponding author upon request.

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**Author contributions** H.D. and L.Z. conceived the experiment. L.Z. performed the measurements. F.W. provided theoretical models. L.Z. and Z.Z. fabricated the device. L.Z. and H.D. performed data analysis. S.H. deposited the silver mirror. Y.-H.C. grew the bottom DBR mirror. K.W. and T.T. grew hBN single crystals. H.D. and S.R.F. supervised the projects. L.Z. and H.D. wrote the paper with inputs from other authors. All authors discussed the results, data analysis and the paper.

**Competing interests** The authors declare no competing interests.

**Additional information**

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Extended Data Fig. 1 | hBL twist angle. Twist angle of the hBL described in the main text, measured by second-harmonic generation (SHG) spectroscopy. 

**a.** Polarization-dependent (SHG) signal measured on the ML WS$_2$ (green open circles) and MoSe$_2$ (blue filled squares) regions of the hBL, and the corresponding fits with sinusoidal functions (green and blue solid lines).

**b.** The SHG signal from the ML WS$_2$, ML MoSe$_2$, and hBL regions, measured with the same experimental configurations. The suppressed SHG signal from the hBL, resulting from destructive interference, indicates that the stacking order is H-stacking. The twist angle is determined to be $56.5^\circ \pm 0.8^\circ$. 
Extended Data Fig. 2 | Temperature dependence of moiré exciton. Temperature dependence of the moiré excitons $X_1$ and $X_2$ measured from a separate hBL prepared on a sapphire substrate. The black dashed lines are guides for the eyes. The two exciton states can be well resolved up to 200 K, which could exclude charged excitons or excitons trapped by defects.
Extended Data Fig. 3 | Schematic of the device. a. Schematic of the device showing the different layers of a heterostructure embedded inside a microcavity that consists of a bottom DBR and a top silver mirror. b. Microscope image of the hBL on top of the DBR mirror, taken before depositing the PMMA layer and the silver mirror. c. Thickness of each layer of the device.
Extended Data Fig. 4 | Photoluminescence from the moiré polariton. Left, angle-resolved photoluminescence spectrum of an hBL in a cavity, excited by a continuous-wave laser with energy of 2.3 eV and power of 50 μW. To enhance the visibility of states at higher energy, the emission intensity above 1.607 eV is magnified five times. Right, simulated angle-resolved absorption, which agrees well with the measurement.
Extended Data Fig. 5 | Transition from strong coupling to weak coupling driven by thermal broadening. a, Transition from strong coupling to weak coupling, measured from the temperature dependence of $\Omega_1$ (red open circles) and ($\gamma_c + \gamma_{X1})/2$ (purple triangles) (a) and $\Omega_2$ (red open circles) and ($\gamma_c + \gamma_{X2})/2$ (purple triangles) (b). $\gamma_c = 2.7$ meV is constant with temperature. $\Omega_1$ and $\Omega_2$ drop below the average linewidth at about 100 K, showing the transition to the weak-coupling regime. $\Omega_1$ and $\Omega_2$ are extracted by fitting the angle-resolved white-light reflection spectra with equation (1) and the error bars correspond to the 95% confidence interval. $\gamma_{X1}$ and $\gamma_{X2}$ are measured independently from the bare hBL, and the error bars correspond to the 95% confidence interval of the Lorentzian fit.
Extended Data Fig. 6 | Time-resolved photoluminescence of ML exciton, ML trion, hBL excitons and hBL trion. 

**a, b**, Time-resolved photoluminescence spectra measured by a streak camera for ML MoSe$_2$ (**a**) and hBL WS$_2$–MoSe$_2$ (**b**). (The hBL data are collected from a sample different from that discussed in the main text, which is not integrated with the microcavity.) The different resonances are labelled with white arrows, including the ML exciton (ML-X), ML trion (ML-T), moiré exciton at higher energy (hBL-X$_2$), moiré exciton at lower energy (hBL-X$_1$) and moiré trion (hBL-T).

**c, d**, Time-resolved decay of ML-X (**c**) and hBL-X$_1$ (**d**), obtained by integrating the spectrum in the range labelled by the red rectangles in **a, b**. The red solid lines in **c, d** are fits with a single exponential decay function. The photoluminescence decay time for ML-X and hBL-X$_1$ is 6.7 ps and 8.0 ps, respectively.
Extended Data Fig. 7 | Strong nonlinearity measured in another device.
a, Angle-resolved white-light reflection spectra taken at 5 K on the second sample. White solid lines are fits using the coupled-oscillator model. Dashed white lines are fitted energies of the uncoupled cavity photon and excitons. 
b, Power-dependent reflection spectra for the lower polariton (left) and middle polariton (right). c, Shift of polariton energies versus carrier density (logarithmic scale) obtained from b. d, Extracted nonlinear coefficients for lower polariton (red circles) and the calculations using fitted polariton energies (solid line). The error bars on the energy data correspond to the 95% confidence interval of the Lorentzian fit. The error bars of g correspond to the 95% confidence interval of the fit using \( g(n) = |\frac{dE(n)}{dn}| \).
Extended Data Fig. 8 | Profile of the laser used for nonlinearity characterization. Profile of the pulsed laser (red dot) used for the nonlinearity measurement of moiré polaritons, compared with the moiré excitons X₁ and X₂ (blue dots) and moiré polaritons (black dots). The laser has a negligibly small tail on X₂ and UP.
Extended Data Fig. 9 | Dipole–dipole interaction strength as a function of twist angle $\theta$. $U_{dd}^{(0)}$ and $U_{dd}^{(1)}$ are the onsite and nearest-neighbour interaction strength, respectively.
Extended Data Fig. 10 | Effect of moiré exciton X₂ on the nonlinearity of polaritons. Calculated shifts of hBL LPs and hBL MPs as a function of $\Omega^2$. When $\Omega^2$ changes by 10%, hBL LPs and hBL MPs shift by only 0.16 meV on average, which is negligible compared to the shift induced by exciton $X_1$ (up to 2 meV; Fig. 3c).