



Full Length Article

van der Waals epitaxy of SnS film on single crystal graphene buffer layer on amorphous SiO₂/Si

Yu Xiang^{a,b,*}, Yunbo Yang^{a,b}, Fawen Guo^{a,b}, Xin Sun^{a,b}, Zonghuan Lu^{a,b},
Dibyajyoti Mohanty^{a,c}, Ishwara Bhat^{a,c}, Morris Washington^{a,b}, Toh-Ming Lu^{a,b},
Gwo-Ching Wang^{a,b}

^a Center for Materials, Devices, and Integrated Systems, Rensselaer Polytechnic Institute, 110, 8thStreet, Troy, NY 12180-3950, USA

^b Department of Physics, Applied Physics and Astronomy, Rensselaer Polytechnic Institute, 110, 8thStreet, Troy, NY 12180-3950, USA

^c Department of Electrical, Computer and Systems Engineering, Rensselaer Polytechnic Institute, 110, 8thStreet, Troy, NY 12180-3950, USA

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ABSTRACT

Conventional hetero-epitaxial films are typically grown on lattice and symmetry matched single crystal substrates. We demonstrated the epitaxial growth of orthorhombic SnS film (~500 nm thick) on single crystal, monolayer graphene that was transferred on the amorphous SiO₂/Si substrate. Using X-ray pole figure analysis we examined the structure, quality and epitaxy relationship of the SnS film grown on the single crystal graphene and compared it with the SnS film grown on commercial polycrystalline graphene. We showed that the SnS films grown on both single crystal and polycrystalline graphene have two sets of orientation domains. However, the crystallinity and grain size of the SnS film improve when grown on the single crystal graphene. Reflection high-energy electron diffraction measurements show that the near surface texture has more phases as compared with that of the entire film. The surface texture of a film will influence the growth and quality of film grown on top of it as well as the interface formed. Our result offers an alternative approach to grow a hetero-epitaxial film on an amorphous substrate through a single crystal graphene buffer layer. This strategy of growing high quality epitaxial thin film has potential applications in optoelectronics.

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1. Introduction

Tin mono-sulfide (SnS) has drawn interests among photovoltaic cells (PVCs) research community worldwide [1,2]. The advantages of SnS film include an optical energy band gap of 1.3 eV which is close to the optimum value required for efficient light absorption, a high optical absorption coefficient of $>10^4 \text{ cm}^{-1}$ above the photon energy threshold (~1.3 eV) [3], low cost, and non-toxic. SnS also has other applications in photodetectors [4], gas sensors [5], and Li-ion batteries [6]. One of the major challenges in the SnS thin film based PVCs is producing high quality materials [7]. Attempts have been made to grow high quality epitaxial SnS film by using lattice-matched (LM) single crystal substrates such as MgO(001) [8] and Al(100) [9]. In this conventional hetero-epitaxy the lattice mismatch and thermal mismatch between the chosen overlayer and the substrate materials are usually small. The large lattice mismatch

and strong chemical bonds at the interface may have several consequences: (1) Strain develops at the interface. (2) Various structural defects may nucleate in the overlayer. (3) The chemical inhomogeneity may be introduced in the overlayer. (4) The intermixing between the overlayer and substrate may occur at the interface. If the dangling bonds on a substrate are not fully passivated, the surface reconstruction and lattice relaxation could happen. This could affect the growth mode and structural quality of an overlayer grown on it later.

Limited choices of LM single crystal substrates, possible structural imperfect overlayer, and chemical bondings at the interface of the hetero-junction in conventional hetero-epitaxy led researchers to pursue alternative routes to grow high quality epitaxial SnS film. Apart from being a promising light absorption material, SnS also belongs to a class of materials called layered metal chalcogenide (LMC). The LMC materials have rich fundamental physical and chemical properties and potential applications in electronics and optoelectronics, which has stimulated intense research activities world-wide in recent years [10]. A LMC material consists of a stacking of two-dimensional (2D) MXs layers where M is a metal

* Corresponding author at: Center for Materials, Devices, and Integrated Systems, Rensselaer Polytechnic Institute, 110, 8thStreet, Troy, NY 12180-3950, USA.

E-mail address: xiangy@rpi.edu (Y. Xiang).

from groups IV, V or VI and X is a chalcogen atom from S, Se or Te. A unique characteristic of this class of materials is that the chalcogen atoms are chemically saturated and, as a result, the weak inter-layer interactions are dominated by the van der Waals force [11]. The van der Waals force nature of the interaction between SnS layers makes it possible to grow van der Waals epitaxies (vdWEs) [12]. Compared to conventional hetero-epitaxies, the van der Waals substrate may not give rise to large strain in the overlayer, and therefore high density structural and chemical defects may not develop in the overlayer and at the interface [13]. Thus, the lattices between the overlayer and the substrate could have a large mismatch and the overlayer could be incommensurate with the substrate at the interface [14]. The lift of this restriction on lattice match greatly broadens the choices of materials used in hetero-epitaxy systems. One can imagine the growth of either layer [15] or even non-layer [16–18] epitaxial films or non-planar nanostructures [19] on the van der Waals substrates such as graphene.

All these merits of vdWEs make it a very favorable method to grow high-quality SnS film. SnS film has been successfully grown on transferred double-layer, polycrystalline graphene buffer layer on single crystal GaAs(100) or amorphous SiO₂/Si substrates [15,20]. The double-layer, polycrystalline graphene was grown by chemical vapor deposition (CVD). These previous reports indicate that the SnS films grown on transferred double-layer, polycrystalline graphene on an amorphous substrate or a GaAs(100) substrate have narrower rocking curves and stronger out-of-plane orientations compared with that on substrates without the transferred double graphene layer. However, the in-plane epitaxy appears to contain many orientation domains.

In this work, we report the growth and characterization of high-quality SnS (~500 nm thick) films using thermal evaporation of SnS powders onto the homemade single crystal graphene which has one orientation domain and polycrystalline commercial graphene which has two dominant orientation domains with 30° rotation relative to each other. We will refer to these two substrates as the single crystal graphene and the polycrystalline graphene in the following text. We characterized the morphology and surface roughness of graphene and SnS film using atomic force microscopy (AFM) and scanning electron microscopy (SEM). The grain size and misorientation were measured by electron backscatter diffraction (EBSD). The graphene symmetry and number of orientation domains were examined by reflection high energy electron diffraction (RHEED) 2D reciprocal space mapping. The structure and texture of the bulk SnS films were measured by X-ray diffraction (XRD) and X-ray pole figure. For structure and texture near the film surface, RHEED was used. The in-plane epitaxial relationship at the interface of SnS and graphene was determined by combining XRD and RHEED. The vibrational and electrical properties of the films were measured by Raman spectroscopy and Hall measurement, respectively.

2. Experimental

2.1. Buffered substrates

2.1.1. Commercial polycrystalline graphene

The polycrystalline graphene buffered SiO₂/Si substrate used in our experiment was a commercial product (graphene-supermarket.com). Monolayer, polycrystalline graphene on 1 cm × 1 cm silicon wafer (p-doped) with <100> orientation and a silicon oxide coating (~150 nm) was prepared by the company following the procedure developed by Li et al. [21]. and then transferred onto the silicon wafer using the “PMMA-mediated” approach [22,23]. 95% of the surface area is covered with monolayer graphene, while 10–30% of this area is also covered with

bilayer graphene islands. Prior to the SnS film deposition the morphology and grain orientations of this commercial graphene substrate were characterized by using SEM, AFM, and RHEED. See figures later.

2.1.2. Homemade single crystal graphene

The homemade graphene was first epitaxially grown on single-crystalline Cu(111)/sapphire substrates with a low pressure chemical vapor deposition method. The single-crystalline Cu(111) films were prepared on sapphire(0001) substrates by DC sputtering and post-deposition annealing. The graphene layer was then transferred onto a 50 nm thick SiO₂ layer on Si(100) substrate [18]. The structure of the graphene layer was determined to be single crystal through azimuthal RHEED reciprocal space mapping [24].

2.2. SnS film preparation

According to the vapor phase diagram, the SnS evaporates concurrently during deposition by thermal evaporation [25]. This makes thermal evaporation a desirable method to grow SnS thin film. The advantage of using thermal evaporation of SnS is that thermal evaporation introduces negligible disorder in the single layer graphene as compared with other deposition techniques such as sputtering, e-beam, and pulsed laser depositions as shown by Raman spectra [26]. The SnS film was grown on graphene/SiO₂/Si(100) substrate by evaporating SnS powder (Sigma Aldrich, purity ≥ 99.99%) placed in an alumina coated tungsten basket with the top opening diameter of about 1 cm (R.D. Mathis). The substrate was mounted about 15 cm above the SnS source. The base pressure of the vacuum chamber was 5–7 × 10⁻⁷ Torr. Before a deposition, the substrate was heated to 280 °C from the backside with a ramping rate of 20 °C/min using a double ended halogen lamp (Ushio, FCL, J120V-500 W) with a rectangular reflector (8 cm × 12 cm) installed inside the vacuum chamber. A schematic of the experimental setup of the thermal evaporator is shown in Fig. 1(a). During the deposition, the temperature of the substrate surface was maintained at 283–286 °C, which was monitored using a type K thermocouple attached to the substrate surface. The deposition rate was about 0.6–0.8 Å/s according to a quartz crystal monitor (QCM) with a gold coated quartz monitor crystal (Inficon, 008-010-G10). After the deposition, the lamp was turned off immediately. The substrate and the deposited SnS film were cooled down naturally. Both SnS films of about 500 nm thickness were deposited simultaneously on the polycrystalline and single crystal graphene substrates using this system. Fig. 1(b) shows a schematic of the layered structure of the SnS on graphene/SiO₂/Si(100) sample.

2.3. Characterization techniques used

The surface morphologies of the synthesized SnS films were imaged using an AFM (PSI XE100) in contact mode and a field emission scanning electron microscope (SEM) (ZEISS SUPRA 55). The AFM tip (µmash, HQ: CSC17/AL BS) used had a tip radius of 8 nm, a force constant of 0.18 N/m, and a resonant frequency of 13 kHz. To characterize the crystal structure and the average grain size of the SnS films in the vertical direction, X-ray diffraction (XRD) was carried out using an X-ray diffractometer (Bruker D8 Discover). The Cu Kα X-ray source used had a wavelength of 1.5405 Å. A point detector with a 0.01° scanning step size and a 0.1 s counting time at each step was used to collect the X-ray diffraction theta-2theta (θ-2θ) scans. The scanning step size for the rocking curve was 0.002°. The incident beam, detector, and anti-scatter slits used all had a 0.6 mm width. XRD pole figure measurements were carried out using the same X-ray diffractometer with a 1° step size in both azimuthal and chi directions.

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