Field-effect transistors and intrinsic mobility in ultra-thin MoSe$_2$ layers

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We report the fabrication of back-gated field-effect transistors (FETs) using ultra-thin, mechanically exfoliated MoSe$_2$ flakes. The MoSe$_2$ FETs are $n$-type and possess a high gate modulation, with On/Off ratios larger than 10$^6$. The devices show asymmetric characteristics upon swapping the source and drain, a finding explained by the presence of Schottky barriers at the metal contact/MoSe$_2$ interface. Using four-point, back-gated devices we measure the intrinsic conductivity and mobility of MoSe$_2$ as a function of gate bias, and temperature. Samples with a room temperature mobility of $\sim$50 cm$^2$/V·s show a strong temperature dependence, suggesting phonons are a dominant scattering mechanism.

Transition metal dichalcogenides (TMDs) are materials characterized by a $\text{MX}_2$ formula where $\text{M}$ stands for a transition metal (Mo, W), and $\text{X}$ stands for a chalcogen (S, Se or Te)\textsuperscript{1,2}. The transition metal dichalcogenides are layered materials, consisting of layers with an X-M-X structure. Within each layer the M-X bonds are covalent, while separate layers are bonded via Van der Waals interaction. Recent studies\textsuperscript{3-6} proved that micromechanical exfoliation, employed to isolate graphene monolayers\textsuperscript{7} is also an effective method to obtain thin flakes of various TMDs. The layers can be indentified\textsuperscript{3-6} using optical microscope thanks to the thickness dependent contrast on SiO$_2$/Si substrates. Various TMD-based electronic devices have been demonstrated, with MoS$_2$ being investigated most extensively to date. Examples include MoS$_2$ monolayer $n$-type field effect transistors (FETs)\textsuperscript{8,9}, MoS$_2$ phototransistor\textsuperscript{10}, MoS$_2$ bilayers chemical sensors\textsuperscript{11}, logic gates, memory cells and amplifiers using large MoS$_2$ flakes\textsuperscript{12-14}. In addition, top-gated $p$-type FETs using monolayer WS$_2$\textsuperscript{15}, and ambipolar back-gated FETs using multilayered WS$_2$\textsuperscript{16} have been demonstrated. The TMD-based FETs generally possess a high On/Off ratio, larger than 10$^6$. Such semiconductors can be scaled down to a 0.7 nm thickness monolayer, which renders them attractive as an ultra-thin body for aggressively scaled FETs. The larger band-gap by comparison to graphene can ensure a large On/Off ratio.

In this study we examine the electron transport in another TMD, namely MoSe$_2$, a semiconductor with an indirect band-gap of 1.1 eV\textsuperscript{15,16} in bulk, which increase to 1.55 eV and become direct in monolayer and bilayer\textsuperscript{17,18}. We discuss the realization of field-effect transistors using ultrathin MoSe$_2$ flakes mechanically exfoliated on SiO$_2$/Si substrates. The MoSe$_2$ FETs are $n$-type, and possess a good gate control, with On/Off ratios larger than 10$^6$. Four-point, gated devices allow us to measure the intrinsic conductivity, and extract the mobility of MoSe$_2$. The temperature ($T$) dependence reveals that the mobility increases significantly with decreasing the temperature, suggesting that phonon scattering dominates at room temperature.

The MoSe$_2$ flakes used here are produced by micromechanical exfoliation using commercially available MoSe$_2$ powder (Materion Inc.) with grain size < 44 $\mu$m (mesh -325). The MoSe$_2$ flakes are exfoliated on a 285 nm-thick SiO$_2$ film, thermally grown on highly doped $n$-type Si (100) wafers ($N_D > 10^{20}$ cm$^{-3}$). The exfoliated flakes are identified using optical microscope, and their thickness measured by atomic force microscopy (AFM). Figure 1(a) shows an optical micrograph of a MoSe$_2$ flake on SiO$_2$/Si substrate, while Fig. 1(b) shows the flake topography, probed by AFM, illustrating layered stacking. The typical flake size is 1-3 $\mu$m, and their thickness ranges between 3 to 80 nm. Individual MoSe$_2$ flakes typically exhibit terraces with various thicknesses, with a terrace surface roughness ranging from 0.2 to 0.6 nm. The optical contrast of the MoSe$_2$ on 285 nm thick SiO$_2$ substrate allows identification of flakes as thin as 4.8 nm (7 layers)\textsuperscript{19,20}.

To assess the MoSe$_2$ quality we performed X-ray diffraction (XRD) on powder samples. The powder XRD pattern, shown in Fig. 1(c) matches with the 2H-Drysadallite patterns\textsuperscript{21} ensuring our material is characterized by hexagonal 2H-MoSe$_2$ structure with space group $D_{6h}^4$ (P6$_3$/mmc)\textsuperscript{22}, and in agreement with previous MoSe$_2$ powder XRDs\textsuperscript{22,23} studies. The full width at half maximum (FWHM) of the <002> peak is 0.223$^\circ$. To further assess the exfoliated MoSe$_2$ flakes we performed $\mu$-Raman spectroscopy using a Renishaw InVia Raman microscope. Figure 1(d-e) show Raman spectra using 442 nm [Fig. 1(d)] and 532 nm [Fig. 1(e)] excitation wavelengths on a MoSe$_2$ flake. The spectrum of Fig. 1(d), acquired with an incident laser power of 30 mW and 650 nm spot size presents four peaks at 169, 242, 285 and 352 cm$^{-1}$, corresponding to the $E_{1g}$, $A_{1g}$, $E_{2g}^1$ and $A_{2u}^1$ modes respectively, as reported by Sekine et al.\textsuperscript{24} and more recently by Tongay et al.\textsuperscript{18}. We note that the $A_{2u}^1$ mode (352 cm$^{-1}$) is an infrared active mode, not present in Raman scattering in bulk samples. The emergence of this mode in Raman scattering acquired on small flakes suggests a breakdown of inversion symmetry, possibly because of the substrate. A similar observation has been made recently in a Raman spectroscopy study of Bi$_2$Te$_3$ nanolayers\textsuperscript{25}. The Raman spectrum of Fig. 1(e), acquired with a 5 mW incident laser power shows similar peaks as Fig. 1(d) data, but with a higher intensity of the $A_{1g}$ peak (242 cm$^{-1}$) with respect to the other modes. This observation is consistent with previous studies which show that the $A_{1g}$ mode is better resolved at longer excitation wavelengths.\textsuperscript{24}
The FWHM of $E_{1g}$, $A_{1g}$, $E^{1}_{2g}$ and $A^{2}_{2g}$ Raman peaks of Fig. 1(d) are 5.5, 5.2, 7, and 5 cm$^{-1}$, respectively. The $A_{1g}$ peak FWHM of Fig. 1(e) is 3.5 cm$^{-1}$, assuming in all cases a Lorentzian fit. The higher FWHM values when using a 442 nm excitation wavelength is explained by the higher incident laser power. Previous Raman spectroscopy studies on MoS$_2$ shows the same trend, explained by enhanced thermal effects.$^{5,26}$

In order to electrically probe an exfoliated MoSe$_2$ flake, we select a single terrace with uniform thickness to define the active region of the field-effect transistor. Electron-beam lithography (EBL) combined with reactive ion etching using Cl$_2$ are then used to define the device active region. The metal contacts are defined by a second EBL step followed by 80 nm-thick Ni evaporation and lift off. The highly doped Si substrate serves as back-gate for the MoSe$_2$ FETs. Several two- and four-point back-gated field effect transistors (FET) were investigated in this study.

The output and transfer characteristics of an FET fabricated on a 5.8 nm thick MoSe$_2$ flake (~ 8 layers), with channel length $L$ = 1.8 $\mu$m and width $W$ = 0.8 $\mu$m are shown in Fig. 2. The measurements presented in this paper are carried out in vacuum ($\sim$10$^{-7}$ torr), and in dark. In each measurement the source contact is grounded, while the drain is biased. Figure 2(a) shows transfer characteristics defined as drain current ($I_D$) vs. drain voltage ($V_D$) measured at various back gate voltages ($V_G$) at room temperature. The $I_D-V_D$ dependence is mostly linear, and does not show saturation at high drain bias, in contrast to what is expected in a conventional FET. Moreover the $I_D-V_D$ data exhibit a slight super-linear behavior at low drain bias, which suggests that the electrons are injected through a Schottky barrier at metal (Ni)-semiconductor interface. Figure 2(b) shows the transfer characteristic ($I_D-V_G$) measured at two different drain biases $V_D$ = 50 mV and $V_D$ = 1 V. The device shows an n-type behavior, and is depleted from free carriers for $V_G < 0$ V. The threshold voltage of this device, $V_T \approx 0$ V does not appear to change noticeably with $V_G$, consistent with a long-channel device. The $I_{on}/I_{off}$ ratio at $V_D = 1$ V curve is larger than 10$^6$, similar to reported On/Off ratio values for MoS$_2$, WS$_2$, and WSe$_2$ devices$^{8,15,16}$, and explained by the large energy gaps that characterize this family of materials.

To further probe the electron injection in MoSe$_2$, in Fig. 3 we show two sets of output characteristics measured on the same two-point back-gated MoSe$_2$ FET, but using a different source contact in each data set. For the same $V_G$ value, different $I_D$ values are obtained depending on which physical contact is used as source. This asymmetry is $I_D-V_D$, data further confirms the presence of a Schottky barrier at the metal-semiconductor contact. As a result the electron injection depends not only to the device geometry, e.g. contact area, but also on the electric field across the metal/MoSe$_2$ interface, and therefore will be sensitive to MoSe$_2$ flake thickness, SiO$_2$ dielectric thickness, as well as gate and drain bias. Schottky barrier contacts are common place for other nano-electronic devices, such as carbon nanotube (CNT)$^{27}$ and nanowire devices$^{28}$. The presence of non-ohmic contacts affects adversely the device performance by reducing the On state current and prevents a quantitative analysis of the device characteristics. Most importantly, extracting the intrinsic MoSe$_2$ mobility from data such as that of Fig. 2 is difficult.
To probe the intrinsic mobility of MoSe$_2$ flakes, we fabricate four-point back-gated devices, which allow conductivity measurements without contributions from the contact resistance of the metal-semiconductor Schottky barriers. The inset of Fig. 4 (a) shows an AFM image of a four-point MoSe$_2$ device. The outer contacts labeled $S$ and $D$ serve as source and drain, respectively. The inner contacts ($V_I$, $V_2$) are used as voltage probes, and have a limited overlap with the MoSe$_2$ flake to minimize screening of the gate-induced charge density in the channel. The measured channel conductance ($G$) is defined as $G = I_D/(V_I-V_2)$. Figure 4(a) shows the $G$ vs. $V_G$ data measured at different $T$ values from 298 to 78 K. For $V_G$ values lower than a threshold voltage ($V_T$), $G$ remains vanishing. Above threshold, the $G$ increases with $V_G$, with an approximately linear dependence. As the temperature is reduced, $V_T$ shifts progressively towards higher voltages.

![Fig. 2. (a) $I_D$ vs. $V_D$ measured at different $V_G$ values. The data show a super-linear behavior at low $V_D$ suggesting the presence of a Schottky barrier at the metal MoSe$_2$ contact. (b) $I_D$ vs. $V_G$ traces measured at $V_D = 50$ mV (solid squares), and $V_D = 1$ V (open circles) with $I_{sw}/I_{D}$ > 10$^6$ at $V_D = 1$ V.](image)

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To offset the $V_T$ shift with $T$, Fig. 4 (b) shows $G$ vs. $V_G$-$V_T$, at different $T$ values. Figure 4(b) data show a noticeable increase of the $dG/d(V_G-V_T)$ slope with decreasing $T$. The intrinsic mobility can be thus extracted as $\mu = (L/W) dG/dV_G C_{a/o}^{1/2}$, where $C_{a/o} = 1.2 \times 10^{-8}$ F cm$^{-2}$ is the capacitance of the 285 nm-thick bottom SiO$_2$ dielectric; $L$ and $W$ denote the length and width of the active device area, respectively. The inset of Fig. 4 (b) shows the mobility dependence on temperature for three different devices. The room temperature mobility is as high as 50 cm$^2$/V-s, and increases almost four fold when reducing the temperature to 78 K. In two dimensions, charged impurity scattering causes a temperature independent mobility in the degenerate limit, as in the case of graphene, and a $\mu \propto T$ dependence for a non-degenerate two-dimensional electron system. Acoustic phonon scattering generates a $\mu \propto T^2$ dependence, and optical phonon scattering, including polar optical phonons, cause a stronger $T$-dependence in TMDs. A functional fit of the form $\mu^{-1}(T) = A + BT^2$ to the $\mu$ vs. $T$ data of Fig. 4(b) inset yields an exponent $\nu$=2.1 for the highest mobility sample, suggesting that phonon scattering dominates in this sample. Temperature dependent Hall mobility measurements carried out on bulk MoSe$_2$ samples also report an increased mobility with reducing temperature.

Lastly we address the contact resistance in our devices. Having measured the MoSe$_2$ resistivity using four-point devices, we can subtract the flake intrinsic resistance from the measured source-to-drain resistance in order to estimate the contact resistance. The room temperature contact resistance values range between 200 k$\Omega$ at large, $V_G$-$V_T = 35$ V gate overdrive, and increase to 6 M$\Omega$ as $V_G$ approaches $V_T$. Reducing the temperature leads to an increase in contact resistance. The strong $V_G$ dependence of the contact resistance provides further evidence for the presence of a Schottky barrier at the metal/MoSe$_2$ interface, an obstacle which will have to be overcome in order to increase the On current.
In summary, we demonstrate n-type field-effect transistors on ultra-thin MoSe$_2$ flakes, showing high $I_{on}/I_{off}$ ratios. We probe the intrinsic mobility as a function of temperature using gated four-point device structures, and show that the mobility increases significantly with lowering the temperature, which suggest phonon scattering plays a dominant role at room temperature.

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