Strain control of exciton-phonon coupling in atomically thin semiconductors

Iris Niehues¹, Robert Schmidt¹, Matthias Drüppel², Philipp Marauhn², Dominik Christiansen³, Malte Selig³, Gunnar Berghäuser³, Daniel Wigger², Robert Schneider¹, Lisa Braasch¹, Rouven Koch¹, Andres Castellanos-Gomez⁴, Tilmann Kuhn², Andreas Knorr³, Ermin Malic⁵, Michael Rohlfing², Steffen Michaelis de Vasconcellos¹, and Rudolf Bratschitsch^{1}*

¹Institute of Physics and Center for Nanotechnology, University of Münster, D-48149 Münster, Germany

²Institute of Solid State Theory, University of Münster, D-48149 Münster, Germany

³Institut für Theoretische Physik, Nichtlineare Optik und Quantenelektronik, Technische Universität Berlin, D-10623 Berlin, Germany

⁴Materials Science Factory, Instituto de Ciencia de los Materiales de Madrid (ICMM-CSIC), Madrid, E-28049, Spain.

⁵Chalmers University of Technology, Department of Physics, SE-412 96 Gothenburg, Sweden

*e-mail: Rudolf.Bratschitsch@uni-muenster.de

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Abstract

Semiconducting transition metal dichalcogenide (TMDC) monolayers have exceptional physical properties. They show bright photoluminescence due to their unique band structure and absorb more than 10% of the light at their excitonic resonances despite their atomic thickness. At room temperature, the width of the exciton transitions is governed by the exciton-phonon interaction leading to strongly asymmetric line shapes. TMDC monolayers are also extremely flexible, sustaining mechanical strain of about 10 % without breaking. The excitonic properties strongly depend on strain. For example, exciton energies of TMDC monolayers significantly redshift under uniaxial tensile strain. Here, we demonstrate that the width and the asymmetric line shape of excitonic resonances in TMDC monolayers can be controlled with applied strain. We measure photoluminescence and absorption spectra of the A exciton in monolayer MoSe₂, WSe₂, WS₂, and MoS₂ under uniaxial tensile strain. We find that the A exciton substantially narrows and becomes more symmetric for the selenium-based monolayer materials, while no change is observed for atomically thin WS₂. For MoS₂ monolayers, the line width increases. These effects are due to a modified exciton-phonon coupling at increasing strain levels because of changes in the electronic band structure of the respective monolayer materials. This interpretation based on steady-state experiments is corroborated by time-resolved photoluminescence measurements. Our results demonstrate that moderate strain values on the order of only 1% are already sufficient to globally tune the exciton-phonon interaction in TMDC monolayers and hold the promise for controlling the coupling on the nanoscale.

Uniaxial mechanical strain alters the lattice constants and reduces the symmetry in single crystals, leading to significant changes in the electronic band structure. The controlled application of strain to a material is therefore a powerful tool for tailoring its electronic and optical properties. For example, strain significantly increases the charge carrier mobility in the traditional semiconductors silicon and germanium¹. Strained germanium nanomembranes even exhibit an indirect to direct band gap transition². Mechanical strain is also well-known to change the electronic phases of complex oxides. In that way, their fundamental bulk electronic properties such as superconductivity or magnetism can be strongly influenced^{3–5}. Strain values on the order of a few percent are readily achievable by growing the oxides on lattice-mismatched substrates⁶. In contrast, two-dimensional materials, such as graphene or transition metal dichalcogenides (TMDCs), hold the promise for tunable strain control due to their atomic thickness and large flexibility^{7,8}. In particular, the optical properties of these materials can be strongly modified^{9,10}. For example, the fundamental resonances in TMDCs, called excitons, strongly downshift in energy with the application of uniaxial tensile strain^{11–21}. Strain engineering might open the door to tune also the widths of the excitonic resonances which are exceedingly difficult to control by other means. At cryogenic temperatures, they were successfully narrowed down by reducing inhomogeneous broadening with encapsulation techniques^{22–24}. At room temperature, however, the line width of the excitons in TMDCs is dominated by the electron-phonon interaction^{25,26}, which poses a fundamental lower limit on the line width. Here, we demonstrate that the electron-phonon interaction in 2D TMDCs can be engineered by mechanical strain. We find that the excitonic line width in WSe₂ and MoSe₂ monolayers can be substantially reduced by applying uniaxial tensile strain. In contrast, the line width stays constant for monolayer WS₂ and increases

for MoS₂. The main reason for these observed trends is the exciton-phonon coupling due to the change of the band structure under mechanical strain.

TMDC monolayers are micromechanically exfoliated²⁷ onto polydimethylsiloxane (PDMS) and identified by reflection and photoluminescence (PL) microscopy. Subsequently, the atomically thin crystals are stamped on a flexible polycarbonate (PC) substrate of 500 μm thickness²⁸ and covered with a 160 μm thick PDMS layer to avoid slippage of the TMDC monolayers during the straining experiments^{29,30}. Reversible uniaxial tensile strain is applied to the monolayers in a controlled way by bending the substrate⁹.

We perform photoluminescence and absorption measurements of four different TMDC monolayers (MoSe₂, WSe₂, WS₂, and MoS₂) at room temperature to investigate the effect of mechanical strain on the A exciton. As previously reported⁹, uniaxial tensile strain leads to a linear redshift of the A exciton energy for all materials in the strain range of less than 1.5 % (see supporting information Fig. S1 and S2). To highlight the influence of strain on the excitonic line shape, we normalize all PL spectra and shift them to the maximum of the A exciton (Fig. 1a-d). Exemplary spectra are shown for minimum (purple), medium (pink), and maximum (orange) strain.

Figure 1: Photoluminescence (PL) spectra of TMDC monolayers under uniaxial tensile strain. a)-d) Normalized PL spectra of the A exciton for monolayer MoSe₂ (a), WSe₂ (b), WS₂ (c), and MoS₂ (d) for three exemplary strain levels. The spectra are horizontally shifted to pin the A exciton peak at the same position. e-h) Full width at half maximum (FWHM) line width of the spectra for increasing strain levels for the different materials. For MoSe₂ (e) and WSe₂ (f) the line width is decreasing with increasing strain ((-4.44±0.20) meV/% and (-9.16±1.35) meV/%, respectively) and the line shape becomes more symmetric, while for WS₂ (g) the line shape stays

almost constant (-1.61 ± 0.86 meV/%). For MoS₂ (**h**), the line width increases with increasing strain ($+13 \pm 1$ meV/%).

In the unstrained case, the PL line shape is asymmetric for all four monolayer materials with a steeper slope on the high-energy side compared to the low-energy side. The line shape is dominated by asymmetric phonon sidebands due to the strong exciton-phonon coupling. The sidebands originate mainly from the emission and absorption of optical phonons³¹. For monolayer MoSe₂, the PL intensity of the low-energy side decreases with increasing tensile strain, while the high-energy side remains unchanged (Fig. 1a). For WSe₂ this effect is much more pronounced (Fig. 1b), and in addition, a narrowing on the high-energy side is observed. Consequently, the shape of the PL spectra for both materials becomes more symmetric with increasing strain. In strong contrast, no significant change of the line shape is observed for the WS₂ monolayer (Fig. 1c), and for MoS₂ the line shape even becomes more asymmetric due to a slightly increased PL intensity on the low-energy side (Fig. 1d).

To quantify the effect of strain on the exciton-phonon coupling in the TMDC monolayers, the full width at half maximum (FWHM) of the A exciton line is depicted in Fig. 1e-h for increasing uniaxial tensile strain levels. The value of the FWHM is determined by fitting the asymmetric line shape, which is described in detail in the supporting information in Fig. S3. For MoSe₂ (**e**) and WSe₂ (**f**) the line width decreases linearly with increasing strain in the examined range. The extracted gauge factors are (-4.44 ± 0.20) meV/% and (-9.16 ± 1.35) meV/% for monolayer MoSe₂ and WSe₂, respectively. For monolayer WS₂ (**g**), the line width remains almost unchanged at 25 meV (the gauge factor is -1.61 ± 0.86 meV/%). In contrast, for monolayer MoS₂ (**h**), the line width increases with $(+13 \pm 1)$ meV/%.

To verify the trends observed in emission, we also investigate the optical absorption using the same samples for all four atomically thin materials in Fig. 2. The optical absorption is derived from light transmitted through the TMDC monolayer T_{ml} normalized by the transmission through the substrate T_{sub} . The absorption spectrum is calculated via $A_{\text{ml}} = 1 - \frac{T_{\text{ml}}}{T_{\text{sub}}}$, considering that the reflectivity of the monolayer is negligible compared to the absorption at the excitonic resonances³⁴. Similar to the photoluminescence spectra, we find a prominent redshift of the A exciton resonance with gauge factors being in excellent agreement with the extracted values from the PL measurements and with calculations (see supporting information Fig. S2). In Fig. 2a-d the absorption spectra are presented for three different strain levels (minimum in purple, intermediate in pink, and maximum in orange) for the four materials. Again, the spectra are normalized and plotted against the detuning with respect to the A exciton maximum position to facilitate the comparison between spectra acquired at different strain levels highlight the effect of strain on the line shape.

Figure 2: Absorption spectra of TMDC monolayers under uniaxial tensile strain. a)-d) Absorption spectra for monolayer MoSe₂ (a), WSe₂ (b), WS₂ (c), and MoS₂ (d) for 3 exemplary strain levels. The spectra are horizontally shifted to pin the A exciton peak at the same position. e)-h) FWHM line width of the absorption spectra for increasing strain levels for the different materials. For MoSe₂ (e) and WSe₂ (f) the FWHM is decreasing with increasing strain ((-4.05±0.78) meV/% and (-9.25±1.44) meV/% respectively) and the spectral shape becomes more symmetric, while for WS₂ (g) no change is observed (-0.16±0.88 meV/%). Again, in MoS₂ (h) the line width is increasing (+10±2 meV/%) and the spectra become more asymmetric.

Contrary to light emission in Fig. 1, the high-energy side of the excitonic resonance is more pronounced in absorption. This is because the phonon creation processes appear at higher energies in absorption³⁵. For the light emission spectra, this broadening appears on the low-energy side, because of its photon-emission character (see supporting information Fig. S4). In Fig. 2e-h the line width of the A exciton is plotted for all measured strain levels. As in the PL measurements, for MoSe₂ (e) and WSe₂ (f) a prominent decrease of the line width with increasing strain is observed with gauge factors of (-4.05 ± 0.78) meV/% and (-9.25 ± 1.44) meV/%, respectively. Again, for WS₂ (g) the line width is constant with (-0.16 ± 0.88) meV/%, and for MoS₂ (h) an increase of $(+10 \pm 2)$ meV/% is observed. The changes of the line shape of the A exciton with strain from emission (PL) and absorption measurements are in excellent quantitative agreement, which confirms the validity of our measurements. Only for monolayer MoS₂, the extracted change of the line width is stronger in the PL measurement compared to absorption (13 meV/% vs. 10 meV/%). We attribute this to the possible weak influence of the trion on the PL on monolayer MoS₂, which slightly broadens the spectra (see supporting information Fig. S5)³². However, a clear line broadening of the A exciton with uniaxial tensile strain is visible in both the PL and absorption measurement for monolayer MoS₂, while in the latter one the influence of the trion is negligible (see supporting information Fig. S4). In addition, all observed changes in the four investigated materials are reversible with strain (see supporting information Fig. S1 and S2).

We attribute the decrease or increase of the line widths of the A excitons under uniaxial tensile strain mainly to two different effects in the monolayers: changes in the electronic band structure and change of the phonon energies. They lead to a modification of the exciton-phonon coupling and alter the intravalley and intervalley scattering rates. Fig. 3 schematically depicts the different

exciton energy alignments and phonon scattering channels with and without strain for all four materials.

In monolayer MoSe₂ (Fig. 3a), intervalley scattering is very unlikely, because the indirect KA exciton has a much larger energy than the KK exciton. Additional strain shifts it to even higher values. Therefore, intravalley scattering of the excitons with phonons solely governs the excitonic line width at room temperature³¹. It leads to the pronounced asymmetry in the line shape at the high-energy side of the absorption spectrum (see supporting information Fig. S6a for details). The observation that the A exciton line shape of monolayer MoSe₂ becomes more symmetric and narrower with increasing strain can be explained by a decrease of the exciton-phonon coupling. By applying uniaxial strain to the monolayer, the longitudinal-optical phonon bands split into two branches, with one branch shifting towards higher and the other towards lower energies^{36,37}. The calculated splitting of the phonon branches is on the order of 3 to 10 cm⁻¹ at 2% of uniaxial strain³⁶, which has been confirmed by experiments³⁸. The model described in Ref.²⁶ demonstrates that the coupling strength increases with lower phonon energies and decreases with higher phonon energies. Therefore, the strain-induced shift to lower/higher phonon energies renders exciton scattering with phonons more/less probable. At the same time, the phonon population increases/decreases, which increases/decreases the scattering probability^{26,31}. Since our measurements show a line narrowing with increasing uniaxial tensile strain, we conclude that the influence of the energetically higher phonon branches, which decrease the scattering processes, dominates. For acoustic phonons, previous calculations predict no significant change in the dispersion at 1% of strain. Only at much higher strain values (>7%) the slope of the phonon branches shift to lower values^{39,40}. Overall, our measurements indicate that intravalley scattering

becomes less probable in monolayer MoSe₂ with increasing strain and thus the reduction of the exciton linewidth is due to a decrease of the exciton-phonon coupling upon straining.

In contrast to MoSe₂, intervalley scattering processes play an important role for WSe₂ monolayers. Excitons are effectively scattered by phonons from the bright KK to the dark KA exciton state, because the KA state lies energetically below the KK exciton state at zero strain³¹ as sketched in Fig. 3b. While intravalley scattering broadens the high-energy side of the A exciton (similar to MoSe₂), intervalley scattering processes lead to an additional broadening of the low-energy side of the spectra (Fig. S6b). As a consequence, both sides of the A exciton line are broadened at zero strain^{26,31}. With increasing strain, our measurements indicate that for monolayer WSe₂ both the low- and high-energy side of the A exciton narrows (Fig. 1b), which demonstrates that strain also influences intervalley scattering. This narrowing can be tracked down to the change in the band structure of the WSe₂ monolayer under the influence of uniaxial tensile strain, where the KA state approaches the energy of the KK state (Fig. 3b and supporting information Fig. S7). Since both scattering processes (intervalley and intravalley) contribute about equally to the line width of monolayer WSe₂ at room temperature²⁶, the measured narrowing gauge factor of monolayer WSe₂ (-9 meV/%) becomes about twice as large as for MoSe₂ (-4 meV/%).

Figure 3: Schematic drawing of the exciton-phonon scattering mechanisms in monolayer TMDCs without and with applied uniaxial tensile strain. In the unstrained case (left column) intravalley scattering dominates the spectral shape of MoSe₂ (a) and MoS₂ (d), while for WSe₂ (b) and WS₂ (c) also intervalley scattering plays an important role, because the KA state lies energetically below the KK state. With applied strain (right column) the band structure and the exciton-phonon coupling changes in a different way for the four materials. For all materials, the intravalley scattering decreases. Furthermore, the KA state lies energetically higher than the KK state. Therefore, the intervalley scattering decreases in tungsten-based materials (b, c). For sulfur-

based materials (c, d) also the Γ K exciton plays a role, which is decreasing with strain and opens a new scattering channel.

In monolayer WSe₂ (and WS₂) the spin-forbidden dark exciton states lie energetically below the bright ones^{41–43}. They have a negligible effect on the excitonic line shape of bright excitons, because the required spin-flip process occur on a much slower time scale compared to phonon-assisted processes^{31,44}. Therefore, the overall narrowing trend of the A exciton with increasing strain can be witnessed in both MoSe₂ and WSe₂ monolayers, although both materials exhibit a different spin ordering in the conduction band.

In WS₂ (Fig. 3c) and MoS₂ (Fig. 3d) monolayers the situation for the KK and the KA exciton and their coupling to phonons is the same as for the selenium-based counterparts. But in sulfur-based materials a third exciton has to be taken into account, the one formed by the hole located in the Γ valley and the electron in the K valley. For WS₂ this dark transition lies energetically slightly above the bright KK exciton, and in MoS₂ slightly below⁴⁵. This opens a new scattering channel for excitons in MoS₂ even at zero strain, which can explain the observed broad line width together with the high density of defects⁴⁶ leading to substantial inhomogeneous broadening in this material.

In monolayer WS₂ (Fig. 3c), surprisingly, no change of the exciton line width with increasing strain is measured. An important clue to understand this behavior is the much weaker exciton-phonon coupling compared to the other TMDCs. Sulfur-based TMDCs exhibit less efficient exciton-phonon scattering compared to selenium-based compounds because of the higher phonon energies⁴⁷. Indeed, the exciton line width of monolayer WS₂ is already rather small at zero strain (25 meV), compared to monolayer MoSe₂ and WSe₂ (39 meV and 36 meV). Furthermore, the indirect Γ K exciton decreases in energy upon applied strain, because of an upshift of the valence

band at the Γ point, which may open a new scattering channel (see Supporting Information Fig. S7). Therefore, there are now two counteracting contributions of increased (Γ K exciton) and decreased (K Λ and KK exciton) phonon scattering. The almost unchanged line shape of monolayer WS₂ with increasing strain seems to be due to this peculiar situation where the two contributions are balanced.

Finally, the measured increase of the line width of the A exciton in monolayer MoS₂ can be understood by an upshift of the valence band at the Γ point. The Γ K exciton energy decreases and leads to enhanced exciton-phonon scattering into the Γ K state (Fig. 3d). Already at around 0.1% tensile strain, monolayer MoS₂ undergoes a direct to indirect transition¹³. The additional intervalley scattering increases under strain, and this broadening of the line width overcompensates the effect of the decrease in intravalley scattering in MoS₂.

To corroborate the change of the exciton-phonon coupling directly in the time domain, we perform time-resolved photoluminescence (TRPL) measurements for various strain levels (see the Supporting Information for experimental details) for MoSe₂, WSe₂, and WS₂ monolayers. For monolayer MoS₂, the dynamics is too fast to be resolved by the experimental setup. In Fig. 4a-c the PL dynamics are shown for three different strain values for MoSe₂, WSe₂, and WS₂, respectively.

Figure 4: Time-resolved photoluminescence of TMDC monolayers under uniaxial tensile strain. **a)-c)** Time-resolved photoluminescence measurements for monolayer MoSe₂ (**a**), WSe₂ (**b**), and WS₂ (**c**) for 3 different strain levels (dots) with fits (solid lines). The instrument response function (IRF) is drawn in black. **d)-f)** PL decay rate extracted from **a)-c)** for increasing strain levels and low power (purple circles) and high power (orange triangles). For MoSe₂ (**c**) and WSe₂ (**d**) the decay rate is increasing with increasing strain ((0.21 ± 0.04) ns/% and (0.15 ± 0.01) ns/% respectively, while for WS₂ (**f**) no change is observed.

For both MoSe₂ (Fig. 4a) and WSe₂ (Fig. 4b), the decay of the PL signal of the A exciton becomes faster with increasing strain. To analyze the TRPL data in detail, the measured curves are fitted with a single exponential decay and including the exciton-exciton annihilation process. For each strain level, the PL decay is recorded for two laser powers to show the influence of the optically excited exciton density (see Supporting Information for more details). The corresponding extracted decay rates are displayed in Fig. 4c-d. For MoSe₂ (**d**) the PL decay rate increases with (0.21 ± 0.01) ns⁻¹/% and for WSe₂ (**e**) with (0.15 ± 0.01) ns⁻¹/% with increasing strain. In general, PL decay rates are strongly influenced by scattering of excitons out of the light cone, either by

intravalley scattering⁴⁸ or intervalley scattering^{26,49}. Both the intra- and intervalley exciton-phonon scattering into dark states become less efficient with increasing strain. Therefore, more excitons remain within the light cone, which results in a faster PL decay. The increase of the decay rates for MoSe₂ and WSe₂ with increasing strain therefore nicely corresponds with the narrowing of their line widths. In the same way, the constant PL decay rate for monolayer WS₂ is in good agreement with the time-integrated PL measurements, where no change in the line width of WS₂ is observed with increasing strain. In WS₂ monolayers no significant change in the exciton-phonon coupling is observable for the considered strain levels. For monolayer MoS₂, the decay time of the A exciton is in the order of a few picoseconds at zero strain⁵⁰ and below the time resolution of our experimental setup. We would expect a decrease of the decay rate with increasing strain from our considerations above, deduced from our integrated PL measurements.

In summary, our results highlight the influence of mechanical strain of the exciton-phonon interaction in atomically thin TMDCs. We find that the A exciton line substantially narrows and becomes more symmetric for selenium-based TMDCs, because of the weakening of the exciton-phonon coupling with increasing strain. Indeed, at room temperature, a moderate amount of mechanical strain may be used to strongly narrow excitonic lines on the order of 10 meV/%. We envision that our newly found intrinsic line narrowing mechanism could be readily exploited without a strain apparatus, by growing the atomically thin TMDCs directly onto different substrates at high temperatures and inducing strain in the monolayers by the different thermal expansion coefficients of TMDC monolayer and substrate⁵¹. For WS₂ monolayers, no change in the excitonic line width and life time is observed due to the already weak exciton-phonon coupling in this material. In MoS₂, the line width increases, because of the change from a direct to an indirect semiconductor under tensile strain and the enhanced electron-phonon coupling. In contrast to these

global strain phenomena, it has been shown that local strain plays an important role for the formation of single-photon emitters in monolayer TMDCs^{10,52–55}. Hence, our work also provides first clues towards understanding the influence of strain on the fundamental exciton-phonon coupling in these nanoscopic light emitters and thereby paves the way to tailoring local strain environments on the nanoscale.

ASSOCIATED CONTENT

Supporting Information.

- Photoluminescence under strain: A exciton resonance gauge factor
- Absorption under strain: A exciton resonance gauge factor
- Fitting of the PL and absorption spectra
- Asymmetry of PL and Absorption
- Valley positions under uniaxial strain
- Time resolved PL measurements (methods)
- Phonon contributions to the line width at zero strain

AUTHOR INFORMATION

Corresponding Author

Email: Rudolf.Bratschitsch@uni-muenster.de

ORCID

Rudolf Bratschitsch: 0000-0002-2368-2548

Andres Castellanos-Gomez: 0000-0002-3384-3405

Steffen Michaelis de Vasconcellos: 0000-0003-3584-0635

Iris Niehues: 0000-0001-7438-2679

Robert Schmidt: 0000-0002-8856-3347

Malte Selig: 0000-0003-0022-412X

Author Contributions

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ABBREVIATIONS

PL photoluminescence, TRPL time resolved photoluminescence, TMDC transition metal dichalcogenide

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ToC Figure