Highly-Oriented Atomically Thin Ambipolar MoSe₂ Grown by Molecular Beam Epitaxy

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Transition metal dichalcogenides (TMDCs), together with other two-dimensional (2D) materials have attracted great interest due to the unique optical and electrical properties of atomically thin layers. In order to fulfill their potential, developing large-area growth and understanding the properties of TMDCs have become crucial. Here, we used molecular beam epitaxy (MBE) to grow atomically thin MoSe₂ on GaAs(111)B. No intermediate compounds were detected at the interface of as-grown films. Careful optimization of the growth temperature can result in the growth of highly aligned films with only two possible crystalline orientations due to broken inversion symmetry. As-grown films can be transferred onto insulating substrates allowing their optical and electrical properties to be probed. By using polymer electrolyte gating, we have achieved ambipolar transport in MBE-grown MoSe₂. The temperature-dependent transport characteristics can be explained by the 2D variable-range hopping (2D-VRH) model, indicating that the transport is strongly limited by the disorder in the film.

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Atomically thin two-dimensional (2D) materials have shown great potential because of their interesting electrical and optical properties.¹-⁴ Potential applications in flexible electronics and the possibility to further extend their range of applications by integrating them into heterostructures⁵ motivate scientists to develop a reliable way to grow large-area 2D materials.⁶-⁹ While chemical-vapor-deposition (CVD) can on one hand result in high-quality 2D materials and 2D heterostructures with sharp and clean interfaces¹⁰, the toxicity of some of the precursors introduces challenges, and the lack of in-situ characterization and monitoring during growth can lead to non-reproducible results. On the other hand, using molecular-beam-epitaxy (MBE) to grow atomically thin transition metal dichalcogenides (TMDCs) and other 2D materials via van der Waals epitaxy¹¹-¹⁶ has several potential advantages. The deposition can be well controlled using high-purity elemental sources, direct heterostructure growth can be achieved, and the growth in an ultra-high vacuum environment limits the amount of impurity atoms. In addition, the quality of the MBE-grown film can be monitored during growth in-situ using reflection-high-energy-electron-diffraction (RHEED). The growth of different TMDCs on various substrates has been recently demonstrated,¹⁷-¹⁹ and the band structures and thin film morphology have been intensively investigated using angle-resolved photoemission spectroscopy and scanning tunneling microscopy.²⁰-²² However, reports on the electrical properties of atomically thin layers grown by MBE²³ are rare and indicate that the material quality needs to be improved further. Starting from this point, it is crucial to optimize epitaxial growth and to investigate the optical and electrical properties of MBE-grown atomically thin TMDCs.

Here, we report on the use of MBE to grow atomically thin MoSe₂ on GaAs(111)B down to nominal monolayer (ML) thickness, exhibiting high optical quality confirmed by photoluminescence (PL) and Raman spectroscopy. We find that using GaAs(111)B as the growth substrate results in a high degree of control over lattice orientation of the domains forming the polycrystalline film. The features of van der Waals epitaxy were thoroughly investigated by X-ray photoelectron spectroscopy (XPS). Using scanning transmission electron microscopy (STEM), we have observed that depending on the growth temperature, polycrystalline MoSe₂ films can be composed of grains with either a 15° or a 60° misorientation angle. Both configurations are predicted to be energetically stable by density functional theory (DFT) calculations. We further address the electrical properties of highly-oriented nominal ML MoSe₂, and demonstrate ambipolar transport behavior using polymer electrolyte gating.
Temperature-dependent measurements indicate that the transport is limited by the disorder and can be explained using the 2D variable range hopping (2D-VRH) model.

Atomically thin MoSe$_2$ films were grown on GaAs(111)B substrates by MBE and the growth was monitored in-situ using a RHEED camera. Atomic hydrogen was used to effectively remove native oxide at low temperature without As desorption, and a clean GaAs(111)B surface with sharp RHEED streaks along the GaAs[11-2] azimuth was observed (see Figure 1a). As the growth progresses, GaAs-

![Figure 1. Growth of atomically thin MoSe$_2$ by MBE. (a) RHEED patterns of GaAs(111) along the GaAs[11-2] azimuth at growth start. (b) Half-half transition along the GaAs[11-2] azimuth. (c) Nominal monolayer (ML) MoSe$_2$ observed along MoSe$_2$[10-10] and MoSe$_2$[11-20] azimuths at growth end after 22 min. (e) Mo 3d and (f) Se 3d core-level spectra in XPS. (g) Comparison of Raman spectra from MBE-grown and exfoliated ML MoSe$_2$. (h) Photoluminescence of transferred ML MoSe$_2$ and exfoliated ML MoSe$_2$. Inset shows optical image of the transferred film. The sample was grown at 470 °C.](image)
related streaks gradually fade out and are replaced by a new set of faint streaks with a smaller spacing, indicating the formation of MoSe$_2$. A RHEED pattern from the MoSe$_2$ film with a half-layer coverage is shown on Figure 1b, with GaAs and MoSe$_2$ streaks having similar intensities. GaAs-related streaks completely vanish as a full film is grown, while MoSe$_2$ streaks become more intense Figure 1c. The sharp streaks in Figure 1c and Figure 1d indicate the epitaxy with MoSe$_2[10-10]$/GaAs[11-2] and MoSe$_2[11-20]$/GaAs[10-1], respectively. The van der Waals epitaxy thus provides a reliable way for developing large-scale 2D materials on different substrates.19,26

Films grown on GaAs(111)B were examined ex-situ using x-ray photoelectron spectroscopy (XPS). Figure 1e shows the core level spectrum of the Mo 3d range. The binding energies of Mo 3d$_{5/2}$ and Mo 3d$_{3/2}$ peaks are 229.2 eV and 232.4 eV, respectively. An additional AsLMM peak with a lower binding energy of 227.2 eV is also observed. The Se 3d core level spectrum in Figure 1f shows a Se 3d$_{5/2}$ peak at 54.9 eV and an Se 3d$_{3/2}$ peak at 55.8 eV, demonstrating the existence of Mo-Se bonds. A consistent binding energy shift is observed in the core level spectra of Ga 3d and As 3d between pristine GaAs substrate and as-grown MoSe$_2$, implying that charge transfer takes place at the interface (see Supplementary Section 1 for more details). On the other hand, the peak positions and the high quality of fitting shows that no intermediate compounds exist at the interface, as expected from van der Waals epitaxy.

The Raman spectrum of nominal ML MoSe$_2$/GaAs(111) shown on Figure 1g clearly shows the MoSe$_2$ A$_{1g}$ mode at 238.7 cm$^{-1}$ which is comparable with that of exfoliated ML MoSe$_2$ on 270 nm SiO$_2$. Photoluminescence spectroscopy was also used to investigate the optical properties of ML MoSe$_2$. In order to avoid the optical quenching effect from the substrate,18,27 the as-grown film was transferred onto a 270 nm SiO$_2$/Si chip with the optical image shown in the inset of Figure 1h and Figure S2. The 1.58 eV peak recorded at room temperature is clearly shown in Figure 1h and is comparable with the exfoliated ML flake in terms of the peak position. Despite inevitable existence of wrinkles and folded regions due to the imperfect transfer process which might introduce defects and peak broadening, the full-width-half-maximum of 37 meV is comparable to that of exfoliated ML flakes ($FWHM = 27$ meV).

The films were transferred onto the TEM grids and investigated using Cs-corrected STEM in order to examine their morphology. Figure 2a shows a low-magnification HAADF-STEM image of MoSe$_2$ grown at 470°C. Inset is the corresponding FFT image showing two sets of spots. (b) High-magnification image of an incomplete bilayer (BL) comprised of ML domains with two orientations denoted as ML(A) and ML(B). (c) Low-magnification HAADF-STEM image of MoSe$_2$ grown at 530 °C. Inset shows the corresponding FFT image with a single set of diffraction patterns. (d) High-magnification of ML region with schematic of atom positions. Bright spots correspond to double Se atoms (e) Intensity histogram of the image shown in (c). (f) Intensity profile along the slice shown in (c). (g) Intensity profile of the slice from (d).

Figure 2. Morphology of MBE-grown MoSe$_2$ films. (a) Low-magnification HAADF-STEM image of MoSe$_2$ grown at 470°C. Inset is the corresponding FFT image showing two sets of spots. (b) High-magnification image of an incomplete bilayer (BL) comprised of ML domains with two orientations denoted as ML(A) and ML(B). (c) Low-magnification HAADF-STEM image of MoSe$_2$ grown at 530 °C. Inset shows the corresponding FFT image with a single set of diffraction patterns. (d) High-magnification of ML region with schematic of atom positions. Bright spots correspond to double Se atoms (e) Intensity histogram of the image shown in (c). (f) Intensity profile along the slice shown in (c). (g) Intensity profile of the slice from (d).
MoSe$_2$/GaAs (111)B superlattices are very similar, indicating types of MoSe$_2$/GaAs (111)B superlattices by considering MBE-grown MoSe$_2$ on highly oriented pyrolytic graphite, before the first MLs complete. Similar morphologies of voids in the film indicates that BLs start to form orientation at this point, since these would result in a set of layers with an interlayer twist of 15° and could present an interesting material system for studying the effect of interlayer twist on electrical and optical properties of 2D semiconductors. Overall, formation energies of oriented and misoriented MoSe$_2$/GaAs (111)B superlattices are related to Rutherford scattering which increases with the atomic number ($Z$). Different intensities in the image can be attributed to different layer thickness. Figure 2b shows the intensity histogram with dashed lines that correspond to intensities in ML and BL regions. The intensity profile along the slice of interest is plotted in Figure 2c, showing the thickness varying from ML to BL. Figure 2d shows a high-magnification image of a ML region with the intensity profile along a slice of interest shown in Figure 2f, where the positions of Mo and Se atoms can be assigned to periodically varying intensities. The ML has a 2H structure with a lattice constant estimated to be 3.29 ± 0.03 nm, which is in line with the bulk lattice constants reported in literature. To further confirm the long-range uniformity of the MoSe$_2$ film, we have measured second harmonic generation (SHG) from as-grown MoSe$_2$/GaAs (111)B and suspended MoSe$_2$ on TEM grids, Figure S7. Polar plots of SHG intensity show six-fold symmetry, while the PL intensity and Raman $A_{1g}$ peak position and intensity maps indicate a high degree of uniformity (see Supplementary Section 4).

We now focus on the electrical properties of 0, 60° MoSe$_2$ with nominal ML thickness. We use electrical double-layer transistors (EDLT) in order to access a wide range of electrostatically induced doping levels and to reduce Shortky barrier heights at contacts, allowing efficient electron and hole injection using the same contact material. Figure 3 shows the schematic of the device. The polymer electrolyte PS-PMMA-PS:[EMIM]-[TFSI] is spin-coated on top and allows us to reach high carrier densities, increases the efficiency of carrier injection, making it ideal for achieving ambipolar regime of operation. We also include a back gate, allowing charge carrier modulation in the semiconducting film at temperatures below the freezing point of the polymer electrolyte ($T \sim 200$ K). Devices without a polymer electrolyte show a very poor transistor behavior (see Supplementary Section 5). The dependence of the EDLT on the polymer electrolyte voltage $V_{PE}$ exhibits a clear ambipolar behavior close to room temperature ($T \sim 280$ K) (Figure 3b). We find a current $I_{on}/I_{off}$ ratio of $10^4$ and $10^2$ for the $n$ and $p$ sides, respectively. The maximum current density on the $n$-side, with the value of ~1 μA/μm is two orders of magnitude larger than on the $p$-side, possibly due to intervalley scattering of electrons, while the off current remains at pA levels. The subthreshold swing calculated from the linear

![Figure 3. Ambipolar transport in MoSe$_2$ EDLT. (a) Schematic of the MoSe$_2$ EDLT in a dual-gate geometry. The back-gate is used to modulate the charge density in the 2D channel around the value set by the reference electrode with the applied voltage $V_{PE}$. (b) Channel current as a function of $V_{PE}$ showing ambipolar behavior.](image.png)
region is for both sides ~390 mV/dec. Field-effect mobilities can be extracted from four-contact devices by freezing the polymer electrolyte at 200 K and performing a back-gate sweep (see Supplementary Section 6) which allows the charge carrier concentration in the 2D semiconductor to be modulated around the value set by $V_{PE}$ prior to freezing the electrolyte. The extracted electron mobility $\mu_e$ is $\sim 0.28 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ and hole mobility $\mu_h$ is $\sim 0.28 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$. The mobility values are significantly lower than those of CVD-grown MoSe$_2$. These results indicate that charge carrier transport is strongly influenced by the disorder in the film.

In order to elucidate the dominant transport mechanism in MBE-grown atomically thin MoSe$_2$, we have performed electrical measurements as a function of temperature, polymer electrolyte and back-gate voltage. Figure 4a shows the sheet conductivity $G_{sh}$ as a function of $V_{PE}$ based on the four-contact device shown in the inset. The $V_{PE}$ applied for $p$-side needs to be pushed to $V_{PE} > -4 \text{ V}$ to reach the same value of $G_{sh}$ of $n$-side, indicating strong electron doping or Fermi level pinning to the conduction band. A drop in $G_{sh}$ takes place at $V_{PE} > 3 \text{ V}$ and can be attributed to the electrolyte-induced disorder which is commonly observed in experiments involving EDLTs. The sweep at 280 K can then provide a reference curve for different doping levels that can be achieved by changing $V_{PE}$. Once the doping level at a given $V_{PE}$ is stabilized at 280 K, we freeze the electrolyte down to 200K with a cool-down rate of 1 °C/min. The $V_{PE}$ is then disconnected at 200 K after the electrolyte was completely frozen so that $G_{sh}$ can be stabilized (see Supplementary Section 7). The $G_{sh}$ during each cooldown was recorded down to 12 K with a cooling rate of 0.5 °C/min. The process was reversible after a mild annealing by ramping the temperature to 333 K.

The $G_{sh}$ of MoSe$_2$ at a given $V_{PE}$ monotonically decreases with decreasing temperature, showing semiconducting behavior for both sides. The dependence above 80 K follows the 2D-VRH model with the relation $G_{sh} \propto \exp[-(T_0/T)^{3}]$ where $T_0$ is the characteristic temperature. A linear fit of ln $G_{sh}$ to $T^{-1/3}$ is plotted at each given $V_{PE}$, demonstrating the validity of the 2D-VRH mechanism for both sides, Figure 4b and c. The VRH mechanism is usually observed in disordered systems and the results imply that the transport of MoSe$_2$ EDLT is strongly influenced by the voids and the nanometer-scale grains in the film.

Charge carriers are strongly scattered and tend to hop between different conductive paths. The localization length can be changed in a small range by changing $V_{PE}$, i.e., changing the doping level. The dependency is evident by extracting $T_0$ from the fits to the 2D-VRH model with the value of slopes $s$ extracted from Figure 4b and Figure 4c, where $T_0 = s^3$. The values of $T_0$ decrease by more than one order of magnitude with the increase of $|V_{PE}|$ because of the increase of carrier densities in the material, thus screening the disorder along the conductive paths. The localization length $\xi_{loc}$ can be extracted using the expression $\xi_{loc} = \sqrt{\frac{138}{8} k_B D T_0}$, where $k_B$ is the Boltzmann constant and $D$ the density of states (see Supplementary Section 8). The results are plotted in Figure 4d and e for $p$ and $n$ sides, respectively. Holes have a slightly lower $\xi_{loc}$ with $\xi_{loc}$ reaching a maximum value of ~4 nm at $V_{PE} = -4.2 \text{ V}$. The $n$-side on the other hand shows tunable $\xi_{loc}$ up to ~9 nm with $V_{PE} = 2.6 \text{ V}$. All values of $\xi_{loc}$ have the same order of magnitude as the grain size shown in STEM images, indicating that the 2D-VRH transport is linked to the disorder in the as-grown MoSe$_2$. Future MBE-based growth efforts...
will have to concentrate on increasing the grain size in order to improve the film quality.

In conclusion, we have grown atomically thin MoSe$_2$ using MBE. Films show a high degree of alignment due to the van der Waals interaction with the GaAs substrate and can be transferred to insulating substrates for further optical and electrical transport studies. We realize electrolytically gated transistors based on transferred ML MoSe$_2$ films. Electrical transport follows 2D-VRH model due to the disorder in film, with localization length comparable to the grain size.

**METHODS**

**MBE Setup and Material Growth**

The growth was carried out in an Omicron MBE (Lab 10) with a \( \sim 10^{-10} \) mBar base pressure. Cleaved \( 1 \times 1 \) cm$^2$ GaAs(111)B substrates were outgassed at up to 500 °C for at least 30 min. The native oxide was removed from the surface of GaAs(111)B by heating it to 350 °C under a flux of atomic hydrogen. Hydrogen molecules were dissociated by a tungsten filament with Joule heating at 70 W and were introduced into the chamber via a leak valve. The procedure lasted 30 min or more at base pressure \( \sim 3 \times 10^{-7} \) mBar resulting in sharp streaks in RHEED. A Kondsen cell and an electron beam source (EFM-3 from Omicron) were used for Se and Mo evaporation, respectively. The flux rates were calibrated using a quartz crystal microbalance, and the flux ratio of Se/Mo was optimized to be \( \sim 40 \) for growth. A RHEED camera (Staib Co.) was used to monitor the growth *in-situ*. The growth temperature was optimized in the 470 – 530 °C temperature range. Post annealing at up to 550 °C was performed in Se atmosphere. Higher temperature leads to GaAs decomposition and increasing surface roughness.

**XPS and Raman Spectroscopy**

The XPS spectra were obtained *ex-situ* in a commercial KRATOS AXIS ULTRA system, and C1s core level peak at 284.8 eV was used for the reference. Peak identification and fitting was performed in PHI MultiPak processing software. Raman analysis was performed using a Horiba LabRAM HR800 system using a 532-nm-wavelength green laser with spot size \( \sim 4 \) μm. The laser power was kept below 4 mW during all measurements. We used a 1800 lines/mm grating and have calibrated the system using the polycrystalline Si peak at 520 cm$^{-1}$. The PL was measured in a home-built setup using a 488 nm laser (Coherent) for excitation.

**STEM Microscopy and Analysis**

The STEM experiments were performed on a FEI Titan Themis 300 double Cs corrected microscope at an acceleration voltage of 80 kV in order to minimize beam damage. The scanning probe had a 28 mrad semi-convergence angle allowing resulting in a resolution close to 1 Å. The data was acquired under annular dark field conditions using an annular detector with a collection half-angle between 40 and 200 mrad.

**Material Transfer and Device Fabrication**

As-grown films were coated with PMMA and immersed into 30% KOH$_{aq}$ at 90 °C. The detached PMMA layer with the as-grown film was then transferred to a beaker with deionized water several times to remove excess KOH$_{aq}$ and was transferred onto a degenerately doped p$^+$ Si chip covered by 270 nm SiO$_2$ or TEM grids. PMMA was removed in acetone. Palladium was used for electrical contacts in a standard MBE-based e-beam lithography process. A second e-beam lithography was performed, followed by O$_2$/SF$_6$ plasma etching in order to define the device geometry. To fabricate an EDLT, the electrolyte PS-PMMa-PS:[EMIM]-[TFSI] was spin-coated onto the device and soft-baked at 60 °C for 10 min. A more detailed description is available elsewhere.$^{33,44}$ Electrical measurements were carried out using an Agilent 5270B SMU and Keithley 2000 DMM. Cyclic measurements were performed in a Janis closed-cycle cryogen-free cryostat.

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**Supporting Information Available**

Supplementary figures and discussion related to XPS spectroscopy, TEM imaging, DFT calculations, SHG and Raman mapping, field-effect transistor characterization and variable range hopping transport mechanism. This material is available free of charge *via* the Internet at http://pubs.acs.org.

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