**Molecular Beam Epitaxy of atomically thin 2D dichalcogenide Van der Waals semiconductor heterostructures**

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**Abstract**

Atomically thin 2D semiconductors offer solutions for nanoelectronic device scaling and a number of low power versatile applications. There are a number of other 2D semiconductors such as the group IV-dichalcogenides (e.g. $HfSe_2$, $ZrSe_2$) which remain unexplored. Due to Van der Waals heteroepitaxy [1] and the variety of energy gaps and electron affinities, VIB/IVB dichalcogenide heterostructures offer the possibility of sharp, defect–free heterointerfaces and type-II band alignments [2] with potential applications in vertical broken gap 2D-2D TFETs. In this work, we show that using molecular beam epitaxy (MBE) of selenides [3] it is possible to grow good crystalline quality $MoSe_2/HfSe_2$ ($ZrSe_2$) heterostructures on large area (up to 2 inch) w-AlN(0001)/Si(111) substrates. The availability of 200 and 300 mm AlN/Si substrates creates the prospect for Si compatible processing in compliance with the semiconductor industry standards. All materials require a low temperature deposition step, followed by in-situ UHV annealing at high temperature. Epitaxy can start either with $MoSe_2$ in direct contact with AlN substrate followed by $HfSe_2$ or with the reverse order without jeopardizing the quality of heterostructure bilayer. In situ RHEED (Fig. 1) and HRTEM (not shown here) reveal that the 2D heterostructures, despite the relatively large lattice mismatch, are epitaxially grown on AlN such that the [11-20] in-plane crystallographic direction of all materials are aligned. Films of a few layers of $MoSe_2$ and $HfSe_2$ are continuous with smooth surfaces over substrate area of at least 2 inches as seen by *in-situ* UHV-STM and SEM characterization (Fig. 2). Using *in-situ* XPS, the films are found to be very close to the ideal stoichiometry. Using Kraut's method, the valence band offsets are calculated to be (VBO~0.45 eV and CBO~0.9 eV) which deviate from the ideal band alignment as estimated by DFT [2]. The electronic valence band structure of 4 ML $ZrSe_2$ (Fig. 3) and $HfSe_2$ was imaged by *in-situ* HeI-ARPES and found to be in good agreement with our DFT calculations using GGA / v.der Waals potentials and spin orbit coupling in the scalar approximation.

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**References**

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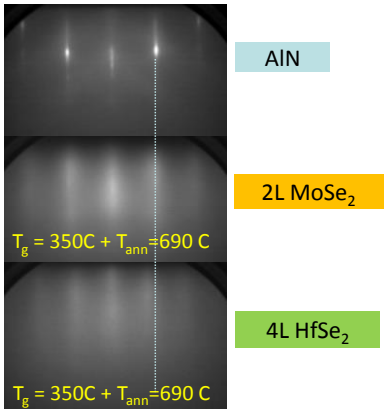


Fig. 1: RHEED of the $HfSe_2/MoSe_2$/AlN structure along the [11-20] azimuth

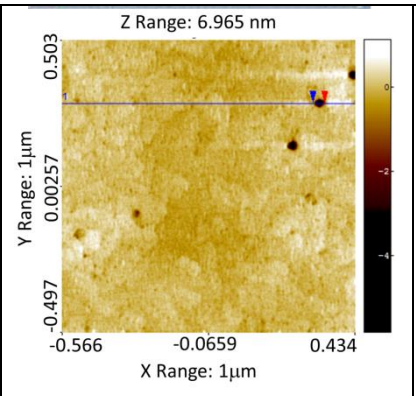


Fig. 2: UHV-STM of the $HfSe_2/MoSe_2$/AlN structure over an area 1x1 $\mu m^2$, with an RMS surface roughness of ~0.3 nm

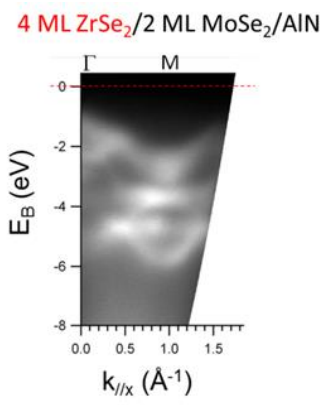


Fig. 3: ARPES of the valence band structure of $ZrSe_2/MoSe_2$/AlN