Excitation-dependent luminescence of mono- and few-layer MoS_2

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Introduction

2D materials are very promising in terms of application in microelectronics, including optoelectronics. For 2D MoS_2, there is still limited information about the influence of optical excitation conditions (e.g., laser power in a broad range and different energies) on the formation and behavior of the trion emission component, finely separated from the A exciton band. In this paper, the behavior of trion and exciton photoluminescence (PL) was studied under various laser excitation conditions, i.e., energy and power, in monolayer and few-layer MoS_2 flakes grown on SiO_2/Si.

Methods

The thicknesses of of MoS_2 triangular flakes grown on SiO_2/Si by chemical vapor deposition was determined with AFM and Raman spectroscopy (fig. 1a). To obtain μ-PL and μ-Raman spectra and mappings, the laser excitations of 532 and 405 nm were focused on sample to a spot of ~3 μm diameter. The PL maps (fig. 1b) present the PL intensity of each band with a mapping step of 1 μm.

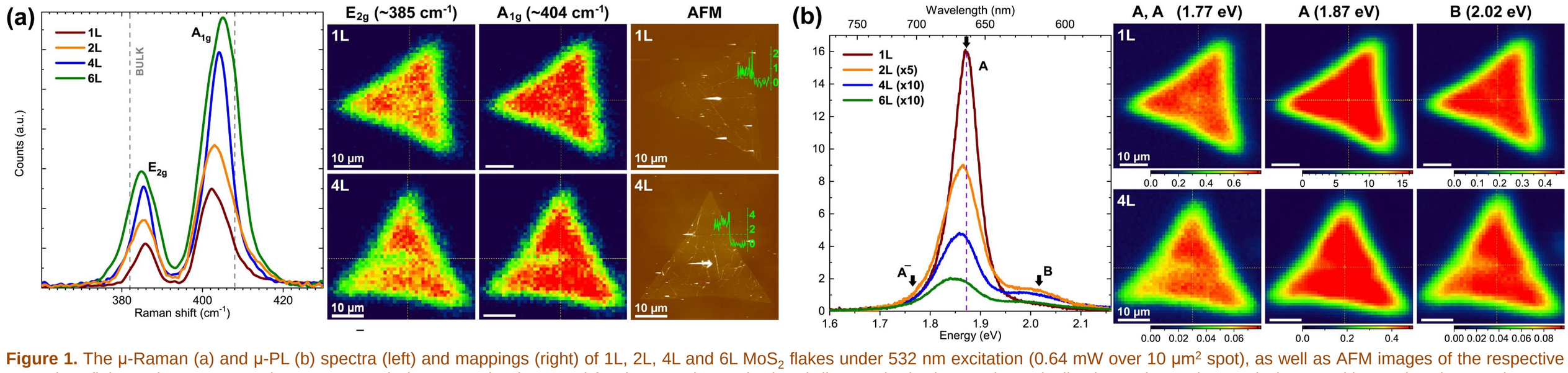

Figure 1. The μ-Raman (a) and μ-PL (b) spectra (left) and mappings (right) of 1L, 2L, 4L and 6L MoS_2 flakes under 532 nm excitation (0.64 mW over 10 μm^2 spot), as well as AFM images of the respective 1L and 4L flakes. The arrows on the spectra mark the spectral points used for the mappings. The band diagram in the inset schematically shows the exciton and trion transitions. The plots on the AFM images show the estimated thickness in nanometers along the dashed lines.

Results and Discussion

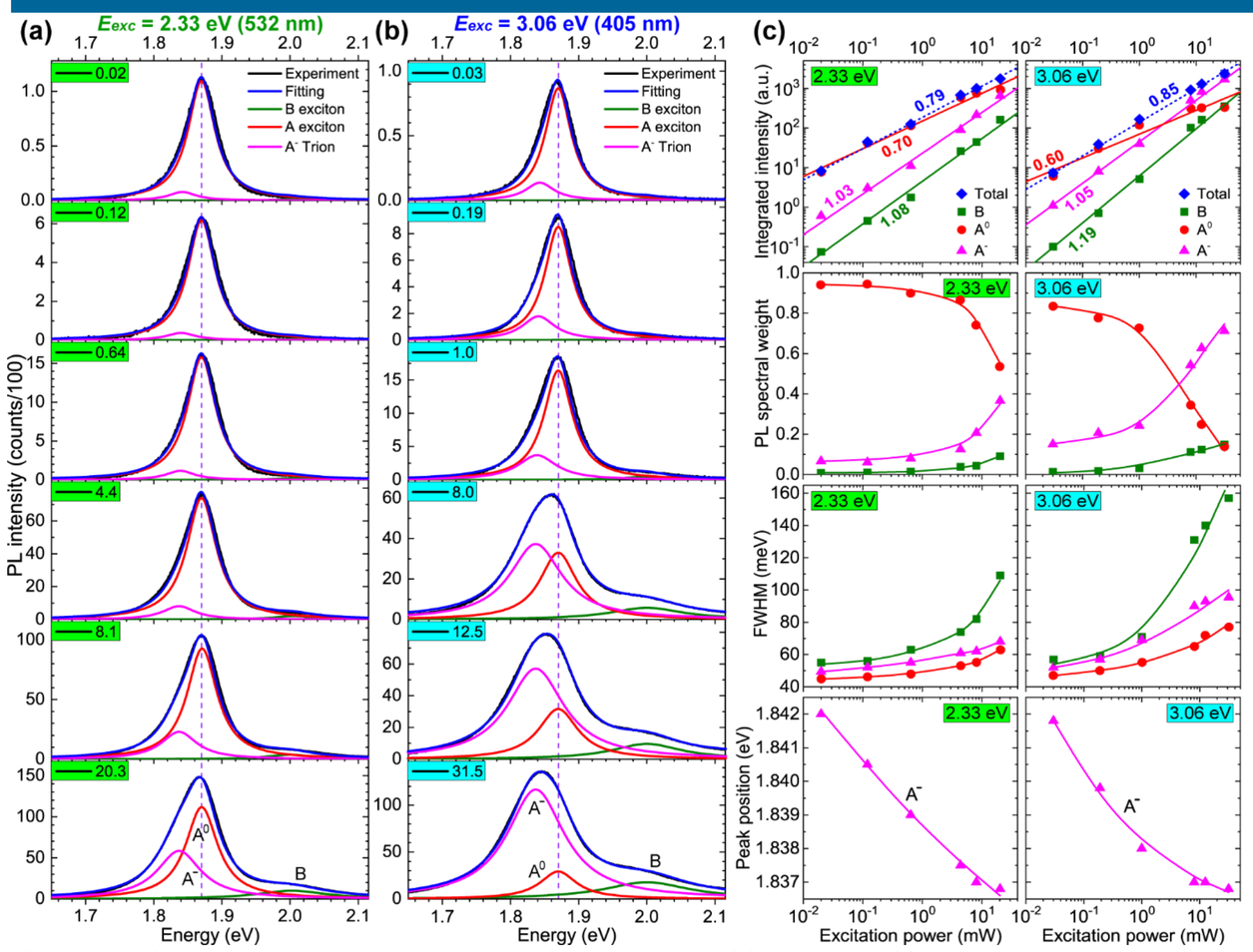

Figure 2. The μ-PL spectra of 1L MoS_2 flake excited by (a) 532 and (b) 405 nm at different laser powers (mW) and fitted by three Lorentzian functions related to the A and B excitons and the trion components. (c) Parameters of the spectra and their components versus excitation power.

The enhancement of trion binding energy leads to a strong intensification of the trion component. Thereby, the intense A exciton/trion PL band redshifts and becomes more asymmetric when increasing power of excitation at both energies of 2.33 or 3.06 eV. Simultaneously, the B exciton contribution to the spectrum also increases. Such a behavior of an enhanced formation of the trions and B excitons is explained by a rate of photogenerated electron-hole pairs leading to a higher population of nonequilibrium electrons. In the measurements under the higher excitation energy of 3.06 eV, the PL redshift is more prominent, because the trion component is more intense compared to the A band. This is explained considering a higher absorption of MoS_2 at higher energies.

Using Lorentzian functions, the PL spectra were deconvoluted to the 3 separated bands corresponding to A and B excitons as well as trion, and the band parameters were analyzed (fig. 2, 3). The trion spectral weight and dissociation energy are observed to increase with the number of layers, being correlated with an increase in nonequilibrium electron density. We propose the presence of many-body effects explaining this unusual dependence in CVD-MoS_2 on a *n*-type SiO_2/Si substrate. The analysis shows a faster intensification of the trion component compared to the A exciton with increasing laser power. The trion binding (dissociation) energy varied from 28 to 33 meV in monolayer MoS_2 and from 32 to 46 meV in few-layer MoS_2, when increasing power of excitation light. The phenomenon is found to be more prominent when increasing energy of excitation from 2.33 to 3.06 eV.

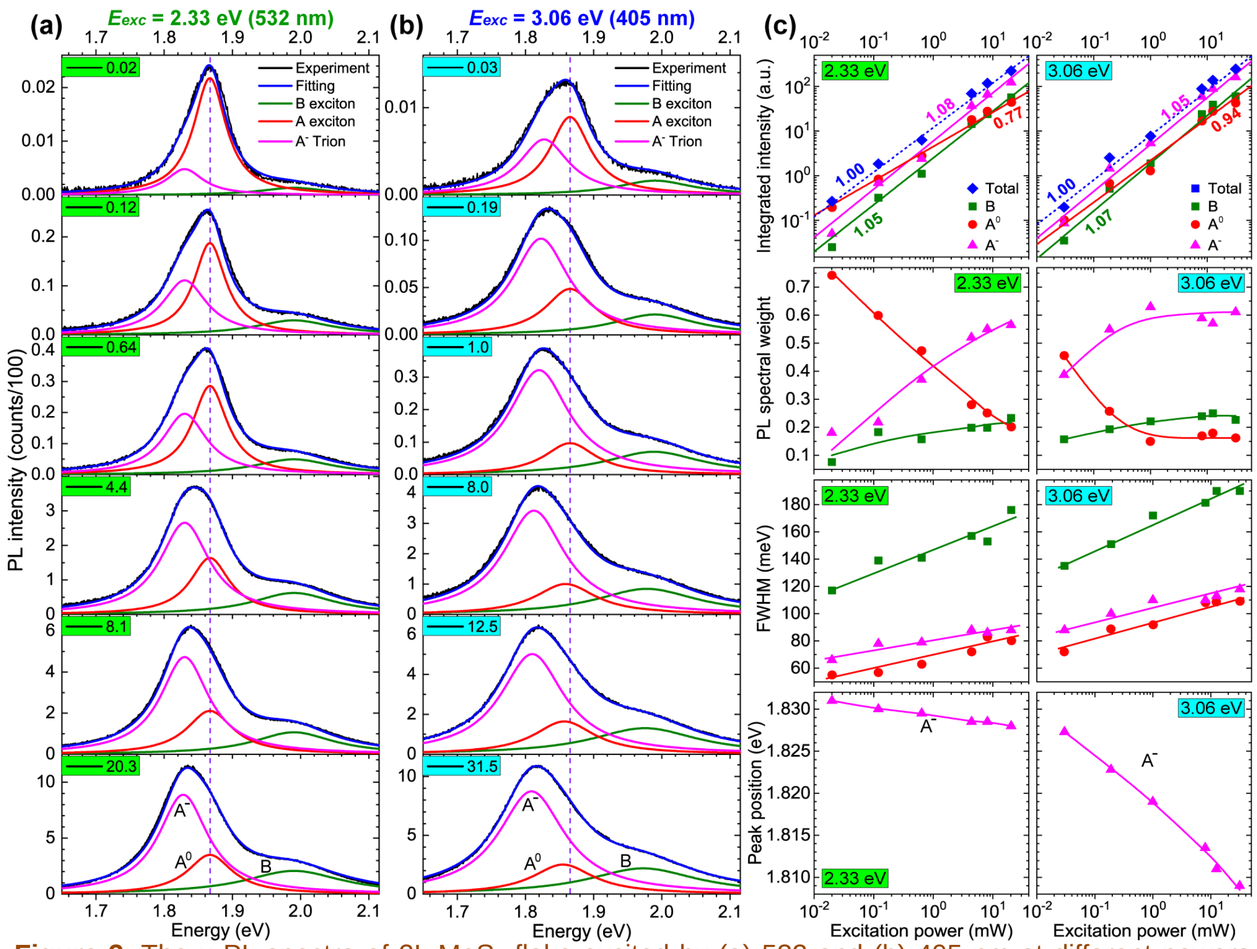

Figure 3. The μ-PL spectra of 2L MoS_2 flake excited by (a) 532 and (b) 405 nm at different powers (mW) and fitted by three Lorentzian functions related to the A and B excitons and the trion components. (c) Parameters of the spectra and their components versus excitation power.