Investigation of Photoluminescence Intensity from MBE–Grown $MoSe_2$ Monolayers Towards the Performance of the Exfoliated Samples.

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Until recent discoveries [1] the ultimate optical quality of the molybdenum diselenide monolayers was restricted to samples mechanically exfoliated from bulk crystal and encapsulated in hexagonal boron nitrate flakes. Such structures exhibit very distinct, narrow–line excitonic resonances in low–temperature photoluminescence as well as sharp reflectivity spectra. Recently, similarly high optical quality was achieved due to special growth via molecular beam epitaxy technique on exfoliated hexagonal boron nitre flakes that provide a super flat and smooth substrate for the monolayer growth.

In our work, we investigate the photoluminescence intensity of MBE–grown samples and compare them to the high–quality mechanically exfoliated $MoSe_2$ monolayer.

First, we use a time–resolved measurement of exciton lifetime in $MoSe_2$.[2] Although a typical neutral and charged exciton states decay in exfoliated $MoSe_2$ monolayers is slow enough for measurement to be performed on a streak camera, we find that the MBE–grown monolayers exhibit a much shorter lifetime, making the streak camera unsuitable for this application. Therefore, we applied the excitation correlation spectroscopy pump–probe technique, yielding superior time resolution. It resulted in identifying the two important non–radiative relaxation channels – one active always (even in cryogenic temperatures) and the other which activates with a temperature rise. We attribute the high efficiency of non–radiative channels in MBE–grown samples to lattice imperfections, which may stem from selenium vacancies.[3]

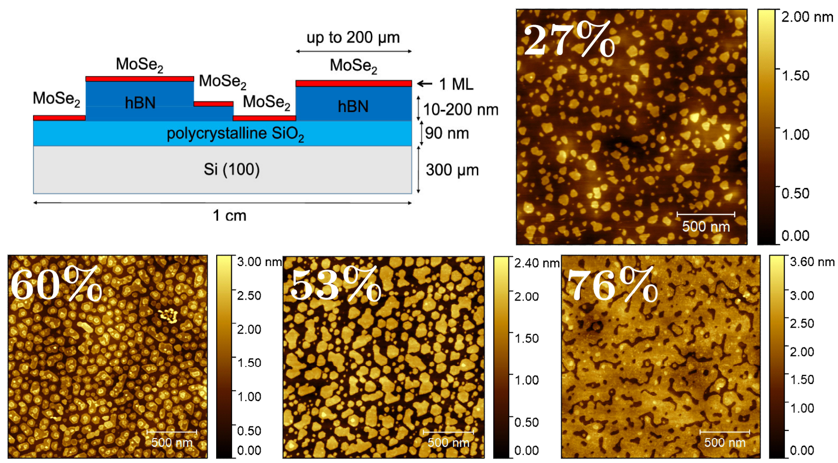

Fig. 1. Vertical sample cross–section [1] (top left) and AFM images of several MBE–grown $MoSe_2$ samples with different monolayer coverage factor.

Further, we take into account the variable monolayer coverage factors for MBE–grown samples (Fig 1). We use an atomic force microscope to measure the area occupied by the monolayer and account for this parameter in the samples benchmark. Another crucial effect that needs to be addressed when observing PL or Raman scattering intensity is interference. The laser excitation efficiency, and the Purcell effect, both depend on the thickness of the underlying boron nitride flake. To confirm these measurements we performed interference simulations that consider the h–BN flakes thicknesses estimated by reflectivity spectra, and AFM scans. We find that usually grown samples follow the same pathway of the quantum efficiency being 10 times lower than the one typical to the exfoliated samples.

Finally, we show that calibration and optimization of the MBE growth process gives a sample achieving PL intensity as high as a quarter of the exfoliated one, and still, it maintains its large–scale homogeneity.

References

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[3] S. M. Poh et al. *ACS Nano*, **12**, 8 (2018)