Exciton spectroscopy of transition metal dichalcogenides grown by MBE on hBN

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Monolayer transition metal dichalcogenides (TMDs) are two-dimensional materials with exceptional optical properties such as high oscillator strength, valley related excitonic physics, efficient photoluminescence, and several narrow excitonic resonances. However, above effects have been so far explored only for structures produced by techniques involving mechanical exfoliation and encapsulation in hBN inevitably inducing considerable large-scale inhomogeneity. On the other hand, techniques which are essentially free from this disadvantage, such as molecular beam epitaxy (MBE), have to date yielded only structures characterized by considerable spectral broadening, which hinders most of interesting optical effects.

We report for the first time on the MBE-grown TMD exhibiting narrow and fully resolved spectral lines of neutral and charged exciton (see Fig. 1, Ref. 1). Moreover, our MBE-grown TMD exhibits unprecedented high spatial homogeneity of optical properties, with variation of the exciton energy as small as 0.16 meV over a distance of tens of micrometers. Our recipe for MBE growth [1,2] is presented for $MoSe_2$ and includes extremely slow growth rate, the use of atomically flat hexagonal boron nitride (hBN) substrate and the annealing at very high temperature. Importantly, good optical properties are achieved for as-grown sample, without any post growth exfoliation and encapsulation in hBN. This novel recipe opens a possibility of MBE growth of TMD and their heterostructures with optical quality, dimensions and homogeneity required for optoelectronic applications.

We compare structural and optical properties of $MoSe_2$ grown on exfoliated hBN to properties of various TMDs grown on various substrates (2D, 3D, polycrystalline) [2,3]. This reveals particularly high diffusion parameters of transition metals on hBN, role of distribution of orientation of TMD grain domains, role of tendency to merge grains or form bilayers and 3D structures on substrates with various symmetry. Interestingly, good optical quality $MoSe_2$ can be obtained for various deformations of a monolayer, but only on hBN substrates (see Fig. 1).

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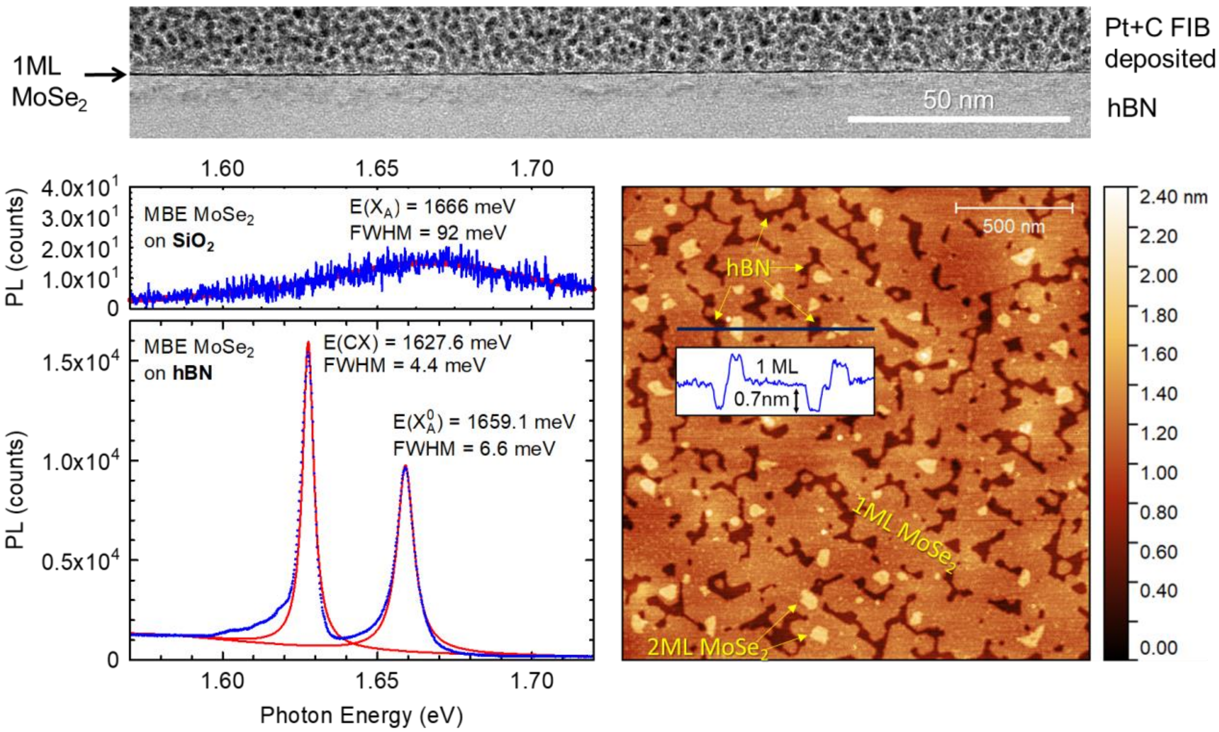

Fig. 1. Narrow excitonic lines presented for the first time for MBE-grown TMD. TEM cross-section of studied $MoSe_2$ monolayer grown on hBN is shown in (a). Low temperature (T = 10 K) PL spectra (blue curves) of MBE $MoSe_2$ grown on SiO_2 (b) and hBN (c) both produced in the same growth process. Only for $MoSe_2$ grown on hBN it is possible to resolve neutral exciton line X_A^0 and charged exciton line CX. Red curves show Lorentizan fits with following parameters: 1659.1 meV energy and 6.6 meV FWHM for the neutral exciton of $MoSe_2$ grown on hBN; 1627.6 meV energy and 4.4 meV FWHM for the charged exciton of $MoSe_2$ grown on hBN; and 1666 meV energy and 92 meV FWHM of $MoSe_2$ grown on SiO_2. Insert in (c) shows PL spectrum in wider spectral range. (d) AFM image of the sample shown in (a,c), with the almost complete monolayer, only in a very small part covered by the second layer. Size of the scanned area is 2 μm × 2 μm. The inset shows the height profile with 1 ML and 2 ML of $MoSe_2$ and uncovered hBN. Ref. [1].