

Extremely High Excitonic g Factors in 2D Crystals by Alloy-Induced Admixing of Band States

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Monolayers (MLs) of semiconducting transition metal dichalcogenides emit light very efficiently and display rich spin-valley physics, with gyromagnetic (g) factors of about -4 . Here, we investigate how these properties can be tailored by alloying. Magneto-optical spectroscopy is used to reveal the peculiar properties of excitonic complexes in $\text{Mo}_x\text{W}_{1-x}\text{Se}_2$ MLs with different metal concentrations. We show that the alloys feature extremely high g factors for neutral excitons, that change gradually with the composition up to reaching values of the order of -10 for $x \approx 0.2$. First-principles calculations quantitatively identify the alloy-induced mixing between different conduction band valleys as the underlying mechanism originating the anomalous composition dependence of the neutral exciton g factor. The theoretical framework also suggests a high strain sensitivity of the alloys, making them promising candidates for tailor-made optoelectronic devices.

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Introduction—Semiconducting transition metal dichalcogenides (TMDs) and monolayers (MLs), such as MoS_2 , MoSe_2 , WS_2 , WSe_2 , and MoTe_2 , exhibit several intriguing optical properties [1–12]. In particular, the optical response of TMDs is dominated by excitonic effects [8,13–16], and, moreover, the MLs can be divided into two subgroups: bright MLs with a bright optically active exciton ground

state (MoSe_2 and MoTe_2) and darkish MLs whose exciton ground state is spin forbidden and thus optically dark (MoS_2 , WS_2 , and WSe_2) [8,10,11,17–29].

In TMD MLs, the lowest energy optically active excitons (referred to as A excitons) have energies that span the spectral range from 1.15 eV for MoTe_2 [7] up to 2.1 eV for WS_2 [27]. Continuous energy tuning within this range can be achieved by mixing two transition metal atoms (i.e., Mo and W) or two chalcogen atoms (e.g., S and Se), which induces the creation of TMD alloys with adjustable stoichiometry ratio (e.g., $\text{Mo}_x\text{W}_{1-x}\text{Se}_2$ and $\text{WS}_{2x}\text{Se}_{2-2x}$) [30]. Such compositional tuning opens new possibilities for absorbing and emitting light at desired wavelengths, which is crucial for optoelectronics applications [31–35]. The opportunities enabled by alloying can be further exploited in fields such as valleytronics [36], which employs local energy extrema, i.e., the so-called valleys, to encode and

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process binary information, with applications for quantum computers [37–41].

Bright A excitons in TMD ML are formed by an electron-hole pair originating from the inequivalent $\pm K$ valleys of the hexagonal Brillouin zone, and are associated selectively to $\sigma^\pm$ circularly polarized light [8,42,43]. The energy degeneracy of excitons at the $\pm K$ valleys can be lifted by applying a magnetic field perpendicular to the ML plane giving rise to the excitonic Zeeman effect described by the excitonic Landé g factor (g) [44,45]. In an external field (B), the g -factor value is related to the energy splitting between excitons from the $\pm K$ points with $\sigma^\pm$ polarizations, i.e., $\Delta E(B) = E_{\sigma^+}(B) - E_{\sigma^-}(B) = g\mu_B B$, (μ_B is the Bohr magneton). Theoretically, the magnetic field couples to both the spin and orbital parts of the Bloch states, and thus the excitonic g factor reflects the energy difference between the conduction (CB) and valence (VB) bands in both the $\pm K$ valleys [44–46].

For TMD MLs, the g factors are approximately equal to -4 [13,19,21,23,25,27,44,47–50] for the A excitons. Moreover, while a significant variation in the ML dielectric environment affects such physical quantities as the exciton binding energies and therefore the diamagnetic shift of excitons [51–55], the g -factor value stays insensitive [55,56]. For the WSe₂ ML with almost equal ratio of S and Se atoms, the excitonic g factor is close to -4 [57], similarly to the WS₂ and WSe₂ MLs. On the other hand, investigating the g factors of MLs of Mo-W selenide alloys could be of particular interest, since these alloys combine bright and darkish MLs.

In this Letter, we investigate the magneto-optical properties of excitonic complexes in Mo_{*x*}W_{1-*x*}Se₂ MLs encapsulated in hexagonal BN (*h*-BN) with different ratios of Mo and W atoms and compare the results with those achieved for WSe₂ and MoSe₂ MLs. Using low-temperature ($T = 10$ K) photoluminescence (PL) experiments carried out in external out-of-plane magnetic fields up to 30 T, we extract the g factors of the neutral (X) and charged (T) excitons. While the measured T g factors for Mo_{*x*}W_{1-*x*}Se₂ MLs vary from around -3.5 to almost -5 and are thus comparable to those of MoSe₂ and WSe₂ MLs, the g factors for the X transitions change gradually from about -4 for MoSe₂, to about -4.5 when the Mo concentration is $\sim 85\%$ then -6 when the Mo concentration is $\sim 70\%$, to about -7 when it reaches $\sim 50\%$, and even up to about -10 for Mo concentrations of $\sim 20\%$, to then go back to -4 in WSe₂. This striking tunability of the g factor is verified by first-principles calculations of the band structures and angular momenta of MoSe₂ and WSe₂ MLs and their alloys. The calculated values of the g factors show a trend similar to the experimental ones and also reveal an additional increase and decrease under the application of compressive or tensile biaxial strains, respectively. Our studies indicate that the alloying of TMD MLs is an efficient mechanism to enhance the g factors of neutral excitons, up to values that have only been observed for interlayer excitons in TMDs heterostructures (HSs) with nearly 0° or 60° twist

angles so far [58–62]. Because of the much simpler fabrication process of MLs compared to TMD HSs with specific twist angles, alloy MLs open new avenues as potential candidates for valleytronic and quantum devices [63].

Investigated samples—A series of *h*-BN-encapsulated Mo_{*x*}W_{1-*x*}Se₂ MLs with five different ratios between Mo and W atoms MoSe₂ and WSe₂, was prepared by exfoliation and dry transfer techniques, see Methods section of Supplemental Material (SM) [64] for details. The precise stoichiometry of each crystal was measured by scanning electron microscopy (SEM) with energy dispersive x-ray analysis (EDX). Each alloyed crystal was measured at multiple points, giving the following average Mo concentrations (*x*): $(22.5 \pm 1.9)\%$, $(45.8 \pm 1.7)\%$, $(49.40 \pm 0.81)\%$, $(67.79 \pm 0.94)\%$, and $(85.41 \pm 0.63)\%$. We further performed SEM EDX mapping measurements to verify the crystal homogeneity; see Sec. S1 of SM [64]. The crystal with 49% Mo concentration was grown by chemical vapor transport, while the other crystals were purchased from HQ graphene. Our experiments show very consistent results between grown and purchased samples, with the grown sample being characterized by a higher homogeneity.

Optical properties of the alloys—Low-temperature example PL spectra of MoSe₂, WSe₂ MLs and of the Mo_{*x*}W_{1-*x*}Se₂ alloys are shown in Fig. 1(a). The optical properties of the MoSe₂ and WSe₂ MLs originate from the bright [19] and dark [4,10,20,25,26] characters of the ground excitonic states, respectively. The PL spectrum of the MoSe₂ ML [19,22,25,44,98,99] consists of only two emission lines, while the WSe₂ spectrum is rather complex, since it is composed of several lines [11,19,20,23,24,65–76]. Following previous works, the X and T lines can be ascribed to the neutral and charged exciton [8], respectively; see Sec. S4 of SM for details [64]. The PL spectra of the Mo_{0.85}W_{0.15}Se₂, Mo_{0.68}W_{0.32}Se₂, and Mo_{0.49}W_{0.51}Se₂ MLs are similar to those of MoSe₂ ML; i.e., two emission lines can be distinguished. A small energy shift of the X and T lines is apparent in the transition from MoSe₂ ML to alloys with decreasing Mo concentration up to 46%, and, thus, by analogy, we attribute them correspondingly to the neutral and charged excitons; see Sec. S2 of SM for details [64]. The PL spectra measured on the Mo_{0.85}W_{0.15}Se₂, Mo_{0.68}W_{0.32}Se₂, Mo_{0.49}W_{0.51}Se₂, and Mo_{0.46}W_{0.54}Se₂ (the latter is not shown in Fig. 1) MLs indicate that the quality of the investigated MLs is very high, similar to that of the MoSe₂ ML (the X linewidths are about 4–5 meV); see Sec. S4 of SM for details [64]. This suggests that while the overall arrangement of Mo and W atoms in these MLs is random, the Mo/W ratio stays at a fixed level at least in the spatial area comparable to the diameter of the excitation spot (i.e., 1 μ m). This contrasts to the PL spectrum of the Mo_{0.23}W_{0.77}Se₂ ML, where the X linewidth reaches the value of about 11 meV. This points to a lower quality of the Mo_{0.23}W_{0.77}Se₂ ML, probably associated with a larger variation of the concentration of Mo atoms below the spatial area of excitation (i.e., higher disorder of

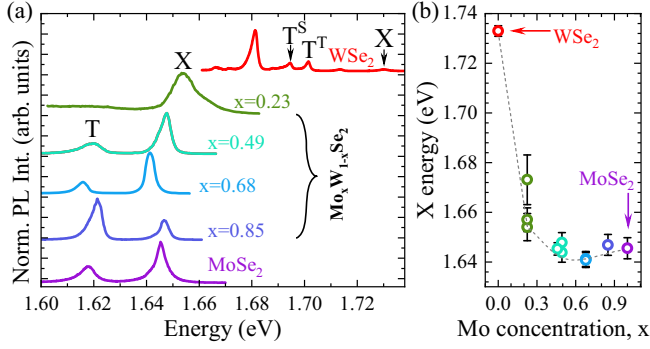


FIG. 1. (a) Low-temperature ($T = 10$ K) example PL spectra of MoSe_2 , WSe_2 , and $\text{Mo}_x\text{W}_{1-x}\text{Se}_2$ MLs encapsulated in h -BN flakes. (b) Extracted energy of neutral exciton (X) as a function of molybdenum (Mo) concentration. The dashed lines are guides to the eye.

the atoms at the nanoscale), as reported for WSe_2 MLs [57]. We performed a detailed analysis of the temperature- and power-dependent PL spectra measured on MoWSe_2 MLs, as discussed in Sec. S2 of SM, that confirm the attributions of the X and T lines [64].

Indeed, the alloy composition can efficiently tune the exciton energy. Figure 1(b) presents the neutral exciton energy as a function of the Mo composition in MoWSe_2 MLs extracted from all studied samples. As the Mo composition increases from the WSe_2 side, the neutral exciton (X) energies demonstrate two distinct trends. Initially, the X energies decrease significantly, with a pronounced drop observed for the $\text{Mo}_{0.23}\text{W}_{0.77}\text{Se}_2$ ML. This indicates that even small variations in Mo concentration (below 23%) lead to substantial changes in exciton energies. Beyond this critical point, the exciton energies vary at a much lower rate. A similar trend of the X energy has previously been reported for MoWSe_2 MLs [77,100–102].

Slight variations in the exciton energies between different works can be attributed to concentration inhomogeneities and possible strains induced during the sample preparation. Consistently, by probing different MLs exfoliated from the same crystal, we observed X energy variations of almost 4 meV [103,104]. A larger variability, up to almost 20 meV, was only observed for $\text{Mo}_{0.23}\text{W}_{0.77}\text{Se}_2$ MLs, i.e., for the crystal characterized by a larger inhomogeneity, as also deduced by the broader line shape and by SEM EDX measurements. Furthermore, for small Mo concentrations, the energy is highly dependent on the concentration, causing the observed energy variability.

Magneto-optical response—We now focus on the effects of out-of-plane magnetic fields on the alloyed MLs. Figure 2(a) shows the helicity-resolved PL spectra of an h -BN-encapsulated $\text{Mo}_{0.49}\text{W}_{0.51}\text{Se}_2$ ML in magnetic fields up to 30 T. Indeed, the application of the magnetic field results in a clearly observable Zeeman effect, which manifests itself as a splitting of the two counter-circularly-polarized components ($\sigma^\pm$).

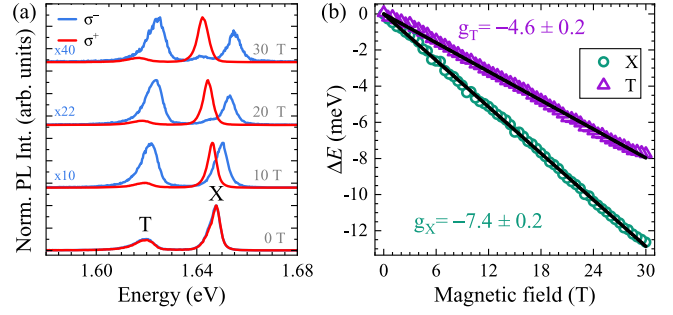


FIG. 2. (a) Helicity-resolved PL spectra of an h -BN-encapsulated $\text{Mo}_{0.49}\text{W}_{0.51}\text{Se}_2$ ML at $T = 4.2$ K measured at selected values of the applied out-of-plane magnetic field. The red (blue) color corresponds to the σ^+ (σ^-) polarized spectra. The measurements were performed under excitation energy of 2.41 eV and power of 2.5 μW . The σ^+ -polarized spectra were normalized to the X intensity, while the σ^- -polarized spectra were multiplied by scaling factors to make them better visible. The spectra are vertically shifted for clarity. (b) Magnetic-field evolutions of the energy differences (ΔE) between the two circularly polarized split components of the X and T transitions. The solid lines represent fits according to the equation described in the text.

The X and T bands are fitted by Lorentzian functions to extract the Zeeman splitting [ΔE ; Fig. 2(b)] and to derive information on the intensity to calculate the degree of circular polarization, shown in SM [64], Sec. S3. Indeed, the PL spectra reveal a large intensity variation for the σ^+ and σ^- components when increasing the field. For both the X and T bands, the lowest energy transitions (σ^+) gain intensity, whereas the highest energy transitions (σ^-) lose intensity with increasing field.

From the Zeeman splitting data shown in Fig. 2(b), we can quantify the g factor through linear fits: $g_X = -7.4 \pm 0.2$ and $g_T = -4.6 \pm 0.2$. Although the g factor for the trion is similar to those of the MoSe_2 and WSe_2 MLs (about -3.9 to -4.3), the g factor for the X line is almost 2 times larger than for the parent MLs (around -4.0 ; see Sec. S4 of SM for further details [64]).

In Fig. 3, we summarize the experimental values of the g factors extracted for the neutral and charged excitons measured on the MoSe_2 , WSe_2 , and $\text{Mo}_x\text{W}_{1-x}\text{Se}_2$ MLs with different x . The complete magneto-optical data, comprising magneto-PL spectra, Zeeman splitting analysis, and degree of circular polarization, of the parent compounds (MoSe_2 and WSe_2) and of the alloys ($\text{Mo}_x\text{W}_{1-x}\text{Se}_2$) are reported in SM [64], Secs. S4 and S5, respectively. Two main effects can be appreciated. First, the g factors for the X lines change gradually from almost -4.5 for the $\text{Mo}_{0.85}\text{W}_{0.15}\text{Se}_2$ ML through -6 for the $\text{Mo}_{0.68}\text{W}_{0.32}\text{Se}_2$ ML, to about -7 for MLs with almost equal compositions of Mo and W atoms, up to reaching a value of the order of -10 for the ML with $\sim 23\%$ of Mo atoms. In the latter sample, the possible influence of local strain and compositional variations is addressed in Sec. S6 in SM [64] showing the robustness of the observed exceptionally high g -factor

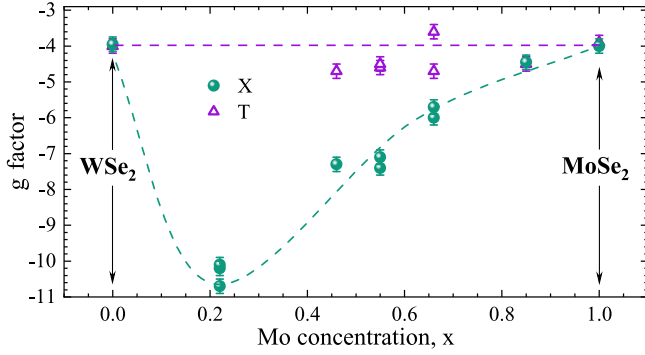


FIG. 3. Experimental values of g factors extracted for the neutral and charged excitons measured on the MoSe_2 , WSe_2 , and MoWSe_2 MLs with different Mo concentration. The dashed curves are guides to the eye.

value. Second, we note that the corresponding T g factors are in the range of around -3.5 to almost -5 , similar to the values of around -4 found in standard TMD MLs [27,44,78], leading to a substantial difference in the g factor values found for the X and T lines. This is indeed in contrast with the almost identical values reported for standard TMD MLs [27,44,78].

First-principles calculations—To elucidate the origin of the exciton g factor's striking behavior, we performed density functional theory (DFT) calculations using the modern methodology for g -factor evaluation, based on the summation-over-bands approach for the orbital angular momentum of Bloch states [45,79,105,106]. We are aware that our DFT calculations provide the underestimated value of a band gap, but the relevant valley-Zeeman physics does not depend on it [45,105,107]. Particularly, we model the $\text{Mo}_x\text{W}_{1-x}\text{Se}_2$ systems using 2×2 supercells, allowing us to probe Mo concentrations of $x = 0.25, 0.50, 0.75$.

We used experimental in-plane lattice constants of $a_{\text{MoSe}_2} = 3.288 \text{ \AA}$ [108] and $a_{\text{WSe}_2} = 3.282 \text{ \AA}$ [109]. The lattice constants of the alloys are interpolated linearly; that is, $a_0(x) = xa_{\text{MoSe}_2} + (1-x)a_{\text{WSe}_2}$. Additionally, we explore different values of a_0 to account for typical uncertainties, such as the choice of the exchange-correlation functional within DFT [110,111], spread of the experimentally determined values [108,109,112–116], and possible disorder effects due to alloying and strain [117–120] (which can reach values as high as 2% in TMD MLs [80]). The lattice parameters we considered here are within the 1% range. The computational details are given in SM [64], Sec. 7.

Let us first look at the electronic properties of the $\text{Mo}_x\text{W}_{1-x}\text{Se}_2$ alloys. Figure 4 shows the calculated electronic band structures near the K point for different values of the Mo concentration, with the color code indicating the degree of wave function localization at the W and Mo atoms. While the VBs gradually change their composition and spin splitting as a function of x , the scenario is quite different for CBs. Particularly, in the conduction bands, we observe 8 energy branches, 2 originally from the K point, and 6 originating from the 2 spin branches of the CBs from the Q valleys, as depicted in Sec. S7.1 of SM [64]. The vertical black arrows in Fig. 4 indicate the relevant bands involved in the (spin conserving) lowest energy direct optical transitions with circular polarization at the K point. We note that the folded Q bands do not participate in the optical processes but, due to their nearly resonant energetic alignment with the K point bands, they contribute to the wave function hybridization by modifying the orbital composition.

The calculated g factors for the lowest energy transitions are displayed in Fig. 5(a). Our calculations reveal that, for

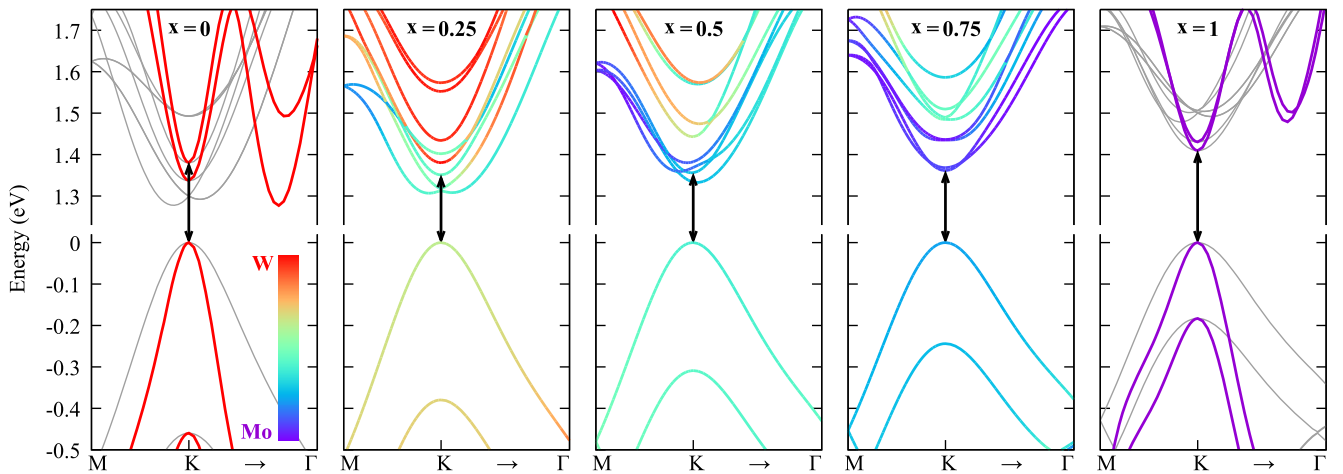


FIG. 4. Electronic band structures calculated for the studied $\text{Mo}_x\text{W}_{1-x}\text{Se}_2$ MLs with $a_0(x)$. The color code reveals the wave function localization at the W and Mo atomic sites. Band structures using the 2×2 supercells for pure MoSe_2 and WSe_2 crystals are shown with thin gray lines for comparison. The lowest energy interband transitions with circular polarization are indicated with vertical arrows. The unfolded band structures are shown in Fig. S7.2 of SM [64].

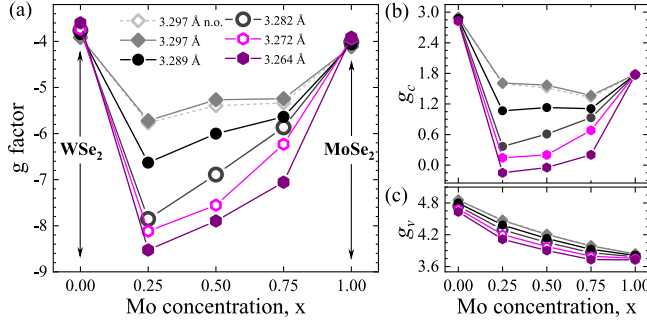


FIG. 5. (a) Exciton g factors calculated from first principles. Colored lines correspond to different lattice constants of MoSe_2 . The nonmonotonic dependence reproduces the experimental trend shown in Fig. 3. The dashed lines indicate the results without optimization of the atomic positions (n.o.), which has a minor impact on the final values. Calculated g factors for the (b) conduction g_c and (c) valence g_v bands at the K point, allowing us to pinpoint the electronic origin of the nonmonotonic dependence observed in the exciton g factor.

intermediate values of $x = 0.25, 0.50, 0.75$, the exciton g factor is significantly enhanced and reaches the most negative value at $x = 0.25$. This nonmonotonic dependence revealed by our first-principles calculations is in excellent agreement with the experimental trends shown in Fig. 3. Moreover, the calculated trend is observed regardless of the choice of the lattice parameter. However, the distinct nonmonotonic feature is more pronounced for the lowest value of $a = 3.264$ Å for $x = 0.25$. In fact, the sensitivity of the g -factor trend with respect to the chosen in-plane lattice parameter suggests that external strain can be employed to efficiently alter the valley-Zeeman features of alloyed TMDs, even much more strongly than what has been reported for their parent MLs [107,121,122]. Note that further insights into the progressive evolution around $x = 0.25$ and random alloys may be obtained in future work using larger supercells.

To understand the nonmonotonic dependence of the exciton g factor on x , let us look at the calculated g factors of the conduction (g_c) and valence (g_v) bands, shown in Figs. 5(b) and 5(c), respectively. We readily identify that g_v decreases monotonically and remains in between the values of g_v of the parent compounds. In contrast, g_c displays a distinct nonmonotonic behavior, with calculated values reaching magnitudes smaller than the g_c values of the parent compounds depending on the chosen value of the lattice parameter. This nonmonotonic dependence of g_c reflects the nontrivial changes in the orbital nature of the CBs in the K valleys (mostly d_{z^2} orbitals) due to the mixing with conduction bands from Q valleys (mostly $d_{x^2-y^2} + d_{xy}$ and p orbitals). Interestingly, for larger values of the in-plane lattice parameter (with Mo and W atoms further apart), the Q and K conduction bands decouple and, consequently, the g_c acquires a monotonic dependence (see Sec. S7.3 of SM [64]). These dramatic changes in g_c

revealed by our first-principles calculations indeed demonstrate that engineering the wave function hybridization in TMD alloys to enhance exciton g factors is much more efficient than using proximity effects in van der Waals heterostructures, as recently reported in WS_2 and graphene systems [123].

Conclusions—In this Letter, we outlined striking valley properties of TMD alloys. In particular, we experimentally observed a high valley polarization close to unity for excitonic complexes in the $\text{Mo}_x\text{W}_{1-x}\text{Se}_2$ alloys, and a highly stoichiometry-dependent behavior of the exciton g factor. Specifically, we found that the g factors for neutral excitons exhibit a strongly nonlinear dependence from about -4 in the MoSe_2 and WSe_2 MLs, up to -10 for $x \approx 0.2$. Our first-principles calculations successfully reproduced the experimental trends and identified the origin of such strong g -factor modulation due to the orbital mixing in the conduction band. We also revealed that the g -factor values can be significantly altered by strain. Our results thus show that TMD alloys represent a unique, yet not much explored, material platform with remarkable valley-tunable properties that can be manipulated on demand.

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Data availability—The data that support the findings of this article are openly available [124].

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