Excitonic characteristics of singlelayer MoS_2

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Abstract

Complementary to the gapless material graphene, the transition-metal dichalcogenide molybdenite (MoS_2) is a promising two-dimensional layered semiconductor for future ultrathin nanoelectronic and optoelectronic devices. Recent experiments have revealed the transition from an indirect to a direct-gap semiconductor, going from the bulk material to the monolayer regime [1]. Subnanometer thickness, large direct bandgap in the visible range and ultrafast carrier dynamics make singlelayer MoS_2 interesting for devices like transistors, ultrafast optical switches or photovoltaic applications.
Here, we present photoluminescence and Raman measurements on single- and few-layer MoS_2 flakes. The samples are produced by the well-known transparent tape liftoff method from natural molybdenite. To identify monolayer regions on the substrate, we first characterize them in an optical microscope. For further determination of the layer number, we apply scanning Raman spectroscopy and atomic force microscopy [2, 3]. The observation of a frequency shift in the Raman spectrum of the interlayer shear mode induced by a change in the layer number allows us to use a mapping of this mode frequency for the assignment of the number of layers in few-layer samples (fig. 1).
Via photoluminescence (PL) measurements on the singlelayer regions of our samples, we investigate the different behavior of the so-called A and B excitons. These bound electron-hole-pairs arise from transitions from the spin-orbit split valence band to the conduction band at the K-point of the Brillouin zone with energies of 1.8 eV (A) and 2.0 eV (B). We see a difference in the temperature-induced energy shift for the two excitons, going from 4 K to room temperature. We also infer an energy transfer between A and B excitons dependent on the excitation power.
Recently, coupled spin-valley-physics in monolayer MoS_2 due to symmetry reasons was predicted [4]. Based on an inverse spin occupation of the split valence band at the K_+ and the K_- valley in the Brillouin zone, a lot of interesting new effects are expected using the valley index as a new degree of freedom in this system. We can see the conservation of the helicity of the incoming laser light in the PL if the excitation energy lies between the A and B exciton energy, which might be a basic effect of coupled spin-valley-physics (fig. 2).
Furthermore, we could generate monolayer regions out of few-layer flakes via intense focused laser radiation. Thereby, monolayer MoS_2 samples can be designed in shape and size as desired.
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References

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Figures

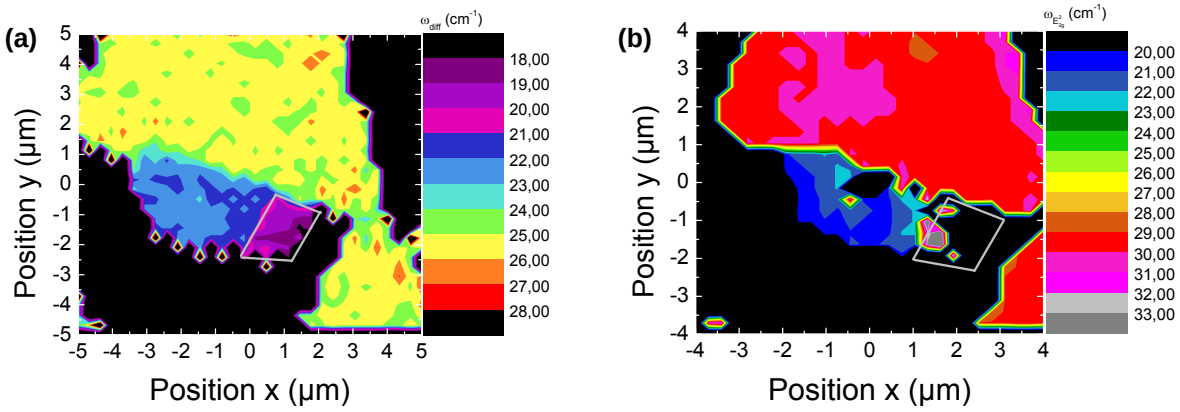

Fig. 1: (a) Raman mapping of the frequency difference of the E^1_{2g} and the A_{1g} mode. Purple color indicates a monolayer region. (b) Raman mapping of the interlayer shear mode on the same sample as in (a). The frequency of this mode is strongly dependent on the layer number and is therefore suitable for sample characterization.

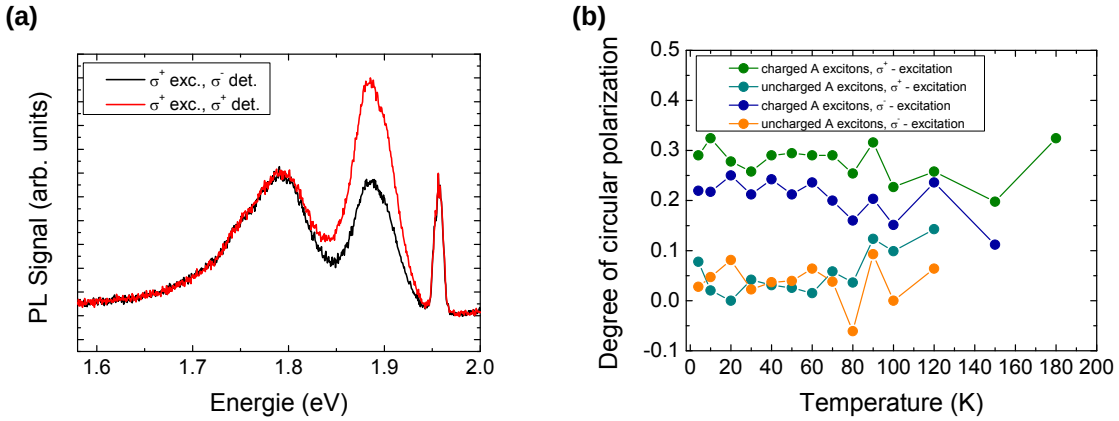

Fig. 2: (a) Photoluminescence spectra of singlelayer MoS_2 at 20 K under circularly polarized near resonant excitation and helicity dependent detection. We see a conservation of the helicity of the incoming laser light. (b) Degree of circular polarization under circularly polarized near resonant excitation for sample temperatures from 4K up to 180K.