Comparison between optical properties of oxidized and non-oxidized MoS_2 monolayer

Maya Isarov, Jenya Tilchin, Georgy I. Maikov, Efrat Lifshitz
Technion-Israel Institute of Technology, Haifa, Israel
mayai@tx.technion.ac.il

Arend van der Zande
Columbia University, New York, USA

MoS_2 is a semiconductor material with hexagonal lattice structure. Bulk MoS_2 has an indirect band gap while a single monolayer behaves like a direct semiconductor. In comparison with graphene thickness of one atom, the MoS_2 monolayer has thickness of three atoms, giving the monolayer more stiffness. MoS_2 monolayer features make it a good candidate to serve as a component in optical triggered switches. [1]
In this work MoS_2 monolayers were grown on top of SiO_2 substrate (with courtesy to Dr. van der Zande from Columbia University for the preparation of the samples). The properties of the samples were examined on a resolution of a single monolayer utilizing combined method, comprised of atomic force microscopy (AFM) and confocal spectroscopy (photograph of the set-up is given in Fig.1). This combination eliminates average effects found in a measure of an ensemble and uncover the properties of individual monolayers.
The emission spectra were recorded at 4 K under cw and picosecond pulsed non-resonant excitation with various laser powers and presence of magnetic fields of various strengths, as well as at different polarized detection filtering. We had obtained two different types of behavior of the monolayers under light exposure (Fig.2). The first one possesses mainly a strong emission band with typical peak energy at ~1.9 eV and a lifetime 300-500 psec (Fig.2 right inset).The second type has an additional low energy shoulder at 1.75-1.85 eV, with a bi-exponential lifetime, τ=17 and 175 nsec, as shown in Fig.2 left inset. The emission shoulder can be correlated with a bound exciton formation on surface defects, while the absence of the shoulder could refer to non-oxidized surface [3], where oxygen substitutes the sulfur vacancies. Furthermore, the main band is shifted up to 21 meV to a new position under illumination with time (see dependent plot in Fig.3). This shift can be related to a transfer of a neutral exciton to a charged exciton after certain irradiation exposure. Recently, Mak *et al*.[2] had shown the activation of this charged emission channel by applying a gate voltage on a monolayer. By examining five different layers we obtained that high excitation power reduces the time for layer charging, as shown in Fig.3 right inset. A typical exposure time evolution of these two states throughout 600 sec is presented in Fig.3 middle inset. At 120 sec there is equal intensity exchange between the two states. The shift to the charged state is irreversible. After 18 hours at dark, the spectrum exhibits slight red shift and broadening, as shown in Fig.3 left inset.
Since the charging effect possesses irreversibility, we preliminary exposed the layer to intense laser and then recorded the power dependent emission. All of the monolayers showed a sub-linear dependence, suggesting the existence of different emission channels. Also, they exhibit an additional high energy band, blue shifted by 155 meV above the main band (Fig.2). This band is in good agreement with the spin-orbit interaction splitting of the valence band edge.
The polarization measurements at zero magnetic field reveal a slight linear and circular polarized components of the main emission peak. Fig.4.a represents emission intensity dependence under circular polarization detection. The broad structure of the emission line suggests that the exciton recombines throughout a number of the acoustic and optic phonon channels. This admixture of the emission channels breaks a pure polarization identity of the recombination channel. The polarization measurements at 8 T reveal energy difference of ~2.5 meV between circular polarizations (Fig.4.b). Future measurements and investigation shall be done for better understanding of MoS_2 monolayers features.

References:

[1] K. F. Mak *et al*., Physical Review Letters 105, 136805, 2010
[2] K. F. Mak *et al*., Nature Materials 12, 207, 2013
[3] G. Plechinger *et al*., Phys Status Solidi RRL 6 (3), 126, 2012

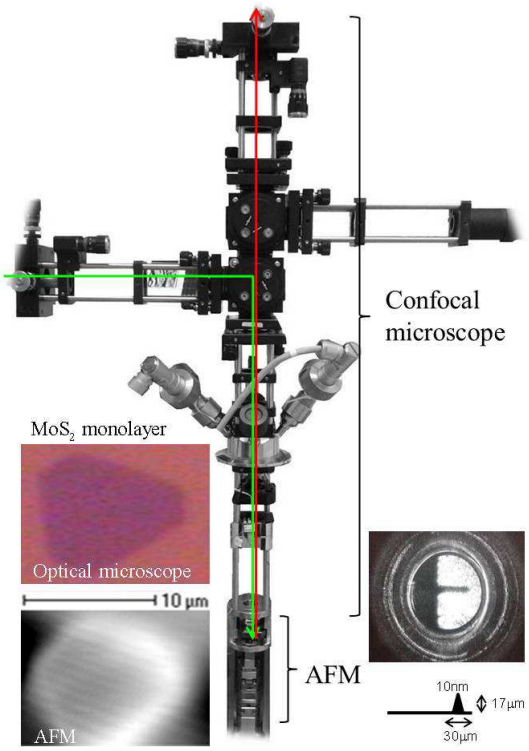

Figure 1: Combined AFM-Confocal Microscope system.

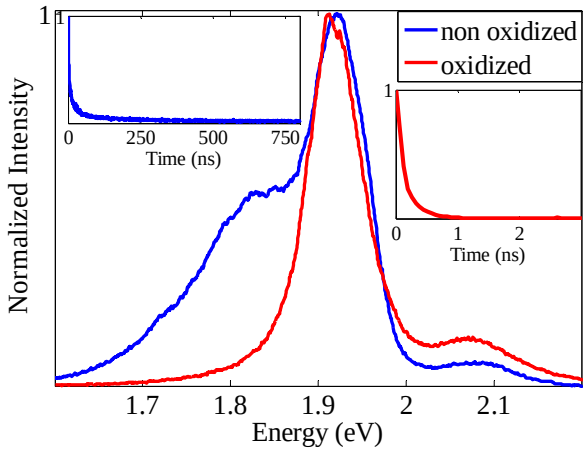

Figure 2: PL spectra of oxidized (red) and non-oxidized (blue) monolayers.Right inset: PL-decay curve of the main band at ~ 1.9 eV. Left inset: Low energy band between 1.75 – 1.85 eV.

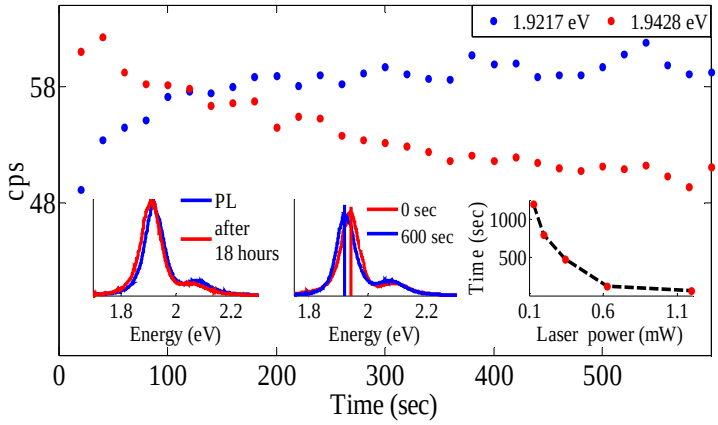

Figure 3: Intensity dependence on exposure time of oxidized monolayer, measured at two different energies (see legend).Left inset: PL vs. PL after 18 hours at dark of the same monolayer. Middle inset: PL recorded at 0 and 600 sec under irradiation. Right inset:Interception time between neutral and charged states of 5 different monolayers at various laser powers.

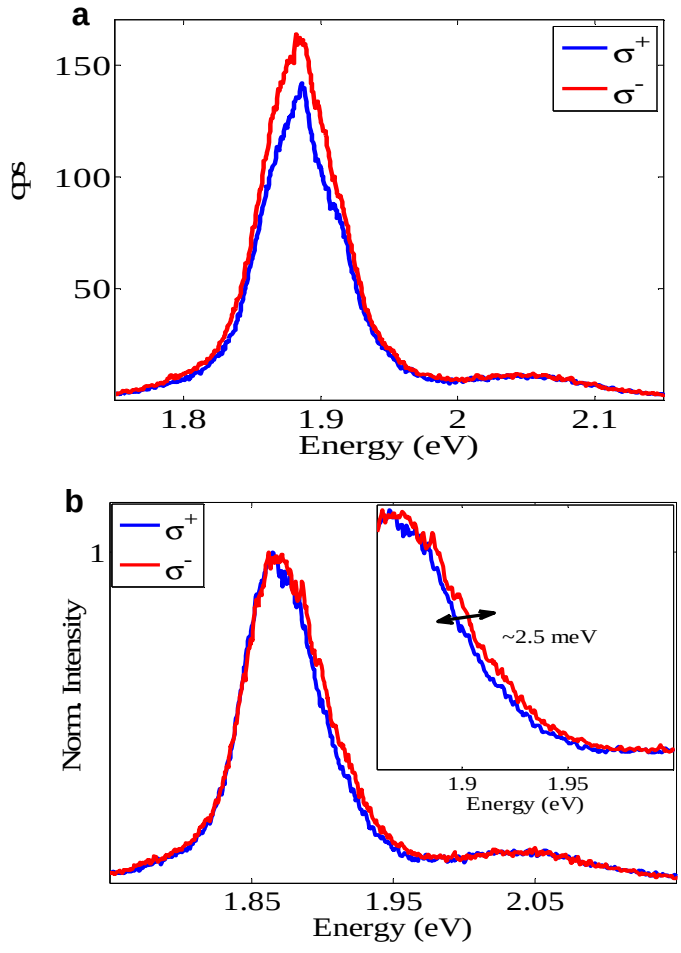

Figure 4: Circular polarized emission: a. At zero magnetic field. b. At 8 T. Inset: Energy difference between σ^+ , σ^-.