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Growth and examination of non-linear electrical behavior of bulk lead-tin-selenide

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ABSTRACT

The design and development of topological insulators (TIs) as well as novel Dirac materials including Weyl semimetals has expanded a new field of condensed matter physics due to their potential applications in spin-electronics, quantum computing, and thermoelectrical device. Various new materials have been proposed and investigated, however, Pb and Sn based topological insulators are a top choice mainly due to the tunable nature of chemical potential, easy transformation from trivial insulating to topologically nontrivial behavior, and relatively low temperature growth. Earlier this material has been widely investigated in the bulk form as well as in its nanocrystalline form. Pb/Sn ratio is a great factor that control the morphology as well as phase purity of the material. In this present study, we investigate the many different substitutional solid solutions of Sn, Pb, and Se of varying compositions and their associated morphology. The phase and structural analyses of selected samples were also performed with the help of the X-Ray diffractometer and Raman spectrophotometer, to confirm the purity of the material, prior to electrical examination. The non-linear behavior of bulk sample was systematically studied.

1. Introduction

In order to study the new science as well as the intrinsic band structure of Dirac and Weyl materials and to realize novel and the high-performance device functionalities, high quality samples of uniform and large grains with less crystallographic defects, are required. Topological insulators (TIs) are a new class of materials characterized by a conducting surface band gap and an insulating band gap in bulk, which means that electrons can travel only at the surface of the material. However, the unique characteristic of TIs is that the surface states of these materials are symmetry protected by particle number conservation and time reversal symmetry [1,2]. Due to time reversal symmetry protection, the surface characteristics of TIs are immune to time-reversal perturbances such as non-magnetic impurities and crystal defects [3]. Therefore, due to its distinct properties, TIs find potential applications in spintronic and quantum computation [4,5]. Materials like Bi_2Se_3, Bi_2Te_3, $Bi_{1-x}Sb_x$, HgTe/$Hg_{1-x}Cd_xTe$ are identified as the typical TIs [3,6–10].

Recently, materials identified as topological crystalline insulators (TCIs) in which the topological protection arises from the crystal symmetries [11–13]. In contrast to TIs, the conducting surface gap band in TCIs are only on the highly symmetric crystal planes such as crystal planes {110} and {111} [11–13]. Considerable research on TCIs has been reported in literature [14–20]. SnTe was the first material discovered to show TCI properties from first principle calculations [13]. However, excessive p-type doping due to active Sn vacancies make it difficult to study the TCI behavior experimentally [17]. It was found that $Pb_{1-x}Sn_xSe$ can be tuned to yield n-type or p-type material during the crystal growth [17]. This makes $Pb_{1-x}Sn_xSe$ alloy suitable for experimental study of the TI state. The existence of Dirac cones was experimentally shown on the (001) crystal plane of $Pb_{1-x}Sn_xSe$. Also, the electronic structure of $Pb_{1-x}Sn_xSe$ can be tuned by applying external electrical field [21] and elastic strain [13]. This result suggests that TCIs can be used in tunable electronic and spintronic devices [12]. In recent years substitutional solid-solutions of IV-VI semiconductors like $Pb_{(1-x)}Sn_xSe$, where $x = 0.23$ have exhibited a great promise as topological insulators [14,16].

Since, the signal from the surface is suppressed by the bulk signal, it is difficult to examine the surface properties of TCIs. Therefore, nanostructured TIs are ideal to investigate the surface topological behavior experimentally since the carrier transport from the bulk is effectively suppressed due to large surface-to-volume ratio [22–24]. Nanostructured TIs are important for

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understanding the quantum mechanics phenomena and its application as nanoscale electronic and spintronic devices [22–25]. From this perspective, it is important to develop the growth techniques for high quality single crystalline nanostructured TIs. Various nanostructured TIs with morphologies such as nanoribbons [22], nanowires [26], nanoplates [27], films [28] have been successfully made using the vapor deposition technique by optimized processing parameters. However, in the case of TCIs, controlling the morphologies of TCIs nanostructures is very important for the fact that topological surface states are also dependent on highly symmetric crystal surfaces [29]. New methods of producing uniformity over the interrogated volume in terms of purity, phase, and crystallographic orientation are also essential. Previous studies have shown the controlled synthesis of single crystalline SnTe nanostructures by adjusting the processing parameters of vapor deposition. A few works have been reported for the growth of important nanostructured $Pb_{1-x}Sn_xSe$ TI [14–16]. This may be due to fact that the complicated growth conditions may be required due to its intricate stoichiometry. It should be noted that $Pb_{1-x}Sn_xSe$ exhibits a peculiar surface state containing four Dirac cones.

Pb-Sn-Se alloy, that shows a composition dependent physical and electronic property, exhibits the inversion of conduction and valence bands [30]. The bandgap of material also depends on its stoichiometry [31]. Hence it is essential to focus on growth methods to produce oriented thin film, crystals, as well as polycrystalline samples with fewer imperfections, over areas sufficiently large to be probed either with free-space beams or with the use of antenna structures (in case of THz applications). Studies indicated that $Pb_{1-x}Sn_xSe$ crystallizes in the rock-salt structure having an electronic bandgap less than 0.29 eV ($E_g \leq 0.29$ eV). In addition, $Pb_{1-x}Sn_xSe$ alloy undergoes band inversion when Sn content exceeds a critical value x_c [32]. The band inversion is found to be temperature dependent. When the temperature is below the critical gap-inversion temperature T_{inv} in the samples $x > x_c$, band inversion occurs [32]. It is shown that $Pb_{1-x}Sn_xSe$ undergoes transformation from a trivial insulator to a TCI associated when the band inversion happens [17]. This suggests $Pb_{1-x}Sn_xSe$ could be used in the tunable electronic and spintronic devices by manipulating the composition and/or temperature parameters. Therefore, optimization of processing parameters for the synthesis of newly discovered topological behavior nature, it is very important considering the composition and temperature effects on the surface.

Previous studies reported the synthesis of nanostructured $Pb_{1-x}Sn_xSe$ in various forms using a vapor phase deposition method on silicon (Si) substrate. The effect of substrate on the $Pb_{1-x}Sn_xSe$ was studied earlier and showed to be a critical factor on the morphology during the growth of the crystalline $Pb_{1-x}Sn_xSe$ alloy [33] [17]. It was found that the substrate surface electronic structure is an important factor that affects the vapor phase deposition process and final morphology of $Pb_{1-x}Sn_xSe$ alloy.

Three-dimensional (3D) topological Weyl semimetals (TWSs) are the exotic class of quantum matter showing Weyl fermions as quasistatic excitations and Fermi arcs on the surface. Time-reversal symmetry (TRS), relativistic effects (spin-orbit coupling) and an inverted conical band structure ensures the preservation of these

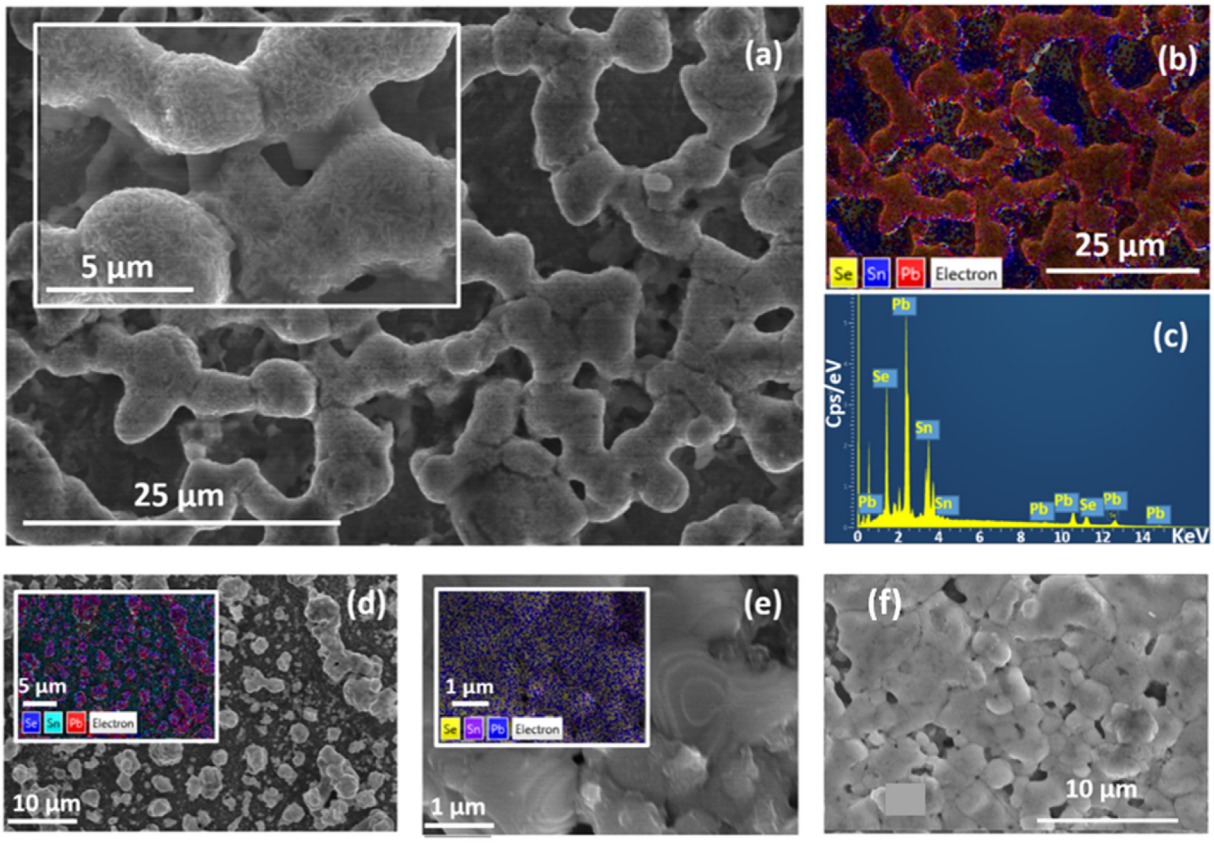

Fig. 1. SEM images and EDS maps of $Pb_{1-x}Sn_xSe$ films. (a) for Pb: Sn ratio 2.5:1) the clusters of grains can be observed which are longer than 20 μm and about ~5 μm in width; (b) associated EDS map and (c) spectrum showing the presence of Pb, Sn, Se in thin film. (d) Pb: Sn ratio is reduced to ~ 1.5: 1, a complete change in morphology was observed in SEM (EDS map was also shown in inset).; (e) Further lowering of the Pb: Sn ratio (~1.3:1) causes the formation of uniform flat grains with maximum size of ~ 6–10 μm; (f) another view of film (Pb: Sn ratio ~ 1.3: 1) as seen in SEM.