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Electronic Supplementary Information: Exposing the trion's fine structure by controlling the carrier concentration in hBN-encapsulated MoS_2

Magdalena Grzeszczyk,[*a] Katarzyna Olkowska-Pucko,[a] Karol Nogajewski,[a] Kenji Watanabe,[b] Takashi Taniguchi,[c] Piotr Kossacki,[a] Adam Babiński,[a] and Maciej R. Molas[a]

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Sample fabrication

The investigated MoS_2 monolayers and hBN flakes were fabricated by two-stage PDMS-based mechanical exfoliation of naturally occurring bulk crystal. Initially, the hBN thin films were exfoliated onto a 90 nm SiO_2/Si substrate and annealed at 200°C. That non-deterministic approach provides the best quality of the substrate surface. Subsequent layers were transferred deterministically using a microscopic system equipped with a *x-y-z* motorized positioners. The assambled structures were annealed at 160°C for 1.5 hour in order to ensure the best layer-to-layer and layer-to-substrate adhesion and to eliminate a substantial portion of air pockets on the interfaces between the constituent layers.

Sample characterization

Sample characerization was done by the use of Raman scattering mapping and atomic force microscopy (AFM) imaging. Raman spectra were obtained by using micro-Raman LabRAM HR system with a 100x objective lens and an excitation source of 532 nm laser light. The scattered light was detected in a backscattering configuration. The lateral resolutions was estimated to be below 1 μm.

The roughness and strain distribution in studied sample has been examined using tapping mode with Solver Nano system, equipped with a professional 100 micron CL piezotube scanner with low noise capacitance sensors. Root mean square roughness for area with monolayer MoS_2 for all characterized samples did not exceed 1 nm (within 5 μm by 5 μm area).

* *E-mail: Magdalena.Grzeszczyk@fuw.edu.pl*
[a] *Institute of Experimental Physics, Faculty of Physics, University of Warsaw, 02-093 Warsaw, Poland;*
[b] *Research Center for Functional Materials, National Institute for Materials Science, 305-0044, Japan*
[c] *International Center for Materials Nanoarchitectonics, National Institute for Materials Science, 305-0044, Japan*

Experimental setup

The PL, PLE, and RC experiments were performed using a λ=515 nm (2.41 eV) continuous wave (CW) laser diode, a tunable CW single-mode DCM dye laser, and a 100 W tungsten halogen lamp, respectively. The studied samples were placed on a cold finger in a continuous flow cryostat mounted on *x-y* manual positioners. The excitation light was focused by means of a 100x long-working distance objective with a 0.55 numerical aperture producing a spot of about 1 μm diameter in PL/PLE measurements and 4 μm diameter in RC measurements. The signal was collected via the same microscope objective, sent through a 0.75 m monochromator, and then detected by using a liquid nitrogen cooled charge-coupled device camera. The excitation power focused on the sample was kept at 70 μW during all measurements to avoid local heating, except for experiments carried out as a function of excitation power.

Magneto-optical experiments were performed in the Faraday configuration using a free-beam-optics arrangement in a superconducting coil producing magnetic fields up to 10 T. Samples were placed in a helium gas atmosphere at the temperature of 5 K. The excitation light was focused by a single aspheric lens to a spot smaller than 2 μm in the case of laser and 6 μm for the white light illumination. The lens was mounted on the x-y-z piezoelectric stage inside the cryostat. The circular polarizations of the emission were analyzed using a polarizer and a $\lambda/4$ waveplate placed directly in front of the spectrometer.

Reflectance contrast in magnetic field

Due to the complexity of resolving the individual trion-releated emission features in helicity-resolved PL spectra shown in the main text, we additionally performed reflectance contrast measurements. As mentioned in the main text: two absorption-type resonances can be easily distinguished for the negative trions in the RC spectra, T_1 and T_2, ascribed to the intravalley spin-singlet

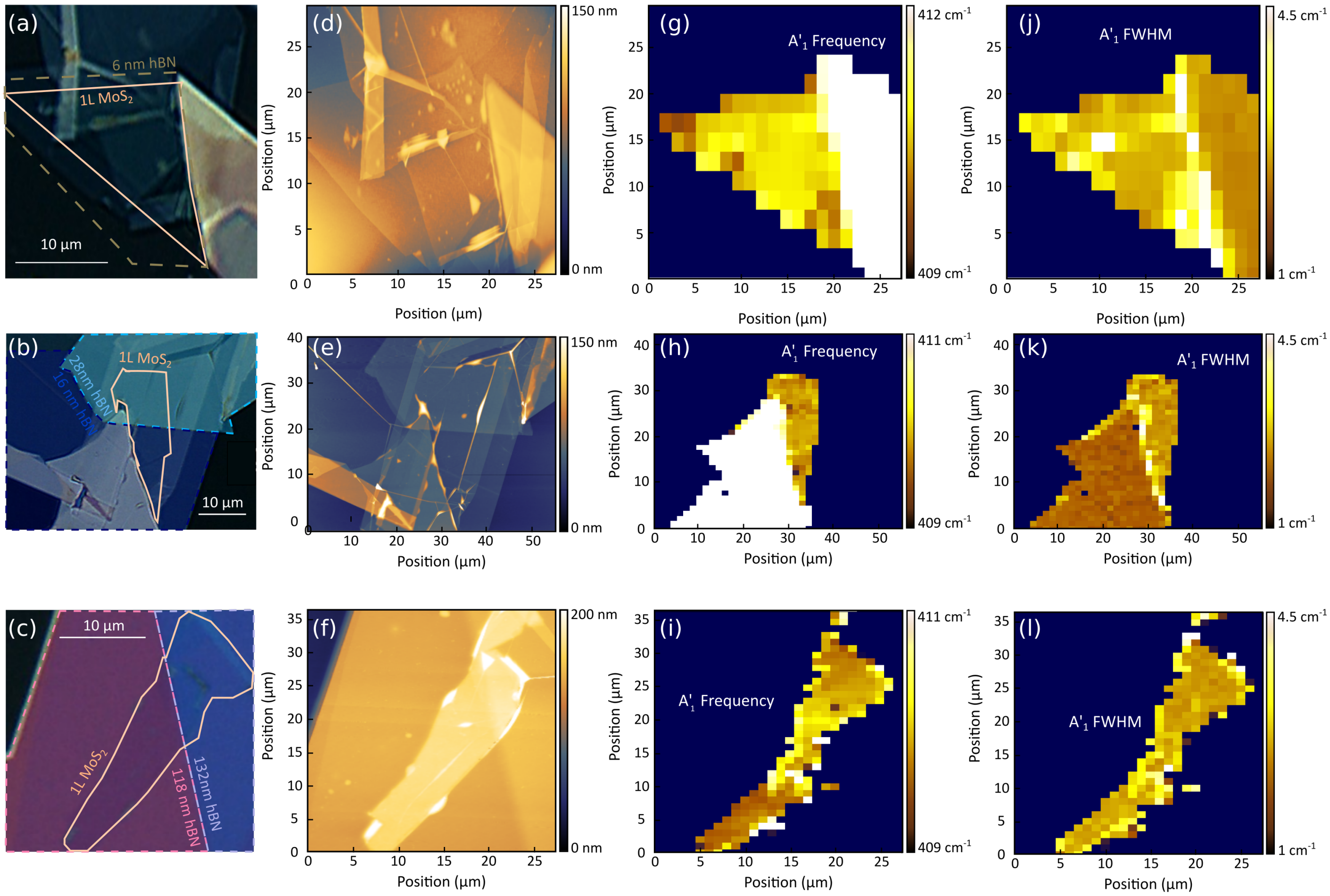

Fig. 1 (a)-(c) Optical images, (d)-(f) AFM topography images, Raman scattering map of A'_1 peak (g)-(i) frequency and (j)-(l) full width at half maximum (FWHM) for selected samples.

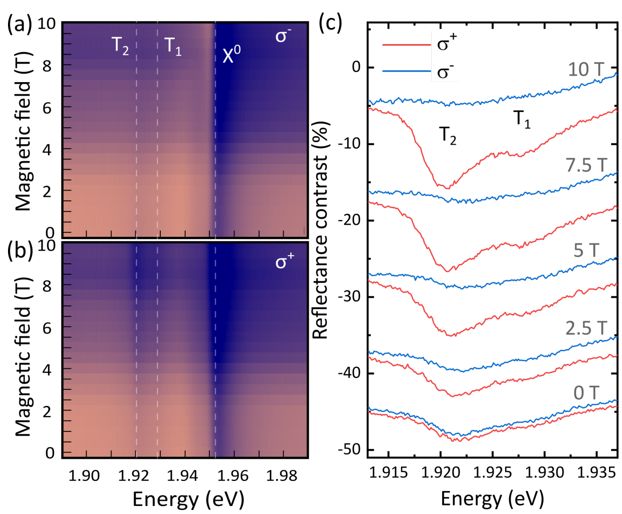

Fig. 2 False-color map of the helicity-resolved low-temperature reflectance contrast measured on the encapsulated MoS_2 monolayer with the 6 nm-thick bottom hBN for (a) σ^- (b) σ^+ polarized light. The white dashed lines are indicated as a guide to the eye. (c) Reflectance contrast spectra of trion-related emission at selected magnetic fields. The red (blue) colour correspond to the σ^+ (σ^-) polarized light. The spectra are vertically shifted for clarity.

and intervalley spin-singlet, respectively.[1–3]

As presented in Fig. 2, similar to the results of PL experiments, with σ^- polarization as the magnetic field increases, the features associated with trions weaken to the point of being indistinguishable. In the opposite photon polarization, σ^+ both resonances gain intensity with increasing magnetic field. The observed evolution for the T_1 and T_2 line is analogous to that observed in the RC spectra reported in Ref.[3].

Notes and references

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