Tuning exciton emission in MOCVD grown MoS_2 monolayers: The role of charge transfer from the substrate

Henrik Myja[1]
Pawan Kumar[2], Benjamin Groven[2], Steven Brems[2], Henry Medina Silva[2], Pierre Morin[2], Tilmar Kümmell[1], Gerd Bacher[1]
[1]*Werkstoffe der Elektrotechnik, University Duisburg-Essen, Bismarckstr. 81, 47057 Duisburg, Germany*
[2]*IMEC, Kapeldreef 75, 3001 Leuven, Belgium*
henrik.myja@uni-due.de

Manufacturing (opto-)electronic devices based on 2D materials relies on the development of methods to deposit single-crystalline transition metal dichalcogenides (TMDs), such as metal-organic chemical vapor deposition (MOCVD). Currently, the electrical performance of TMD monolayers (MLs) deposited via MOCVD is limited by grain boundaries (GBs), vacancies or interface states that reduce charge carrier mobility, affect charge carrier concentration or favour nonradiative recombination [1]. Low temperature photoluminescence (PL) spectroscopy has successfully been used, e.g. to assign bound exciton emission to sulphur vacancies [2] or to reveal the impact of the growth substrate and its surface termination onto the population of in-gap defect states [3, 4]. Nevertheless, it remains elusive how microscopic defect structure originates from the growth and how it can be conceived from PL signatures. We employ temperature- and ambient-controlled PL to reveal defect mechanisms in MoS_2 on sapphire substrates and after transfer to device-grade Si/SiO_2 substrates. 1.2 ML of MoS_2 is deposited on 2-inch c-plane sapphire by MOCVD from $Mo(CO)_6$ and H_2S precursors using an industry-standard 200 mm CVD reactor [5]. We compare polycrystalline (pc) and single-crystalline (sc) ML of MoS_2 with stark difference in number density and type of GBs [5, 6]. Dark-field transmission electron microscopy does not detect high-angle and mirror GBs in the sc MoS_2 ML, as opposed to the pc MoS_2 ML with an average domain size of ~1 µm. Surprisingly, the room temperature PL signal of the sc MoS_2 is dominated by low intensity and charged exciton emission, while the pc MoS_2 lead to an intense neutral exciton emission. After transfer to SiO_2 this difference completely disappears demonstrating the dominant role of the substrate rather than the grain size on the emission process. Low temperature PL studies reveal defect-bound exciton emission that is substantially different between the sc and pc MoS_2 on sapphire. The sc MoS_2 exhibits a very weak defect PL with some contribution of near band edge emission, while in case of the pc MoS_2 the emission is controlled by an intense defect bond. We attribute these findings to a different sapphire termination [7] resulting from surface reconstruction during the growth, that affects the charge transfer between the substrate and the MoS_2 ML.

References

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Figures

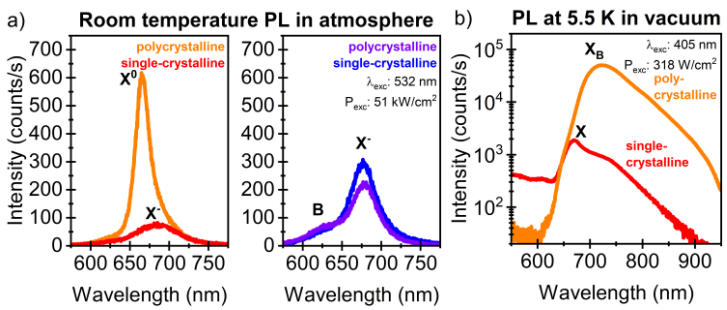

Figure 1: PL spectra of MoS_2 monolayers at room temperature (left) and at 5.5 K (right)