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The investigation of thermal properties on multilayer Sb_2Te_3/Au thermoelectric material system with ultra-thin Au interlayers

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ABSTRACT

The manipulation of heat transport across multilayer thin films with metal-semicounductor interfaces is of great interest for thermoelectric material optimization. Here we fabricated Sb_2Te_3/Au multilayer films with different Au thickness by magnetron sputtering. We demonstrated that the thermal conductivity of the system can be facilely manipulated by simply changing the Au layer thickness, where an optimal thickness (5 nm) value exists with the lowest thermal conductivity (~0.44 $Wm^{-1}K^{-1}$, 44% of the pure Sb_2Te_3 thin film thermal conductivity). It has been proved that the decreased thermal conductivity was mainly attributed to the strong electron–phonon coupling in a metal-nonmetal multilayered system with Au layer thickness larger than 5 nm, where the Two Temperature Model (TTM) predicts the experimental data perfectly. It was also proposed that the grain boundary effect may dominiate the phonon scattering when the Au layer is in a discountinuous form (<5 nm).

1. Introduction

Thermoelectric materials can convert heat into electricity without moving part and operate over a large range of temperatures. Therefore, it shows huge application potential for the next-generation electricity generator and thermoelectric cooler [1–4]. Antimony telluride (Sb_2Te_3) and their alloys have been extensively studied due to their promising effect in the range near the room temperature [5]. It is well-known that the efficiency of thermoelectric materials is characterized by the dimensionless figure of merit ZT ($ZT = \sigma S^2/\kappa$), where σ is electric conductivity, S the Seebeck coefficient and κ the thermal conductivity, respectively [6]. In the past decades, tremendous effort has been dedicated to the power factor (σS^2) optimization [7,8]. More recently, manipulation of thermal conductivity of the TE materials aroused the researchers' interest [9–12]. The existence of traditional Sb_2Te_3 material is in the form of bulk structure, which limits its thermoelectric conversion efficiency [13]. A large number of researches have shown that by introducing nanostructure (e.g. nanowires [14,15], multilayers

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[16,17], nano-composites [18,19]), the thermoelectric efficiency of the materials is expected to enhance. Because of the exist of interfaces in multilayer structure, the thermal conductivity in this structure will have a very small value [20,21]. which because of the interface scattering of phonons is the considered to be the major contributor [11].

Specifically, the control of heat transport across multilayer films with inserting metal layers is of fundamental interest as an important method for thermal conductivity manipulation. In this paper, we report on the fabrication of nano-scale Sb_2Te_3/Au multilayers (ML) thin films by alternate depositing solid antimony telluride and gold onto a Si (100) substrate via magnetron sputtering method. We control the thickness of the antimony telluride to a constant value about 13 nm per layer, and change the thickness of the gold from 1 nm to 20 nm per layer. Samples with different thickness of gold layer in 10 periods were obtained successfully. The thermal conductivity of multilayer films is investigated according to classic heat conductivity model (CHCM) and two-temperature model (TTM) A new physical model is proposed to analyze the thermal conductivity when the Au less than 5 nm.

2. Experimental section

The multilayer films consisting of thin layers of Au and Sb_2Te_3 were prepared by alternate sputtering of the two components onto a silicon wafer in a high vacuum magnetron sputtering (Kurt J.Lesker, PVD 75) system at room temperature. The targets were Sb_2Te_3 (99.999% purity) and Au (99.999% purity). Before sputtering the substrates were thoroughly cleaned we selected the N<100> type high resistance (5000 Ω cm) silicon wafer as substrate. It was socked in B.O.E solution (HF: NH4F = 1: 6) for 5 min to etch off the surface oxide layer, then silicon wafer was ultrasonically cleaned in acetone and ethanol for 10 min respectively. After cleaning, this wafer was dried with nitrogen gas and baked in 80 °C air oven for 15 min. Then we transferred it into the sputtering chamber and fixed on the holder. The distance between the substrate and targets was 20 cm. The base pressure was 3.5×10^{-7} Torr and the sputtering pressure was 3 mTorr. The substrate rotated at a speed of 20 rpm to ensure the deposition uniformity. The deposition was performed at a power level of 20 W in radio frequency (RF) mode for Sb_2Te_3, and at 25 W in direct current (DC) mode for Au, respectively. The deposition rate of Sb_2Te_3 was ~0.375 Å/s while that of Au was ~0.48 Å/s calibrating by a quartz crystal monitor. The thicknesses of Au layer were designed to be 0 nm, 1 nm, 3 nm, 5 nm, 10 nm, 20 nm and that of Sb_2Te_3 was kept to a fixed value of 13 nm. The description [Sb_2Te_3/Au (x nm)] is designated as the multilayer sample with different Au thickness.

The differential 3ω method is often employed in the thermal conductivity characterization of film using a simple one-dimensional heat conduction analysis to determine the thermal conductivity of the thin film. The details of thermal conductivity measurement can be found elsewhere (reference). At room temperature the accuracy of this method can approach ±10% [22]. In this method, a thin silver wire with dimensions of 2 mm (length) × 20 μm (width) × 200 nm (thickness) is deposited through micro-fabrication process to be used simultaneously as a heater and thermometer (Fig. 1). In order to promote the adhesion between the film and the heater line, we recommended titanium as the connection layer. Since the multilayer films are electrically conductive, the silver wire must be insulated from the film to avoid current leakage. The electrical insulation is provided by ~500 nm SiO_2 film deposited electron beam evaporation. In order to avoid the error produced by other factors, we also deposited a reference sample without the under testing film (a silicon with the same pre-processing, insulating layer and heater line). The current at a certain frequency of ω was produced by a Keithley 6221 source of voltage was extracted through a Stanford Research 830 lock-in amplifier within the range of 100–1000 Hz.

3. Results and disscussion

3.1. Periodic multilayer film structure characterization

The cross-plane structure of the MLs was examined using a Zeiss ULTRA55-36-69 SEM. Fig. 2 (a) shows the schematic model of the samples. Fig. 2b–d show the cross-section SEM images of Sb_2Te_3/Au multilayer films. It can be observed that the multilayer films exhibit a well-defined periodic layered nanostructure.

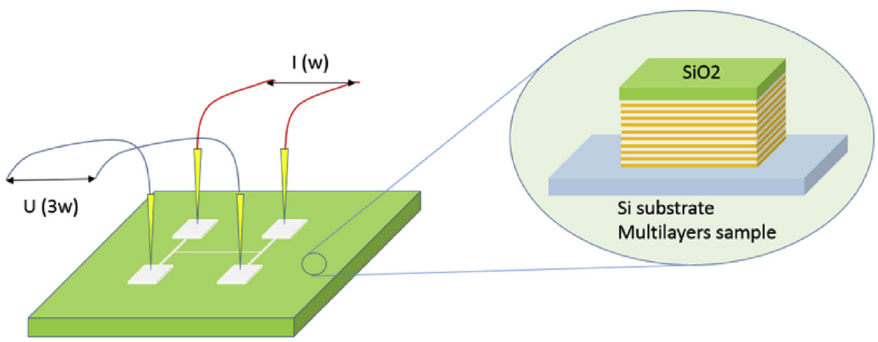

Fig. 1. Schematic of the differential 3ω method.