Comparison of optical properties of $MoSe_2$ monolayers; samples grown by MBE technique vs mechanically exfoliated one.

Mateusz Raczyński[1], Tomasz Kazimierczuk[1], Wojciech Pacuski[1] and Piotr Kossacki[1]

[1]*Division of Solid State Physics, Institute of Experimental Physics, Faculty of Physics, University of Warsaw, ul. Pasteura 5, 02-093 Warsaw, Poland*

Semiconducting transition metal dichalcogenides (s–TMD) exhibit strong optical response within the limit of a single atomic layer. It was first observed for molybdenum disulfide, which exhibits strong photoluminescence due to direct bandgap.[1] The state of the art s–TMD monolayer samples are produced by the process of mechanical exfoliation, which became famous with the first graphene sample produced by this method.[2] The bulk crystals that are used for mechanical exfoliation can be grown with low defects concentration within their volume, especially when compared to defects concentrated on their surfaces. Therefore, the subsequent thinning of the bulk–grown sample with an adhesive tape allows for the exfoliation of atomically thin layered crystals (top–down approach). The amount of defects in such layers is similarly small as in the case of the original bulk crystal. For that reason – the surface of the sample is the sample – epitaxial techniques (bottom–up approach) were expected to be always inferior when compared to the manually exfoliated ones from the bulk crystal. The breakthrough happened with $MoSe_2$ monolayer sample grown by molecular beam epitaxy on high–quality substrate of hexagonal boron nitride.[3] It showed the possibility of producing TMD monolayer sample with an optical response comparable to the best – state–of–the–art – exfoliated ones. The development of TMD monolayer MBE–growth technique is crucial for industrial application due to its unprecedented spatial homogeneity and repeatability unachievable by the technique based on manual mechanical exfoliation. Moreover, MBE–grown $MoSe_2$ monolayers are folds– and ripples–free, and those are the intrinsic features of mechanically exfoliated samples that are very hard to get rid of.

In our work, we investigate the differences in optical properties of MBE-grown and mechanically exfoliated samples. We show that the photoluminescence intensity variation in MBE–grown samples can be explained by three major factors – the interference effects within the h–BN substrate, incomplete surface coverage by epitaxial monolayer (measured with atomic force microscope) and the radiatively non-active edge of $MoSe_2$ flakes. Additionally, we performed polarisation–resolved magnetospectroscopy and time–resolved measurements of exciton photoluminescence. We extract the exciton intervalley scattering time of $\sim$16 ps that is universal for both types of samples – exfoliated and MBE–grown. We also show variations of the charged exciton formation time among different samples caused by the changes in intrinsic carrier density.

[1] K. F. Mak, C. Lee, J. Hone, J. Shan, T. F. Heinz, *Phys. Rev. Lett.* **105**, 13 (2010).

[2] K. S. Novoselov, D. Jiang, F. Schedin, T. J. Booth, V. V. Khotkevich, S. V. Morozov, and A. K. Geim, *PNAS* **102**, 30 (2005)

[3] W. Pacuski, M. Grzeszczyk, K. Nogajewski, A. Bogucki, K. Oreszczuk, J. Kucharek, K. E. Połczyńska, B. Seredyński, A. Rodek, R. Bożek, T. Taniguchi, K. Watanabe, S. Kret, J. Sadowski, T. Kazimierczuk, M. Potemski, and P. Kossacki, *Nano Lett.* **20**, 5 (2020).