Low-temperature photocarrier dynamics in single-layer MoS₂ flakes

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Abstract

The dichalcogenide MoS_{2} , which is an indirect-gap semiconductor in its bulk form, was recently shown to become an efficient emitter of photoluminescence [1,2] as it is thinned to a single layer, indicating a transition to a direct-gap semiconductor due to confinement effects. The material is a layered structure of weakly coupled, covalently bonded two-dimensional sheets, and it can be prepared, just as graphene, using chemical or mechanical exfoliation techniques.

Here, we present temperature-dependent and time-resolved photoluminescence (PL) studies [3] of single-layer MoS₂ flakes, some of which have been covered with thin dielectric layers (HfO₂ or Al₂O₃) using atomic layer deposition (ALD). The flakes are prepared from bulk MoS₂ by transparent tape liftoff. For initial characterization, optical, atomic-force and Raman microscopy are used (Fig.1). We observe that the PL peak position of individual single-layer MoS₂ flakes fluctuates by about 25 meV at room temperature. Mild annealing at 150°C in vacuum leads to a homogenization of the PL peak position. For as-prepared flakes, we clearly see two PL peaks at low temperatures, which we may assign to bound and free exciton transitions (Fig. 2a). Oxide-covered flakes only show the free exciton peak, indicating that bound excitons form due to adsorbates at the flake surface (Fig. 2b). [4]. Temperaturedependent PL measurements indicate that the oxide-covered flakes are strained due to different thermal expansion coefficients of MoS_2 and the oxides. This observation also indicates that the flakes strongly adhere to the oxide layers. Strain strongly influences the band structure in single-layer dichalcogenides and may even cause a metal-insulator-transition [5]. In time-resolved PL measurements using highly nonresonant, pulsed excitation, we observe very fast photocarrier recombination on the few-ps timescale at low temperatures, with increasing photocarrier lifetimes at higher temperatures due to increased exciton-phonon scattering (Fig. 2c). Excitons which are scattered out of the light cone by phonons first have to relax their momentum before they are able to recombine radiatively. Financial support by the DFG via GRK 1570, SFB689, SPP1285, and KO3612/1-1 is gratefully

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Figures



Fig. 1: (a) Optical micrograph of a thin MoS_2 flake. (b) AFM scan of area marked in (a) by orange square. (c) False color plot of the intensity of the $MoS_2 E_{2g}^1$ Raman mode.



Fig. 2: (a) Temperature-dependent PL spectra of single-layer MoS_2 . (b) PL spectra of bare and oxidecovered single-layer MoS_2 measured at low temperatures. (c) Temperature-dependent Time-resolved PL traces of single-layer MoS_2 .