Electronic Supplementary Material (ESI) for Nanoscale.

Supplementary Information

Interlayer excitons in bilayer MoS_2 under uniaxial tensile strain

Iris Niehues, Anna Blob, Torsten Stiehm, Steffen Michaelis de Vasconcellos, and Rudolf Bratschitsch

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

Layer-dependent optical differential transmission of MoS_2

Figure S1 depicts the differential transmission (1-Transmission) of MoS_2 crystals of different thickness. The flakes are mechanically exfoliated from a natural molybdenum crystal (2D semiconductors) onto PDMS placed on a glass slide. The data is shifted vertically for clarity.

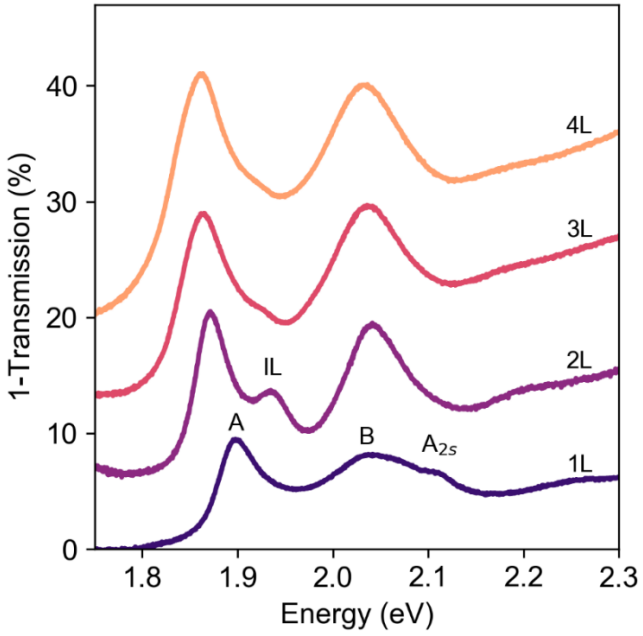

Figure S1: Layer-dependent differential transmission spectra (plotted as "1-Transmission") of MoS_2 crystals of different thickness on PDMS on glass substrate. The spectra are shifted vertically for clarity. The A and B excitons are clearly visible in all spectra. In addition, the first excited state of the A exciton (A2s) can be identified in the monolayer. For the MoS_2 bilayer, the interlayer exciton (IL) is clearly visible.

The energies of the A and B excitons are extracted from the measurement and listed in table T1. The energy of the A exciton resonance shifts to lower energy with increasing layer number [1]. Together with the white-light reflectivity and SHG image, the number of layers of the MoS_2 flakes can be determined. For the monolayer, a third maximum is visible as a shoulder on the high-energy side of the B exciton at 2.096 eV and is identified as the first excited state of the A exciton (A_{2s}). For thicker crystals the A_{2s} exciton shifts closer to the B exciton due to increased dielectric screening and is therefore no longer discernible. In the bilayer, the interlayer exciton between the A and B exciton at 1.933 eV is clearly visible. It becomes less prominent again for thicker layers being only visible as a small shoulder next to the A exciton transition in the tri- and four-layer spectra. In order to verify the existence of the interlayer resonance in MoS_2 bilayers, we measured differential transmission spectra of bilayers exfoliated from natural MoS_2 crystals from two different suppliers (hq graphene and SPI supplies) and on three different substrates: PDMS, polycarbonate and glass substrate. We could observe the interlayer exciton in all cases.

Table 1: Energies of the A and B excitons for different layer thicknesses of MoS_2. The values are extracted from the spectra shown in Fig. S1.

Layer number	A exciton energy (eV)	B exciton energy (eV)
Monolayer	1.989	2.037
Bilayer	1.872	2.042
Trilayer	1.864	2.038
Fourlayer	1.862	2.038

Fitting of the differential transmission spectra

Figure S2 shows the differential transmission spectrum of a MoS_2 bilayer on PC substrate without externally applied strain. The spectrum is fitted with four lines, one for each resonance, the A, B, IL and C exciton, respectively. Due to the asymmetric line shape of the exciton resonances in TMDCs, we choose a Pearson IV distribution to empirically fit the spectra [2]. In the unstrained case, the A, B and IL excitons are at 1.857 eV, 2.025 eV and 1.918 eV, respectively. The full width at half maximum (FWHM) line width of the A and IL exciton is nearly identical with (48±2) meV and (52±7) meV, while the B exciton has width of (80±10) meV. The oscillator strength of the interlayer exciton is (26±10) % of the corresponding A exciton, which is in agreement with calculations [3] (19%). To extract the exciton energies with strain, the spectra are

fitted for each strain level as described above. Since the exciton resonances overlap, the line width and oscillator strength have quite large error bars. We therefore restrict our analysis with strain to the energy positions of the excitons.

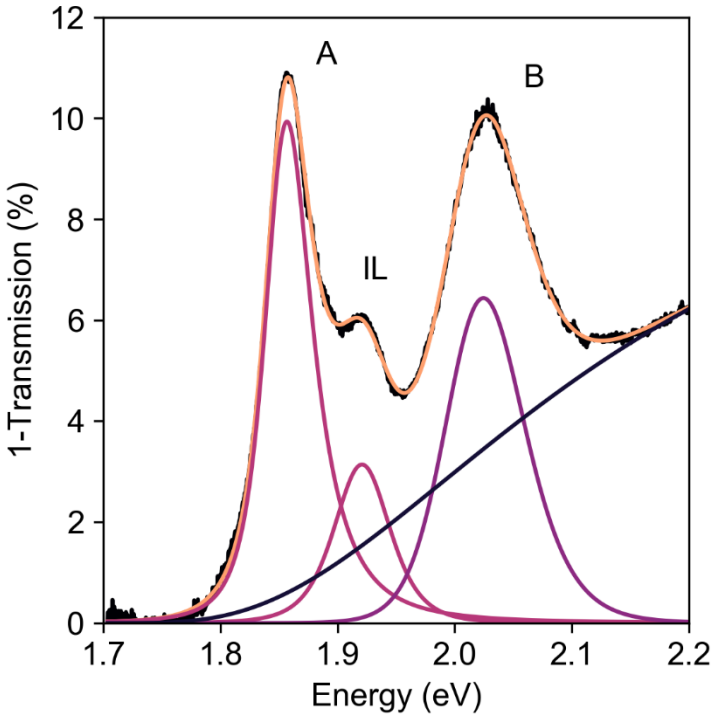

Figure S2: Fitting of the differential transmission spectra for the MoS_2 bilayer without externally applied strain. The thick black line represents the experimental data, the yellow line the fit curve. Pink and dark blue lines are the fit curves for the individual excitonic resonances.

Spatially resolved differential transmission spectra

Figure S3a) shows the white-light transmission of the investigated 2H-MoS_2 bilayer. The direction of applied strain is indicated by the black arrows. The bilayer is strained along its longer side. Spatially resolved differential transmission spectra are recorded all along the vertical pink line a) and shown in b) for the maximum strain value of 1.6%. Therefore, the strain transfer from the PC substrate to the MoS_2 bilayer is mapped in this image. The vertical dashed lines show the excitonic energy positions for the A, B and IL excitons of the bilayer without externally applied strain. Bright yellow parts in the image (above 100 µm and around 25-30 µm) are associated with

the absorption of thicker MoS_2 layers. When strain is applied to the MoS_2 bilayer the exciton resonances undergo a shift to lower energies. It was shown for monolayers that the strain transfer is spatially dependent [4, 5] and increases form the crystal edges towards the center, leading to inhomogeneities of the strain-dependent shift in the measured spectra. At cracks occurring during the straining, the excitonic resonances stay at their unstrained positions since every crack creates new edges of the crystal. This effect can be clearly seen also in the bilayer at 70 µm in the spatially resolved spectra of Fig. S3b. The small, thicker part of the MoS_2 crystal in the region of 25-30 µm has no visible effect on the strain transfer to the bilayer. The orange dash (orange line in a), b)) marks the position where the spectra in the main text are analyzed. At this position the excitons show a maximum shift of 78 meV, which is spatially independent over 30 µm. This observation indicates that the strain applied to the PC substrate is well transferred to the MoS_2 bilayer at our investigated sample position.

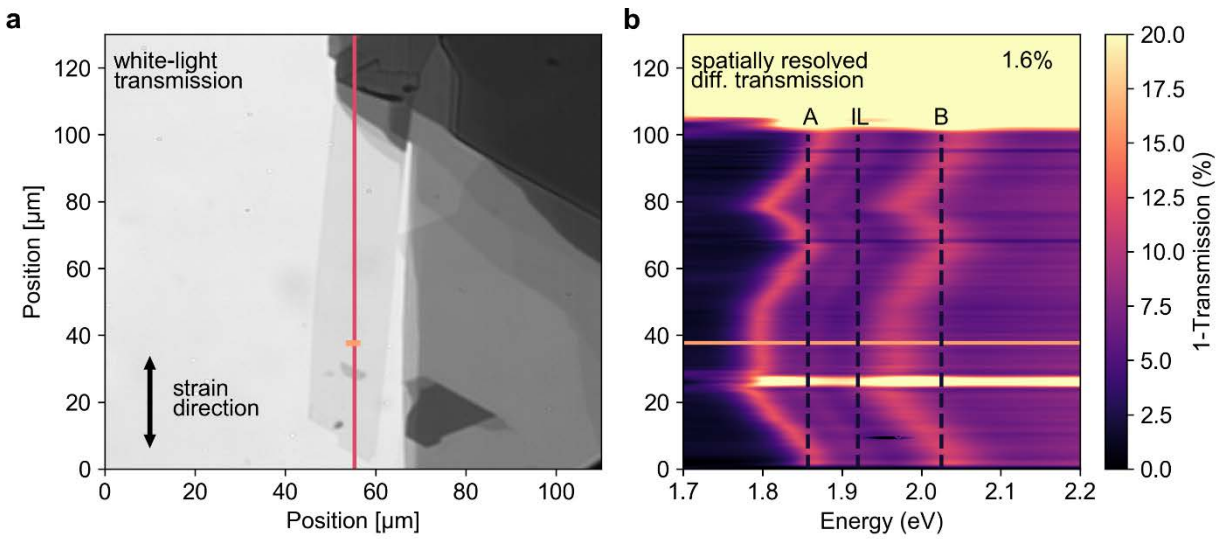

Figure S3: Strain transfer from the PC substrate to the MoS_2 bilayer. a) White-light transmission image of the investigated bilayer. Differential transmission spectra are recorded along the vertical pink line in strain direction (indicated by the vertical black arrow). The orange horizontal dash indicates the position where the spectra from the main text are analyzed. b) Spatially resolved differential transmission spectra along the vertical pink line in a) at 1.6% of uniaxial strain, which shows the strain transfer from the PC substrate to the MoS_2 bilayer. The horizontal orange line indicates the position, where the spectra of Fig. 1 are extracted. The vertical dashed lines mark the excitonic positions without externally applied strain.

REFERENCES

1 R. Frisenda, Y. Niu, P. Gant, A. J. Molina-Mendoza, R. Schmidt, R. Bratschitsch, J. Liu, L. Fu; D. Dumceno, A. Kis, D. Perez de Lara, and A. Castellanos-Gomez, *J. Phys. D: Appl. Phys.*, 2017, **50**, 074002

2 D. Christiansen, M. Selig, G. Berghäuser, R. Schmidt, I. Niehues, R. Schneider, A. Arora, S. Michaelis de Vasconcellos, R. Bratschitsch, E. Malic, and A. Knorr, *Phys. Rev. Lett.*, 2017, **119**, 187402

3 I. C. Gerber, E. Courtade, S. Shree, C. Robert, T. Taniguchi, K. Watanabe, A. Balocchi, P. Renucci, D. Lagarde, X. Marie, and B. Urbaszek, *Phys. Rev. B*, 2019, **99**, 035443

4 R. Schmidt, I. Niehues, R. Schneider, M. Drüppel, T. Deilmann, M. Rohlfing, S. Michaelis de Vasconcellos, A. Castellanos-Gomez, and R. Bratschitsch, *2D Mater.*, 2016, **3**, 021011

5 I. Niehues, A. Blob, T. Stiehm, R. Schmidt, V. Jadriško, B. Radatović, D. Čapeta, M. Kralj, S. Michaelis de Vasconcellos, and R. Bratschitsch, *2D Mater.*, 2018, **5**, 031003