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1D van der Waals Materials in 2D Form

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Abstract

Selenium and Tellurium are 1D van der Waals materials with a helical atomic chain structure, which are tended to form 1D nanowire structures. Recently, we have demonstrated a physical vapor deposition process and a substrate-free solution phase process to synthesize large-area, high-quality 2D single crystal Se and Te films, respectively. Experimental demonstrations of 1D van der Waals materials have been presented by Raman spectroscopy under strain and magneto-transport. The as-fabricated Te field-effect transistors demonstrate large on/off ratio (~10^6), high field-effect mobilities (~700 cm^2/Vs), and excessive drain current of ~600 mA/mm at 100 nm channel length, which are comparable or superior to transistors made of other experimentally accessible 2D semiconductors. In great contrast to black phosphorus and other environmentally sensitive 2D crystals, 2D Se or Te thin film has a pronounced air-stability.

1. Introduction

Trigonal selenium (t-Se) or tellurium (t-Te) is a one-dimensional elemental semiconductor material, in which Se or Te atoms are covalently connected in a spiral chain along c axis with two adjacent atoms rotated by 120°. As shown in Figure 1a and b, all chains are stacked together in its radical direction by weak van der Waals interactions to form hexagonal structure. The t-Se or t-Te has attracted extensive attentions due to its interesting properties such as high carrier mobility, high photoconductivity, high piezoelectricity, thermoelectricity and nonlinear optical responses. Due to the anisotropic chain-like crystal structure, Se or Te tends to form 1D-structures, such as nanowires, nanotubes and nanobelts. These 1D structures with high ratio of edges to bulk always exhibit poor electrical transport behaviors due to the localized states and contacts, and the issue of electrical noise would be critical with decreasing size as described by Hooge's rule. To expand its applications in high-performance electronic and optoelectronic devices, it would be useful to expand the Se or Te nanostructures to two-dimension form.

2. Experimental Results and Discussions

Synthesis of Multi-layer Selenene 2D Film

The multi-layer selenene flakes were grown by PVD [1]. Selenene and Tellunene are the terms to represent single layer of Se or Te 2D films. The growing setup is illustrated in Figure 1c. Se powder was placed in a quartz tube as the precursor. Plan-view SEM image of selenene flakes as grown on Si substrate is shown in Figure 1d, in which inclined flakes (as indicated by arrows) with density over 50% were uniformly distributed on the surface of polycrystalline Se films. Figure 1e shows the representative morphology of the selenene flakes at high magnification. Most of the flakes exhibit saw-like structure with zigzag edges on single narrow side. The average width of flakes is about 8 μm, and the maximum length of the flakes could reach up to 50 μm. In SEM images, the flakes are almost electron transparent, suggesting that they are very thin. It is worthy being noted that the 10% of selenene flakes have bi-lateral zigzag edges, exhibiting feather-like structure (Figure 1f). The growth mechanism study for these 2D selenene flakes will be discussed in the following section.

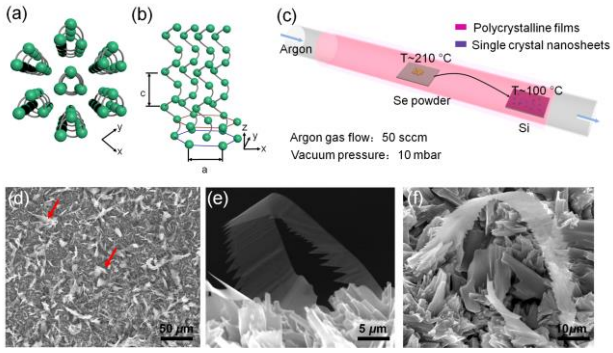

Figure 1 | PVD-grown large-area selenene and material characterization. (a-b) Atomic structure of selenium. (c) Schematic diagram of the PVD method. (d) Low-magnification SEM image of as-prepared selenene on Si (111) substrate. (e) Enlarged view of typical Se flakes with saw-like structure and (f) feather-like twin structure.

Synthesis of Multi-layer Tellunene 2D Film

The samples were grown through the reduction of sodium tellurite (Na_2TeO_3) by hydrazine hydrate (N_2H_4) in an alkaline solution at temperatures from 160-200 ^oC, with the presence of crystal-face-blocking ligand polyvinylpyrrolidone (PVP). The amount of hydrazine is maintained at a high level to ensure the complete reduction of TeO_3 into Te. A controlled, slow release of the Te source was achieved under the alkaline condition for maintaining the Te concentration, critical for the kinetic control of the 2D growth. The Te thin films were then transferred onto a SiO_2/Si substrate after a post-synthesis Langmuir-Blodgett assembly process, and been used for fabrication process [2].

1D van der Waals Materials versus 2D van der Waals Ones

We choose multi-layer Tellunene as an example. Raman spectroscopy measurements have been performed under strains along different principle axes. Pronounced strain

response along c-axis is observed due to the strong intra-chain covalent bonds, while no strain response is obtained along a-axis due to the weak inter-chain van der Waals interaction. Magneto-transport results further verify its anisotropic property, resulting in dramatically distinct magneto-resistance behaviors in terms of three different magnetic field directions. Specifically, phase coherence length extracted from weak antilocalization effect, $L_\phi \sim T^{-0.5}$, claims its 2D transport characteristics when an applied magnetic field is perpendicular to the thin film. In contrast, $L_\phi \sim T^{-0.33}$ is obtained from universal conductance fluctuations once the magnetic field is along c-axis of Te, indicating its nature of 1D transport along the helical atomic chains. Our studies, which are obtained on high quality single crystal tellurium thin film, appear to serve as strong evidences of its 1D van der Waals structure from experimental perspectives. It is the aim of this paper to address this special concept that differs from the previous well-studied 1D nanowires or 2D van der Waals materials [3].

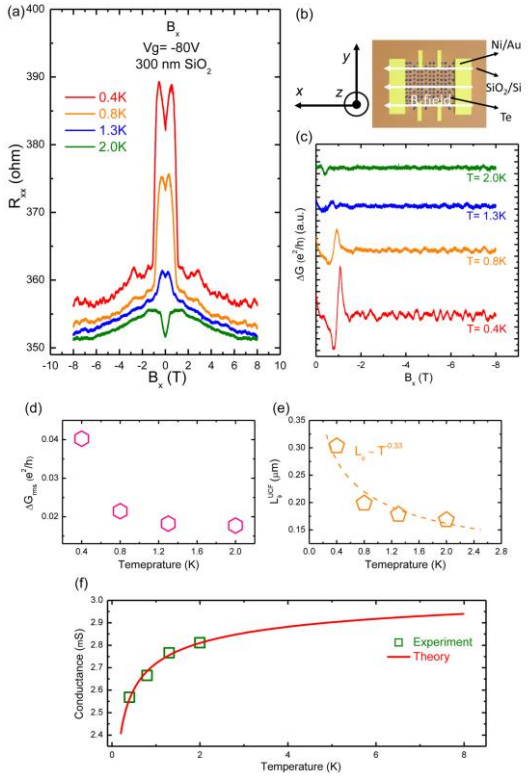

Figure 2 | Magneto-transport of Te in B_x direction. (a) Magneto-resistance of Te thin film with varied B_x field for different temperatures. The sample has a back-gate bias of -80 V, where the SiO_2 thickness is 300 nm. (b) Device scheme configuration and magnetic field indicator. (c) $\Delta G(T)$ curves with varied B_x field. The UCF effect is robust, and persisting to increased temperature up to 2 K. (d) ΔG_{rms} changes with different measured temperatures. (e) The phase coherence length has demonstrated a strong power-law behavior, and dashed line indicates theoretical simulation $L_\phi \sim T^{-\gamma}$ with a power exponent $\gamma = 0.33$. Our experimental results match the theoretical prediction that carrier scattering would give L_ϕ proportional to $T^{-0.33}$ in the 1D system. (f) Temperature dependence of the conductance at zero magnetic field. The red solid line is theoretical prediction, and green squares are experimental data.

Device Demonstration and Characterizations

Field-effect transistors can be demonstrated on 2D films of Se or Te. Te has a suitable bandgap (0.35 eV versus 1.6 eV for Se), high hole mobility (>700 $cm^2/V\bullet s$ at room temperature) and outstanding air-stability which guarantee it to be an ideal channel material for constructing high-speed p-type transistors with great scaling potential. Large-scale few-layer tellurium films (colloquially named as tellurene) with atomic flat surface which provides a platform to build field-effect transistors We explored high-performance tellurene transistors integrated onto high-k dielectric hafnium zirconium oxide and achieved maximum drain current around 1.1 A/mm, which is so far highest record among all the low dimensional material field-effect transistors.

3. Conclusions

We have successfully synthesized large-area few-layer air-stable Se or Te 2D films by PVD or solution based synthesis. 1D van der Waals nature of these 2D films has been confirmed by Raman spectroscopy under strain and low temperature magneto-transport. High performance field effect transistors have been demonstrated on this new type of 2D films showing its high carrier mobility.

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