Electrical control of excitons and trions in MoS_2 monolayer and bilayer crystals

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Ultrathin crystals based on transition metal dichalcogenides form a new fascinating class of materials. Large direct band gaps allow for optoelectronic device concepts, the sub-nanometer thickness is attractive for ultrathin flexible electronics and the unique band structure could evoke new components for spintronics or valleytronics [1]. MoS_2 flakes that can be realized easily by mechanical exfoliation have seen intensive research during the last couple of years, yet main aspects are still unclear. A central question concerns the possibility to manipulate the emission properties by e.g. electric fields. Indeed, charged excitons (trions) that are well known from semiconductor quantum dots could be observed in electrically contacted flakes recently up to room temperature [2]. These findings are limited to monolayers up to now. However, few layer structures have seen growing experimental attention: They are a kind of a link between true two dimensional and bulk semiconductors, and some controversy exists whether significant photoluminescence can be expected from bilayers [3-5]. Up to now no detailed information is available at all on the exciton-trion control in MoS_2 bilayers.

In our contribution, we study the interplay between trions and excitons in MoS_2 bilayers in comparison with monolayers. Flakes were exfoliated mechanically and deposited on Si/SiO_2 (90 nm) substrates. The thickness (layer number) is determined by Raman spectroscopy. Micro-photoluminescence measurements at T = 5 K (Fig. 1) reveal at first a common feature between mono- and bilayer emission: In contrast to the first theories, the dominating emission in both structures stems from the *direct* band gap (A transition), and in both cases, this emission is split into a higher energy line A^0 and a lower energy line A^- with Lorentzian line profile and an energy separation of $\Delta E = 25$ meV. As the MoS_2 flakes are intrinsically negatively doped, we attribute A^0 to an exciton and A^- to a negatively charged trion [2]. For the first time, we see these two lines not only in monolayers but also clearly separated in bilayers, due to the relatively small linewidth of 30 meV (exciton) and 50 meV (trion). However, significant differences are recorded as well: In monolayers a strong influence of defect-bound excitons is found, while these are absent in bilayers and instead emission from the indirect band gap is visible, in agreement with theory.

In order to elucidate the exciton-trion interplay in detail, contact lines to the flakes were defined by electron beam lithography and evaporation of Ti-Au. The Si substrate serves as back gate contact. Micro-PL measurements were performed under variation of the gate voltage U_g which leads to an agglomeration (depletion) of excess electrons for positive (negative) U_g. In PL emission, this effect can be monitored easily in the bilayer structures, where the emission shows a clear shift from the trionic to the excitonic component, if U_g changes from positive to negative values (Fig. 2, right). In contrast, the monolayer shows a high disposition to defect luminescence: A switching is here only possible from trion emission to bound excitons, when U_g is decreased (Fig.2, left). To suppress the bound states, the analysis was expanded up to 300 K. At room temperature, we actually find an opposite behaviour: Switching between exciton and trion can now be observed easily in the monolayer. In the bilayer only exciton and indirect transition can be detected independent of U_g, indicating less robust trions in bilayer as compared to monolayer MoS_2.

References

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Figures

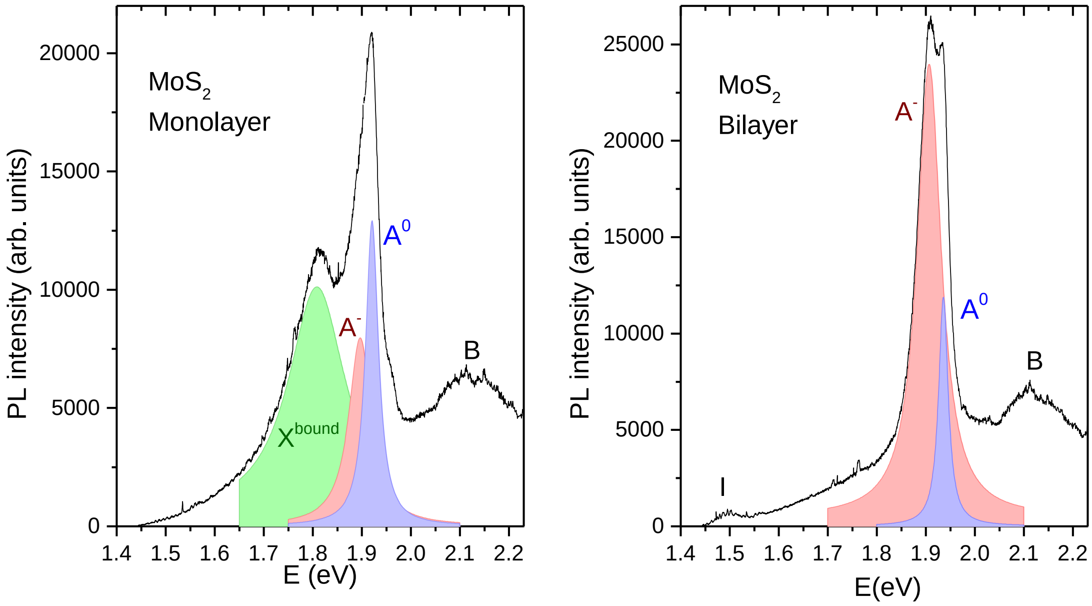

Fig 1.: Photoluminescence from MoS_2 monolayer and bilayer. The symbols represent A^0; exciton, A^-: negative trion, B: transition between conduction band and split-off valence band, I: indirect band gap, X^{bound}: exciton bound to defects.

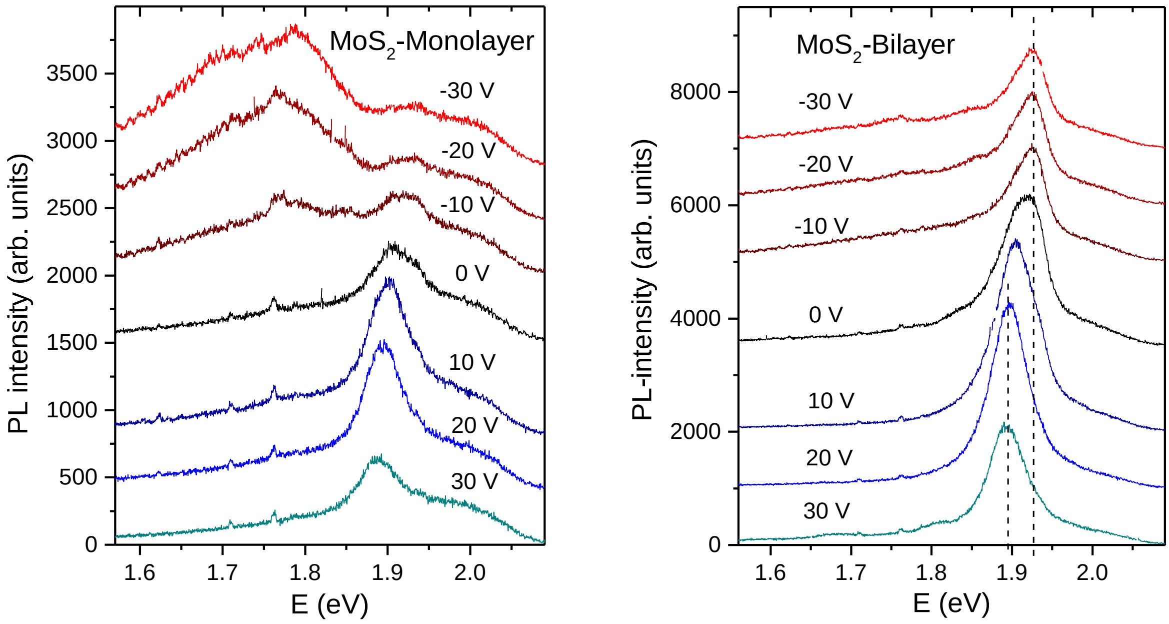

Fig 2.: Photoluminescence spectra from MoS_2 monolayer and bilayer in dependence of the gate voltage.