

Valley addressable exciton-polaritons in atomically thin MoSe₂

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While conventional semiconductor technology relies on the manipulation of electrical charge for the implementation of computational logic, additional degrees of freedom such as spin and valley offer alternative avenues for the encoding of information. In transition metal dichalcogenide (TMD) monolayers, where spin-valley locking is present, strong retention of valley chirality has been reported for MoS₂, WSe₂ and WS₂ while MoSe₂ shows anomalously low valley polarization retention. In this work, chiral selectivity of MoSe₂ cavity polaritons under helical excitation is reported with a polarization degree that can be controlled by the exciton-cavity detuning. In contrast to the very low circular polarization degrees seen in MoSe₂ exciton and trion resonances, we observe a significant enhancement of up to 7X when in the polaritonic regime. Here, polaritons introduce a fast decay mechanism which inhibits full valley pseudospin relaxation and thus allows for increased retention of injected polarization in the emitted light. A dynamical model reproduces the detuning dependence through the incorporation of the cavity-modified exciton relaxation, allowing an estimate of the spin relaxation time in MoSe₂ which is an order of magnitude faster than those reported in other TMDs. The valley addressable exciton-polaritons reported here offer robust valley polarized states demonstrating the prospect of valleytronic devices based upon MoSe₂ embedded in photonic structures, with significant potential for valley-dependent nonlinear polariton-polariton interactions.

Single layers of TMDs are two-dimensional (2D) direct band-gap semiconductors which exhibit pronounced exciton resonances with binding energies of up to 0.55 eV [1–3]. The 2D nature of TMDs gives rise to strongly confined excitons with Bohr radii of around 1 nm, leading to a large oscillator strength of the excitonic transition with absorption as strong as 15% [1]. Recently, strong exciton-photon coupling has been reported in van der Waals crystals such as MoSe₂, WSe₂, WS₂ and MoS₂ embedded in optical microcavities [4–8]. The observation of exciton-polaritons in TMDs creates opportunities in engineering the polariton-polariton interaction [9–11] in 2D materials, which can potentially be used for the development of active nanophotonic devices through integration of thin films into photonic crystal cavities and waveguides [12]. In addition, TMD polaritons have additional degrees of freedom, where the valley pseudospin and finite Berry curvature can be utilised in new valleytronic implementations [13].

In TMDs, the combination of time-reversal symmetry, strong spin-orbit coupling and the absence of crystal inversion symmetry leads to the coupling of spin and valley degrees of freedom [14, 15]. At both the conduction and valence band edges, the electron spin orientation is locked to the valley index allowing direct optical initialization of the valley pseudo-spin in the non-equivalent K and K' valleys of the hexagonal Brillouin zone with σ^+ / σ^- helicities respectively, as shown in Fig. 1a and Fig. 1b. Upon optical excitation the valley polarization can be directly probed through the degree of circular polarization of the photoluminescence (PL) which is defined

as $\rho = (I_{co} - I_{cross})/(I_{co} + I_{cross})$ where I_{co} (I_{cross}) is the co-polarized (cross-polarized) component to the excitation. Extensive polarization-resolved PL studies of MoS₂, WSe₂ and WS₂ report high initialized degrees of circular polarization, indicating strong chiral selectivity of excitonic states which are robust from inter-valley relaxation [16–20]. Kerr rotation measurements on WSe₂ report exciton valley relaxation times of around 6 ps attributed to exciton intervalley transfer caused by precession in effective magnetic fields due to the long range electron-hole exchange interaction [21–25]. The large valley polarization degrees observed in continuous-wave (cw) photoluminescence (PL) studies are then possible since the exciton lifetime of around 2-5 ps also occurs on an ultrafast timescale [5, 26, 27]. In stark contrast, A-excitons in MoSe₂, which have a comparable lifetime, show very low circular polarization degrees of < 5% [28]. The exact mechanism leading to this anomalous polarization behaviour of MoSe₂ is currently unclear, but would be consistent with an unconfirmed sub-ps valley relaxation time [28].

In this article we report on the valley addressability of exciton-polaritons in MoSe₂ embedded in tunable microcavities. A strong dependence of the polarization degree on the exciton-cavity detuning is reported along with significant enhancement compared to the bare exciton and trion polarization degrees. Due to interaction with disorder and localisation in the film, excitons in the bare film undergo efficient (< 0.1 ps) scattering between low and high k-vectors where efficient depolarization occurs due to the Maialle-Silva-Sham (MSS) mechanism - val-

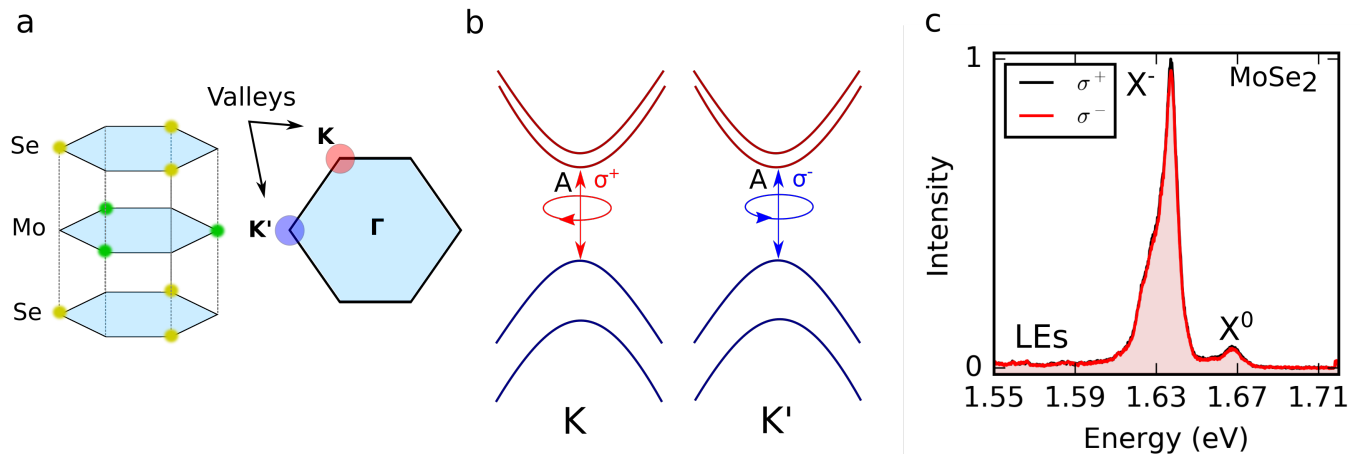


FIG. 1. **Spin-valley locking of excitons in MoSe₂.** **a** Illustration of the hexagonal lattice structure of MoSe₂. Optical band gaps are formed at the inequivalent K and K' valleys at the edge of the Brillouin zone. **b** Sketch of the valley dependent optical selection rules at the band edges. In the K (K') valley, the optical transitions couple to σ^+ (σ^-) polarized light. **c** PL spectra of MoSe₂ under σ^+ excitation showing A-exciton and trion emission in co- (black) and cross- (red) polarized detection.

ley relaxation induced by scattering in the presence of exciton LT-splitting from the electron-hole exchange interaction [22–25]. This leads to extrinsic valley depolarization and the observed low polarization degrees of the bare exciton and trion emission in MoSe₂. Intrinsic depolarization due to the exciton-phonon interaction will also contribute to depolarization but the effect of disorder in current samples is dominant since the inhomogeneous broadening of 10-20 meV is much larger than the homogeneous linewidth. When in the strong exciton-photon coupling regime, the polariton wavefunction (1 μ m) is much greater than the typical lengthscale of exciton disorder (10s of nm), leading to significant reduction of polariton scattering by the exciton disorder potential [29]. In addition, polaritons are also detuned from the exciton reservoir states (lower and middle polariton branches) or have a photonic component (upper polariton branch) so there is a non-zero net relaxation of high k-vector exciton states to the polariton states. This relaxation to the polariton branches inhibits full valley pseudospin relaxation in the exciton reservoir resulting in an increased polarization degree of the polariton emission. A dynamical model reproduces this dependence allowing estimates of the valley pseudospin relaxation time for bare excitons and trions to be 0.15 ps and 1 ps respectively. This sub-ps exciton intervalley relaxation is an order of magnitude faster than those reported for other TMDs providing strong evidence that the valley chirality in MoSe₂ is lost due to extremely fast pseudospin relaxation of excitons. The MSS depolarization mechanism discussed here may be suppressed for bright excitons in TMDs with lowest energy dark exciton states, if the relaxation to dark states occurs on a comparable sub-ps timescale to disorder scattering. This provides an explanation for the discrepancy in polarization behavior of MoSe₂ when compared with WSe₂ and

WS₂ [30–32] as well as MoSe₂, which was recently shown to have a lowest energy dark exciton state [33]. The optically bright nature of the lowest energy exciton transitions in MoSe₂ combined with narrow linewidths make MoSe₂ the most suitable TMD for reaching the high polariton density regime and the achievement of nonlinear TMD polariton phenomena. The demonstration of retention and control of valley polarization and suppression of intervalley relaxation for MoSe₂ polaritons opens new avenues for the exploration of spin/valley dependent polariton-polariton interactions in TMD microcavity systems as well as the development of valleytronic devices based upon TMDs embedded in photonic structures.

RESULTS

The van der Waals (VDWs) heterostructure utilised in this study consists of a single monolayer of MoSe₂ encapsulated on both sides with thin films of hexagonal boron nitride (hBN) placed on the surface of a planar distributed Bragg reflector (DBR). PL spectra under non-resonant excitation at 1.946 eV at 4.2 K for MoSe₂ are shown in Fig. 1c. Two characteristic peaks ascribed to neutral (X^0) and charged (X^-) exciton resonances are observed. In addition, localised emitters (LEs) are present on the low energy shoulder of the trion and below [34, 35]. Under σ^+ excitation the polarization resolved PL exhibits low retention of injected valley pseudospin of around 5% and 2% for the exciton and trion respectively, in agreement with previous reports [28].

The tunable cavity is formed through introducing a top concave DBR into the optical path using nanopositioners and bringing the two DBRs to a total optical cavity length of around 2.9 μ m [36, 37]. Full details

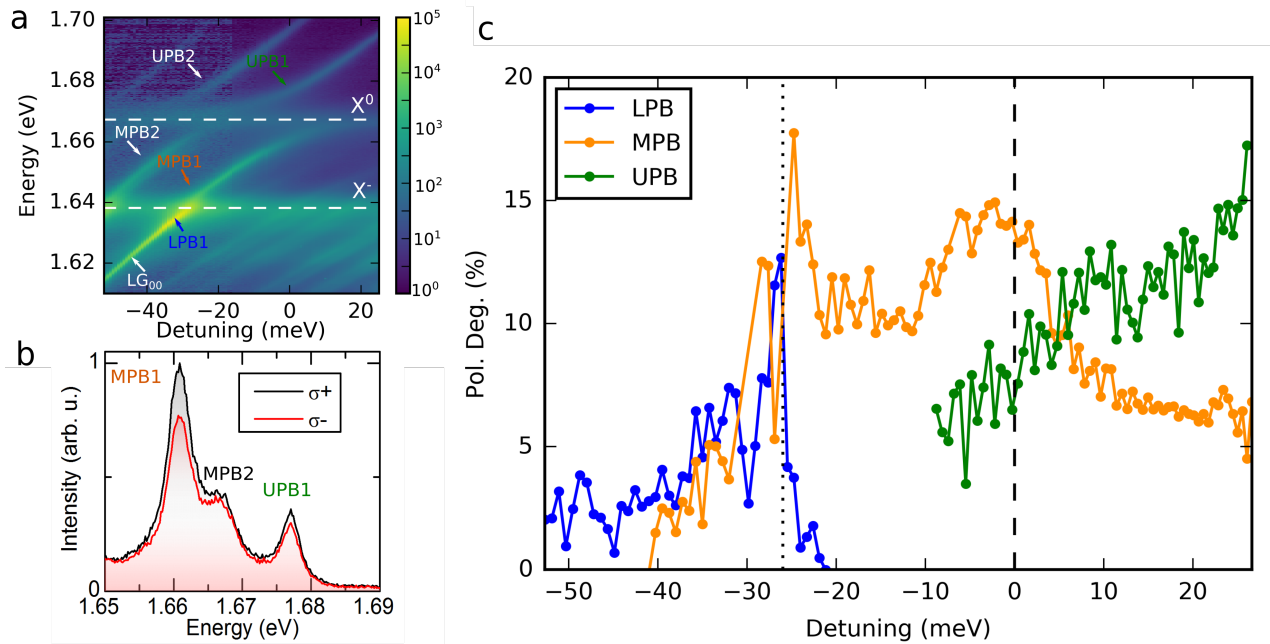


FIG. 2. **Valley addressable exciton-polaritons in MoSe₂.** **a** Colourmap of PL spectra as a function exciton-photon detuning ($\Delta = E_c - E_{X^0}$), where E_c and E_{X^0} are the tunable cavity and exciton energies, showing clear anticrossing with X^0 with a Rabi splitting of 15.2 ± 0.1 meV. The onset of strong coupling is present for the trion with a coupling strength of 1.3 ± 0.1 meV. **b** PL spectra at zero exciton-photon detuning with σ^+ excitation. Clear retention of circular polarization is seen to varying degrees in all peaks. **c** Circular polarization degree of the polariton branches as a function of exciton-photon detuning. The vertical dashed and dotted lines correspond to zero trion-cavity and zero exciton-cavity detuning.

of the cavity can be found in [5]. The cylindrical symmetry of the formed hemispherical cavity introduces a harmonic-like photonic confinement leading to the formation of Laguerre-Gaussian (LG) cavity modes. A clear anticrossing is observed in PL when the modes are tuned through resonance with X^0 as shown for the longitudinal mode in Fig. 2a. Coupling to the higher order modes is also present within the scan but all following discussion refers to the polariton branches formed due to coupling with the longitudinal mode. Fitting the polariton peak positions with a coupled oscillator model gives a Rabi splitting of 15.2 ± 0.1 meV for X^0 . Analysis and fitting of the lineshape when close to resonance between the cavity mode and trion reveals the onset of strong coupling with a coupling strength of around 1.3 ± 0.1 meV as shown in Supplementary Fig. S1. Here the trion-polariton peaks are not fully resolved at resonance due to the cavity-trion Rabi splitting being comparable to the inhomogeneously broadened polariton linewidths - indicating that we are at the crossover point between the weak and strong coupling regimes. In reflectivity and transmission, a clear splitting at resonance and the formation of trion-polariton branches has previously been shown [5]. In the following discussion we take the peak below the trion as the lower polariton branch (LPB), the peak between the trion and exciton as the middle polariton branch (MPB) and the peak above the exciton as the

upper polariton branch (UPB), as labeled in Fig. 2a and customary for a strongly coupled 3-level system.

The retention of valley pseudospin of the polaritonic system was probed through excitation with non-resonant σ^+ circularly polarized light at 1.946 eV, close to a DBR stopband minimum. Fig. 2b shows the PL spectra at zero exciton-photon detuning for co- (black) and cross-polarized (red) collection, where clear retention of valley polarization for the MoSe₂ polariton branches occurs indicating an injected imbalance in the polariton valley populations. In-situ tunability of the cavity resonance allows the dependence of the polarization degree on detuning to be probed through control of the mirror separation. The result for each polariton branch is plotted in Fig. 2c where the LG₀₀ cavity mode is tuned through resonance with X^- and X^0 as in Fig. 2a, and the polarization degree is calculated from the peak intensities of fitted spectra in co- and cross-polarized components. Spectra corresponding to a full range of detunings are shown in Supplementary Fig. S2 and Fig. S3. For the LPB we see a low polarization degree when strongly negatively detuned and an abrupt increase to around 15% when close to resonance with X^- , before falling when positively detuned to the trion resonance. For detunings $\Delta > -20$ meV, coupling between the trion to high order modes masks the effect of coupling to the longitudinal mode on the LPB so is neglected in the following analy-

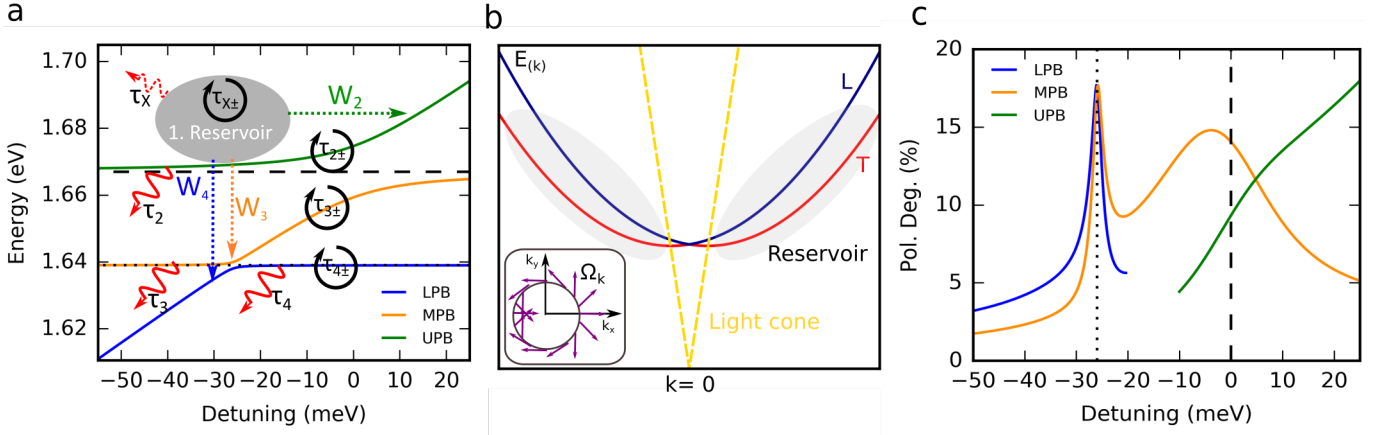


FIG. 3. **Cavity modified relaxation dynamics.** **a** Scheme of the relevant states and relaxation dynamics. **b** Illustration of the spin-splitting of the exciton dispersion into linearly polarised states with dipole moment parallel (L) and perpendicular (T) to the in-plane wavevector. Yellow dashed lines mark the light cone. Inset shows the orientation of the effective magnetic field Ω_k which induces spin precession in the exciton reservoir. **c** Calculated circular polarization degree of the three polariton branches as a function of detuning using exciton and trion spin relaxation times of $\tau_{X\pm} = 0.15$ ps and $\tau_{\pm} = 1$ ps respectively. The vertical dashed and dotted lines correspond to zero trion-cavity and zero exciton-cavity detuning.

sis. The MPB polarization degree is initially low at negative detunings before rising to a maximum close to zero trion-cavity detuning, before dropping and increasing to a maximum of around 15% when slightly negatively detuned from X^0 , before falling to the bare exciton level at positive detunings. Finally the UPB shows an almost linear increase in polarization degree with detuning to a maximum value of around 17% at the maximum probed positive detuning of $\Delta = +25$ meV. For all polariton branches, the maxima of polarization degree are significantly enhanced in comparison to the bare exciton and trion resonances. Similar behaviour is measured for a second MoSe₂ sample with polarization degrees of up to 20% as shown in Supplementary Fig. S5.

DISCUSSION

To describe the processes taking place in the system with both exciton and trion resonances coupled with a single cavity mode we consider the dynamical process represented in Fig. 3a. Excited valley polarized carriers relax to the reservoir of high k -vector excitons. The exciton depolarization occurs through the MSS mechanism [22–25]. The exciton valley pseudospin precesses about the k -dependent effective magnetic shown in the inset of the Fig 3b which is induced by the exciton exchange coupling. This precession combined with random disorder scattering leads to an exciton valley pseudospin relaxation with a characteristic time $\tau_{X\pm}$. Reservoir excitons with non-radiative lifetime τ_X , then relax towards the light-cone and form polaritons with a relaxation rate W_i . Further valley pseudospin relaxation can then occur in the polariton branches with a timescale $\tau_{i\pm}$ be-

fore decay out of the cavity with a polariton radiative lifetime τ_i . The resultant rate equations can be found in the Theoretical Model section below. The exciton-photon detuning affects the circular polarization degree of the polariton emission via the excitonic (x_i) and trionic (t_i) fractions of the polariton states determining the scattering rates towards these states, while the lifetimes of the states are affected via their photonic fraction (p_i). The following dependences for the scattering rates to the middle and lower polariton branches ($i = 3, 4$), derived in Supplementary Note 1, are given by:

$$W_i = W_0(x_i + t_i)p_i \quad (1)$$

where W_0 is the exciton energy relaxation rate. The scattering rates have to be proportional to the exciton fraction, and the exciton and trion are summed over, where we assume the presence of an extra electron does not change the relaxation mechanism. If the branch is completely excitonic, it becomes degenerate with the exciton reservoir, and the scattering, due to interaction with disorder, towards the branch becomes balanced by an inverse scattering rate from the branch. The imbalance between the two scattering rates (forward and backward) $W_{X \rightarrow i} - W_{i \rightarrow X} \propto 1 - e^{-(E_X - E_i)/k_B T} \approx (E_X - E_i)/k_B T$ is therefore proportional to the photonic fraction in the first order approximation, hence $W_i \propto p_i$.

The UPB is a special case, as it is always resonant with some states of the exciton reservoir at high k -vectors. It is therefore possible for high momentum excitons to scatter directly to the photon-like UPB states due to interaction with disorder. The corresponding scattering rate is therefore only proportional to the photonic fraction, while the excitonic one does not play any role: i.e. the

exchange between the excitonic reservoir and the excitonic fraction of this branch is always strongly balanced. Moreover, since the excitons do not need to relax their energy, and need only to change their momentum, the characteristic time of this process is that of momentum relaxation (pseudospin relaxation), and not that of the energy relaxation. Therefore, we assume that:

$$W_2 \propto p_i / \tau_{X\pm} \quad (2)$$

Another consideration for the UPB is that the direct scattering from the reservoir not only drains it, shortening its lifetime and increasing the average polarization degree, but also brings strongly polarized particles from higher energy states directly into the resonant polariton states. Since the spin precession rate is linearly proportional to k -vector, we assume that the polarization degree in the reservoir decreases linearly with energy, from 100% at the injection point ($E_X + 300$ meV) down to ρ_X at E_X . This polarization degree can then decay in the UPB before being emitted out of the system, giving a renormalization $\tau_{2\pm} / (\tau_{2\pm} + \tau_2)$, derived in Supplementary Note 2. In order to reproduce the data the ratio of UPB polariton population coming from this direct scattering from the reservoir compared to indirect relaxation (Eq. 1) is required to be 1 : 1.

We take the exciton and trion lifetimes as $\tau_{X0} = 5.3$ ps and $\tau_T = 12$ ps [5] and assume that the non-radiative exciton reservoir lifetime is comparable to the exciton PL-lifetime ($\tau_X = 5.3$ ps). The photon lifetime measured from the cavity at large negative detuning is $\tau_p = 1.3$ ps. Finally we take the exciton energy relaxation rate to be $W_0 = 1/0.15$ ps to fit the data; an upper limit of 1 ps can be justified from the resolution limited rise-time for MoSe₂ excitons in recent time resolved measurements [27]. The polarization dependence on detuning, shown in Fig. 3c, is well reproduced using exciton and trion valley relaxation times of $\tau_{X\pm} = 0.15$ ps and $\tau_{T\pm} = 1$ ps respectively.

The enhancement in polarization degree of the lower and middle polariton branches relative to the bare monolayer case is due to cavity modified relaxation which allows MoSe₂ excitons to relax from the reservoir into polariton states quickly, inhibiting complete spin relaxation in the reservoir. In these polariton states, which are formed with excitons around $k \approx 0$, the exciton LT-splitting is small and hence the spin relaxation from the excitonic component of the polariton is around 20 times slower than the bare exciton, as explained below in the Theoretical Model section, much slower than the polariton radiative lifetime, allowing lower and middle polariton branch particles to radiatively decay without further significant depolarization, and with higher retention of valley pseudospin. When the photonic fraction of the polariton is high, relaxation into these polariton branches is less efficient causing larger accumulation of particles

in the reservoir and hence low polarization degree. At high excitonic fractions, scattering from the reservoir into these polariton branch is efficient, but the low photonic fraction, which determines the radiative decay and hence the imbalance between forward and backwards scattering between polariton and reservoir states, leads to a longer time spent in the reservoir and hence low polarization of emitted light. At intermediate detunings, and roughly equal exciton and photon fractions a maximum in polarization degree is seen which is a compromise of efficient relaxation due to the exciton component and efficient radiative decay due to the photonic component. The UPB high valley polarization is due to direct scattering of high momentum excitons through interaction with disorder, populating the UPB with highly polarized particles which decay quickly due to their photonic component.

In conclusion, we observe clear retention of valley polarization of MoSe₂ exciton-polaritons. The strong dependence on exciton-trion-photon components can be well described by a dynamical model that incorporates modified relaxation and state lifetimes arising from strong coupling with the cavity mode. We estimate that the valley pseudospin relaxation time for MoSe₂ excitons is sub-ps, indicating that the low polarization retention of MoSe₂ is due to fast precession of exciton pseudospin due to the MSS mechanisms. This depolarization mechanism due to disorder scattering is likely suppressed in TMDs with low energy dark states due to the additional fast decay channel, providing insight into the anomalous behavior of MoSe₂ in comparison to WSe₂, WS₂ and MoS₂. The spin-valley locking inherited by TMD polaritons from their excitonic component offers new avenues or valley-dependent polariton-polariton interactions [38] where the populations of polaritons in K and K' valleys can be closely controlled with resonant circularly polarized excitation.

METHODS

Theoretical Model

The Boltzmann equations for the populations of the various states are given as:

$$\frac{dn_{X\pm}}{dt} = P_{\pm} - \frac{n_{X\pm}}{\tau_X} - n_{X\pm} \sum_{i=2}^4 W_i \mp \frac{n_{X+} - n_{X-}}{\tau_{X\pm}} \quad (3)$$

$$\frac{dn_{i\pm}}{dt} = W_i n_{X\pm} - \frac{n_{i\pm}}{\tau_i} \mp \frac{n_{i+} - n_{i-}}{\tau_{i\pm}}, i = 2, 3, 4 \quad (4)$$

Here $n_{X\pm}$ and $n_{i\pm}$ are the excitation and polariton populations in the K and K' valleys respectively, where i runs from 2 to 4 and denotes the UPB, MPB and LPB. $\tau_{i\pm}$

is the spin relaxation time of the polariton branch given below, and P_+ is the pumped valley polarized population of particles. Solving these coupled rate equations in the steady state allows to find the solutions for the circular polarization degrees of the exciton-polariton branches:

$$\rho_i = \frac{\tau_{i\pm}\tau_{X\pm} \left(1 + \tau_X \sum_{j=2}^4 W_j\right)}{(2\tau_i + \tau_{i\pm}) \left(\tau_{X\pm} + 2\tau_X + \tau_X\tau_{X\pm} \sum_{j=2}^4 W_j\right)} \quad (5)$$

For the exciton reservoir, the circular polarization degree is given by:

$$\rho_X = \frac{\tau_{X\pm} \left(1 + \tau_X \sum_{i=2}^4 W_i\right)}{\tau_{X\pm} + 2\tau_X + \tau_X\tau_{X\pm} \sum_{i=2}^4 W_i} \quad (6)$$

The circular polarization degree for the polariton branches and for the system in general is given by the balance of the exciton spin relaxation time and exciton energy relaxation time (total scattering rate out of the exciton reservoir). If the average time the particle spends in the reservoir is longer than the spin relaxation time, the circular polarization degree will be low.

The coupling of the exciton, trion, and photon modes is described by a coupled oscillator model, where the eigenenergies and the Hopfield coefficients are obtained from the matrix

$$\begin{pmatrix} E_X & 0 & V_X \\ 0 & E_T & V_T \\ V_X & V_T & E_C \end{pmatrix} \quad (7)$$

where $E_X = 1.667$ eV and $E_T = 1.639$ eV are the exciton and the trion energies, E_C is the photon energy, which varies from 1.720 to 1.580 eV. The Rabi splitting for exciton is taken as $2V_X = 15.2$ meV, while for the trion we take $2V_T = 1.3$ meV. Diagonalizing this matrix, one obtains the three polariton branches as shown in Fig. 2a. The corresponding eigenvectors give the exciton, trion, and photon fractions x_i , t_i , p_i . For the polariton radiative lifetimes, the dependence can be written as:

$$\frac{1}{\tau_i} = \frac{x_i}{\tau_{X0}} + \frac{t_i}{\tau_T} + \frac{p_i}{\tau_p} \quad (8)$$

where τ_p , τ_{X0} and τ_T are the photon, exciton and trion lifetimes respectively. Finally, for the polariton spin relaxation times we take the following dependence:

$$\frac{1}{\tau_{i\pm}} = \alpha \frac{x_i}{\tau_{X\pm}} + \beta \frac{t_i}{\tau_{T\pm}} + \frac{p_i}{\tau_{p\pm}} \quad (9)$$

where we assume different valley pseudospin relaxation times for excitons, $\tau_{X\pm}$, and trions, $\tau_{T\pm}$. The coefficients α and β are required since the polariton spin relaxation due to the excitonic component is reduced in comparison to the bare flake exciton and trion. In the bare flake efficient scattering between low k (low LT-splitting) and high k (large LT-splitting) excitons occurs due to scattering on disorder or localisation giving rise to fast depolarization. Since polaritons are formed from low-k excitons and scattering due to disorder is reduced for the polaritons we expect the contribution to the polariton depolarization from the exciton component to be less than the bare exciton spin relaxation time. Assuming a thermal population of excitons in the reservoir and a polariton wavefunction $\approx 1 \mu\text{m}$ [39] and a linear dependence of the effective magnetic field with in-plane wavevector [23, 25] we estimate $\alpha \approx \beta \approx 0.05$. We also take into account spin relaxation of the photonic component, $\tau_{p\pm} = 15$ ps due to the finite TE-TM splitting of the ground state mode [40] (see Supplementary Fig. S6).

Sample Preparation

Monolayer sheets of MoSe₂ and thin films of hexagonal boron nitride (hBN) were obtained through mechanical exfoliation of bulk crystals. A thin film of hBN was transferred onto the DBR using standard transfer techniques before a monolayer of MoSe₂ was transferred onto its surface. The sample was then encapsulated with a thin film of hBN. Bulk crystals were acquired from HQGraphene.

Optical Measurements

Optical measurements were performed with samples held in a helium bath cryostat system at a temperature of 4.2K. Top and bottom DBRs were attached to XYZ nanopositioners with additional goniometer stages allowing tilt control of the bottom DBR. Optical excitation of the bare monolayer was possible by removing the top DBR from the optical path. All μ -PL experiments were performed with a continuous-wave (cw) excitation using a 638 nm laser diode, focused onto the sample with an achromatic lens. Polarization resolved measurements were performed using a combination of linear polarizer and a quarter waveplate in the excitation path, and quarter wave-plate, half-wave plate and linear polarizer in the collection path. PL was collected by focusing onto a single mode fibre which was guided into a 0.75m spectrometer and a high sensitivity charged couple device.

Microcavity

The tunable microcavity with embedded TMD monolayer is formed using an external concave mirror to produce a zero-dimensional tunable cavity [39]. The formed cavity electric-field profile is shown in Supplementary Fig. S7 with the monolayer placed at an electric-field antinode, and nanopositioners are used to control the cavity spectral resonance energy. The nominal radius of curvature of the concave mirror is 20 μm leading to a beam waist on the planar mirror of around 1 μm [39].

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AUTHOR CONTRIBUTIONS

S.D and T. P. L carried out optical investigations. Sample fabrication was performed by F. W. A. A. P. T. designed and fabricated the concave mirrors. D. D. S. and G. M carried out theoretical analysis. S. D. analysed the data and prepared the manuscript with contributions from all co-authors. J. M. S., K. N., M. S. S., D. N. K., and A. I. T. provided management of various aspects of the project. D.N.K proposed the idea of using an open-access microcavity system, developed by his group, for polariton studies with TMD monolayers. A. I. T. conceived and oversaw the project.

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