MBE Growth of Transition Metal Dichalcogenides

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3D materials such as silicon have been the workhorse of the semiconductor industry for decades. However, as transistor technology approaches nanoscale, the performance of these materials is seriously impacted by short-channel effects. In contrast, 2D van der Waals materials hold several distinct advantages, including relative immunity from short-channel effects and a lack of dangling bonds. Monolayer Transition Metal Dichalcogenides (TMDs) have been shown to exhibit modest and direct bandgaps, making them ideal semiconductors. Field effect transistors (FETs) fabricated utilizing exfoliated TMDs have already exhibited high On/Off ratio, small hysteresis and small subthreshold swing, and high mobilities. Exfoliated materials are typically of high quality but aren't scalable. While methods like Chemical Vapor Deposition can grow these materials to scale more economically, Molecular Beam Epitaxy (MBE) can deposit large-area films with atomically precise thickness, as well as precisely control the composition of deposited films, making it an ideal method for studying the transport properties of TMDs. While the growth of TMDs on c-sapphire is common in chemical vapor deposition (CVD), its use in MBE growth is uncommon due to the large lattice mismatch between TMDs and c-sapphire. Growth on c-sapphire by MBE requires temperatures 900°C and higher in ultra-high vacuum in order to make oriented films [1], without which, a randomly oriented polycrystalline film is obtained. [2,3] Our films are grown with precise thickness control, and are highly crystalline and uniform. The

aggressive heating that is required to obtain oriented films causes chalcogenide vacancies to accumulate in the film, which has been demonstrated in MoS₂ films with annealing temperatures as low as 600°C. The accumulation of these vacancies can lead to increased scattering of charge carriers and shorter exciton lifetimes. We will discuss the optical and transport properties of the films.

Figure 1: Second harmonic generation as a function of polarization angle (left) and reflection high-energy electron diffraction (right) pattern for 13 Layer MoSe₂ grown on c-sapphire.

[1] M. Nakano, Y. Wang, Y. Kashiwabara, H. Matsuoka, and Y. Iwasa, Layer-by-Layer Epitaxial Growth of Scalable WSe2 on Sapphire by Molecular Beam Epitaxy, Nano Lett. 17, 5595 (2017).

[2] M. T. Dau et al., Millimeter-scale layered MoSe2 grown on sapphire and evidence for negative magnetoresistance, Applied Physics Letters 110, 011909 (2017).

[3] A. Roy, H. C. P. Movva, B. Satpati, K. Kim, R. Dey, A. Rai, T. Pramanik, S. Guchhait, E. Tutuc, and S. K. Banerjee, Structural and Electrical Properties of MoTe2 and MoSe2 Grown by Molecular Beam Epitaxy, ACS Appl. Mater. Interfaces 8, 7396 (2016).

Supporting 1: RHEED Oscillations for 6 Layer polycrystalline MoSe2 grown on c-sapphire.

Supporting 2: AFM of 13 Layer MoSe₂, illustrating low surface roughness and step heights corresponding to ML thickness.

Supporting 3: RHEED (top), SHG (bottom left), and AFM (bottom right) for 13 layer WSe2 grown on c-sapphire.