



Power factor investigation of RF magnetron sputtered c-GeSbTe thin film



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ABSTRACT

Germanium antimony tellurium (GeSbTe) thin film has been deposited by the RF magnetron sputtering from the initial material target of the 1:1:1 atomic ratio. The GeSbTe thin film was annealed by furnace at 473 K, 523 K, 573 K and 623 K samples for 1 h under ultra-high vacuum. The samples were analyzed by X-ray diffraction (XRD), field-emission scanning electron microscopy (SEM), and energy dispersive spectroscopy (EDS) to study their structure and chemical composition. The carrier concentration, charge mobility, electrical, Seebeck coefficient and power factor are reported. The as-deposited thin film showed amorphous and consequently cubic structure (c-GeSbTe) after annealing treatments. In addition, the film thickness was rapidly decreased with increasing annealing temperature. The as-deposited thin film had the atomic ratio of 1:0.6:0.7, and after annealing treatments became 1:0.9:0.9. The c-GeSbTe thin film annealed at 523 K showed the highest mobility, lowest electrical resistivity, and the highest power factor of $8.31 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$, $3.25 \times 10^{-5} \text{ } \Omega \text{ m}$, and $0.81 \times 10^{-4} \text{ W m}^{-1} \text{ K}^{-2}$, respectively.

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1. Introduction

Thermoelectric thin films are potential candidates for applications in microelectronics, for example, temperature sensors, micro-coolers, and micro-power generators [1–2]. The efficiency of thermoelectric materials is to produce thermoelectric power that heavily relies on a dimensionless figure of merit as the following:

$$ZT = \frac{S^2 T}{\rho \kappa}, \quad (1)$$

where S , T , ρ , and κ are the Seebeck coefficient, absolute temperature, electrical resistivity, and thermal conductivity, respectively. The thermoelectric performance has been improved by increasing the Seebeck coefficient, and decreasing electrical resistivity and thermal conductivity [3–4]. From previous publications, the thermal conductivity is said to be reduced by a phonon scattering,

i.e., a mass fluctuation scattering, a crystallite boundary scattering, and an interface scattering in thin films or multilayer systems [5]. In addition, the lowest electrical resistivity and thermal conductivity, phase change materials, i.e., Germanium antimony tellurium (Ge–Sb–Te) system have also become the thermoelectric materials of interest [6]. Moreover, in modern technology, Ge–Sb–Te chalcogenide ternary compounds are widely used because of their stable phase transformation between amorphous and crystalline phases [7]. The Ge–Sb–Te system has studied the several possible compositions of GeSb_2Te_4 , GeSb_4Te_7 , and $\text{Ge}_2\text{Sb}_2\text{Te}_5$ shown in a Ge–Sb–Te phase diagram [8–9]. The $\text{Ge}_2\text{Sb}_2\text{Te}_5$ has been the preferred thermoelectric material with high Seebeck coefficient and low electrical resistivity [10–11]. Nevertheless, other GeSbTe compositions, for example, Sb_2Te_3 –GeTe and $\text{Ge}_x\text{Sb}_y\text{Te}_{1-(x+y)}$ are also promising materials for thermoelectric applications [8], but have not been fully explored.

The aim of this work is to deposit the GeSbTe thin film by a radio frequency (RF) magnetron sputtering from the initial GeSbTe material of a 1:1:1 atomic ratio and annealing treatments at difference temperature. In particular, we report on the effects of the annealing treatments to the crystalline structures, physical morphologies, atomic compositions and thermoelectric properties.

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2. Material and methods

The GeSbTe thin film was deposited using RF magnetron sputtering (AJA International, Inc., ATC 2000-F). A Ge:Sb:Te material (commercial, Mercantile Hi-Tech Co., Ltd.) at 1:1:1 atomic ratio with 99.99% purity, 0.635 cm thickness, and 5.08 cm diameter was used as a sputtering target. Silicon wafers with 1- μm thick silicon dioxide were used as substrates. The substrates were pre-cleaned by sonication method, and then with plasma treatment for 15 min in a vacuum state. During the film deposition, the GeSbTe thin films were prepared by the deposition conditions as shown in Table 1, at ambient temperature. After the film deposition, the as-deposited GeSbTe thin films were annealed under ultra-high vacuum at 473 K, 523 K, 573 K, and 623 K, each for 1 h. The obtained GeSbTe thin films were investigated by several instrumentation techniques. First, their phase identifications were analyzed by X-ray diffraction (Shimadzu, XRD6100). Physical morphologies, i.e., surface topologies, cross sections, and thickness (d), were investigated by high-resolution scanning electron microscopy (SEM) with an acceleration voltage of 200 kV (JEOL, JSM-5410). Their carrier concentration (n) and charge mobility (μ) were measured by Hall Effect measurements (Ecopia, HMS-3000) based on van der Pauw four-point probe method at room temperature. The electrical resistivity was determined from the n and μ values. Finally, the Seebeck coefficients were measured by a steady state method at room temperature. The power factor (P) was calculated from electrical resistivity and the Seebeck coefficient to represent thermoelectric performance of the obtained films.

3. Results and discussion

Results from XRD patterns of the as-deposited and annealed GeSbTe thin films are shown in Fig. 1. The as-deposited thin film shows in an amorphous structure while the annealed film shows in a cubic structure (c-GeSbTe), as confirmed by major XRD peaks of (111), (200), (222), and (220) planes according to PDF# 054-0484 data [12]. These XRD peaks have been demonstrated in the crystallinity and the crystal structure of intermediate states during the amorphous-crystalline solid-state transformation within sequentially annealing temperature [7–11,13]. The crystallite sizes (D), lattice strains (ε), and lattice parameters of c-GeSbTe thin films were evaluated from a width at half maximum (FWHM) of XRD patterns, based on the Scherrer's equation as the following:

$$D = \frac{K\lambda}{\beta \cos\theta}, \quad (2)$$

$$\varepsilon = \frac{\beta}{4 \tan\theta}. \quad (3)$$

where K , λ , and θ are a dimensionless shape factor, the wavelength of the $\text{CuK}\alpha$ radiation, and the Bragg angle, respectively. The annealing treatments were effected to the crystallite sizes and the lattice strains of the GeSbTe thin films according to the (200) peaks, as seen in Table 2. The results indicated the highest crystallite size and the smallest lattice strains from the annealing temperature of 573 K. The calculated

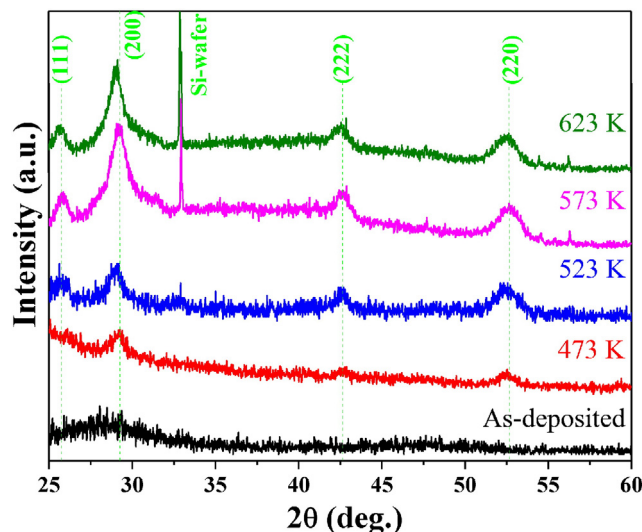


Fig. 1. XRD pattern of as-deposited and annealed GeSbTe thin film.

lattice parameter of the thin films, based on Bragg's law is 6.038 Å, and was found to be independent to the annealing temperature, similar to the other publication [12].

The top-view and cross-sectional SEM images of the GeSbTe thin film are shown in Fig. 2. In Fig. 2(a), the as-deposited thin film showed a smooth surface, which is generally associated with amorphousness, as previously confirmed by XRD. In Fig. 2 (b)–(e), in the annealing treatment in thin films gradual changes of the surface topologies are clearly observed, where the surface roughness and porosity of the films were increased. The cross-sectional images show the as-deposited thin film with smooth grain structure and a strong interface bonding between the nucleation growth and the SiO_2 film on the silicon substrate, as shown in Fig. 2 (f) [14]. The thin film porosity was considerably observed while the interface bonding instead reduced at annealing treatments, as shown in Fig. 2 (g)–(j). In addition, the annealing treatments also strongly resulted in the decrease of the film thickness from 375 nm (as-deposited) to 230 nm (623 K annealed). These results were associated with a re-evaporation process in the film material and the change in the film composition, because of the annealing. In Fig. 3, the observed film thicknesses were plotted along with an exponential fit to predict the film re-evaporation, as affected from the annealing treatments to the as-deposited GeSbTe thin film.

The EDS spectra represented relative atomic compositions of Ge, Sb, and Te in the as-deposited and annealed thin films are shown in Fig. 4. Their percentage ratios of the atomic composition of Ge:Sb:Te are 43:27:30 (as-deposited), 38:31:31 (523 K annealed), and 36:33:31 (623 K annealed), and plotted on a ternary atomic ratio diagram, as seen in Fig. 5. We recalled that the initial GeSbTe target obtained the Ge:Sb:Te atomic ratio of 1:1:1. The normalized atomic ratio of the as-deposited film shows approximately 1.0:0.6:0.7, which is a result of different sputtering yields of the Ge, Sb, and Te materials, as seen in Fig. 5. Such different sputtering yields of different materials therefore

Table 1
Deposition conditions of GeSbTe thin film.

Base pressure (Pa)	3.20×10^{-5}
Operating pressure (Pa)	0.67
Ar flow rate (sccm)	30
DC power (W)	50
Substrate temperature (K)	300
Deposition time (min)	60
Substrates	SiO_2/Si wafer
Annealing temperature (K)	473, 523, 573 and 623
Annealing time (min)	60

Table 2
Crystallographic data of GeSbTe thin film.

Sample	200 peak (2θ)	FWHM (2θ)	Crystallite size, D (nm)	Lattice strain, ε (%)
Ref. [12]	29.570			
473 K	29.057	1.192	14	1.00
523 K	28.993	0.965	17	0.81
573 K	29.054	0.838	20	0.71
623 K	29.211	0.981	17	0.82

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