Ultrafast Exciton and Charge Carrier Dynamics in Monolayer MoS_2 Measured with Time-resolved Spectroscopic Ellipsometry

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Monolayer MoS_2 belongs to the Transition Metal Dichalcogenide (TMD) family with interesting optical properties like excitons with exceptionally high binding energies in the range of hundreds of meV [1] which allows us to study exciton physics at room temperature. MoS_2 is a promising candidate for future electronic and photonic devices like transistors [2] or LEDs [3]. We commercially purchased monolayer MoS_2 film on a double side polished c-cut sapphire substrate. The quality of our sample was investigated with Laser Scanning Microscopy. We found a closed, homogeneous film with a low amount of defects. The nature of the defects was investigated using Raman-spectroscopy, photoluminescence and imaging ellipsometry. The defects turned out to be either nucleation centers of multilayer MoS_2 or areas (roughly 15x15 μm^2) with a high defect density which unintentionally n-doped our sample. The optical properties of our sample were probed with Spectroscopic ellipsometry, which gave us access to the dielectric function in the spectral range from 0.5 to 6.5 eV. To describe the features found in the MoS_2 dielectric function we modeled it with 7 oscillators. The resulting dielectric function aligns well with the reported literature values and extend its knowledge into the UV spectral range. To access the exciton dynamics at ultrashort times we employed time-resolved spectroscopic ellipsometry [4], a pump-probe technique. We were able to measure the temporal evolution of the dielectric function for delay times up to 5 ns in a spectral range from 1.75 - 3.5 eV with a temporal resolution of 80 fs. Figure 1 shows the changes of ϵ_2 when the pump excites the A-exciton at an energy of 1.85 eV. A steep reduction in the value of ϵ_2 can be observed at 1.85 eV and 2.0 eV which we attribute to the excitation of the A- and B-exciton. We can also observe the formation of negatively charged trions which leaves free holes in the valence band which can then scatter to the Γ-Point and block the C-exciton. This results in a decrease of ϵ_2 at 2.85 eV. Furthermore new transitions from lower valence bands are now possible which leads to an increase of ϵ_2 at 1.75 eV and 2.5 eV. We further performed measurements exciting also the B- and C-exciton as well as the electronic bandgap. From this dataset, under comparison with band structure data, we derive a comprehensive picture on the carrier dynamics in monolayer MoS_2, compromising the formation of trions as well as hole scattering from the K to the Γ-Point of the BZ.

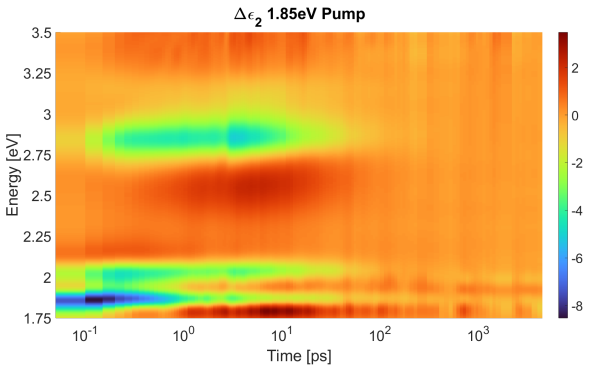

Fig. 1. Dynamic Changes in ϵ_2 after excitation at 1.85 eV excitation

References

[1] Soohyung Park et al., 2D Materials **5** 025003 (2018)
[2] B. Radisavljevic; A. Radenovic; J. Brivio; V. Giacometti and A. Kis, Nature Nanotechnology **6**, 147-150 (2011).
[3] Shen, C.-C.; Hsu, Y.-T.; Li, L.-J.; Liu, H.-L., Applied Physics Express 2013, **6**, 125801
[4] Richter et al. Rev. Sci. Instrum., **92**, 033104 (2021)