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Transition metal dichalcogenide and hexagonal boron nitride heterostructures grown by molecular beam epitaxy

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ABSTRACT

Heterostructures coupling transition metal dichalcogenides (TMDs) and insulating hexagonal boron nitride (h-BN) were grown by molecular beam epitaxy (MBE) demonstrating the unique opportunities for fabricating all 2D heterostructures with the desired band alignments for novel nanoelectronic devices. Structural and chemical characterization of the TMDs and h-BN was conducted via reflection high energy electron diffraction (RHEED), X-ray diffraction (XRD), transmission electron microscopy (TEM), scanning tunneling microscopy/spectroscopy (STM), and X-ray photoelectron spectroscopy (XPS).

1. Introduction

Two-dimensional (2D) TMDs are promising materials for future devices to extend the field effect transistor (FET), such as tunnel FETs. TMDs are layered materials similar to graphite that can be metallic or semiconducting depending on the composition [1]. Hexagonal boron nitride (h-BN), a dielectric, has recently emerged as another interesting 2D material for implementation in novel nanoelectronic devices [2]. Because of its wide band-gap, h-BN can be used as a dielectric interlayer for heterostructures coupling layers of graphene or TMDs enabling new all-2D heterostructures (Fig. 1). Most of the 2D materials device research to date has been performed using exfoliated, geological TMD materials, such as MoS_2. However, these geological TMDs are riddled with native defects [3] (Fig. 2), extrinsic impurities, and damage induced by the violent exfoliation process. Additionally, the exfoliation process results in flake sizes <1000 μm^2 limiting the research potential of that technique [4]. These issues highlight the critical need for bottom up synthesis of 2D materials to enable large-area, low-defect TMDs and heterostructures. The hexagonal crystal lattices of TMDs and h-BN coupled with van der Waals interactions make them ideal for heterostructure growth without the introduction of extended defects resulting from lattice mismatch as observed in cubic semiconductor materials. Additionally, interdiffusion normally observed in superlattices of II–VI or III–V compounds is reduced in these layered materials. This opens the door for the fabrication of novel nanoelectronics such as vertical broken-gap tunnel FETs and the BiSFET [5].

2. Experiments

TMD and h-BN films were grown on ~1 × 1 cm^2 mechanically exfoliated HOPG, h-BN, and MoS_2 substrates purchased from SPI supplies and HQ Graphene. The growth was accomplished in a unique multi-chamber MBE at UT-Dallas. Each of the VG-Semicon growth chambers are linked together with a UHV transfer tube system (Fig. 3) operating at a base pressure of about 10^{-11} mbar. TMD growth is performed in a V80H growth chamber equipped with two vertical e-beam evaporators enabling the growth of high melting temperature metals such as Hf, Ti, Mo, and W in addition to effusion cell evaporation of the chalcogens. The two e-beam evaporators allow for TMD heterostructure growth as well as mixed transition metal TMDs, providing significant flexibility in material and device design. Each growth chamber is equipped with *in situ* RHEED allowing us to assess the growth

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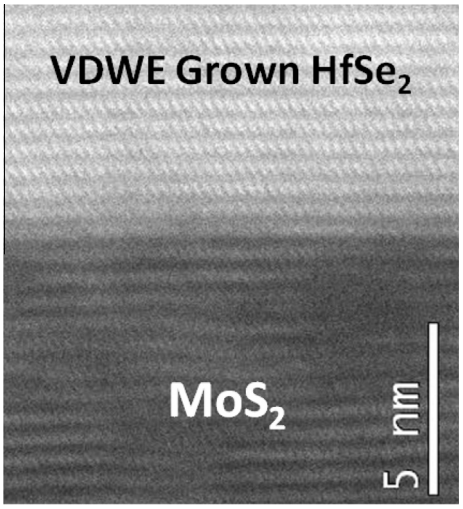

Fig. 1. HAADF-STEM image of $HfSe_2$ grown on MoS_2 using VDWE in the Hinkle laboratory.

quality in real-time. The chamber is also equipped with $ZnCl_2$ and nitrogen plasma sources for doping and surface functionalization.

The III–V chamber is also a V80H with recently upgraded hardware and software to provide state-of-the-art growth capabilities including In, Ga, As, B, Al, and N compounds as well as *in situ* H-cleaning and Be and Si doping. The *in situ* growth of hexagonal boron nitride (h-BN) on TMDs is highly promising and provides another 2D material (in this case a wide Eg insulator) to further heterostructure device design options. The B is evaporated using a high temperature effusion cell while ultra-high purity (99.9999%) N_2 gas is used with the Veeco UNI-Bulb RF plasma source that is water cooled and provides outstanding plasma stability. The Group IV chamber, a V80S, is used to grow Si, Ge, and Sn epitaxial films, SiSnGe compounds, and strained heterostructures. The vertical growth chamber in this system incorporates electron-beam evaporators and effusion cells for Sb and B doping. Preparation chambers with high temperature heating stages are available for each material system.

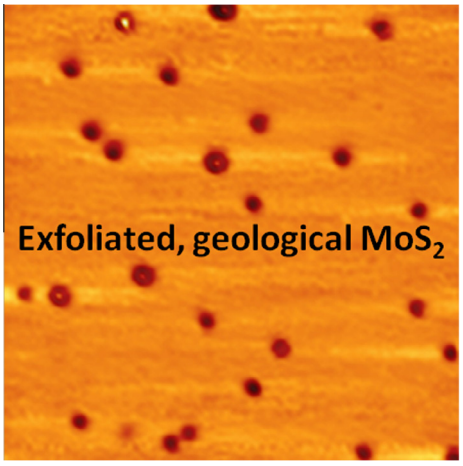

Fig. 2. STM image of exfoliated, geological MoS_2 demonstrating the high defect density that is common in these samples. These defects and the resultant device variability motivates the need for bottom up growth of TMDs. Image conditions were 100 × 100 nm, V_{bias} = 0.4 V, I_t = 1.2 nA.

3. Results and discussion

Fig. 1 shows a high angle annular dark field (HAADF)-scanning transmission electron microscopy (STEM) image of MBE grown $HfSe_2$ on MoS_2, an example of the 2D heterostructures that we have achieved through van der Waals epitaxy (VDWE) [6]. We have demonstrated layered films that have atomically abrupt interfaces without misfit dislocations associated with a lattice mismatch as large as 41% [7]. RHEED data (Fig. 4) shows the starting HOPG (left) and ~1 ML of $HfSe_2$ growth on HOPG (right). The spacing of the reciprocal lattice streaks indicates the growth of unstrained $HfSe_2$ revealing that the TMD immediately has its own lattice constant. This is consistent with TMDs grown by VDWE on other inert hexagonal materials and opens the door for minimized defect heterostructures without concern for substrate lattice matching. The RHEED image also indicates rotational alignment with the hexagonal substrate and flat 2D growth. This rotational alignment

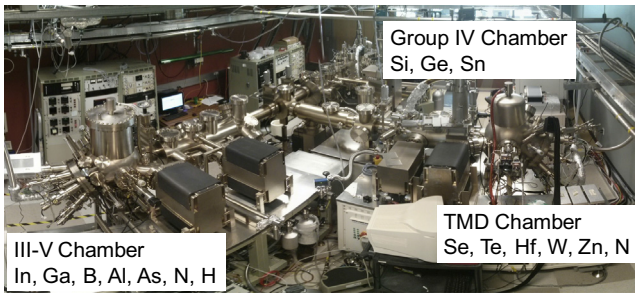

Fig. 3. Interconnected multi-chamber MBE system located in the Hinkle laboratory at UT-Dallas.

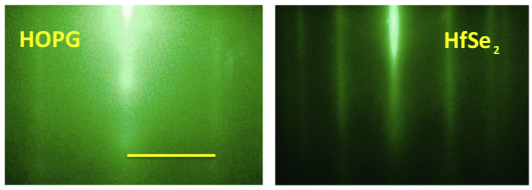

Fig. 4. Left: RHEED of starting HOPG substrate. Right: ~1 ML of $HfSe_2$ grown on HOPG. The reciprocal lattice spacing indicates the growth of unstrained $HfSe_2$.

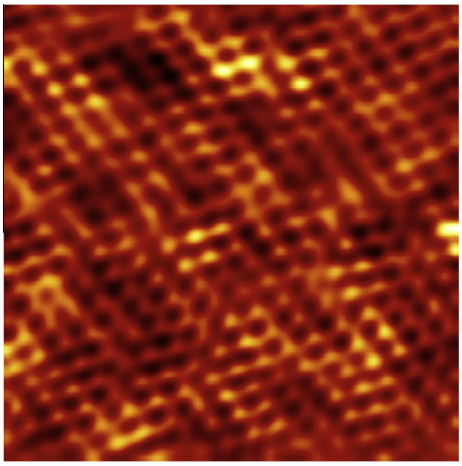

Fig. 5. STM image showing atomic resolution of the MBE grown $HfSe_2$. Image conditions were 5 × 5 nm, V_{bias} = 0.2 V, I_t = 2 nA.