Moiré-Trapped Interlayer Triions in a Charge-Tunable WSe₂/MoSe₂ Heterobilayer

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Transition-metal dichalcogenide heterobilayers offer attractive opportunities to realize lattices of interacting bosons with several degrees of freedom. Such heterobilayers can feature moiré patterns that modulate their electronic band structure, leading to spatial confinement of single interlayer excitons (IXs) that act as quantum emitters with C_3 symmetry. However, the narrow emission linewidths of the quantum emitters contrast with a broad ensemble IX emission observed in nominally identical heterobilayers, opening a debate regarding the origin of IX emission. Here we report the continuous evolution from a few trapped IXs to an ensemble of IXs with both triplet- and singlet-spin configurations in a gate-tunable 2H-MoSe₂/WSe₂ heterobilayer. We observe signatures of dipolar interactions in the IX ensemble regime which, when combined with magneto-optical spectroscopy, reveal that the narrow quantum-dot-like and broad ensemble emission originate from IXs trapped in moiré potentials with the same atomic registry. Finally, electron doping leads to the formation of three different species of localized negative triions with contrasting spin-valley configurations, among which we observe both intervalley and intravalley IX triions with spin-triplet optical transitions. Our results identify the origin of IX emission in MoSe₂/WSe₂ heterobilayers and highlight the important role of exciton-exciton interactions and Fermi-level control in these highly tunable quantum materials.

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I. INTRODUCTION

Van der Waals heterobilayers consisting of vertically stacked monolayer transition-metal dichalcogenide semiconductors (TMDs) form atomically sharp interfaces with type-II band alignment [1,2]. This band alignment enables the formation of interlayer excitons (IXs)—electron-hole Coulomb bound states between electrons and holes spatially separated in different monolayers. The reduced overlap of the electron and hole wave functions gives rise to long IX radiative lifetimes (compared to intralayer excitons) [3,4] that can be further tailored by the momentum mismatch between the carriers [5]. The spatial separation of the IXs carriers also results in a large permanent electric out-of-plane dipole moment, which enables a large tunability of the

exciton energy by externally applied electric fields [6–8]. The combination of long lifetimes and large binding energies [9,10] position IXs in TMD heterostructures as an exciting platform to explore many-body exciton-exciton phenomena such as dipolar interactions of IXs in the low-density regime [11,12] or the high-density regime, where signatures of coherent excitonic many-body quantum states and high-temperature exciton (boson) condensation have been predicted [13–15] and observed [16,17]. The robust and long-lived IXs in TMD heterobilayers also offer novel opportunities to realize atomically thin optoelectronic devices such as lasers [18] and excitonic transistors that can operate at room temperature [19].

Beyond the large permanent dipole moment and strong Coulomb interactions, the compelling concept of a moiré superlattice [20] emerges in TMD heterobilayers due to the lattice mismatch and any relative twist angle between the constituent monolayers. In MoSe₂/WSe₂ heterobilayers, the moiré superlattice results in a periodic potential landscape for IXs [21] (with a periodicity that depends on the relative crystallographic alignment of the layers) in which three trapping sites with three different local atomic registries arise [22–24]. Experimental evidence of IXs

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trapped in a moiré-induced potential has been reported in MoSe₂/WSe₂ heterobilayers with twist angles near 0°, 21.8°, and 60° at cryogenic temperatures [8,25,26]. These localized IXs present emission linewidths below 100 μ eV [8,25,26] and photon antibunching [8], clear hallmarks of quantum-confined excitons. Moreover, the trapped IXs show well-defined magneto-optical properties: The g factors of the trapped IXs depend on the relative valley alignment (i.e., stacking configuration) between the layers hosting the carriers, while their optical selection rules are determined by the atomic registry of the trapping site [25,26]. Together, these magneto-optical properties provide compelling evidence for the moiré potential as the origin of the IX trapping. The important role the moiré potential plays in the confinement of IXs is further supported by the twist-dependent IX diffusion in TMD heterobilayers, in which the diffusion length of the IX ensemble depends on the moiré periodicity [27,28]. However, the narrow emission linewidths observed for single trapped IXs contrast with the broad photoluminescence (PL) spectra observed in similar MoSe₂/WSe₂ heterostructures [4–7,16,29–31], which show IX emission bands with linewidths of 4–6 meV in the cleanest samples [6,16,30,32], 2 orders of magnitude broader than single trapped IXs. Such contrasting observations have opened a debate regarding a unified picture of the nature of IX emission in TMD heterobilayers, and in particular, in the prototypical heterostructure: MoSe₂/WSe₂ heterobilayers [33]. To date, clear experimental evidence which can marry these two regimes (narrow-linewidth trapped IX vs broad-linewidth ensemble IX) is missing. Further, gate doping of MoSe₂/WSe₂ heterobilayers has been shown to lead to the formation of charged IX (trions) with spin-singlet and spin-triplet configurations [7,34]. However, the trapped or delocalized nature of IX trions, and all their possible spin-valley configurations, have yet to be addressed.

Here, we investigate the magneto-optical properties of IXs in a gate-tunable MoSe₂/WSe₂ heterobilayer. We tune the density of IXs by scanning the excitation power over 5 orders of magnitude and report a clear and continuous evolution from the narrow emission of single trapped IXs to broad ensemble IX peaks, for which IXs with both spin-triplet and spin-singlet configuration are observed. In the high-excitation-power regime, we observe an energetic blueshift of the IXs due to repulsive dipolar interactions. We estimate the density of IXs from the power-dependent evolution of the IX PL and find that, even at the highest excitation powers employed in our measurements (approximately 80 μ W), the estimated density of optically generated IXs is 2 orders of magnitude smaller than the estimated density of moiré traps (approximately 4×10^{12} cm⁻²) in our heterostructure. Polarization-resolved PL measurements under a vertical magnetic field confirm that the narrow and broad IX PL present identical magneto-optical properties, confirming

that both the quantum-dot-like and broad PL emission from IXs arise from IXs trapped in moiré potentials with the same atomic registry. Moreover, we investigate the formation of negatively charged IX trions and find that the trion creation originates from on-site charging of the trapping potentials. The magneto-optical properties of the negatively charged IXs reveal three different negative trion species with contrasting spin-valley configurations. Interestingly, we observe both intervalley and intravalley IX trions with spin-triplet optical transitions, a consequence of the absence of a dark exciton state in MoSe₂/WSe₂ heterobilayers. The identification of the various neutral and charged exciton species provides new insight into multiple peaked IX spectra in heterobilayers, while the identification of the localized nature of the IX ensemble emission in MoSe₂/WSe₂ heterobilayers demonstrates the important role of the moiré potential in their magneto-optical properties. These findings highlight the necessity to consider the spatial pinning of the IXs to the moiré lattice in order to achieve an accurate description of many-body exciton-exciton phenomena in TMD heterobilayers.

II. MAGNETO-OPTICS OF SPIN-SINGLET AND SPIN-TRIPLET NEUTRAL IXs

Figure 1(a) shows a sketch of the dual-gated heterobilayer device we employ, which consists of a monolayer MoSe₂ and a monolayer WSe₂ vertically stacked with a twist angle $\Delta\theta \sim 56.4 \pm 0.2^\circ$ ($2H$ stacking). The twist angle in our heterobilayer is beyond the theoretically proposed critical angle for lattice reconstruction [35–37], ensuring minimal moiré domain formation. The heterobilayer is encapsulated by hexagonal boron nitride (h -BN). Graphene layers act as electrical contacts for the top, bottom, and heterobilayer gates (see Ref. [8] for more details). Via voltage-dependent reflectance spectroscopy, we observe clear signatures of strongly correlated electronic states in both the conduction (CB) and valence bands (VB) at numerous fractional filling values of the moiré superlattice (see Supplemental Material [38] Note 1). Our observations, consistent with recent reports of correlated insulating states in angle-aligned WSe₂/WS₂ heterobilayer samples [39–42], confirm the quality of our heterobilayer sample and the formation of a moiré superlattice. However, in-depth analysis of this experiment is beyond the scope of the current manuscript. Next, we investigate the evolution of the low-temperature ($T = 4$ K) confocal PL spectrum as a function of the IX density (with all gates grounded). The density of optically generated IXs in our sample can be varied by changing the power of the continuous-wave excitation laser (P_{exc}) [43]. We excite resonantly to the $1s$ state of the intralayer A exciton in monolayer MoSe₂ ($\lambda = 759$ nm), and scan P_{exc} over 5 orders of magnitude. Figure 1(b) shows a color plot of the full evolution of the PL spectrum at a representative spot in the grounded heterobilayer for $0.4 \text{ nW} \leq P_{\text{exc}} \leq 80 \mu\text{W}$, while Fig. 1(c) presents line cuts extracted from Fig. 1(b) for

FIG. 1. Power dependence of moiré-trapped IXs. (a) Sketch of the dual-gated $\text{WSe}_2/\text{MoSe}_2$ heterobilayer. Graphite layers are used as contacts for top, bottom, and heterobilayer, while h -BN layers ($D = 18$ nm) are used as dielectric spacers. (b) Color plot with the full evolution of the low-temperature ($T = 4$ K) confocal PL spectrum of a representative spot in the undoped heterobilayer for $0.4 \text{ nW} \leq P_{\text{exc}} \leq 80 \mu\text{W}$ in logarithmic scale. (c) Line cuts extracted from the color plot in (b) for P_{exc} with different orders of magnitude. The spectra are shifted vertically and normalized by the values indicated in the figure for visualization purposes. (d) Schematics of the spin-valley configuration of the proposed optical transitions with the corresponding selection rules for H_h^h atomic registry [24]. (e) Magnetic field dependence of IX PL in the low- and high-excitation regimes (bottom and top panels, respectively) for σ^+ - and σ^- -resolved confocal collection (left and right panels, respectively). (f) Magnetic field dependence of the Zeeman splitting measured for IX_T^0 and IX_S^0 at high P_{exc} (top panel) and for a representative moiré-trapped IX at low P_{exc} .

P_{exc} of different orders of magnitude. At low-excitation powers ($P_{\text{exc}} \leq 10 \text{ nW}$), the PL spectrum reveals several discrete narrow emission lines with energies in the range 1.39–1.41 eV, consistent with recently reported values for neutral IXs trapped in the moiré potential landscape [25,26]. Magneto-optical studies of $\text{WSe}_2/\text{MoSe}_2$ heterobilayers with $2H$ stacking have shown that the moiré-trapped IXs arise from optical transitions involving the lowest spin-orbit-split CB of MoSe_2 at $\pm K$ [8,25,26]. This observation leads to spin-triplet optical transitions for the trapped IXs [see Fig. 1(d)]. Although such spin-flip transitions are normally forbidden in monolayer TMDs [44], they can be brightened due to the selection rules dictated by the resulting interlayer atomic registry of the heterostructures, as theoretically predicted [24] and experimentally shown [6,31,34,45].

For the lowest excitation powers employed, only a few tens of IXs are generated (approximately 10 to 40 depending

on the spatial position in the sample). Figure S1 in the Supplemental Material [38] shows the full evolution of the PL spectrum measured at a second spatial location of the heterobilayer in a similar P_{exc} range. The PL measured at both spatial locations shows the same overall behavior under increasing excitation power. First, the emission intensity of the few discrete narrow lines saturates with increasing power [see Supplemental Material Figs. S2(a) and S2(b) [38]], hallmarks of few-level quantum-confined systems. Simultaneously, as P_{exc} increases, more trapping sites are populated with IXs, and we progressively lose the ability to resolve individual spectral lines, since they merge into an IX ensemble band. The IX ensemble band blueshifts with increasing P_{exc} . At excitation powers of approximately $2 \mu\text{W}$, a second IX emission peak appears at higher energy and continuously blueshifts with increasing P_{exc} . For the highest P_{exc} used in our experiments ($80 \mu\text{W}$), the two IX peaks exhibit linewidths between 5 and 10 meV (depending

on the spatial location on the sample) and consistently exhibit an energy splitting of approximately 24 meV.

To unambiguously identify if these peaks arise from band-edge states at $\pm K$ and disentangle the spin-valley configuration of each IX emission band, we perform helicity-resolved magneto-optical spectroscopy measurements in the Faraday configuration under linearly polarized (π) excitation at 1.63 eV (759 nm). Figure 1(e) shows the magnetic field (B_z) dependence of IX PL in the low- and high-excitation regimes (bottom and top panels, respectively) for σ^+ - and σ^- -resolved confocal collection (left and right panels, respectively). A clear Zeeman splitting with increasing B_z is observed for the IX PL in both excitation regimes. The low-energy ensemble band (IX_T^0) observed at high P_{exc} shows the same polarization dependence with B_z as the few individually resolved moiré-trapped IXs. However, the high-energy ensemble band (IX_S^0) presents the opposite polarization. Figure 1(f) shows the B_z dependence of the experimental Zeeman-splitting (ΔE_z) values measured for IX_T^0 and IX_S^0 (top panel) and a representative single moiré-trapped IX (bottom panel), as extracted from Lorentzian fits of the experimental data. The Zeeman splitting is defined as $\Delta E_z(B_z) = E^{\sigma^+}(B_z) - E^{\sigma^-}(B_z) = g\mu_B B_z$, with g an effective g factor, μ_B the Bohr magneton, and $E^{\sigma^\pm}$ the B_z -dependent energies of the IX transitions with $\sigma^\pm$ polarization. Linear fits reveal g factors of -15.71 ± 0.05 , 12.24 ± 0.13 , and -15.98 ± 0.02 for IX_T^0 , IX_S^0 , and the moiré-trapped single IX, respectively. Since the carrier spin, the valley index and the atomic orbital involved in the optical transitions are associated with a magnetic moment [25,46–48], the measured g factors provide valuable information about the nature of the transitions. In this regard, the band-picture model presented in Ref. [49] has successfully predicted both the magnitude and the sign of the IX g factors in 2H- and 3R-stacked MoSe₂/WSe₂ heterobilayers [6,8,25,26,31,34] that show excellent agreement with more rigorous theoretical descriptions of the IX Zeeman shifts [50]. Taking into account the reported effective masses of $m_h^* = 0.37 m_0$ and $m_e^* = 0.84 m_0$ for holes in the topmost valence band of WSe₂ [51] and electrons in the lowest CB of MoSe₂ at $\pm K$ [52,53] (with m_0 the free-electron rest mass), we estimate theoretical g -factor values of -15.8 and 11.8 for IX_T^0 and IX_S^0 transitions, respectively [26]. The good agreement between the experimental and calculated g -factor values confirms the spin-valley configurations of both excitonic transitions and corroborates the identification of IX_T^0 and IX_S^0 in the high-excitation regime [see Fig. 1(d) for a level schematic, the optical transitions, and the corresponding selection rules [24]], in agreement with recent works [6,31,34,45]. These results bring to light a striking property of these IX transitions: Unlike counterpart monolayer TMDs [44,54], TMD heterobilayers do not host dark excitons (i.e., spin-forbidden optical transitions). In the rest of the work, we

focus on the properties of the IX ensemble emission peaks with different spin configurations (IX_T^0 and IX_S^0) observed at high IX densities.

III. DIPOLAR INTERACTIONS OF AN ENSEMBLE OF TRAPPED IXs

To understand the link between the individually resolved moiré-trapped IXs and the ensemble IX emission, we focus on the power-dependent blueshifts observed in the PL energy of both IX_T^0 and IX_S^0 . The peak blueshifts originate from the repulsive dipolar exciton-exciton interaction of IXs, which arises as a consequence of the large permanent electrical dipole moment induced by the spatial separation of the exciton carriers [55–58]. The power-dependent energy shifts for IX_T^0 (ΔE_T) and IX_S^0 (ΔE_S) can be expressed as $\Delta E_{S,T}(P_{\text{exc}}) = E_{S,T}(P_{\text{exc}}) - E_{S,T}^0$, with $E_{S,T}(P_{\text{exc}})$ and $E_{S,T}^0$ being the energy of the corresponding exciton species at P_{exc} and vanishing excitation, respectively. Such excitation-dependent energy shifts can be quantitatively estimated using the plate capacitor formula [55,56]:

$$\Delta E_{S,T}(P_{\text{exc}}) = 4\pi N_{S,T}(P_{\text{exc}})e^2 d / \epsilon, \quad (1)$$

where $N_{S,T}$ is the exciton density of the corresponding IX configuration, e is the electron charge, d is the interlayer distance, and ϵ is the dielectric constant [see Supplemental Material [38] Note 3 for the derivation and applicability of Eq. (1)]. Therefore, Eq. (1) allows us to estimate the interlayer exciton density at different excitation powers from the experimentally measured energy blueshift [$N_{S,T} = \Delta E_{S,T} \epsilon / (4\pi e^2 d)$] [7,12,55,56]. For example, from the data shown in Fig. 1(b), we estimate a $\Delta E_T \sim 0.56$ meV at $P_{\text{exc}} = 4$ nW. Assuming an $\epsilon = 7.4\epsilon_0$ [59] (with ϵ_0 the vacuum permittivity), and a d ranging from approximately 0.4 [8] to 1 nm [55], we estimate an $N_T(4 \text{ nW}) \approx (1.8 \times 10^9) - (4.6 \times 10^9) \text{ cm}^{-2}$. The estimated value of N_T suggests the presence of approximately 18–46 trapped IX_T^0 in a spot of about $1 \mu\text{m}^2$, which corresponds roughly to the area of the confocal spot in our measurements. The estimated number of trapped IX_T^0 agrees well with the magnitude of the total number of peaks that can be experimentally resolved at $P_{\text{exc}} = 4$ nW. In a similar way, from the estimated total ΔE_T of approximately 8.06 meV for IX_T^0 in Fig. 1(b), we estimate an $N_T \approx (2.6 \times 10^{10}) - (6.6 \times 10^{10}) \text{ cm}^{-2}$ at the highest excitation powers used in our experiments. For such IX densities, the IXs present an average exciton-exciton distance $\langle r_{\text{IX}} \rangle \sim 42\text{--}67$ nm (see Supplemental Material [38] Note 4 for more details). The estimated $\langle r_{\text{IX}} \rangle$ in our experiments is of the same order of magnitude as the $\langle r_{\text{IX}} \rangle$ inferred in other 2D excitonic systems for which dipolar interactions are typically observed, for example, indirect excitons in III-V quantum wells [56] and MoSe₂/WSe₂ heterobilayers [7,12,55].

The estimated values of N_T and $\langle r_{\text{IX}} \rangle$ shed some light on the nature of the IX PL emission bands observed at high

P_{exc} . The stacking angle in our heterostructure ($\Delta\theta = 56.4 \pm 0.2^\circ$ [8]) gives rise to an estimated density $N_{\text{total}} \sim 1.6 \times 10^{13} \text{ cm}^{-2}$ of moiré-trapping sites with three different local atomic configurations: H_h^h , H_h^X , and H_h^M [22], where H_h^h denotes an H -type stacking with either h the hexagon center, X the chalcogen site, or M the metal site vertically aligned with the hexagon center (h) of the hole layer. N_{total} yields a density $N_{\text{Moiré}} = N_{\text{total}}/3 \sim 4.2 \times 10^{12} \text{ cm}^{-2}$ of moiré sites with the same atomic registry. Therefore, our analysis reveals that the estimated density of optically generated IXs (N_{IX}) is around 2 orders of magnitude smaller than $N_{\text{Moiré}}$; less than 2% of the moiré sites are filled with IXs. We note that the highest N_{IX} achieved in our experiments corresponds to the lowest N_{IX} investigated by Wang *et al.*, who reported an optically driven Mott transition from IXs to a charge-separated electron and hole plasmas in a $3R$ -MoSe₂/WSe₂ heterobilayer with a similar moiré period ($\Delta\theta = 4^\circ$) for $N_{\text{IX}} > 3 \times 10^{12} \text{ cm}^{-2}$ (i.e., for $N_{\text{IX}} > N_{\text{Moiré}}$) [43].

In order to provide an additional degree of confidence in the IX density estimated from Eq. (1), we estimate the magnitudes of N_T and N_S from the power-dependent results shown in Fig. 1(b). Figure 2(a) shows the ratio of integrated PL intensities between the IX_T^0 and IX_S^0 peaks as a function of P_{exc} extracted from Fig. 1(b) (black dots). The inset shows the evolution of the integrated PL intensities for each individual peak in the same P_{exc} range. In the range of P_{exc} for which both peaks coexist, we observe that the intensity ratio decreases from approximately 105 to approximately 5 with increasing P_{exc} . To evaluate the PL intensities of the two IX species peaks quantitatively, we employ a rate equation model (see Supplemental Material [38] Note 5). The solid lines in Fig. 2(a) represent fits of the experimental data for both the PL intensity ratio (red) and the PL intensities of the IX_T^0 (blue) and IX_S^0 (green) peaks to the rate equation model. The fits allow us to confidently estimate the power-dependent relative densities of IXs with spin-singlet and spin-triplet configurations. Next, we show in Fig. 2(b) the experimentally measured energy splitting between IX_S^0 and IX_T^0 (ΔE_{S-T}), where we observe that increasing P_{exc} gives rise to a reduction of ΔE_{S-T} by up to 3 meV, well beyond the uncertainty associated with the experimental determination of the energy splitting. From Eq. (1), the density-dependent ΔE_{S-T} can be calculated as

$$\Delta E_{S-T} = \Delta E_{S-T}^0 + \Delta N_{S-T}(P_{\text{exc}})4\pi e^2 d/\epsilon, \quad (2)$$

with $\Delta E_{S-T}^0 = E_S^0 - E_T^0$ and $\Delta N_{S-T}(P_{\text{exc}}) = N_S(P_{\text{exc}}) - N_T(P_{\text{exc}})$. From Eq. (2), it is straightforward to see that the decrease of ΔE_{S-T} with increasing P_{exc} has its origin in the higher density of IX_T^0 in the range of P_{exc} used in our experiments, which leads to $\Delta N_{S-T}(P_{\text{exc}}) < 0$. Figure 2(b) shows the fit of the experimental ΔE_{S-T} (black dots) to Eq. (2) (red solid line), where we use the relative IX

densities shown in Fig. 2(c) and assume an average d value of 0.7 nm [12]. The inset shows a comparison of the experimental and calculated energy shift for each individual exciton band, where the shaded areas represent the confidence interval corresponding to energy shifts calculated using d values ranging from 0.4 [8] to 1 nm [55]. The good agreement between the experimental and calculated values allows us to estimate the order of magnitude of the absolute densities for IXs with both spin configurations, which are shown in Fig. 2(c).

The black dashed line in Fig. 2(c) shows the estimated $N_{\text{Moiré}}$ for our sample. In agreement with the analysis from the plate-capacitor approximation [Eq. (1)], the results in Fig. 2(c) corroborate that, even for the highest excitation powers used in our experiments, the estimated densities of optically generated IXs are around 2 orders of magnitude smaller than $N_{\text{Moiré}}$. The orange shaded area in Fig. 2(c) represents the estimated $N_{\text{Moiré}}$ for MoSe₂/WSe₂ heterobilayers with stacking angles between 54.4° and 58.4° , showing that this conclusion holds true even for a large range of stacking angles.

Our results represent a bridge between the quantum-dot-like [8,25,26] and the broad PL emission peaks [4–7, 16,29,31] previously observed for IX ensembles in MoSe₂/WSe₂ heterobilayers at different excitation powers. Figure 2(d) shows a sketch depicting the transition between the low- and high-density regimes. At low P_{exc} , only a few IXs (red spheres) are localized in the moiré sites lying inside our circular confocal collection spot (not to scale). The spatial and spectral isolation of these trapped excitons is likely aided by dipolar repulsion, which minimizes the probability for excitons to populate neighboring moiré sites. As P_{exc} increases, more and more optically generated IXs are trapped in the moiré sites, giving rise to a broad PL emission band originating from the ensemble of moiré-trapped IXs. Even at the highest P_{exc} used in this work, less than 2% of moiré sites contain trapped IXs. This behavior agrees well with the magneto-optical properties measured for single confined IXs and the IX_T^0 ensemble PL band. The polarization selection rules of moiré-trapped excitons are dictated by the local atomic registry of the moiré trapping site [22,24]. The same optical selection rules observed for both single confined IXs and the IX_T^0 ensemble PL band indicate that only moiré sites with a local atomic registry H_h^h are responsible for the IX trapping [8,25,26,45]. Moreover, the optical selection rules of the IX_S^0 ensemble PL peaks further corroborate this affirmation, since H_h^h is the only local atomic registry in $2H$ -stacked TMD heterobilayers that results in opposite circularly polarized transitions for IXs with spin-triplet and spin-singlet configurations [24]. We note that for MoSe₂/WSe₂ heterobilayers with $3R$ stacking (i.e., $\Delta\theta \sim 0^\circ$), the magneto-optical selection rules suggest that the PL emission at high-excitation power

FIG. 2. Dipolar interactions of an ensemble of trapped IXs. (a) Ratio of integrated PL intensities between the IX_T^0 and IX_S^0 peaks (IX_T^0/IX_S^0) as a function of P_{exc} extracted from Fig. 1(b) (black dots). The inset shows the evolution of the integrated PL intensities for each individual peak in the same P_{exc} range. The red, blue, and green solid lines represent fits of the experimental data to the rate equation model described in the Supplemental Material [38]. (b) Energy splitting between IX_S^0 and IX_T^0 (ΔE_{S-T}) as a function of the excitation power (black dots). The inset shows the energy shifts of the individual peaks. The red solid line represents a fit of the experimental ΔE_{S-T} to Eq. (2), while the green and blue solid lines are fits of the measured ΔE_{S-T} to Eq. (1), and the shaded areas represent the uncertainty interval corresponding to d values ranging from 0.4 [8] to 1 nm [55]. (c) Estimated power-dependent densities of IXs with spin-singlet (green) and spin-triplet (blue) configurations. The thickness of the lines indicates the confidence interval estimated from the fits to the experimental data. The black dashed line indicates the density of moiré traps estimated for our heterostructure, while the orange shaded area represents the density of sites for MoSe₂/WSe₂ heterobilayers with stacking angles between 54.4° and 58.4°. (d) Sketch depicting the transition between the low- and high-excitation power regimes for IXs (red spheres) trapped in the moiré sites of a MoSe₂/WSe₂ heterobilayer. At low P_{exc} , only a few IXs are localized in the moiré sites lying inside the circular confocal collection spot (not to scale). As P_{exc} increases, more and more optically generated IXs are trapped in the moiré sites, giving rise to a broad PL emission band originating from an ensemble of moiré-trapped IXs. Even at the highest P_{exc} used in this work, less than approximately 2% of moiré sites contain trapped IXs.

arises from spin-singlet and spin-triplet IXs with a local stacking registry R_h^X [6,34].

IV. SPIN-VALLEY CONFIGURATIONS OF NEGATIVE IX TRIONS IN 2H-MoSe₂/WSe₂

In this section, we investigate the formation of negatively charged trapped IXs with both spin-triplet and spin-singlet configuration. The Fermi energy in our WSe₂/MoSe₂ heterobilayer can be continuously tuned with an external gate voltage V_g . The color plots of Figs. 3(a) and 3(b) show the effects of electron doping on the PL of the trapped IXs at low- ($P_{\text{exc}} = 20$ nW) and high- ($P_{\text{exc}} = 40$ μ W) excitation powers, respectively. The V_g -dependent evolution of

the IX PL shows the same overall behavior for both low- and high-IX-density regimes, and resembles the doping dependence of the intralayer A exciton of monolayer MoSe₂ measured in a sample region where the MoSe₂ does not overlap with the WSe₂ layer (see Supplemental Material [38] Note 6). For $0 < V_g \lesssim 0.1$ V, the PL spectrum is dominated by neutral excitons: IX_T^0 at low IX density [Fig. 3(a)], and both IX_T^0 and IX_S^0 at high IX density [Fig. 3(b)]. At high IX density, the IX_T^0 and IX_S^0 ensemble peaks present linewidths of approximately 5.2 and 7 meV, respectively, on par with the cleanest MoSe₂/WSe₂ samples [6,16,30]. For $V_g \gtrsim 0.1$ V, new redshifted peaks appear at energies approximately 7 (approximately 4) meV below the IX_T^0 (IX_S^0) peaks, indicating the formation of

FIG. 3. Interlayer exciton trions in WSe₂/MoSe₂. (a),(b) Gate-voltage-controlled PL of IXs in the neutral ($0 < V_g \leq 0.1$ V) and electron-doping regime ($V_g \geq 0.1$ V) at $B_z = 0$ T, for $P_{\text{exc}} = 20$ nW (a) and $P_{\text{exc}} = 40$ μ W (b) at 4 K. In (b), the PL intensity is multiplied by a factor 20 in the spectral range delimited by the vertical dashed area for visualization purposes. (c) Line cuts extracted from the color plots in (a) and (b) (black and red solid lines, respectively) in the neutral ($V_g = 0$ V) and electron-doping regimes ($V_g = 0.6$ V). The spectra are normalized by the values indicated in the figure, and the line cuts at $V_g = 0.6$ V are shifted vertically for visualization purposes. The dashed vertical lines indicate the central energies of the different ensemble peaks, illustrating the power-induced blueshift [$\Delta E(P_{\text{exc}})$] and the energy splittings arising from the binding energies of the charged IXs with spin-triplet (ΔE_{IX_T}) and spin-singlet (ΔE_{IX_S}) configuration.

negatively charged IX trions with different spin configurations [see line cuts in Fig. 3(c)]. The measured energy differences between the neutral and charged exciton peaks are attributed to the binding energies of the charged IXs, in good agreement with recently reported values for MoSe₂/WSe₂ heterostructures with 3R stacking [7,34]. The new trion peaks coexist with IX_T^0 and IX_S^0 for a short range of applied voltages until IX_T^0 and IX_S^0 eventually vanish with increasing V_g . We note the observation of IX trions in the quantum emitter (low-excitation) regime is novel. At high-excitation powers, the PL spectrum in the n -doped region shows broad emission shoulders in both the low- and high-energy tails of the IX trion peak with spin-triplet configuration. In order to spectrally resolve the emission bands, we plot the PL of IXs for n doping ($V_g = 2$ V) as a function of P_{exc} [see Fig. 4(a)]. Figure 4(b) shows the PL spectrum acquired for an intermediate P_{exc} value of 2 μ W at $V_g = 2$ V, as indicated by the white dashed line in Fig. 4(a). At this excitation power, we clearly resolve four emission peaks. The brightest peak in the spectrum (at approximately 1.39 eV) corresponds to intervalley IX trions with spin-triplet optical transitions ($\text{IX}_{T,\text{inter}}^-$). At higher emission energies, we observe two additional peaks with an energy splitting of approximately 12 meV. These two peaks show a similar integrated PL intensity across the entire range of P_{exc} (see Supplemental Material [38] Note 7), suggesting that their corresponding charge configurations involve the same band-edge energy levels. We attribute the low-energy and high-energy peaks of this doublet to intravalley ($\text{IX}_{T,\text{intra}}^-$) and intervalley ($\text{IX}_{S,\text{inter}}^-$) trions with spin-triplet and spin-singlet configuration, respectively. Figure 4(c) shows a schematic representation of the charge

configurations for $\text{IX}_{T,\text{inter}}^-$ (left), $\text{IX}_{T,\text{intra}}^-$ (middle), and $\text{IX}_{S,\text{inter}}^-$ (right) with optical transitions that involve a hole in the topmost valence band of WSe₂ at K .

The PL spectrum in Fig. 4(b) also shows a broad emission peak at lower energies than $\text{IX}_{T,\text{inter}}^-$ (IX_T^-). Although its origin is not yet fully understood, this feature has also been recently observed in the reflectivity of n -doped monolayer WSe₂ [60,61] and the PL of n -doped MoS₂ [62,63] and attributed to either a Mahan-like exciton [62] or an exciton-plasmon-like excitation [60,61]. We find that contrary to the behavior of $\text{IX}_{T,\text{inter}}^-$, $\text{IX}_{S,\text{inter}}^-$, and $\text{IX}_{T,\text{intra}}^-$, the integrated emission intensity of IX_T^- increases with the carrier concentration. Moreover, increasing the electron concentration also leads to a linear increase of the energy splitting between IX_T^- and $\text{IX}_{T,\text{inter}}^-$ (see Supplemental Material [38] Note 8).

Finally, the charge configurations for the different charged exciton species depicted in Fig. 4(c) highlight a striking difference between $\text{IX}_{S,\text{inter}}^-$ and $\text{IX}_{T,\text{intra}}^-$. Unlike $\text{IX}_{S,\text{inter}}^-$, in which the photon emission arises from electron-hole recombination between the topmost VB and the top spin-split CB, the absence of a dark exciton ground state in MoSe₂/WSe₂ heterostructures enables the electron-hole recombination between the topmost VB and the bottommost spin-split CB for $\text{IX}_{T,\text{intra}}^-$. This behavior contrasts with negative trions in W-based TMDs, in which both intravalley and intervalley negative trions present electron-hole recombination involving the same top spin-split CB [64]. Moreover, since $\text{IX}_{S,\text{inter}}^-$ is the only trion species in which the electron-hole recombination involves the top spin-split CB, the V_g -dependent relative integrated PL intensities of

FIG. 4. Interlayer exciton trions in $WSe_2/MoSe_2$. (a) Color plot with the full evolution of the PL spectrum of negative IX trions for $0.2 \text{ nW} \leq P_{exc} \leq 40 \mu W$ at $V_g = 2$ V in logarithmic scale, in which four different peaks can be resolved (as indicated by the arrows). (b) PL spectrum acquired for the intermediate P_{exc} value indicated by the white dashed line in (a) ($2 \mu W$) at $V_g = 2$ V, fitted to four Lorentzian peaks corresponding to various exciton species: $IX_{T,inter}^-$ (blue), $IX_{T,intra}^-$ (gray), $IX_{S,inter}^-$ (green), and $IX_{T'}^-$ (red). (c) Schematic representation of the charge configurations for $IX_{T,inter}^-$ (left), $IX_{T,intra}^-$ (middle), and $IX_{S,inter}^-$ (right) showing the optical transitions that involve a hole in the topmost valence band of WSe_2 at K . (d) Magnetic field dependence of the IX trions PL for σ^+ - (purple) and σ^- -polarized (orange) collection in the range $0 \leq B_z \leq 5$ T under $V_g = 1$ V and $P_{exc} = 40 \mu W$. (e) Zeeman splitting of each IX trion species (dots) as extracted from fits of the experimental data shown in (d). The solid lines represent linear fits of the experimental data.

the different trion species should provide an estimate of the energy splitting between the spin-split CBs in $MoSe_2$ [34]. We observe that the PL intensity from $IX_{S,inter}^-$ overtakes the combined PL intensity from $IX_{T,inter}^-$ and $IX_{T,intra}^-$ at $V_g \sim 6.1$ V (see Supplemental Material [38] Note 9), which corresponds to an energy splitting of approximately 21 meV, in good agreement with the calculated spin-orbit splitting of $MoSe_2$ CBs at $\pm K$ (23 meV) [65]. Therefore, the spin-valley configurations depicted in Fig. 4(c) for the different trion states suggest that, contrary to intravalley negative trions in W-based TMDs, although $IX_{S,inter}^-$ and $IX_{T,intra}^-$ involve the topmost CB, they should present different magneto-optical properties.

To investigate the magneto-optical properties of the different trion peaks, we measure their PL as a function of B_z at $P_{exc} = 40 \mu W$ and $V_g = 1$ V, for which both the intensity and linewidth of the different peaks allow us to clearly monitor their emission properties. Figure 4(d)

shows the magnetic field dependence of the IX trions PL for σ^+ - (purple) and σ^- -polarized (orange) collection in the range $0 \leq B_z \leq 5$ T. The results reveal a clear Zeeman splitting for the three IX trion species: A positive vertical B_z field leads to a redshift of the σ^+ -polarized emission for both $IX_{T,inter}^-$ and $IX_{T,intra}^-$, while it induces a slight blueshift of the σ^+ -polarized emission for $IX_{S,inter}^-$. Such contrasting behavior can also be seen in the experimental Zeeman splitting of each IX trion species as shown in Fig. 4(e): $IX_{T,inter}^-$ and $IX_{T,intra}^-$ present negative slopes (negative g factor), whereas $IX_{S,inter}^-$ exhibits a positive one. Linear fits of the measured Zeeman splittings reveal g factors of -15.44 ± 0.06 , -15.1 ± 0.4 , and 12.31 ± 0.11 for $IX_{T,inter}^-$, $IX_{T,intra}^-$, and $IX_{S,inter}^-$, respectively. The extracted values show that $IX_{T,inter}^-$ and $IX_{S,inter}^-$ have g factors with opposite signs and different magnitudes, as previously observed for IX_T^0 and IX_S^0 in the results shown in Fig. 1(f). More importantly, the fits also reveal that $IX_{T,inter}^-$ and $IX_{T,intra}^-$

present g factors with the same sign and very similar magnitude, which confirms that the electron-hole recombination in these trions involve the same CB and VB states, confirming the origin of the $\text{IX}_{T,\text{intra}}^-$ peak. We note that the particular band alignment and optical selection rules in $2H\text{-MoSe}_2/\text{WSe}_2$ heterobilayers allow the formation of intervalley and intravalley IX trions by optical pumping of both the upper and lower CBs of MoSe_2 even in the neutral-doping regime (see Supplemental Material [38] Note 10). These results suggest the importance of revisiting the interpretation of multiple IX emission peaks in previous reports. Finally, the fits reveal a dependence of the trions' g factors with the carrier concentration. Figure S7 in the Supplemental Material [38] shows the evolution of the g factor of the brightest peak in our spectra ($\text{IX}_{T,\text{inter}}^-$) as a function of the electron doping. Similar to the case of the g factor of quasiparticles in a 2D electron gas [66] and intralayer exciton polarons in monolayer TMDs [67–69], we observe a change of g with increasing electron doping. This change in the effective g factor with increasing carrier doping has been attributed to many-body interactions and phase-space filling effects [68,69].

Figure 4(e) also shows the measured Zeeman splitting for the excitonic feature IX_T^- . The observed negative slope in the Zeeman splitting of this peak with increasing B_z reveals similar selection rules to $\text{IX}_{T,\text{inter}}^-$. However, similar to the exact origin of IX_T^- , the extracted g factor of -25.3 ± 0.5 is not yet understood. Finally, we note a quadratic redshift of the central energy of the Zeeman-split peaks of the IX ensemble peaks with increasing magnetic field (see Supplemental Material [38] Note 12), which is absent for low IX densities, highlighting the importance of the strong exciton-exciton interactions at high IX densities.

V. CONCLUSION AND PERSPECTIVES

In summary, we investigate neutral and negatively charged IXs trapped in moiré confinement potentials in a gate-tunable $2H\text{-WSe}_2/\text{MoSe}_2$ heterostructure. The homogeneous linewidths of the ensemble IX peaks we find in our sample (approximately 5 meV for neutral and charged species) are on par with the best reported, confirming the high-quality nature of the sample thanks to $h\text{-BN}$ encapsulation and control of the Fermi level. By scanning the excitation power over 5 orders of magnitude, we tune the density of optically generated neutral excitons and observe a continuous evolution of the trapped IX density, from a few isolated quantum emitters to a large ensemble. In the high-excitation regime, neutral IXs with spin-triplet (IX_T^0) and spin-singlet (IX_S^0) configurations are identified. The IX ensemble peaks energetically blueshift with increasing density due to dipolar repulsion. From this effect, we are able to estimate N_{IX} and find that even at the highest IX density we measure, $N_{\text{IX}} \ll N_{\text{moiré}}$. We discover that the magneto-optical properties of the trapped IXs are identical, regardless of their density: Both the narrow

quantum-dot-like and broad ensemble IX emission originate from IXs localized in moiré potential traps with the same local atomic registry (H_h^h). Moreover, the optical selection rules of the IX_S^0 ensemble further corroborate this affirmation, since H_h^h is the only local atomic registry in $2H$ -stacked TMD heterobilayers that results in opposite circularly polarized transitions for IXs with spin-triplet and spin-singlet configurations.

Next, the Fermi energy is tuned to create negatively charged moiré-trapped IXs, which we observe in both the low- and high-IX-density regimes. To our knowledge, the observation of charged moiré-trapped IXs in the low-density regime is novel, and an exciting avenue for future investigations. Binding energies for the on-site negatively charged IXs are found to be 7 meV (4 meV) for the spin-triplet (spin-singlet) trion configuration. Our magneto-optical measurements at high IX densities reveal a fine structure for negative IX trions with spin-triplet configuration; we clearly resolve intravalley ($\text{IX}_{T,\text{intra}}^-$) and intervalley ($\text{IX}_{T,\text{inter}}^-$) IX trions. We note that, using moderate excitation powers, the formation of intervalley and intravalley IX trions is observed even in the neutral-doping regime, which creates a multi-peaked PL spectrum consisting of six possible neutral and charged IX species (IX_T^0 , IX_S^0 , $\text{IX}_{T,\text{intra}}^-$, $\text{IX}_{T,\text{inter}}^-$, $\text{IX}_{S,\text{intra}}^-$, and IX_T^-). These results suggest it could be fruitful to revisit previous interpretations of multiple-peaked IX spectra from $\text{MoSe}_2/\text{WSe}_2$ heterobilayers in previous reports, particularly those using ungrounded devices and high-excitation powers.

We remark that the unified picture of narrow-linewidth IX emitters and broad ensemble IX peaks reconciles contrasting IX spectra reported from nominally similar $\text{WSe}_2/\text{MoSe}_2$ heterobilayer samples. This unified picture provides further valuable evidence about the nature of quantum emitters in moiré heterostructures, complementing previous results [8,25,26]. So, although the narrow-linewidth peaks have an inhomogeneous energy distribution similar to defect-related single-photon emitters in 2D materials, the properties of the moiré quantum emitters are identical to the ensemble IX and arise from the intrinsic symmetry of the moiré confining potential at the specific atomic registry. Further, in the low-excitation regime, spatial isolation of trapped IXs is likely aided by dipolar repulsion, which minimizes the probability for excitons to populate neighboring moiré sites. Finally, in the high-excitation regime, the results highlight the importance of considering the spatial pinning of the ensemble IXs to the moiré lattice in order to achieve an accurate description of many-body exciton-exciton phenomena in TMD heterobilayers: The dipole interactions will depend on, and can be tuned by, the twist angle.

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Note added in proof.—Recently, two related works on the topic of moiré-trapped interlayer trions have become available [70,71].

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