Multi-particle Excitonic Systems in WSe₂ Grown on hBN by Molecular Beam Epitaxy

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Monolayer transition-metal dichalcogenides (TMDs) exhibit exceptional optical properties useful for optoelectronic applications. However, for industrial use, large-scale homogeneity optical response is needed. To address this demand we utilize molecular beam epitaxy (MBE). In our previous works, we have achieved super-high quality of MBE grown MoSe₂ on hexagonal boron nitride (hBN) [1] and then we have mixed it with Mn ions and showed that it can lead to the induction of 1T' phase of this material [2]. Here, we report on the WSe₂ grown on hBN by MBE, demonstrating high structural and optical quality on a large-scale. For the first time, multi-particle excitonic systems can be observed in WSe₂ samples fabricated in a bottom-up approach. Monolayers of WSe₂ were grown by MBE on exfoliated hBN flakes that are the best substrate known for TMDs growth. Because of its atomically smooth surface, lack of dangling bonds and uncompensated charges, hBN substrates allows to grow the highest quality monolayers both in terms of optical and structural properties. Before TMD growth, hBN flakes were exfoliated from bulk and deposited on a Si substrate with polycrystalline SiO_2 buffer. Growth has been realised in very easy, two - step process: first deposition at relatively low temperature (300 °C) and then annealing at a high temperature (800 °C) under a high Se flux. WSe₂ was investigated by atomic force microscopy and transmission electron microscopy in crosssection to verify their structural properties. Both techniques confirm that most of the TMD material is observed in the form of one monolayer thin flakes, that covers ~50% of the substrate surface. Both techniques confirm hexagonal structure of the material as well as high crystalline quality. We have used room and low temperature photoluminescence measurements to study optical properties of monolayers. WSe₂ features narrow and resolved spectral lines of neutral and charged exciton as well as wide range of localised excitons present in energies lower that charged exciton. Material exhibits high homogeneity of optical properties within micrometres squared. This is the first MBE grown WSe₂ exhibiting such a high resolution of spectral lines.

[1] Narrow excitonic lines and large-scale homogeneity of transition metal dichalcogenide monolayer grown by molecular beam epitaxy on hexagonal boron nitride, 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, P. Kossacki, Nano Letters 20, 3058 (2020).

[2] Molecular Beam Epitaxy Growth of Transition Metal Dichalcogenide (Mo,Mn)Se₂ on 2D, 3D and polycrystalline substrates, J. Kucharek, R. Bożek, W. Pacuski, Materials Science in Semiconductor Processing 163, 107550 (2023)



Fig. 1: a). PL spectrum taken in 10 K with 532 nm excitation laser. b). PL map presenting exciton intensity and c). AFM picture maped in b).