Flipping exciton angular momentum with chiral phonons in MoSe$_2$/WSe$_2$ heterobilayers

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Abstract
Optical selection rules in monolayers of transition metal dichalcogenides and of their heterostructures are determined by the conservation of the z-component of the total angular momentum—$I_z = L_z + S_z$—associated with the $C_3$ rotational lattice symmetry which assumes half integer values corresponding, modulo 3, to distinct states. Here we show, based on polarization resolved and low temperature magneto-optical spectroscopy experiments, that the conservation of the total angular momentum in these systems leads to a very efficient exciton–phonon interaction when the coupling is mediated through chiral phonons. We identify these phonons as the $\Gamma$ point $E''$ modes which despite carrying angular momentum $\pm 1$ are able to induce an excitonic spin-flip of $\mp 2$ thanks to the $C_3$ symmetry. These experiments reveal the crucial role of electron-phonon interaction in the carrier dynamics of group 6 transition metal dichalcogenides.

A useful frame to understand the optical properties of group VI transition metal dichalcogenide (TMDs) monolayers is to consider spin conserving transitions, i.e. recombination of excitons with composing carriers of identical spin [1]. Using spin as the quantum number to describe optical transitions in TMD monolayers, excitons have been classified as bright for spin singlets (parallel spin configuration of electron and hole) and as dark for spin triplets (anti-parallel spin configuration) [2, 3]. Recently has it been realized that the so-called dark excitons emit light that propagates within the layer plane and can be observed using an adequate experimental configuration [4–6]. Following this approach of spin conserving transitions, one also definitely fails to describe interlayer optical transitions in heterobilayers elaborated from distinct individual TMD monolayers [7–9].

Optical emission of interlayer exciton (IX) in TMD heterostructures [10–12] is in first order allowed only for two particular alignment angles, i.e. $\theta \approx 0^\circ$ or $\theta \approx 60^\circ$ [13]. For $\theta = 0^\circ$ the lowest energy exciton in a WSe$_2$/MoSe$_2$ heterobilayer, is a singlet state where the spin of the electron and the hole is conserved during the recombination process. The situation is different for the recombination of IX where the constituent layers have been rotated by 60°. In this case, the large extracted g-factor of $|g| = 16$ [7, 14] is only compatible with a triplet state of the exciton [15, 16]: the involved electronic bands have anti-parallel spin configuration and one would not expect this exciton to be optically bright. The optical activity of such IX is a consequence of the conservation of the total angular momentum as it was recently described by theoretical calculations [17, 18]. The recent observation of chiral phonons in the optical response of TMDs [19, 20] evidences that the total angular momentum is a key point to understand their physical properties but has not been appreciated so far.

Here, based on polarization resolved and low temperature magneto-PL measurements, we illustrate the crucial role of electron–phonon interaction in the carrier dynamics of group 6 transition metal dichalcogenides.
is their total angular moment $J_z$ which provides a correct and extensive description of the light–matter interaction in this class of materials, including in their heterobilayers. Indeed, the dominant part of spin-orbit interaction in TMDs is proportional to $L_z S_z$ and does conserve separately $L_z$ and $S_z$, the components of the orbital and spin angular momentum, perpendicular to the layer plane. Still, a weak residual part of the spin-orbit interaction, allowed by the crystal symmetry, mixes the different spin states and only the total angular momentum $J_z = L_z + S_z$ is a good quantum number. This picture naturally explains why interlayer spin triplet excitons are optically active. A direct consequence of the conservation of the total angular momentum in TMD heterobilayer is revealed by a particularly efficient exciton-phonon coupling mechanism between IX in van der Waals (vdW) heterostructures made of monolayer WSe$_2$ and MoSe$_2$ and chiral optical phonons [22] of the individual constituents, which would not be possible if spin would have been the correct quantum number of the system. The exciton-chiral phonon interaction has been recently described for intralayer excitons in monolayers of WSe$_2$ [19, 20], we here show its effect on interlayer excitons in the more complexe system of heterobilayers of TMDs. This magneto-phonon resonance effect is observed when the Zeeman energy is tuned to the optical phonon energy and is responsible for an extremely efficient relaxation of excitons into the lower Zeeman component when the Zeeman energy of the IX precisely matches the energies of the non-Raman active $E''$ optical phonon modes of WSe$_2$ and of MoSe$_2$. Finally, thanks to the finesse of the exciton-chiral phonon interaction we can further disclose that the underlying potential landscape in the heterobilayer caused by the twist angle between the monolayers affects the properties of the IX through an energy dependent $g$-factor.

Two different heterobilayer samples consisting of stacked MoSe$_2$ and WSe$_2$ monolayers encapsulated in hexagonal Boron Nitride (h-BN) using a dry transfer technique [23] have been fabricated (see supplementary materials (stacks.iop.org/2DM/7/041002/mmedia)). Optical photographs are presented in figure 1(a) and (b). The schematics of the structure is illustrated in the inset of figure 1(c). The rotation angle between the monolayers is of 60°, as verified below by their
excitonic g-factor [14]. The benchmark of both samples is the presence of a peak at 1.4 eV seen in figure 1(c), in addition to the emission peak of intralayer excitons of MoSe\(_2\) and WSe\(_2\) at higher energy [12] (for details of the experimental setup, see the Methods section at the end of the manuscript). The emission peak shown in figure 1(c) is the direct consequence of the type-II band alignment between MoSe\(_2\) and WSe\(_2\) [12, 24–26] and the experimental signature of the recombination of interlayer excitons. It is worth noting that the full width at half maximum (FWHM) of the main peak of 4 meV (sample S1) is close to the intrinsic linewidth of intralayer excitons in TMD monolayers [27] and the differences in the spectrum between both samples can be attributed to varying electrostatic potential landscapes. In agreement with previous reports [14, 28], the IX line shape evolves when the laser power is lowered down in figure 1(d) and (the) narrow emission lines observed in the spectrum of 50 nW excitation originate from excitons trapped in the lateral moiré potential of the heterobilayer (virtually identical results are presented in the Supplementary Information for sample S1).

Figure 2(a) displays the polarization resolved PL spectra of the heterostructure in sample S1 for magnetic fields of 0, 7 and 14 T. In agreement with previous results, the intralayer exciton in MoSe\(_2\) are characterized by a Landé factor \(g \sim -4\) [29] while a significantly larger value is found for the interlayer exciton with \(g \sim -16\). As shown in figure 2(b), the Zeeman effect acting on the IX remains robust up to 30 T and all sharp features accompanying the main PL line follow the same magnetic field dependence as the bright main transition. This means that all features that we observe in this spectral region originate from the energetically most favorable region with the smallest interlayer distance [30] (AB-stacking in our case). This stacking implies that the \(\pm\) K points of one layer are aligned with the \(\mp\) K points of the other layer [31]. The large value of the g-factor of the interlayer exciton is fully consistent with values reported previously in the literature for 60° aligned MoSe\(_2\)/WSe\(_2\) vdW heterostructures [7] and very recent reports have thoroughly identified that the lowest energy exciton with a \(g\)-factor of \(g \sim -16\) belongs to an excitonic triplet state [15, 16].

In order to understand the bright recombination of the spin triplet state, we focus our attention to the schematics of the band structure presented in figure 2(c). Because of the strong spin–orbit coupling in TMDs we associate to each band the total angular momentum \(J_z\) of the different optical excitations. This implies that \(J_z = +3/2\, \mp -3/2\). The light dashed lines indicate the position of the band edges in absence of magnetic field.
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Figure 3. Valley relaxation due to exciton-phonon coupling (a), (b) False color map of the circularly polarized PL emission from IX of S1 measured with a resolution of 10 mT in the vicinity of the two identified resonant magnetic fields. (c), (d) Peak position and intensity (proportional to the size of the symbol) of the polarized IX emission. The excitonic Zeeman splitting is tuned in resonance with two characteristic energies of 22.3 and 22.95 meV for which a very efficient thermalization of the excitons towards the lowest energy component is observed. The inset in panel d sketches the thermalization between the valleys mediated with help of a chiral phonon with \( L = \pm 1 \). (e) Zeeman splitting of the IX as function of the magnetic field, which shows that the energy of the chiral phonons in WSe\(_2\) and MoSe\(_2\) are crossed between 24 and 25 T.

The giant g-factor associated with IX in 60° aligned MoSe\(_2\)/WSe\(_2\) heterobilayer allows one to tune the Zeeman energy up to few tens of meV with magnetic fields available in high magnetic fields infrastructures. In figure 2(b), one can see close to \( B = 24 \) T a nearly complete extinction of the high energy component of the IX with \( \sigma^- \) polarization (indicated by a white arrow). In figures 3(a) and (b) we explore this range of magnetic fields with a much higher field resolution of 10 mT and it appears that the \( \sigma^- \) polarized IX emission disappears at two distinct values of the magnetic field, close to \( B = 24.0 \) T and at \( B = 24.6 \) T for S1. Simultaneously, as shown in figure 3(c) and 3(d) for each extinction respectively, the intensity of the lower energy \( \sigma^+ \) component grows and decreases back to its initial value for higher magnetic fields. The excitonic emission does not show any significant energy shift nor broadening during the resonance. Its intensity vanishes in the \( \sigma^- \) branch at these two precise values of the magnetic field for which a very efficient relaxation of the IX is obtained. The resonant magnetic fields correspond to an excitonic Zeeman energy of \( E_Z = 22.30 \) meV and 22.95 meV, carry zero total angular momentum and thus can couple to photons polarized perpendicularly to the monolayer plane [5]. The experimentally observed IX in figures 2(a) and (b) appears to correspond to the IX\(_{-1}\) exciton (green arrow in figure 2(c)). The change of angular momentum for this exciton is \( \Delta J_z = +2 \equiv -1 \) allowing for the creation of a \( \sigma^- \) polarized photon and the Zeeman effect increases the transition energy by \( 8\mu_B B \) when applying a magnetic field, raising a value for the factor of the IX\(_{-1}\) close \( g \sim -16 \) (the negative sign shows that the \( \sigma^- \) component increases in energy under magnetic field). The IX\(_{+1}\) exciton, not observed in our low temperature experiment, would be \( \sigma^+ \) polarized with a g-factor close to \( g \sim +12 \) [15, 16]. Additionally, magneto-optical experiments performed in Voigt geometry, up to \( B = 29 \) T applied in the plane of the sample, did not show any evidence of magnetic brightening [4, 37] of any other IX transition (see Supp. Info.).
respectively. Striking is the narrow field interval where the energy relaxation occurs: its width of ~ 150 mT corresponds to the change in $E_g$ of 150 μeV, as presented in figures 3(a) and (b). This indicates a resonance with a mode with a very well defined energy, which couples IX excitons of different polarizations (i.e. from different valleys). Following our precedent discussion, such a mode reverses also the IX’s angular momentum.

A natural candidate for this mechanism is the emission of $E_2$ chiral in-plane optical phonons, i.e., phonons at the $\Gamma$-point carrying an angular momentum such that $L_z = \pm 1$ [22]. These types of phonon are known to exist at the high symmetry points $K$ and $\Gamma$, where the chirality originates whether from a collective circular motion of the chalcogene atoms around the center of the hexagon in real space [38, 39] or from the superposition of the motion of the degenerated LO and TO modes [22], respectively. K point chiral phonons can transfer both angular momentum $\pm 1$ and a large K momentum creating for instance phonon replicas of dark excitons in monolayers of WSe$_2$ [40]. $\Gamma$ point chiral phonons are known to efficiently couple to excitons in TMD [41], limiting their lifetime as well as the valley coherence. In particular, they produce replicas in the emission spectrum of quantum dot like structures in WSe$_2$ [42]. They are not Raman active in monolayers and we could not detect them in our heterobilayers (see Supplementary Information section S6). The angular momentum $L_z = \pm 1$ [22, 38, 39] carried by this phonon can however be transferred to the electronic system, as sketched in the inset of figure 3(d).$^1$

A similar situation has been also observed in the case of $E_{2g}$ phonons in graphene or in graphite [43–45]. The resonant energies observed experimentally correspond to a single phonon and hence, two-phonon processes involving two K-point chiral phonons can be neglected. The $E''$ phonon energy at the $\Gamma$-point in TMDs, is close to 21.8 meV in WSe$_2$ [46] and to 21.1 meV in MoSe$_2$ [47]. These values are slightly lower than the ones observed but are known to be slightly affected by details of the experimental conditions. The high magnetic fields required to reach this resonance prevent any direct application of this effect but this very efficient scattering mechanism can play a crucial role enabling energy level lifetime engineering in optical devices.

The same phenomenon is observed in sample S2 as shown in figure 4(a). Due to the slightly smaller $g$ factor for S2, the resonant magnetic fields for this sample are $B = 24.25$ T and $B = 24.9$ T (see figure 4(a)). In contrast to S1, the resonance occurs over a broader range of magnetic fields due to different emission components composing the IX emission feature [14, 48], but appears in the form of the specific diagonal feature seen in figure 4(a). The exciton-phonon resonance remains nevertheless extremely sharp for each emission peak composing the broad IX emission, still occurring over a variation of magnetic field as small as 100 mT. Our interpretation of this effect is that each of these components corresponds to an exciton trapped in a fluctuating potential with spatially dependent amplitude. Assuming for S2 the same phonon energies as in sample S1, the finesse of this exciton-phonon coupling enables to resolve the energy dependence of the excitonic $g$-factor, whose slope is $\partial g/\partial E \approx 7.5$ and 8.5 eV$^{-1}$ for the two resonant magnetic fields, respectively. We also present in figure 4(b) the values of $g$-factor extracted close to $B = 24.2$ T together with the zero field IX spectrum (an alternative analysis can be found in the Supplementary Information section S9-10).

$^1$The angular momentum transfer necessary to flip the excitonic spin is $\Delta J_z = \pm 2$ because both the electron and the hole should change their spin. $E''$ phonons can only exchange a unit of angular momentum but because of the $C_3$ symmetry of the lattice, the angular momentum space is defined modulo 3 and this change $\Delta J_z = \pm 1$ angular momentum is equivalent to $\Delta J_z = \mp 2$.  

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**Figure 4. Influence of the moiré potential on the intrinsic properties of TMD heterobilayers** (a) False color map of the $\sigma^\prime$-PL emission from IX of S2 measured with a resolution of 10 mT in the vicinity of the two identified resonant magnetic fields. The magnetic field for every sharp peak is different and the resonance has a diagonal profile. The white line indicates the energy dependence of the excitonic $g$ factor, whose slope is $\partial g/\partial E \approx 7.5$ eV$^{-1}$ (at $B \approx 24.2$ T) and 8.5 eV$^{-1}$ (at $B \approx 24.9$ T) for the two resonant magnetic fields. (b) PL Emission spectrum (left axis) of the IX at $B = 0$ T of S2 with energy dependent $g$-factor value (right axis) extracted from the data of panel a.
The magnitude of the slope as well as the fact that the exciton g-factor is determined by the exciton band energy is consistent with the standard band theory expression for the orbital contribution $g_{\text{orb}}^i$ to the g-factor of a single-electron band $i$ due to virtual transitions to other bands $j$ [49]:

$$g_{\text{orb}}^i = \sum_{j \neq i} \frac{|\langle i | \hat{p}_+ | j \rangle|^2 - |\langle i | \hat{p}_- | j \rangle|^2}{m_0 (\epsilon_i - \epsilon_j)},$$  \hspace{1cm} (1)$$

where $\hat{p}_\pm = \hat{p}_x \pm i \hat{p}_y$ are the components of the momentum operator, $\epsilon_i, \epsilon_j$ are the band energies and $m_0$ is the free electron mass. Note that the bands are predominantly localized on one of the layers, i.e. none of the energy differences $\epsilon_i - \epsilon_j$ corresponds to the IX transition energy and therefore only an order-of-magnitude estimate can be given. Typically, $|\langle i | \hat{p}_\pm | j \rangle|^2 / m_0$ is several eV, a few times larger than nearest band energy differences. If different components of the luminescence spectrum are due to random band energy variations across the sample, equation (1) gives $\partial g / \partial E$ of the order of a few eV$^{-1}$, in agreement with the experimental observation in figure 4(a).

To conclude, the conservation of the total angular momentum in monolayers and heterobilayers of TMDs not only defines the optical selection rules but also has profound consequences on the electron-phonon interaction. Benefiting of the very high values for excitonic g-factor for 60° aligned heterobilayers, we have tuned the excitonic Zeeman energy in the range of optical phonons with an external high magnetic field and we have shown that chiral phonons efficiently couple to excitons in these heterobilayers. Γ point chiral phonons can transfer angular momentum, which, together with the C$_3$ rotational symmetry, induces an excitonic spin-flip that can be revealed in polarization resolved photoluminescence experiments. Together with the ability to control the interlayer relative angle, this further provides a tool to design heterostructures with tailored electronic properties. The ability to control the lifetime of a quantum state is a key element for designing for instance emitting quantum device and using momentum transfer for this aim is a unique possibility offered by TMDs.

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