Materials Science in Semiconductor Processing 57 (2017) 210–219

Contents lists available at ScienceDirect

Materials Science in Semiconductor Processing

journal homepage: www.elsevier.com/locate/mssp

Characterization and optical properties of bismuth chalcogenide films prepared by pulsed laser deposition technique

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ARTICLE INFO

Keywords:
Chalcogenides
Thin films
Optical characteristics

ABSTRACT

Thin films of Bi-based chalcogenides were prepared by pulsed laser deposition (PLD) technique according to the stoichiometric formula: $Bi_2(Se_{1-x}Te_x)_3$. Their optical properties were studied aiming to find the suitable area of application and the optimum composition amongst the samples under study. X-ray diffraction analysis proved the crystallinity of the deposited samples; in addition, surface roughness and films homogeneity were studied by atomic force microscopy (AFM) confirming the suitability of PLD technique to prepare homogenous and smooth films of the concerned alloys. Absorption coefficient calculations showed higher absorption values of 5×10^5 and 6×10^5 cm^{-1} for Te contents of 90% and 100% in the $Bi_2(Se_{1-x}Te_x)_3$ system respectively. Optical band gap of the concerned films were calculated and found to be in the range of 0.76–1.11 eV, exhibiting comparable values with the previously reported by other authors. Optical studies conformed direct and allowed transitions in all films. Refractive index (n) and dielectric constants (ε_r) and (ε_i) were calculated and studied as a function of the wavelength. Values and behavior of (n), (ε_r) and (ε_i) indicated strong dependence on the composition and the wavelength range.

1. Introduction

Bismuth Selenide (Bi_2Se_3) system is one of the best known topological insulators because it has a gapless single Dirac cone and a bulk band-gap that is larger than other comparable materials. Selenium (Se) vacancies or antisites defects are usually formed when this material is grown, such kind of defects serve as donors and further shift the Fermi energy considerably above the band gap [1]. The compound (Bi_2Se_3) is quite often used in thermoelectric generators, it is an exceptionally good electrical conductor–as good as gold and is transparent to infrared light as well.

On the other hand, Bi_2Te_3 is one of the most state-of-the art efficient materials working near room temperature; in addition, Bi-Te based alloys are just suited for many applications involving small temperature difference, such as harvesters operating between room and human skin temperatures. Accordingly, lots of applications were benefited from such small power, for instance, pacemakers and blood pressure regulators [2]. Furthermore, Bi_2Te_3 based alloys find applications in microelectronics, optoelectronics and electromechanical devices, such as photoconductive targets in TV cameras, IR spectroscopy, IR detectors sensors and memory devices [3–6].

The chalcogenide system $Bi_2Se_{3-x}Te_x$ has long been the subject of a number of investigations in the present time because of the possible applications of these alloys, in particular, in thermoelectric devices, since defects can be obtained by Te doping in Bi_2Se_3.

Using pulsed laser deposition (PLD) to synthesize thin films such as thermoelectric Bi-Chalcogenide materials received only little attention for the preparation of thermoelectric materials. However, the technique allows growing high quality films at lower deposition temperature than any other technique which in turn enables to restore quite easily the stoichiometry of the target [7].

As reported in [8], the thermoelectric performances can be improved through a suitable modification of electron and phonon transport mechanisms which is strongly predicted for low dimensional or nanostructured materials, however, this requires a control of the material structure down to the nanoscale. In [8] they show that pulsed laser deposition provides a good control on the film composition, phase and structure, necessary for a comprehension of the relationship between structure and thermoelectric properties. In other words, the engineering of tailored Bi_2Te_3 thin films with improved thermoelectric properties can be achieved.

The present work aims to prepare thermoelectric thin Bi-chalco-

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http://dx.doi.org/10.1016/j.mssp.2016.10.043
Received 5 July 2016; Received in revised form 18 October 2016; Accepted 21 October 2016
Available online 29 October 2016
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genide films by PLD technique and investigate their optical parameters at different contents of the chalcogenide elements: Se and Te. Bi_2Se_3 and Bi_2Te_3 binary system were studied as well, since tellurium is substituted for selenium in the molecular formula $Bi_2(Se_{1-x}Te_x)_3$, until $Bi_2Se_3$is transformed into Bi_2Te_3.

2. Experimental technique

Thin films the $Bi_2(Se_{1-x}Te_x)_3$ system were synthesized using pulsed laser deposition technique (PLD). Target materials are previously prepared bulk crystalline alloys, prepared by melting method for pure elements, 5N pure (99.999%). PLD was performed with a KrF* excimer laser source (λ=248 nm, τ_{FWHM}=25 ns) operating at a repetition rate of 2 Hz. The incident laser fluence was 3.3 J/cm^2. The targets were prepared from previously synthesized bulk materials. The distance target – substrate was 3 cm. The working pressure was maintained at 4×10^{-4} Pa. A total number of 1000 pulses were applied on the target for the deposition of each layer.

According to studies [9], significant improvements in terms of the properties of the deposited films based on Bi_2Te_3 were resulted from using PLD as a preparation technique. Interestingly enough; the first attempt to produced Bi_2Te_3 thin films via pulsed laser deposition was reported in 1996. In this work, it was found that, the deposited film is Te-deficient at positions close to the incoming laser, as the laser interacts with the plasma plume coming off the target [10].

The thickness of the deposited films was around 2500 Å. Thin films have been characterized by the X-ray diffraction technique, energy dispersive analysis and atomic force microscopy; the transmission spectra of the thin films in the spectral range 400–2700 nm were obtained using a double beam ultraviolet–visible–near infrared spectrophotometer V-670.

3. Results and discussion

3.1. X-ray diffraction (XRD) analysis

X-ray diffraction analysis was carried out to study the crystal structure of $Bi_2(Se_{1-x}Te_x)_3$ films at x=0.00, 0.50 and 1.00. Fig. 1 declares the XRD diffractograms of the PLD synthesized films; the three samples of the concerned films are polycrystalline with different positions for the main peak, which reflects different direction for the preferable orientation of atoms constituting each film which in turn refers to the compositional dependence of the structure on the film's constituents. (01,11), (10,10) and (220) were the preferable direction of atoms orientation for the samples X=0.00, X=0.50 and X=1.00 respectively. Furthermore, it is noteworthy mentioning that the films are not purely single phased of Bi_2Se_3 or Bi_2Te_3, since other phases such as BiTe and Bi_3Te_4 were detected. As in [1], Bi_2Te_3 thin films were deposited by pulsed laser deposition technique, a polyhedral (PH) structure that was composed of 3D triangular and polygonal crystals was observed, and the PH exhibited a diminished density because of the presence of microvoids between crystals. the PH film possessed Bi_4Te_5 phase (JCPDS 22-0115), which was associated with a composition of approximately 51.5 at% Te. However, the degree of purity of the material obtained is relatively high. Thus, the recorded XRD patterns do not show peaks due to the various forms of Bi_2O_3 or bismuth selenium/telluride oxide (Bi_2O_5Se/Te) that could be also formed during the deposition process.

It is worth mentioning here that the structure is significantly dependent on the Te content in the $Bi_2(Se_{1-x}Te_x)_3$ host alloy, furthermore, the binary Bi_2Se_3 sample shows a completely different crystal structure which may result in different optical properties of Bi_2Se_3 from the other samples. Regarding the sample containing 50% Bi_2Se_3-50% Bi_2Se_3, we cannot observe evidence of any phase transformations in this system as all available evidences refer to the existence of a continuous solid solution range between Bi_2Se_3 and Bi_2Te_3 [11]. In other words $Bi_2(Se_{1-x}Te_x)_3$ is a pseudo-binary system, as the distribution coefficient of bismuth is unity [12].

Generally, the XRD peaks are shifted to lower two-theta values indicating that the Te ions are inserted into the Bi_2Se_3 hosting lattice.

As shown in Fig. 2, a notable left shift in the position of the (015) peak (a common peak in the three samples) can be noticed as a result of Te ions incorporation into the Bi_2Se_3 lattice, indicates the compositional dependence of the crystal structure of the samples under the study. Accordingly, impact of Te/Se ratio's variation in the $Bi_2(Se_{1-x}Te_x)_3$ system on the concerned optical properties is expected to be strong.

From X-ray diffraction pattern for maximum intensity peak, the crystallite size (D) of the PLD prepared thin films was calculated using Scherrer's equation:

$$D = \frac{K\lambda}{\beta \cos\theta}$$

Since, D is the grain size, λ is the X-ray wavelength, k is a dimensionless parameter known as shape factor, β is the line broadening at half maximum of intensity and θ is the Bragg angle.

Very small sizes of grains were induced from Scherrer's equation and listed in Table 1, referring to smooth surface and nano-structure nature of the concerned films.

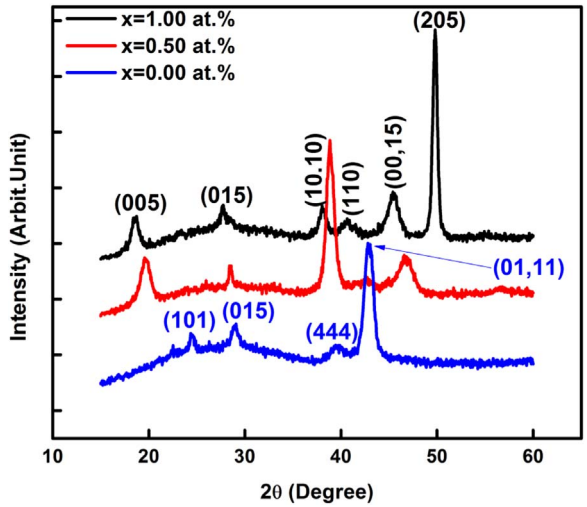

Fig. 1. XRD patterns of $Bi_2(Se_{1-x}Te_x)_3$ thin films prepared by PLD.

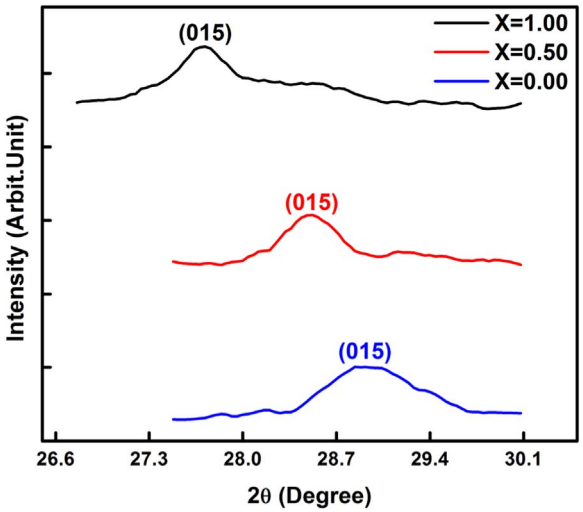

Fig. 2. Compositional dependence of the peak (015) position.

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