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Effect of electronic contribution on temperature-dependent thermal transport of antimony telluride thin film

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ABSTRACT

We study the theoretical and experimental characteristics of thermal transport of 100 nm and 500 nm-thick antimony telluride (Sb_2Te_3) thin films prepared by radio frequency magnetron sputtering. The thermal conductivity was measured at temperatures ranging from 20 to 300 K, using four-point-probe 3-ω method. Out-of-plane thermal conductivity of the Sb_2Te_3 thin film was much lesser in comparison to the bulk material in the entire temperature range, confirming that the phonon- and electron-boundary scattering are enhanced in thin films. Moreover, we found that the contribution of the electronic thermal conductivity (κ_e) in total thermal conductivity (κ) linearly increased up to ~77% at 300 K with increasing temperature. We theoretically analyze and explain the high contribution of electronic component of thermal conductivity towards the total thermal conductivity of the film by a modified Callaway model. Further, we find the theoretical model predictions to correspond well with the experimental results.

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1. Introduction

The efficiency of thermoelectric (TE) devices for energy conversion from heat to electricity is determined by the dimensionless figure-of-merit, ZT [1–5], which is defined as $ZT = S^2\sigma T/\kappa$, where $S^2\sigma$ is the power factor, S is the Seebeck coefficient, σ is the electrical conductivity, T is the absolute temperature, and κ is the total thermal conductivity. High-performance TE materials should therefore have high power factor and low thermal conductivity in order to obtain high ZT values [1–5]. Recently, several research groups, including ours have studied thermal transport of various one-dimensional (1D) nanostructures, such as, nanopillar [6] and nanowires [7–10] with the aim of enhancing the phonon- and electron boundary scattering. However, it is also reported that thermal properties of 1D nanostructure materials depend highly on its dimensionality and morphology, which in turn limits its application as TE devices [11,12]. Moreover, even though 1D materials have low thermal conductivity, it is known that they are not scalable and, hence, not of practical use as TE devices. In order to overcome such limitations of 1D materials, we propose the use of materials with two-dimensional (2D) structures as high-performance TE materials for scalar device applications. Further, we have recently shown that thermal conductivity of 2D magnetite (Fe_3O_4) thin-films (100, 300, and 400 nm in thickness) are ~1.7 to ~11.5 order of magnitude lower, compared to that of bulk materials at 300 K [13]. With these advantages of 2D layers, nanoporous bismuth (Bi) [14] and its alloys, including $Bi_{0.5}Sb_{1.5}Te_3$ [15], Pb-doped Bi_2Te_3 [16], and $Bi_{0.5}Sb_{1.5}Te_3$ and $Bi_2Te_{2.7}Se_{0.3}$ [17] have been proved as the interesting TE materials around room temperature.

Antimony telluride is a narrow band-gap (E_g < 0.2 eV) semiconductor with tetradymite structure (space group $R\bar{3}m$-D^5_{3d}), and is widely used in TE devices such as solid-state thermo-generators and coolers due to its good thermoelectric properties near room temperature [18–21]. In 2006, Lost'ak et al. widely studied the incorporation of Ag in the crystal lattice of Sb_2Te_3 bulk materials [20]. Recently, various deposition techniques such as molecular beam epitaxy [22], atomic layer deposition [23], and radio frequency magnetron sputtering [24] are employed to achieve high TE performance. They showed that these materials can be very useful for enhancing the TE properties for future cooling and power generation device applications [22–24].

However, extensive studies of the temperature-dependent thermal transport properties, including thermal conductivity, of this film are still lacking. Furthermore, since the total thermal conductivity consists of electronic and lattice component, it is important to investigate which component of two actually contribute

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towards the temperature dependence of the thermal conductivity for temperatures ranging from 20 to 300 K [20]. It is also worth mentioning that, there are no theoretical studies on the temperature dependence of lattice and electronic components of thermal conductivity of thin films using the Callaway model.

In this study, we report the temperature dependence of thermal conductivities of Sb_2Te_3 thin film of 100 nm and 500 nm thickness, in the range of 20–300 K, using four-point-probe 3-ω method. We then theoretically analyze the thermal conductivity of the film by a modified Callaway model, and show the high contribution of electronic component to the total thermal conductivity. Our current study on temperature-dependent thermal conductivity of Sb_2Te_3 films has three distinct features in comparison to earlier ones. First, the thermal conductivity of Sb_2Te_3 thin films is demonstrated using 3-ω technique. Second, departing from the conventional approach, the experimental data of thin films were analyzed using a modified Callaway's approach. Third, first proof for the large contribution of the electronic component compared to lattice component of thermal conductivity at room temperature is represented.

2. Experimental details

Antimony telluride thin films of 100 nm and 500 nm thickness were deposited on a SiO_2/Si (001) substrate at room temperature by RF magnetron sputtering with highly pure Sb_2Te_3 as target (99.99% purity). The base pressure of chamber was maintained below $\sim 1.2 \times 10^{-3}$ Pa prior to the deposition. RF power and working pressure for the films were set to ~30 W and 2.6×10^{-1} Pa respectively using argon (Ar) with 99.99% purity. After deposition of the films, the post annealing treatment was performed at 320 °C for 5 min under Ar atmosphere of 1.0×10^5 to 1.0×10^{-2} Pa. The detailed growth processes can be found in our previous publication [25]. The crystal structure and surface morphology of the thin film were characterized by X-ray diffraction (XRD, Rigaku O/MAX-RC), atomic force microscopy (AFM, Auto Probe CP), and by field emission scanning electron microscope (FE-SEM, SIGMA/Carl Zeiss) equipped with energy dispersive X-ray spectrometry (EDX). The thermal conductivity (κ) of the thin film was measured by four-point-probe method 3-ω technique based on the application of an alternating current (AC) with angular frequency, which was first developed by Cahill [26] in 1990. Prior to the thermal conductivity measurements, a thin SiO_2 layer (~100 nm) was deposited onto the Sb_2Te_3 thin film by plasma-enhanced chemical vapor deposition (PECVD) for electrical insulation of the films. A narrow metal strip (Ti/Au = 10/300 nm) consisting of four-point probe electrodes was then patterned onto the sample via conventional photo-lithography process. The patterned four-point-probe electrodes can act as both heater and a sensor to measure the thermal conductivity of thin films [13,26]. The thermal transport measurements were performed in the temperature range of 20–300 K in closed cycle refrigerator (CCR, Janis, USA) system equipped with turbo pump (Edward, UK). The system was electrically shielded to prevent heat loss due to convection and radiation. Fig. 1 shows the schematic diagram of the experimental setup and circuit connections of the 3-ω system for thermal conductivity measurement. In brief, the sample was first attached to a printed circuit board substrate with vacuum grease for mounting inside a CCR system. The source meter (Keithley 6221, USA) was connected to both metallic pads to flow an AC (I_0) as shown in Fig. 1. I_0 with an angular modulation frequency of 1-ω was applied to generate joule heat and temperature fluctuations at a frequency of 2-ω. The resistance of the narrow metal strip is proportional to the temperature that leads to a voltage fluctuation ($V = IR$) of 3-ω across the sample. A lock-in amplifier

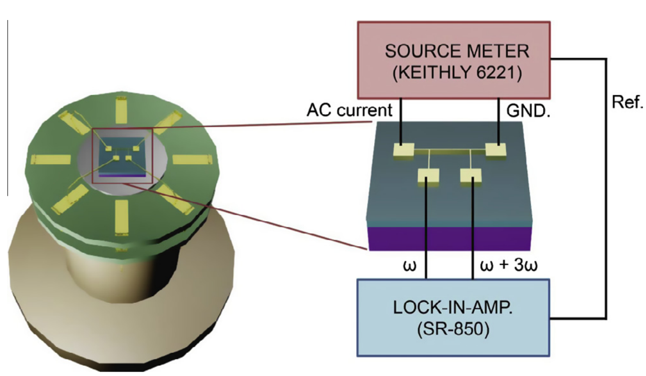

Fig. 1. Schematic diagram of the four-point-probe 3-ω method for thermal conductivity measurements at temperature range of 20–300 K.

(A-B mode, SR-850, Stanford Research System, USA) connected to the two metallic pads in the middle received the 3-ω voltage fluctuation along the narrow metal strip. To evaluate the thermal conductivity of the thin films, a plot of third-harmonic voltage ($V_{3\omega}$) against the natural logarithm of the applied frequencies ($\ln\omega$), which generally shows a linear relationship, was used [13]. Finally, the thermal conductivity can be calculated from the slopes $V_{3\omega}$ versus $\ln\omega$ in the linear region.

3. Results and discussion

Measurements to determine grain sizes, detailed material properties and thermal transport characteristics of Sb_2Te_3 film were carried out using SEM, XRD, EDX, AFM, and 3-ω techniques. Fig. 2(a)–(d) shows cross-sectional and top-view SEM images of a 500 nm-thick Sb_2Te_3 thin film annealed at 320 °C, indicating well crystalized morphology on the substrate as reported in our previous publication [25]. Further, the Fig. 2(c) and (d) show the grain boundaries of the film. To determine the grain sizes of the films from the SEM images (Fig. 2(c) and (d)), the well-known intercept method was adapted [27]. In intercept method, the average grain size (D) is given by, $D = l/n \times M$, where l is the randomly positioned line segments on the objective image, n is the average number of times each line segment intersects a grain boundary, and M is the magnification of objective image. Using this method, we determined the average grain sizes to be ~95.9 ± 2.3 nm in a 500 nm-thick Sb_2Te_3 thin film (inset of Fig. 2(d)). This information is greatly useful to theoretically calculate the total thermal conductivity of the films in modified Callaway model as we discuss in the following section. Fig. 3(a) shows typical 2-theta XRD pattern of the Sb_2Te_3 thin film annealed at 320 °C. Two clear diffraction peaks located at 34.9° and 45°, corresponding to diffraction reflections of (015) and (1010) plane of Sb_2Te_3 film can be seen in Fig. 3(a). This confirms rhombohedral structure (JCPDS No. 71-393, $R\bar{3}m$) of the Sb_2Te_3 thin film, which is also consistent with XRD pattern reported previously for the Sb_2Te_3 films [6, 25, 28, 29]. Moreover, we evaluated a stoichiometry of the 320 °C annealed Sb_2Te_3 thin film using EDX measurement. From the EDX spectrum shown in Fig. 3(b), it can be observed that the peaks of elements Sb and Te have an approximate ratio of 2:3, indicating the stoichiometry of Sb_2Te_3 film, as summarized in Fig. 3(d). In addition, the roughness of the film was examined using AFM image shown in Fig. 3(c). It exhibits smooth morphology with average roughness (R_a) of 7.83 nm and a root-mean-square roughness (R_q) of 9.89 nm as summarized in Fig. 3(d), which is in good agreement in the previous results [25].

Out-of-plane thermal conductivity measurements were made using four-point-probe 3-ω measurements in the temperature range 20–300 K in a vacuum of 5×10^{-5} Torr, which was used for both 1D nanostructures [6–10] and 2D thin films [13,30]. The thermal conductivity was calculated from the 3-ω measurements of the films, by plotting the third-harmonic voltage ($V_{3\omega}$) against the natural logarithm of the applied frequencies ($ln\ \omega$), which in turn results in a linear relationship as shown in Fig. 4(a). In the measurements, the applied frequencies usually provide a suitable current range for an estimation of the $V_{3\omega}$ signal from the sample. This shows that $V_{3\omega}$ estimates are highly dependent on $ln\ \omega$ in the frequency range of 200 to 440 Hz as shown in Fig. 4(a). Further, we verified the dependence of applied current on thermal conductivity of the film and found that the thermal conductivity is independent of the applied current (Fig. 4(a)). We finally calculated out-of-plane total thermal conductivity, from the difference between two $V_{3\omega}$ values. The thermal conductivity (κ) is given by the following equation [13,26]:

$$k = \frac{V_0^3 \cdot \ln(\frac{\omega_2}{\omega_1})}{4\pi l R_0^2 (V_{3\omega_1} - V_{3\omega_2})} \frac{dR}{dT} \quad (1)$$

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