

Limited grain growth in multilayered Bi/Te thin films and the influence on the thermal and electrical conductivity

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ABSTRACT

Bi₂Te₃ alloys were fabricated through thermal annealing of multilayered Bi/Te thin films as well as by co-sputtering of Bi and Te. In both cases, sharp X-ray diffraction peaks of Bi₂Te₃ were evidenced, indicating good crystallinity. It was found that the heterogeneous interface suppressed the grain growth in the multilayered samples considerably, and thus the thermal conductivity was reduced as a result of enhanced phonon scattering at the grain boundaries, but the electrical conductivity changed a little with temperature. However, the ZT figure (0.39) of the multilayered thin films was a little lower than that (0.43) of the single-layer ones at room temperature, suggesting the combined effect of power factor (PF) and thermal conductivity.

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1. Introduction

The worldwide energy shortage and climate crisis have increasingly become a challenge to the sustainable development of humanity [1–3]. Thermoelectric materials (TE) can convert waste heat into electricity directly and have attracted growing attention [4,5]. To make thermoelectric materials with high figure of merit, or called ZT which is defined by $ZT = S^2\sigma T/\kappa$, (where S , σ , κ , and T are the Seebeck coefficient, electrical conductivity, thermal conductivity, and absolute temperature), is highly needed [6]. Tremendous efforts have been adopted, including resonance doping, band gap engineering and nanostructure et al., to enhance the thermoelectric properties. Among them, nanostructure has been proved to be a promising and efficient way to enhance figure of merit around room temperature since the nano-structured configurations can enhance the scattering of phonons at grain boundaries and thus reduce thermal conductivity [7–11]. Various methods such as molecular-beam epitaxy (MBE) [12,13], spark plasma sintering

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(SPS) [14,15], metal organic chemical vapor deposition (MOCVD) [16,17], have been adopted to fabricate nano-structured materials for high-performance thermoelectric devices. For example, ZT value of 2.4 has been achieved in p-type Bi₂Te₃/Sb₂Te₃ superlattice thin film because of quantum confinement [18]. However, time consuming and fancy price make MBE unsuitable for commercial application. In comparison, magnetron sputtering is relatively stable and low cost. In particular, the thin films fabricated by magnetron sputtering are usually compactly textured because of the spiral motion of sputtered atoms in magnetic field and the pinning effect and it is easy to control the film thickness by setting the deposition time [19–22]. Hence, magnetron sputtering has been extensively accepted as one of the popular approaches to fabricate bismuth telluride thermoelectric thin films.

Among all the thermoelectric materials, rhombohedral bismuth telluride, a typical V–VI compound semiconductor, is widely used at room temperature [23–26]. Nevertheless, in-situ heating or post-thermal treatment is required to prepare bismuth telluride compounds during magnetron sputtering [27,28]. This certainly will induce the grain growth, and thus affect the thermoelectric properties of the thin films. From this point of view, it is meaningful to suppress the grain growth during thermal process. In this situation, Bi/Te multilayer thin films has been fabricated by sequential deposition of elemental Bi and Te layers, and exhibited Seebeck coefficient comparable to the single layer Bi₂Te₃ [29,30]. However,

multilayered Te/Bi thin film with limited grain size is still unexplored. In this paper, the Bi_2Te_3 alloys are obtained through thermal annealing of multilayered Te/Bi thin films as well as by co-sputtering of Te and Bi to study the influence of the heterogeneous interfaces on the process of grain growth as well as on the thermoelectric properties.

2. Experimental details

Co-sputtering is used to prepare Bi_2Te_3 alloy thin films, and alternative sputtering is adopted to fabricate five-layer thin films with the stacking sequence of Te/Bi/Te/Bi/Te. The thickness of each sublayer is 40 nm and the total thickness of about 200 nm, the same as that of the co-sputtered thin films. Prior to film deposition, both the substrates and the targets are ultrasonically treated in acetone and ethanol for 10 min, then cleaned by deionized water, and finally dried to remove the contaminants. The base pressure is 1×10^{-4} Pa before deposition, Ar gas flow is 3 sccm and the operating pressure is 0.3 Pa during deposition. The sputtering powers of Bi and Te targets are both 15 W. Bi sub-layer 10 nm in thickness is deposited on the surface in order to reduce the loss of Te through evaporation during heat treatment. After deposition, the samples are annealed at 423 K, 473 K, 523 K and 593 K, respectively, for 2 h to enhance inter-diffusion, to improve crystallinity and to eliminate the influence of the residual gas in thin films. The annealing process is conducted in protective Ar gas with a pressure of 50 Pa.

X-ray diffraction (XRD) (Bruker D8 Advance) using $\text{Cu K}\alpha$ radiation ($\lambda = 1.5403 \text{ \AA}$) is adopted to characterize the phase structure, and field emission scanning electron microscopy (FESEM) (Quanta x50 FEG) and transmission electron microscopy (HRTEM, JEM-2100F) with Electron Dispersive Spectrometer (EDS) are used to characterize the microstructure. The Seebeck coefficient is measured by Physical Property Measurement System (PPMS) (Quantum Design PPMS-9) and, the electrical properties are measured by Thermal Transport System of Cryogenic Limited of U.K (Mini CFM MS-5Tesla-THT). Fig. 1(a) schematically shows the measurement configuration and Fig. 1(b) presents the sample holder of testing equipment. The thin films are deposited on single-crystal silicon substrates and the natural SiO_2 insulating layer can avoid the influence of substrate on thermoelectric properties. Four Ag ($\Phi = 200 \text{ }\mu\text{m}$) wires with good electrical conductivity and excellent thermal conductivity are connected on the film samples through silver colloid. A temperature gradient between the heating and cooling sides was maintained by a heating power W. Both the

Seebeck voltage ΔV and temperature difference ΔT were measured between the $V+$ and $V-$ electrodes, and the Seebeck coefficient $\Delta V/\Delta T$ can be obtained. The measurement of in-plane thermal conductivity in thin films is really complicated. As addressed previously [31,32], the thermal conductivity is almost isotropic owing to randomly orientated grains. Therefore, the in-plane thermal conductivity in the nanocrystalline thin films could be replaced by the out-of-plane one. So we measured the out-of-plane thermal conductivity in the thin films by the 3ω method [33]. Technologically, SiO_2 layer with a thickness of 20 nm is deposited on Bi_2Te_3 thin films, then thin Ag wires are deposited as the heater and thermometer. The thermal conductance is measured by the Thin-Films Thermal Conductivity Test System (TCT – RT, Giant Technology Ltd. Wuhan, China). Generally, the measurement uncertainty comes from the contact resistance between the thin film and the metal bar, the unexpected heat loss and the system error of instruments. Before thermoelectric measurements, the contact resistance was measured by Van Der Pauw method and it exhibited an Ohmic contact between the thin films and the metal bar. So the influence of contact resistance is negligible [34]. The slope model [35] was adopted to analyze the data measured by 3ω method so that the error due to heat loss into substrate can be avoided reasonably. In addition, prior to measurements, a standard sample of 200 nm SiO_2 on Si substrate was utilized to calibrate the system error ($< 5\%$) from the instrument at room temperature. Referencing to the commonly adopted condition [32,36], all the measurements were conducted at a pressure of 10^{-3} Pa.

3. Results and discussion

The chemical composition of the thin films is analyzed by the energy dispersive X-ray (EDS). Table 1 lists the atomic compositions

Table 1

The atomic composition of bismuth and tellurium in the thin films annealed at different temperatures.

	Single - layer		Multilayer	
	Atomic (%) Bi	Atomic (%) Te	Atomic (%) Bi	Atomic (%) Te
As-deposit	36.05	63.95	39.65	60.35
423 K	37.08	62.92	38.93	61.07
473 K	38.97	61.03	40.49	59.51
523 K	41.84	58.16	40.24	59.76
573 K	42.50	57.50	40.79	59.21

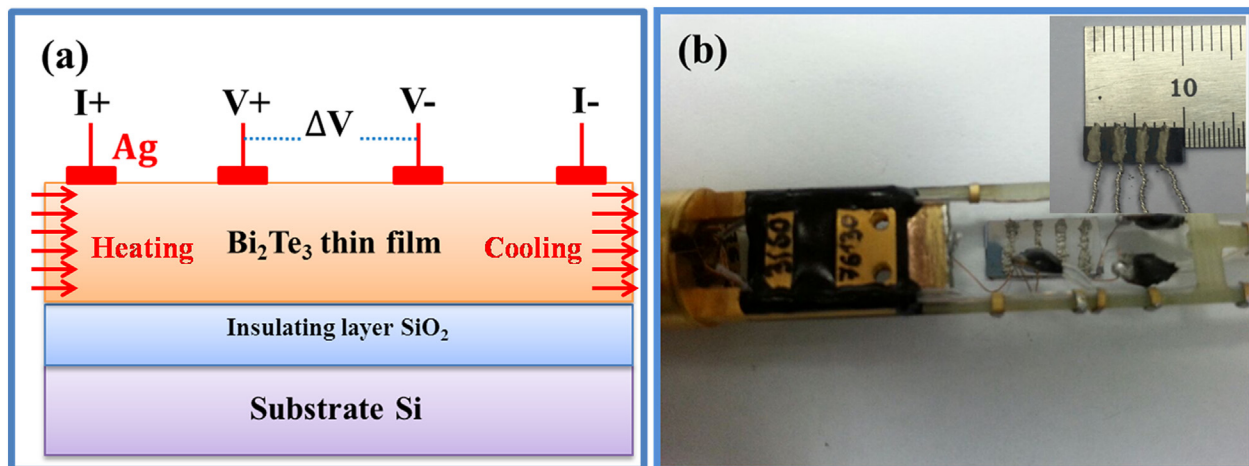


Fig. 1. (a) A schematic cross-section image of the thermoelectric device for measurement of Seebeck coefficient; (b) a device for thermoelectric measurement of Bi_2Te_3 thin films.

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