Control of polarity in multilayer $MoTe_2$ field-effect transistors by channel thickness

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ABSTRACT

In this study, electronic properties of field-effect transistors (FETs) fabricated from exfoliated $MoTe_2$ single crystals are investigated as a function of channel thickness. The conductivity type in FETs gradually changes from n-type for thick $MoTe_2$ layers (above ≈ 65 nm) to ambipolar behavior for intermediate $MoTe_2$ thickness (between ≈ 60 and 15 nm) to p-type for thin layers (below ≈ 10 nm). The n-type behavior in quasi-bulk $MoTe_2$ is attributed to doping with chlorine atoms from the $TeCl_4$ transport agent used for the chemical vapor transport (CVT) growth of $MoTe_2$. The change in polarity sign with decreasing channel thickness may be associated with increasing role of surface states in ultra-thin layers, which in turn influence carrier concentration and dynamics in the channel due to modulation of Schottky barrier height and band-bending at the metal/semiconductor interface.
Keywords: 2D materials, $MoTe_2$, FET

INTRODUCTION

Recent advances and miniaturization in silicon based electronic devices are hindered by the limitation of short channel effects, high contact resistance and high leakage current. These limitations have motivated the researchers to search for alternate materials. Unique electronic and optoelectronics properties of graphene and beyond two-dimensional (2D) materials have shown the potential for next generation electronics.[1] Due to its gapless band structure, graphene-based FETs have a limited application for transistors or logic circuits for low-power applications. Advances and limitations in graphene research attracted researchers towards layered transition metal dichalcogenides (TMDCs). A broad range of bandgap values in TMDCs has paved the path to explore these materials for transistor applications. These layered materials have a common formula MX_2, where M is a transition metal from group IV-VII (Mo, W, Nb, etc.) and X is a chalcogen (S, Se or Te). One layer of MX_2 consists of a hexagonally packed sheet of M atoms that is sandwiched between two sheets of X atoms.[2] Weak van der Waals bonding between the layers enables thinning of TMDCs crystals down to a monolayer by mechanical or chemical exfoliation.

Among all TMDCs, $MoTe_2$ is the only material that can be grown in both semiconducting (2H) and semi-metallic (1T′) thermodynamically stable crystal structures and hence, it lends its potential usefulness for engineering of ohmic contacts and for resistance-switching applications. In the 2H semiconducting phase, band gap values for bulk and monolayer $MoTe_2$ are 0.81 eV (indirect) and 1.13 eV (direct), respectively,[3] which makes it an attractive candidate for optoelectronic devices in the visible to near-infrared spectrum.[4-7] 2H-$MoTe_2$ is intrinsically p-doped but can also exhibit ambipolar behavior.[8] Recently it was demonstrated that $MoTe_2$ FET polarity can be tuned by using dual top gates geometry or by selecting metals with appropriate work functions for drain and source contacts.[9-11]

Here, we demonstrate control over n-, ambipolar and p-type conductivity in $MoTe_2$ back-gated FETs by reducing the channel thickness from thick (above ≈ 65 nm) to medium (between ≈ 60 nm and 15 nm) to thin (below ≈ 10 nm), respectively. The thickness modulated transport properties of $MoTe_2$ FETs open up possibilities for digital and analog

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circuits by providing a guidance for fabricating p-, ambipolar and n-type devices by tuning the channel thickness with no additional steps for extrinsic doping during material and device fabrication processes.

EXPERIMENTAL DETAILS

$MoTe_2$ flakes used in this work were mechanically exfoliated from bulk single crystals that were grown by chemical vapor transport (CVT) method using polycrystalline $MoTe_2$ powder and $TeCl_4$ (ca. 5 mg/cm^3) transport agent sealed in evacuated quartz ampoules. Temperature in the hot and cold zones was 800 °C and 700 °C, respectively. Growth duration of $MoTe_2$ crystals was 140 h. This method produced pure 2H phase of $MoTe_2$ as confirmed by X-ray powder diffraction (XRD), Transmission Electron Microscopy (TEM) and Raman spectroscopy (see Fig. 1 for Raman and elsewhere[12] for XRD and TEM data).

Flakes of different thickness, ranging from ≈ 5 nm to 78 nm, were transferred onto 300-nm-thick SiO_2/Si substrate, where silicon was used as a back-gate electrode and the oxide layer acted as a gate dielectric. Prior to exfoliation, the substrates were ultrasonically cleaned in acetone, 2-propanol and deionized water followed by oxygen plasma cleaning. The exfoliated flakes were located under an optical microscope and were mapped out to predefined alignment marks for source/drain contact fabrication. The source and drain contacts were patterned using conventional photolithography followed by e-beam deposition of Ti(40 nm)/Au(350 nm) bilayer and lift-off process. The devices were thermally annealed in vacuum at 350 ºC for 5 min. AFM measurements were used to verify the thickness, smoothness and uniformity of $MoTe_2$ layers under investigation. All the fabricated devices were characterized by output and transfer characteristics using back-gated FET measurement at room temperature in ambient environment.

RESULTS AND DISCUSSION

The lattice vibrational modes of exfoliated 2H-$MoTe_2$ flakes were identified using Raman spectroscopy with a 532 nm laser source. The spectra for the 2H-$MoTe_2$ (Fig. 1) exhibit characteristic A_{1g} at 170 cm^{-1}, E^1_{2g} at 235 cm^{-1} and B^1_{2g} at ≈ 288 cm^{-1} modes (the latter is active in thin layers only). Comparison of Raman spectra for varying thickness in Fig.1 is in agreement with Raman studies reported in the literature,[4] which confirms the 2H phase and good crystalline quality of $MoTe_2$ layers used in FETs. Inset in Fig. 1 shows plan-view of a typical FET device.

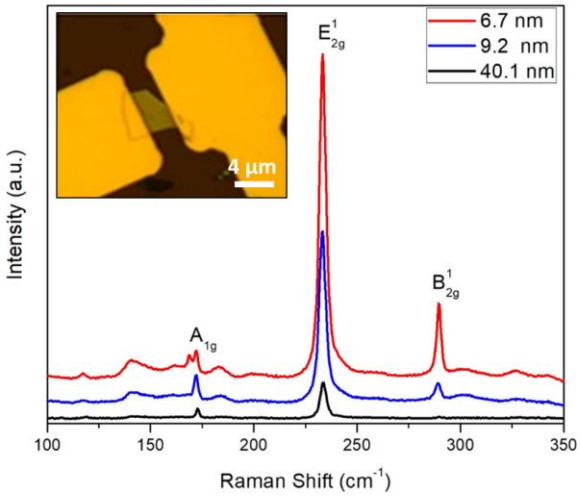

Fig. 1. Raman spectra of $MoTe_2$ flakes of different thickness exfoliated from bulk $MoTe_2$ single crystals grown by CVT with $TeCl_4$ transport agent. Inset shows plan-view optical image of FET device with a 50 nm thick channel.

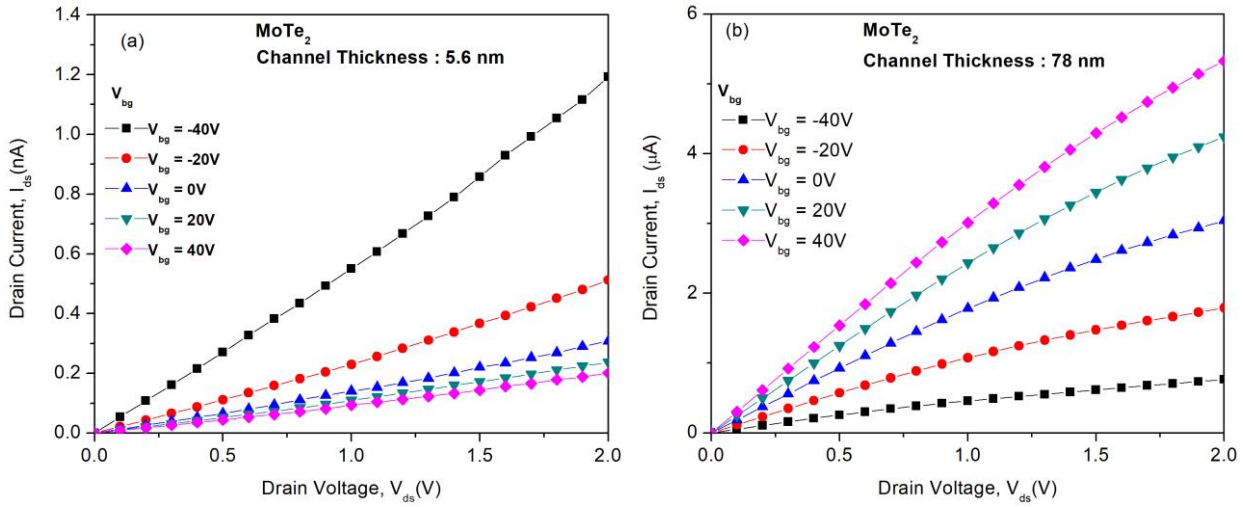

Fig. 2. Output characteristics of $MoTe_2$ FETs under different back-gate voltages, V_{bg}. FETs with channel thickness of (a) 5.6 nm and (b) 78 nm show p- and n- type conductivity, respectively.

To understand the effect of channel thickness on FET transport characteristics, we measured the output (drain current I_{ds} vs drain voltage V_{ds}) and transfer (I_{ds} vs back-gate voltage V_{bg}) characteristics. The results from output and transfer curve show that by adjusting the thickness of the $MoTe_2$ channel, it is possible to let holes or electrons dominate the channel conduction, resulting in *p*- or *n*- type FET behavior. The output curves at varying V_{bg} are shown in Fig. 2(a) for thin, 5.6 nm, and Fig. 2(b) for thick, 78 nm, channels. Clear p-type, i.e., decrease in I_{ds} with increasing V_{bg}, and n-type, increase in I_{ds} with decreasing V_{bg}, transport behavior is observed for thin vs. thick $MoTe_2$ FETs, respectively. Note that although the output characteristics appear to be nearly linear, a small Schottky barrier at the metal/semiconductor junction is expected due to difference between work function of Ti (4.33 eV) and electron affinity of $MoTe_2$ (4.3 ± 0.1 eV).[10, 13]

We further analyzed the transfer behavior of the FETs by I_{ds}-V_{bg} curves at V_{ds} = 2 V, which showed n-, ambipolar and p-type behavior for thick, medium and thin channel, respectively. The gate leakage current I_{gs} in all the devices was negligible in the pA range. The maximum I_{ds} ON/OFF ratio was about 1×10^3, which decreased with increasing channel thickness. The experimental results of transfer curves are shown in Fig. 3. Multiple devices were fabricated to reproduce the transfer characteristic as a function of $MoTe_2$ channel thickness. For channel thicker than ≈ 65 nm FETs showed consistent unipolar n-type behavior. For a medium channel thickness, from ≈ 60 nm to 15 nm, FET behavior was ambipolar, while FETs with very thin channel, from ≈ 10 nm down to 5 nm, were all p-type. Effect of channel thickness on various transport properties in FETs was previously described in black phosphorous, WSe_2, MoS_2, and $MoTe_2$ FETs.[14-18]

In reference to n-type behavior, the n-doping in TMDCs has been observed when using $TeCl_4$ as the transport agent in the CVT growth[19] and during post-growth chloride molecular doping of TMDC compounds.[20] In the present work, since SIMS measurements (not shown here) detected a presence of Cl impurity in the CVT grown $MoTe_2$ crystals, we assume that Cl doping is a dominant factor for n-type behavior in our thick channel devices. The switching of conductivity to p-type in thinner channel FETs suggests that the effectiveness of Cl doping diminishes upon $MoTe_2$ layer thinning. This can be associated with increasing role of surface defects and adsorbates in ultra-thin layers, a phenomenon which is exemplified in[21] for atomically thin MoS_2 FETs. In addition to possible detrimental effect of reduced $MoTe_2$ channel thickness on the n-type doping efficiency, we also speculate that the n- to p- polarity switching in thinner layers may be caused by the modulation of Schottky barrier height and corresponding band alignment and band-bending at the metal/$MoTe_2$ interface.

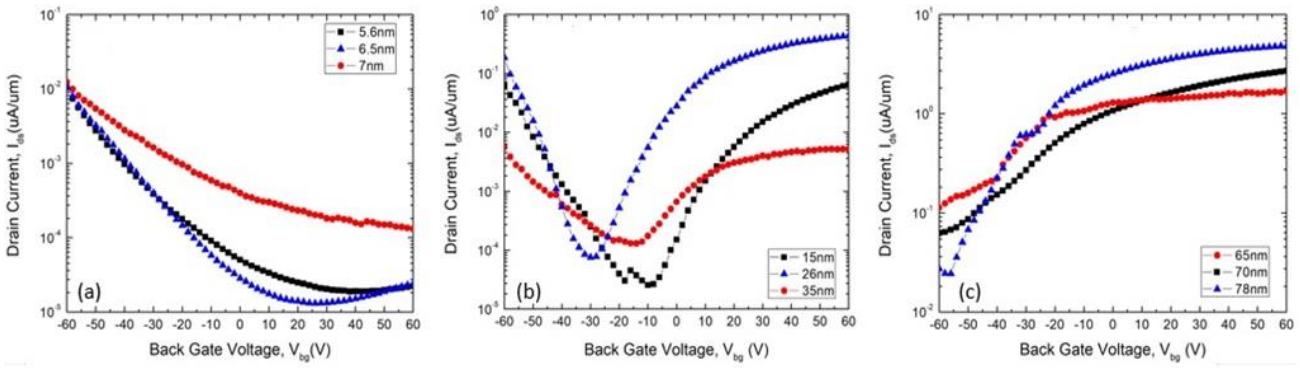

Figure 3. Transfer characteristics at V_{ds} = 2 V of (a) thin (p-type), (b) medium (ambipolar), and (c) thick (n-type) channel FETs. The drain current is normalized by the channel width.

In this study, we also investigated the thickness-dependent field-effect carrier mobility (μ_{FE}) of the fabricated devices, which is extracted from the transfer characteristics shown in Fig. 3. Following equation defines the μ_{FE} of the $MoTe_2$ FET:

$$\mu_{FE} = \frac{dI_{DS}}{dV_{BG}} \left(\frac{L}{W\, C_{OX} V_{DS}}\right), \qquad (1)$$

where C_{ox} is a silicon oxide capacitance per unit area, L is the channel length, and W is the channel width.

Fig.4 shows the overall trend of mobility increasing with increasing channel thickness. The influence of thickness in $MoTe_2$ devices can be related to Coulomb scattering and quantum confinement.[22,23] The scattering of the carriers is weakened by the Coulomb interaction resulting in higher mobility in thicker vs. thinner channel as it was demonstrated for both $MoTe_2$[14] and MoS_2[21] FETs.

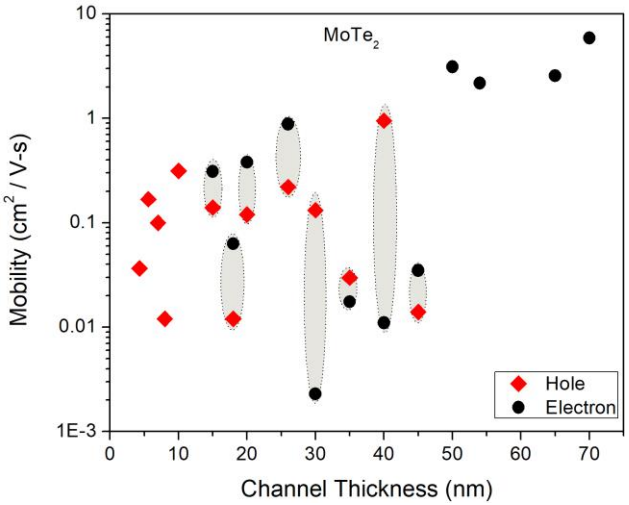

Fig. 4. Field effect mobility of electrons and holes vs channel thickness for $MoTe_2$ FETs. Encircled pairs of data points correspond to the ambipolar devices that exhibit ambipolar conductivity.

CONCLUSION

In this study, we have demonstrated a simple, yet effective way to control the conductivity type in $MoTe_2$ FETs by tuning the channel thickness. FETs were fabricated from the CVT-grown and mechanically exfoliated $MoTe_2$ single crystals. The transport properties were examined for varying channel thickness using the output and transfer characteristics. FETs showed a change in polarity from n-type through ambipolar to p-type with reducing the channel thickness from 78 nm to 5 nm. The n-type conductivity in thick $MoTe_2$ layers is attributed to chlorine doping from $TeCl_4$ transport agent used in the CVT growth. The switch of polarity by thinning the FET channel may be associated with increasing role of surface states in ultra-thin layers, which can influence charge carrier concentration and dynamics in the channel by modulating Schottky barrier height and associated band alignment and band-bending at the metal/semiconductor interface.

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