Highly nonlinear dipolar exciton-polaritons in bilayer MoS₂

Realizing nonlinear optical response in the low photon density limit in solid-state systems has been a long-standing challenge. Semiconductor microcavities in the strong coupling regime hosting exciton-polaritons have emerged as attractive candidates in this context. However, the weak interaction between these quasiparticles has been a hurdle in this quest. Dipolar excitons provide an attractive strategy to overcome this limitation but are often hindered by their weak oscillator strength. The interlayer dipolar excitons in naturally occurring homobilayer MoS₂ alleviates this issue owing to their formation via hybridization of interlayer charge transfer exciton with intralayer B exciton. Here we demonstrate the formation of dipolar exciton polaritons in bilayer MoS₂ resulting in unprecedented nonlinear interaction strengths. A ten-fold increase in nonlinearity is observed for the interlayer dipolar excitons compared to the conventional A excitons. These highly nonlinear dipolar polaritons will likely be a frontrunner in the quest for solid-state quantum nonlinear devices.

Photons are becoming ubiquitous in emerging quantum technologies like quantum communication and metrology due to the ability to propagate long distances while being robust against decoherence. To further extend the span of their utilities, platforms to achieve and implement strongly interacting photons in solid-state systems are highly desirable. Conventional materials do not exhibit nonlinear response at power levels associated with single photons. In this context, remarkable advances have been made in cold atom systems to realize interacting photons at the single-particle level. A strong contender for the generation of strongly interacting photons in the solid-state are exciton-polaritons formed via non-perturbative coupling of cavity photons with excitonic resonances. Although they can be modeled as non-interacting quantum fluids at low densities, beyond a critical density, saturation and short-range exchange interactions become significant and give rise to various phenomena like the appearance of spontaneous coherence, parametric down-conversion, and superfluidity. Strong spatial confinement along with Coulomb interactions give rise to even stronger correlations between polaritons that can no longer be described using a mean-field theory. Such interactions give rise to non-Poissonian statistics of laser transmission, dubbed as polariton blockade. Preliminary evidence of such non-classical correlation was recently observed in confined polariton systems in GaAs but a small ratio of interaction to dissipation rates resulted in only a weak violation of classical correlations.

Transition metal dichalcogenides (TMDCs) have garnered much attention for their exceptional optoelectronic properties and have been used to demonstrate a wide array of fundamental phenomena and technological applications. Furthermore, their two-dimensional (2D) nature allows for the formation of heterojunctions or homojunctions with arbitrary twist angles resulting in emergent properties. Their strong binding energy and large oscillator strength render them capable of forming polaritons at room temperature, which has also been shown to retain the intriguing properties of the 2D excitons. Polariton interactions in TMDCs
under different configurations are being studied extensively. Realization of Fermi polarons, trion polaritons, and excited-state Rydberg exciton-polaritons have shown great potential in their abilities to harness strong polariton interactions. Moiré exciton-polaritons were recently demonstrated using heterobilayers of WS₂/MoSe₂ where the electronic confinement potential arising from the twisted heterostructure was shown to enhance the nonlinearity. However, all the platforms above rely on short-range exchange interactions or phase-space filling that pose a bottleneck in realizing few polariton nonlinearity under current experimental capabilities.

In this context, a very attractive possibility is the use of spatially indirect interlayer excitons (IE) in TMDC heterostructures that can have a permanent dipole moment and hence support highly interacting dipolar polaritons. Formation of polaritons with excitons that possess a permanent dipole moment has been shown to enhance the polariton interactions both in resonant and non-resonant excitation schemes. However, their inherently low oscillator strength creates an impediment in reaching the strong coupling regime without hybridization with a direct intralayer exciton. Furthermore, the large in-built interfacial electric fields make electrical tuning of the energies of the IE far more difficult. Bilayer MoS₂ provides a highly attractive platform to solve these issues. The IE in naturally stacked 2H bilayer MoS₂ remarkably has both an out-of-plane (static) dipole moment and an in-plane (oscillating) dipole moment. Due to the in-plane dipole moment, the IE in MoS₂ bilayer has an oscillator strength of approximately 36% of that of the intralayer A exciton along with strong absorption that is visible up to room temperatures. Moreover, a strong response to DC electric fields has been demonstrated in these systems, thus confirming their dipolar nature.

In this work, we achieve strong coupling of microcavity photons with the IEs (along with intralayer A and B excitons) in bilayer MoS₂. The IE polariton shows 10-fold enhancement of the polariton nonlinearity compared to the intralayer A exciton owing to its dipolar and phase space filling contributions. Such enhanced nonlinear response makes them appealing to realize strongly interacting polaritons in condensed matter systems. In addition, the ease of fabrication and realization of multi-polariton species in this system makes it a practical and fundamentally interesting material for studying polariton physics.

**Results**

**Interlayer exciton nonlinearity**

Figure 1 shows a schematic of the bands that form interlayer exciton (IE) in bilayer MoS₂, IE₁ and IE₂ excitons in which electrons are in layer 1, and layer 2 respectively, are energetically degenerate at zero external bias while the hole is delocalized among both layers. The black arrows in Fig. 1a indicate the transitions that form the IE. This peculiar charge distribution is at the heart of producing both in-plane and out-of-plane dipole moments of IE. Figure 1b shows the charge distribution in real space for IE₁ and IE₂. Owing to the spatial separation of the electron and the hole, it acquires a permanent dipole moment, the nature of which has been a topic of much interest recently. IEs in MoS₂ homobilayer can be thought of as an admixture of B intralayer exciton with an optically dark but electric field tunable charge-transfer exciton, which is typically found in TMDC heterobilayers. As a result, IEs in MoS₂ homobilayer acquire both a strong oscillator strength and electric field tunability. Figure 1c shows the white light differential reflection of the bilayer sample at 7 K under weak illumination. The peaks at 1.9365 eV, 2.000 eV, and 2.1076 eV correspond to the A, IE, and B excitons, respectively. Interestingly, the absorption of the IE is prominent even at room temperature, which provides a straightforward method to identify the MoS₂ bilayer flakes after mechanical exfoliation (see SI Fig. S3).

We probe the nonlinear optical response of the IE and A exciton using resonant pump experiments (see Methods). The relative shift of A exciton and IE so obtained are plotted in Fig. 1d against the estimated density of A exciton and IE, respectively (see Methods for the details of the exciton density calculation). Both the excitons blueshift with increased excitation density, with the IE shift being more pronounced. Exchange interaction is the dominant mechanism responsible for the blue shift for the A exciton, while dipolar interactions dominate the interaction in the IE response as exchange interaction is likely to be small given the larger spatial separation of electron and holes. To better understand the exciton dynamics, we have also performed ultrafast pump-probe spectroscopy on the bilayers (see SI section X for the pump-probe data). In this case, the spectra show a redshift of the resonances accompanied by a nearly identical transient response regardless of which exciton is first pumped (A, IE or B). This can be
understood if carriers rapidly relax out of the K valley to the band minima on a scale faster than the ~200 fs time resolution. This supports our hypothesis that the A and IE transitions are nearly homogeneously broadened with a lifetime dictated by the intervalley scattering rate. This agrees well with recent coherent spectroscopy and numerical calculations performed for bilayers of MoSe231. The above approach is used to estimate the exciton density in Fig. 1d.

Strong coupling of interlayer excitons
Exciton polaritons were realized in the bilayer MoS2 by embedding it in a microcavity. Figure 2a shows the schematic of the structure used in this experiment. It consists of a bilayer MoS2 encapsulated by thin (20 nm) hexagonal boron nitride (hBN), which is sandwiched between a bottom dielectric distributed Bragg reflector (DBR) mirror and a top silver mirror. See SI Note I and Methods for the fabrication details and cavity structure. The cavity is designed such that the bilayer MoS2 flake sits close to an electric field anti-node, allowing us to observe five dispersive modes associated with the different polaritonic states as shown in Fig. 2b. These distinct polariton modes arise due to the hybridization of the cavity photon mode with the various excitonic states present in the bilayer system. We name the polariton branches as pol-1 through pol-5, with pol-1 corresponding to the lowest energy and pol-5 the highest. From the Fig. 2b, we see that the is state of A, the IE, B exciton and 2s Rydberg state of A exciton all strongly couple to the same cavity mode at 7 K. We fit the data with a five-coupled oscillator model where the energy of all the four excitons, the cavity mode, their Rabi splitting, and the effective refractive index of the system are treated as free parameters. The fit results in exciton energies \( E_{A1s} = 1.9323 \pm 0.0003 \text{ eV} \), \( E_{IE} = 2.0014 \pm 0.0001 \text{ eV} \), \( E_{A2s} = 2.078 \pm 0.002 \text{ eV} \), and the Rabi splittings \( \Omega_{A1s} = 40.4 \pm 0.3 \text{ meV} \), \( \Omega_{IE} = 21.4 \pm 0.1 \text{ meV} \), \( \Omega_{A2s} = 13 \pm 0.5 \text{ meV} \), \( \Omega_{IE} = 51 \pm 0.4 \text{ meV} \). The exciton energies obtained through the fit agree well with the experimentally determined exciton energies with slight shifts due to the strain and changes in the dielectric environment in the cavity. In our device, the bare cavity mode is positively detuned from the \( A_{1s} \) exciton by \( \delta = A_{1s} - A < 17 \text{ meV} \). See SI Note II for details on the coupled oscillator model. Figure 2c shows the line cuts of the full k-space data at three different \( k_y \) where the cavity mode is resonant with the A exciton (yellow), IE (red) and B exciton (blue), respectively. We notice that the Rabi splitting of the IE polariton remains observable even at 77 K (see SI Fig. 53).

Nonlinearity of dipolar interlayer exciton polaritons
We measured the white light reflectivity at the IE and A resonance and monitor the energy shifts of the polariton branches, see SI Note V and SI Note VIII for the details of the experiment. Figure 3a shows the energy of the upper and lower branch of the IE polariton as a function of polariton density at the inplane wavevector corresponding to the zero-detuning \( k_y \) (where the cavity mode and exciton energy are degenerate). Two Lorentzian fits at each density is used to obtain the energy of the upper and lower polaritons. SI Note IV discusses the details of the polariton density calculation and SI Note V provides the raw data of the density dependent differential reflection of IE polariton. We can see from Fig. 3 that the lower branch of the IE polariton moves with pump power more than the upper branch. We also observe a simultaneous increase of the magnitude of zero-detuning \( k_y \) along with the reduction of Rabi splitting. This suggests the presence of both exciton energy renormalization and saturation effect. SI Note VII shows the density-dependent Rabi splitting of IE polariton. Taking the derivative of the \( E_{LP} \) with respect to density, we calculate the strength of the nonlinearity, \( g_{LP} \). Since the rate of blue shift saturates at high density (olive curve in Fig. 3a), \( g_{LP}^{IE} \) reduces with polariton density as seen in Fig. 3b. Note that at the lowest powers accessible in our measurement, the IE lower polariton branch already shows a power-dependent blueshift. On the other hand, within the accessible range of the polariton density in our experiment, \( g_{LP}^{IE} \) remains nearly a constant (see SI Note VIII for the blueshift data of the lower polariton branch of A exciton). The obtained strength of the non-linearity in the low density limit for IE and A exciton polaritons are \( g_{LP}^{IE} \approx -10(\pm 2) \text{ meV} \mu m^2 \) and \( g_{LP}^{A} \approx -10(\pm 0.2) \text{ meV} \mu m^2 \) respectively corresponding to 10 fold increase in the nonlinear response for the IE polaritons.

Discussion
We note that the \( g_{LP}^{IE} \) value is comparable to the previous report for A exciton-polariton in monolayer TMDCs30,31. This allows us to compare the nonlinearity of the IE in bilayer MoS2 with that of Rydberg A2s state reported in ref. 20 – we find that the nonlinear response of IE is even
The spectral signature of interlayer Rydberg states are also more susceptible to disorder due to their larger Bohr radius. On the other hand, the spectral signature of interlayer Rydberg states is more due to the combined effect of exciton-exciton interaction and saturation. The error bars in energy represent the uncertainty in determining the peak of the Lorentzian fit (polariton energy) to the reflection data. The energy $E$ (μeV) as a function of the energy of the polariton $\Delta E$ is defined as the local slope of the polariton energy vs. polariton density graph. $g_{LP}$ and $g_{IE}$ are calculated from the lower branch of A exciton and IE polariton, respectively. Note that the $g_{LP}$ of IE polariton -100 ± 2 μeV μm⁻² is almost 10 times larger than the $g_{LP}$ of A exciton polariton -10 ± 0.2 μeV μm⁻². All the power-dependent nonlinear measurements were carried out using a pulsed supercontinuum laser (20 ps pulsewidth) with proper bandpass filter in the input to excite only one polariton species. The polariton density corresponds to the specific branch being excited. The density error bars take into account the error in measuring power and determining the energy of the polariton and linewidth from the Lorentzian fits. The error bars in $g_{LP}$ consider the error in determining the density and the energy of the polariton.

Fig. 3 Comparison of nonlinearity of the IE polariton with a exciton polariton. (a) Energy of the lower and upper branch of the IE polariton as a function of the polariton density at zero detuning $k_0$. The movement of the lower branch is more than the upper branch due to the combined effect of exciton-exciton interaction and saturation. The error bars in energy represent the uncertainty in determining the peak of the Lorentzian fit (polariton energy) to the reflection data. The energy $E$ (μeV) as a function of the energy of the polariton $\Delta E$ is defined as the local slope of the polariton energy vs. polariton density graph. $g_{LP}$ and $g_{IE}$ are calculated from the lower branch of A exciton and IE polariton, respectively. Note that the $g_{LP}$ of IE polariton -100 ± 2 μeV μm⁻² is almost 10 times larger than the $g_{LP}$ of A exciton polariton -10 ± 0.2 μeV μm⁻². All the power-dependent nonlinear measurements were carried out using a pulsed supercontinuum laser (20 ps pulsewidth) with proper bandpass filter in the input to excite only one polariton species. The polariton density corresponds to the specific branch being excited. The density error bars take into account the error in measuring power and determining the energy of the polariton and linewidth from the Lorentzian fits. The error bars in $g_{LP}$ consider the error in determining the density and the energy of the polariton.

larger than the A₂s state by a factor of -2.7. More importantly, we would like to highlight a few critical differences between these two studies. The approximately 4 times enhancement in the $g_{LP}$ for A₂s state in ref. 20 is attributed to the enhanced phase-space filling due to a higher Bohr radius of 2s compared to 1s. However, this enhancement in nonlinearity comes at the cost of reduced oscillator strength. This is overcome in ref. 20 by stacking three layers of monolayer TMDC separated by thin hBN. There is, however, no clear experimental route towards using higher-order Rydberg states beyond n > 2s in TMDCs to get even higher enhancement as the oscillator strength almost vanishes for higher-order Rydberg states. Additionally, the higher-order Rydberg states are also more susceptible to disorder due to their larger Bohr radius. On the other hand, the spectral signature of interlayer exciton in bilayer MoS₂ is visible even at room temperature and was shown to have a significant response to applied out of plane electric field. This provides a platform with the possibility of creating highly interacting dipolar polaritons controlled by external electric fields.

Nonlinear polariton interactions in the present case have contributions from exciton-exciton Coulomb interaction (which includes both dipolar and exchange terms) and phase space filling effect. For intralayer excitons with a small Bohr radius only the exchange term and phase space filling effect is important. On the other hand, for interlayer excitons in traditional quantum well systems, the direct dipole-dipole interaction dominates, and the exchange interaction is neglected due to the separation between charges in the two layers. In contrast in MoS₂ homobilayers, the hole is delocalized across the two layers, and hence there is a finite overlap with the electron wavefunction leading to small but non-zero exchange interactions. Thus in the present experiments, in addition to the saturation effects, both dipolar and exchange interaction contribute to the overall nonlinear response, with the latter being less significant owing to the finite separation between the electron and hole.

To discern the contributions to the polariton nonlinearity due to dipolar and exchange interaction of excitons ($\Delta E_{exc-exc}$) versus phase space filling ($\Delta E_{sat}$) we carry out the following analysis. Figure 4a shows the Hopfield coefficients of all the excitonic components and the cavity photon as a function of $k_0$. This is used to obtain the shift in polariton energy, $\Delta E_{LP}$, as a function of Hopfield cavity photon fraction (C), see Fig. 4b. The interaction strength due to phase space filling can be written in terms of $g_{LP} = 4g_{SAT}(1-C)^{1/2}$, where X is the Hopfield coefficient for the exciton. Although this is a multi-exciton system, the contribution of the other excitons close to the zero-detonating $k_0$ of the IE is small. Thus, we can write $g_{LP} = 4g_{SAT}(1-C)^{1/2}$ which is a non-monotonic function of the cavity Hopfield coefficient as shown in the Fig. 4b inset (red curve). For the case of exchange and dipolar interaction, the interaction strength scales as $g_{LP} = g_{IE}X(1-C)^{1/2}$ which monotonically decreases with increasing C (cyan curve in Fig. 4b inset). We find that the measured nonlinear response of the IE polariton is the highest at an in-plane wave vector larger than zero-detonating $k_0$, as seen in Fig. 4b. We also notice that $\Delta E_{LP}$ changes non-monotonically with the cavity Hopfield coefficient. The tilted parabolic shape of the measured $\Delta E_{LP}$ as a function of $C$ indicates a roughly equal contributions from both $\Delta E_{exc-exc}$ and $\Delta E_{sat}$.

Figure 4c shows a schematic of the polariton energies at two different powers for $\Delta E_{sat} > \Delta E_{exc-exc}$. Due to both the blue shift of the exciton and saturation effect, the lower branch moves with increasing power more than the upper branch. Analyzing the blueshift of the IE lower polariton branch and redshift of the IE upper polariton branch at the zero-detonating $k_0$ we calculate the magnitude of $\Delta E_{exc-exc}$ and $\Delta E_{sat}$ in our system. At the zero-detonating $k_0$ we write the energy shift of IE lower branch as $\Delta E_{LP} = \Delta E_{exc-exc} + \Delta E_{sat}$ and the energy shift of IE upper branch as $\Delta E_{LP} = \Delta E_{exc-exc} - \Delta E_{sat}$. From these equations we obtain $\Delta E_{exc-exc}$ and $\Delta E_{sat}$ as a function of polariton density, see SI Note IX. The ratio of $\Delta E_{sat}/\Delta E_{exc-exc}$ as a function of polariton density is shown in Fig. 4d, indicating that the magnitude of the dipole mediated exciton-exciton interaction nonlinearity and saturation nonlinearity are comparable especially at low polariton densities. Further increase of dipole-dipole interaction by creating imbalance between up and down dipoles (via applied electric field or providing strain) will allow to access regime where dipolar interactions dominate.

We now discuss the origin of the net dipolar interaction arising in our system at the zero electric field. The two ground states of the interlayer exciton are quasi degenerate at the zero electric field and should not possess net dipole moment in an ideal limit. However, as our experiments indicate, dipolar interactions play a crucial role in the 10 fold enhancement in interaction strength we observe for the interlayer exciton-polaritons. This raises questions about the origin of the dipole moment. Structural asymmetry, strain, residual electric fields, and disorder are the likely reasons for breaking the symmetry and resulting in a net dipole moment. Given that the bilayer MoS₂ is sandwiched between two hBN layers of similar thickness, one can rule out the role of structural asymmetry. While local strain and built-in fields could have highly localized effects, we observed similar enhancement over the entire bilayer sample, and hence one can conclude that local strain or fields are also not the key contributors. Finally, if one looks at disorder arising from defects present in the 2D TMDCs, the typical defect densities observed in mechanically exfoliated monolayer MoS₂ is on the order of $10^{12}$–$10^{13}$ cm⁻² as has been shown before. Using the lower bound of $10^{12}$ cm⁻² we can estimate the
The DBR used in our experiment is made of 8 pairs of SiO₂ (104.9 nm) and TiO₂ (64.7 nm) layers and were deposited by radio frequency sputtering on an intrinsic Si chip. MoS₂ and hBN were exfoliated from bulk crystals (from 2Dsemiconductor Inc.) using blue tape (Nitto) and scotch tape respectively. MoS₂ was exfoliated onto PDMS substrate and hBN was exfoliated onto a 300 nm SiO₂/Si substrate. Bilayer MoS₂ flakes were identified by reflection spectroscopy – the additional dip at ~639 nm in the reflection signal at room temperature is the hallmark of bilayer MoS₂. hBN/bilayer MoS₂/hBN heterostructure stacking and transfer were done using the well-known polypropylene carbonate transfer technique. The chosen top and bottom hBN layers were of similar thickness ~20 nm. The final stack was then transferred onto the DBR at a temperature of 150 °C. The chip was kept in chloroform for 12 h to remove the polypropylene carbonate residue. 240 nm poly-methylmethacrylate (PMMA) (495 A4 from Michrochem) was spin-coated to form a 3λ/2 cavity. Finally, a silver layer (40 nm) was deposited by electron-beam evaporation for the top mirror of the microcavity.

**Methods**

**Fabrication details**

The DBR used in our experiment is made of 8 pairs of SiO₂ (104.9 nm) and TiO₂ (64.7 nm) layers and were deposited by radio frequency sputtering on an intrinsic Si chip. MoS₂ and hBN were exfoliated from bulk crystals (from 2Dsemiconductor Inc.) using blue tape (Nitto) and scotch tape respectively. MoS₂ was exfoliated onto PDMS substrate and hBN was exfoliated onto a 300 nm SiO₂/Si substrate. Bilayer MoS₂ flakes were identified by reflection spectroscopy – the additional dip at ~639 nm in the reflection signal at room temperature is the hallmark of bilayer MoS₂. hBN/bilayer MoS₂/hBN heterostructure stacking and transfer were done using the well-known polypropylene carbonate transfer technique. The chosen top and bottom hBN layers were of similar thickness ~20 nm. The final stack was then transferred onto the DBR at a temperature of 150 °C. The chip was kept in chloroform for 12 h to remove the polypropylene carbonate residue. 240 nm poly-methylmethacrylate (PMMA) (495 A4 from Michrochem) was spin-coated to form a 3λ/2 cavity. Finally, a silver layer (40 nm) was deposited by electron-beam evaporation for the top mirror of the microcavity.

**Optical measurement details**

We recorded the angle-resolved reflection spectra using the Fourier space imaging technique. A broadband halogen light source was used for reflection measurements. A supercontinuum pulsed light source (NKT Photonics, repetition rate 80 MHz, pulse duration 20 ps) was used to study the power dependence of the bare excitons and polaritons. Appropriate long pass and short pass filters were used in the input to excite a narrow band around the excitons/polaritons under study. For the IE polariton, the excitation bandwidth of the supercontinuum laser was chosen to be ~40 meV around the IE energy in the...
power-dependent measurements. The set-up was coupled with a Princeton Instruments monochromator with a CCD camera. A 50X, 0.65 numerical aperture objective was used for all the measurements at 7 K in a closed cycle Montana cryostat. The polariton dispersion was revealed by imaging the back aperture of the microscope objective (Fourier plane) onto the camera. The spot size of the laser was 1 μm² on a uniform area of the sample of dimension ~500 μm².

Exciton density calculation in the absence of the top mirror

For the data presented in Fig. 1d, we excite A, IE and B exciton simultaneously at various optical ﬂuences. We have used a supercontinuum laser with a 20 ps pulse width and broadband excitation (1.907 eV to 2.246 eV) scheme for this measurement. At all ﬂuences, each spectrum is ﬁtted with the sum of three Lorentzians to track the evolution of each feature with excitation ﬂuence (see SI Note VI). To calculate the density of individual excitons, we do the following.

We ﬁrst calculate the absorbed power density by the bilayer MoS₂

\[ P_{\text{absorbed}} = \frac{\int_{E_{\text{EX}}}^{E_{\text{MAX}}} I_{\text{laser}}(E) (E - E_X) dE}{A_{\text{beam}}} \]  

(1)

Here \( I_{\text{laser}}(E) \) is the intensity proﬁle of the incident supercontinuum laser, \( L(E - E_X) \) is the absorbance of X exciton around its energy \( E_X \) and \( A_{\text{beam}} \) is the size of the focused laser beam on the sample. We run the integration within 74 meV (53 meV) band around the A exciton (IE) peak energy, respectively. Beyond these bands, the refraction contrast of A exciton and IE are not detectable. The absorbed photon density from the absorbed power density is calculated taking into account the life time of these excitons (45 fs for A exciton 53 fs for IE). We assume all of the absorbed photons form excitons.

Transient reﬂectivity measurements

Transient reﬂection spectroscopy measurements presented in SI Note X were obtained using an ultrafast transient absorption spectrometer (Harﬁa, Light Conversion). An ultrafast optical parametric oscillator (Orpheus-F, Light Conversion) provided the pump pulses and white light continuum generation in a sapphire crystal provided the probe pulse. These were afﬁliated using a pinhole inside a telescope and collinearly sent to a home-built microscope equipped with a 50X objective (NA 0.45). The sample was held at 3.5 K using a closed-cycle cryostat (Montana Instruments) integrated with a microscope. The output lens of the ﬁltering telescope was adjusted to obtain pump and probe spots as large as the bilayer ﬂake. Before its detection by a spectrometer, the pump was removed from the light reﬂected by the sample using a polarizer so that only the probe beam was detected.

Error in estimating density of polaritons

Currently, the density error bars consider the error in measuring power, determining the energy and linewidth of the polariton. However, one primary source of the error is the lifetime of the polaritons. To address this issue we have done pump-probe experiment which validates the use of the linewidth of the polaritons to determine the polariton lifetime. Results of our pump-probe experiment is included in the SI note X. Another likely source of error is estimating efﬁcient photons loading into a cavity. This is because the conventional input-output theory does not account for lossy mirrors, as in our experiment. Also photon loading into microcavities from free space is not efﬁcient due to mode mismatch between the free space laser modes and biorthonormal modes in lossy cavities. However, the relative enhancement of the nonlinearity of IE with respect to A remains unaffected due to the above errors as they exist for both the IE and A exciton polariton.

Data availability

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

References

1. Gisin, N. & Thew, R. Quantum communication. Nat. Photonics. 1, 165–171 (2007).
2. Giovannetti, V., Lloyd, S. & Maccone, L. Quantum-enhanced measurements: beating the standard quantum limit. Science 306, 1330–1336 (2004).
3. Chang, D. E., Vuletić, V. & Lukin, M. D. Quantum nonlinear optics—photon by photon. Nat. Photonics. 8, 685–694 (2014).
4. Lukin, M. D. et al. Dipole blockade and quantum information processing in mesoscopic atomic ensembles. Phys. Rev. Lett. 87, 037901 (2001).
5. Carusotto, I. & Ciuti, C. Quantum ﬂuids of light. Rev. Mod. Phys. 85, 299–366 (2013).
6. Muñoz-Matutano, G. et al. Emergence of quantum correlations from interacting ﬁbre-cavity polaritons. Nat. Materials. 18, 213–218 (2019).
7. Delteil, A. et al. Towards polariton blockade of conﬁned exciton–polaritons. Nat. Materials. 18, 219–222 (2019).
8. Mak, K. F. & Shan, J. Photonics and optoelectronics of 2d semiconductor transition metal dichalcogenides. Nat. Photonics. 10, 216–226 (2016).
9. Zhang, L. et al. Van der waals heterostructure polaritons with moiré-induced nonlinearity. Nature 591, 61–65 (2021).
10. Alexeev, E. M. et al. Resonantly hybridized excitons in moiré superlattices in van der waals heterostructures. Nature 567, 81–86 (2019).
11. Zhang, L. et al. Twist-angle dependence of moiré excitons in ws 2/ mose 2 heterobilayers. Nat. Commun. 11, 1–8 (2020).
12. Tang, Y. et al. Tuning layer-hybridized moiré excitons by the quantum-conﬁned stark effect. Nat. Nanotechnol. 16, 52–57 (2021).
13. Liu, X. et al. Strong light-matter coupling in two-dimensional atomic crystals. Nat. Photonics. 9, 30–34 (2014).
14. Sun, Z. et al. Optical control of room-temperature valley polaritons. Nat. Photonics. 11, 491–496 (2017).
15. Chen, Y.-J., Cain, J. D., Stanev, T. K., Dravid, V. P. & Stern, N. P. Valley-polarized exciton–polaritons in a monolayer semiconductor. Nat. Photonics. 11, 431–435 (2017).
16. Dufferwiel, S. et al. Valley-addressable polaritons in atomically thin semiconductors. Nat. Photonics. 11, 497–501 (2017).
17. Tan, L. B. et al. Interacting polaron-polaritons. Phys. Rev. X. 10, 021011 (2020).
18. Emmanuelle, R. P. A. et al. Highly nonlinear trion-polaritons in a monolayer semiconductor. Nat. Commun. 11, 3589 (2020).
19. Kravtsov, V. et al. Nonlinear polaritons in a monolayer semiconductor coupled to optical bound states in the continuum. Light: Science & Applications 9, 1–8 (2020).
20. Gu, J. et al. Enhanced nonlinear interaction of polaritons via excitonic rydberg states in monolayer wse 2. Nat. Commun. 12, 2269 (2021).
21. Rivera, P. et al. Observation of long-lived interlayer excitons in monolayer mose 2–wse 2 heterostructures. Nat. Commun. 6, 1–6 (2015).
22. Suárez-Forero, D. G. et al. Enhancement of parametric effects in polariton waveguides induced by dipolar interactions. Phys. Rev. Lett. 126, 137401 (2021).
23. Togan, E., Lim, H.-T., Faelt, S., Wegscheider, W. & Imamoglu, A. Enhanced interactions between dipolar polaritons. Phys. Rev. Lett. 121, 227402 (2018).
24. Rosenberg, I. et al. Strongly interacting dipolar-polaritons. Science Advances 4, https://advances.sciencemag.org/content/4/10/eaat8880. https://advances.sciencemag.org/content/4/10/eaat8880.full.pdf (2018).
25. Leisgang, N. et al. Giant stark splitting of an exciton in bilayer mos 2. Nat. Nanotechnol. 15, 901–907 (2020).
26. Lorchat, E. et al. Excitons in bilayer mos 2 displaying a colossal electric field splitting and tunable magnetic response. Phys. Rev. Lett. 126, 037401 (2021).
27. Gerber, I. C. et al. Interlayer excitons in bilayer mos 2 with strong oscillator strength up to room temperature. Phys. Rev. B. 99, 035443 (2019).
28. Peimyoo, N. et al. Electrical tuning of optically active interlayer excitons in bilayer mos2. Nat. Nanotechnol. 16, 1–6 (2021).
29. Deilmann, T. & Thygesen, K. S. Interlayer excitons with large optical amplitudes in layered van der waals materials. Nano lett. 18, 2984–2989 (2018).
30. Erkensten, D., Brem, S. & Malic, E. Exciton-exciton interaction in transition metal dichalcogenide monolayers and van der waals heterostructures. Phys. Rev. B. 103, 045426 (2021).
31. Helmrich, S. et al. Phonon-assisted intervalley scattering determines ultrafast exciton dynamics in mose2 bilayers. Phys. Rev. Lett. 127, 157403 (2021).
32. Stepanov, P. et al. Exciton-exciton interaction beyond the hydrogenic picture in a mose 2 monolayer in the strong light-matter coupling regime. Phys. Rev. Lett. 126, 167401 (2021).
33. Byrnes, T., Recher, P. & Yamamoto, Y. Mott transitions of exciton polaritons and indirect excitons in a periodic potential. Phys. Rev. B. 81, 205312 (2010).
34. Hong, J. et al. Exploring atomic defects in molybdenum disulphide monolayers. Nat. Commun. 6, 1–8 (2015).
35. Shree, S. et al. Interlayer exciton mediated second harmonic generation in bilayer mos2. Nat. commun. 12, 1–9 (2021).
36. Wang, L. et al. One-dimensional electrical contact to a two-dimensional material. Science 342, 614–617 (2013).
37. Carusotto, I. & Ciuti, C. Quantum fluids of light. Rev. Modern Phys. 85, 299 (2013).

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Author contributions
V.M.M., M.K. and B.D. conceived the experiments. B.D. fabricated the device. R.B. helped in the fabrication. B.D., M.K. and P.D. performed the measurements. F.T. did the time resolved pump probe measurements. B.D. did the data analysis with inputs from M.K., P.D. and V.M.M. S.D.L. and S.K.C. gave inputs in the data interpretation. B.D, M.K. and V.M.M wrote the manuscript with comments from all authors.

Competing interests
The authors declare no competing interests.

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