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INVESTIGATION THE IMPACT OF DIFFERENT ANNEALING TEMPERATURES ON STRUCTURAL AND OPTICAL PROPERTIES OF BI-DOPED GETE (BI: 5 %) THIN FILMS FABRICATED BY PVD TECHNIQUE)

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ABSTRACT:

The impact of different annealing temperatures on the Bi-doped GeTe thin films were investigated. The thin films have been prepared by using physical vapor deposition techniques (PVD). The Bismuth (Bi) is doped GeTe with ratio of 5%. Different characterizations techniques have been used to study the different properties of thin films with several annealing temperatures. It has been found that the film thickness decreases as the annealing temperature increases. The XRD patterns show that as-deposited and annealed Bi-doped GeTe films at 150°C,200°C, and 250°C were fully amorphous, while the film annealed at 100 °C was crystalline. FESEM image shows that the structure is amorphous with no grain appearing for the crystallite GeTe compound. Whereas the annealed thin films at 100°C are well-appeared crystallites of GeTe with an average size of (110.64 nm). The thin films are annealed at (150, 200, and 250)°C which reveals that the crystallite or grain is increased. An increase in the annealing temperature has been found to cause a significant shift in the absorption edge toward an extended wavelength and an overall reduction in transmittance. At a wavelength of 1100 nm, the transmittance dropped from 65.25% for as-deposited thin films to 32.57% for annealed thin films at 250 °C. Furthermore, when the annealing temperature rises from 100°C to 250°C, the optical band gap reduces from 0.95 eV to 0.42 eV.

KEYWORDS: Germanium Telluride, Annealing Temperature, Doping, Optical properties, Bismuth .

1. NTRODUCTION

Because of their exceptional qualities, phase-change materials (PCM) continue to be interesting materials for permanent memories optical storage systems. They are also still promising materials for storage class memory (SCMs) and embedded memories in servers with high performance that require a lot of energy. One of those most intriguing media for rewritable usage, such as rewritable optical discs, is the electrical switching that PCM offers [1-3]. Two main issues need to be resolved in order to compete with popular NVM technologies and offer superior efficiency and numerous dependable devices in a variety of memory applications such as first, the memory density of the devices must be raised through reducing the current needed for RESET, and second, the data preservation capability of PCM must be improved by improving the long-term reliability of the state that is amorphous. Therefore, lowering the RESET current is necessary to improve PCM effectiveness, which also facilitates the use of PCMs in any given application. Though many attempts have been attempted to come up with novel compounds to improve the extreme temperatures stability of phase-change technology, it is sufficient for uses in commerce [3].

When Ovshinsky proposed a switch memory in the 1960s, centered around the differences between the crystalline and amorphous phases of multicomponent chalcogenide, the concept of transitioning from an amorphous to a crystalline phase was implemented to store information back then [4,5]. Because of their greater resistance, much greater crystallization temperature

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(Tc), and quicker rate of crystallization, ternary alloys, in particular, on the time line connecting germanium telluride

(GeTe), are a good substitute for germanium antimony telluride (GST) in PCM [6]. Because of their exceptional endurance and quick switching, GeTe compositions are the most dependable and promising for the aforementioned goals [1,4,7]. In addition, germanium telluride (GeTe) has garnered a lot of attention from the industrial and educational communities due to its intriguing electronic and optical properties and adaptable phase shift character [6, 8-11].

However, given that material must be heated to a temperature greater than its Tc for an extended period of time in order for adequate crystallization to occur, the transformation of the amorphous phase to the stable crystalline phase. Melting crystalline material and quickly quenching it afterward solidifies the amorphous phase in this irreversible process. The quenching process is a power-limited operation since sufficient power is needed to raise the material's melting temperature (Tm). In general, PCM's Tm ranges from 500 to 800 ◦C [12] .

The components of Germanium (Ge) and Tellurium (Te) have been organized in an intriguing and sizable initial prototype of the IV and VI groups (4 and 6) of the group's binary alloys family members [13, 14]. Phase shifting memory substances are based on the two components Ge and Te of various alloys that rapidly crystallize from their amorphous state. These sorts of materials are used in industries such as digital versatile disks (DVDs) and memory with random access (RAM) [15-17]. The Curie temperature (TC) of GeTe, an acknowledged ferroelectric substance, is 670 K, strongly dependent on the purity of the sample [18]. GeTe's crystal structure revealed a couple modifications: a low-temperature rhombohedral phase that initially was investigated by Schubert and Fricke, and a high-temperature cubic phase of the NaCl form [18,19].

GeTe has a wide range of the characteristics that include both basic and technological standpoints, making it valuable. However, GeTe is capable of being irreversibly transformed between two phases such as crystalline and amorphous, which can cause significant modifications in its optical and electrical characteristics [20-22]. For instance, substances suitable for this type of application should have high Tc. Thermal reliability is necessary in manufacturing such as automobiles where memory chips are used to locate the region of warm engines. Adding additional elements to the amorphous phase is one method of modifying its Tc. Numerous researchers used this method, adding additional elements to GeTe to raise the Tc. The structure of the material determines its properties, and doping may alter it. Therefore, it is imperative to examine the impact of doping on the structure of GeTe. Because of its straightforward structure and the substance, Bulk GeTe has long been used as a phase-change material prototype [21]. In addition, there are additionally a number of problems with practical applications. These concerns primarily relate to power consumption, amorphous state, and the resistivity contrast of radio frequency equipment. An additional approach to resolving this issue is the doping of elements in GeTe.

Additionally, there are a few explanations for why doping in GeTe is necessary. The first explanation is that adding additional impurities to the welcome, such as nitrogen (N), carbon (C), oxygen (O), SiOx, SiCx, and SiNx, reduced the RESET current in numerous experiments [6, 23, 24]. PCM has minimal heat loss because of its low conductive incorporating. Doping the contaminants raises the substance's resistance to electricity. The additional evidence is that dopant furtherance enhances retention of information performance for up to ten years of fail temperature. The dopant puts together its products in a troubled configuration in PCMs to stack at the grain boundaries, which retains the crystalline grains expanding huge, which is a supporting argument for doping in GeTe for keeping information [25].

However, there are additionally a number of problems with practical applications. These concerns primarily relate to power utilization, amorphous state, and the resistivity contrast of radio frequency devices. In GeTe, doping is one way to address this issue. Bi is just one of the substances that has been demonstrated to be able to alter the type of conductivity in glassy chalcogenide semiconductors (GCS) [26]. Furthermore, it was demonstrated that Bi doping in GeTe resulted in a crystallization time reduction of about 30% as opposed to the undoped one. However, there has not been much research done on how Bi doping affects the characteristics of GeTe thin films [26].

The goal of this work is to synthesize Bi-doped GeTe thin films, which have been prepared in a few studies involving the doping with Bi using the PVD technique. Additionally, research has been done and results obtained regarding the effects of various annealing temperatures on the structural and optical properties of as-deposited and Bi doped GeTe thin films.

2. MATERIALS AND METHODS

Each of the substances used in the current investigation, including powdered germanium (Ge), tellurium (Te), and

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bismuth (Bi), came from the Sigma-Aldrich company and did not require any additional sterilization.

2.1. Cleaning process

A minuscule glass, measuring 25x75x1 mm, was utilized as the substrate for thin-film the process of deposition The substrates are first cleaned by submerging them in chromic acid for a full day. They were subsequently rinsed in acetone and cleaned with water that was distilled. Lastly, they were ultrasonically cleaned with distilled water and stored for later use in an evacuated desiccator.

2.2. Deposition of Bi-doped GeTe thin films

In order to create Bi-doped GeTe thin films, the mixture was first made by mixing Ge and Te powders 1:1 based on their respective atomic weights. As a doping element, 5% bismuth was added to the combination. To create a mixture that was uniform, the GeTe and doped element (Bi) mixture was shaken inside a tiny ampoule. Using the Physical Vapor Deposition Technique (PVD), bi-doped GeTe thin films were deposited in an ultra-high vacuum (~ 2x10-5 mbar). The mixtures were put into boats made of molybdenum. Next, a revolving substrate the holders held 12 fixed substrates. At room temperature, all thin films were geared up.

2.3. Annealing of Bi-doped GeTe thin films

Bi-doped GeTe thin films were deposited and then vacuum-annealed for an hour at various annealing temperatures (100, 150, 200, and 250 degrees Celsius) in a tubular furnace. This was done to investigate the impact of annealing temperature on the optical and structural characteristics of the thin films that were produced.

2.4. Thickness Measurement of Bi-doped GeTe thin films

Optical strategy was used to measure the thickness of as-deposited and annealed Bi-doped GeTe thin films, as schematically illustrated in Figure (1). The formula that follows was used to get the film's thickness (t) [27-29]:

$$t = \frac{\lambda}{2}.\frac{\Delta x}{x} \qquad (1)$$

where x is the fringe width, Δx is the distance between two fringes and λ is the wavelength of the laser beam.

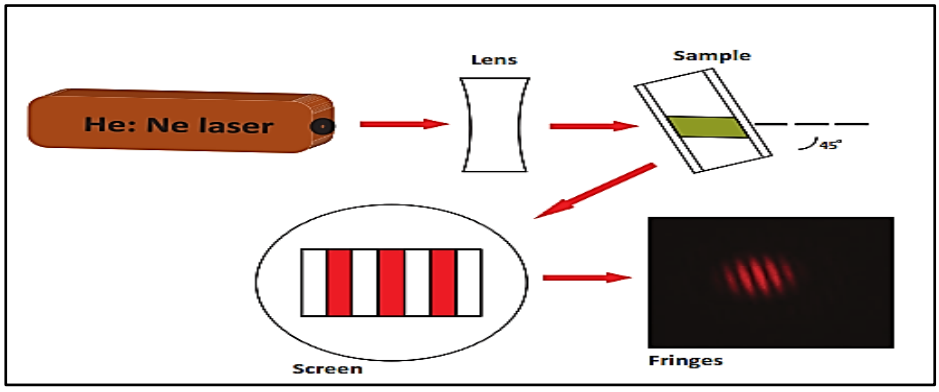

Figure 1: The diagram of the film thickness measurement [29]

2.5. Characterization of the Bi-doped GeTe thin films

The X-Pert Pro PAN analytical X-ray diffraction system with CuKα radiation (1.5406 A°) in the 2θ range (10º–80º) and ZEISS scanning electron microscopy (SEM) were used to characterize the as-deposited and annealed Bi-doped GeTe thin films, respectively. The as-deposited and bi-doped GeTe thin-film optical transmission examinations have been taken in the 200-1100 nm range using a UV/Vis spectrophotometer (JANEWAY 6850). Using the following Tauc equation, the energy bandgap of the as-deposited and bi-doped GeTe thin film was computed.

$$(\alpha h\nu)^2 = B(h\nu - Eg)^n \qquad (2)$$

Where n reflects which kind of transition (equals to 1/2 for enabled oversee transition and 2 for indirect one), and (d) the film thickness. Where: the bandgap energy (Eg), the transmittance (T), the absorption (A), the absorption coefficient (α), and (hü) is the incident photon energy [29, 30].

3. RESULTS AND DISCUSSION

33.1. Thickness Measurement

Figure (2) illustrates the effect of varying annealing temperatures on the thickness of Bi-doped GeTe thin films that are synthesized. It has been observed that when the annealing temperatures rise, the film thickness falls. Annealing typically lowers the concentration of unsaturated and dangling bonds, allowing the structure to relax and approach equilibrium. This

results in a reduction in the density of defects (stress relief) or a decrease in their value (more ordered and dense structure) [31]. Furthermore, when the material is annealed less than the crystallization temperature (Tc, which is approximately 100°C), the amorphous phase experiences an irreversible tensile stress change, which can be understood as the result of material densification. This is followed by an initial linear compressive stress behavior. Then, at Tc, a sharp tensile jump coincides with crystallization, qualitatively consistent with the anticipated rise in density [32].

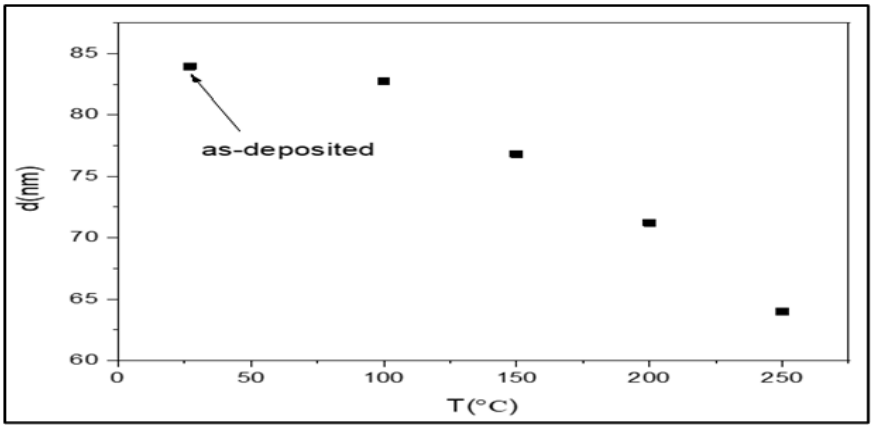

Figure 2: Thickness of Bi-doped GeTe thin films versus annealing temperature.

3.2. Structure Properties of Bi-doped GeTe Thin Film

sigure (3) displays the XRD patterns of bi-doped GeTe at various annealing temperatures. The X-ray patterns reveal an amorphous structure for the films as they are deposited, while the XRD patterns reveal a crystalline structure for all the films that have been annealed, with the exception of the film that was annealed at 100°C (Fig. 3). These findings concurred with those of F.M. Abdel-Rahim and A.H. Moharram [33]. Furthermore, it was also confirmed that thin films have been produced using the Bi-doped GeTe by indexing the resulting patterns and peaks of diffraction as the hexagonal phase of Bi-doped GeTe Germanium Bismuth Telluride to the common spectrum (JCPDS cards No. 98-001-6207).

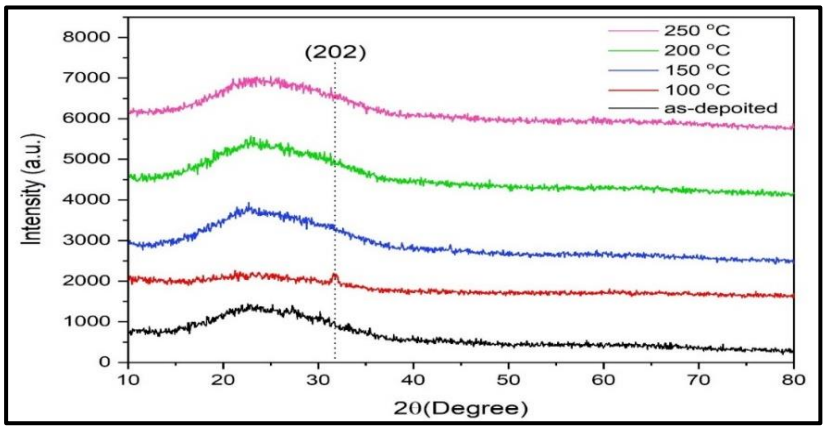

Figure 3: XRD patterns for as-deposited and annealed Bi-doped GeTe thin films.

A rhombohedral structure oriented at a peak position of 2 θ= 31.778, corresponding to the diffraction peak (202) of GeTe thin films, was revealed by analyzing the structure of the annealed sample at 100C. Using the Sherrer formula [34], the average crystalline size was determined to be 11.472 nm.

The rhombohedral GeTe phase that persists in the XRD pattern of bi-doped GeTe thin films can be identified in the XRD patterns of undoped GeTe thin films. In contrast to undoped GeTe thin films, the XRD diffraction peaks of bi-doped GeTe thin films exhibit an associated transition. The peak (202) changes position and angles upward. This phenomenon suggests that Bi atoms occupy lattice sites in GeTe thin films, which causes the interplanar positioning (d) to vary [35].

3.3. Morphological characteristics of Bi-doped GeTe Thin Films

The surface morphology of unannealed (as-deposited) and annealed Bi-doped GeTe thin films at different temperatures is shown in Figure (4).

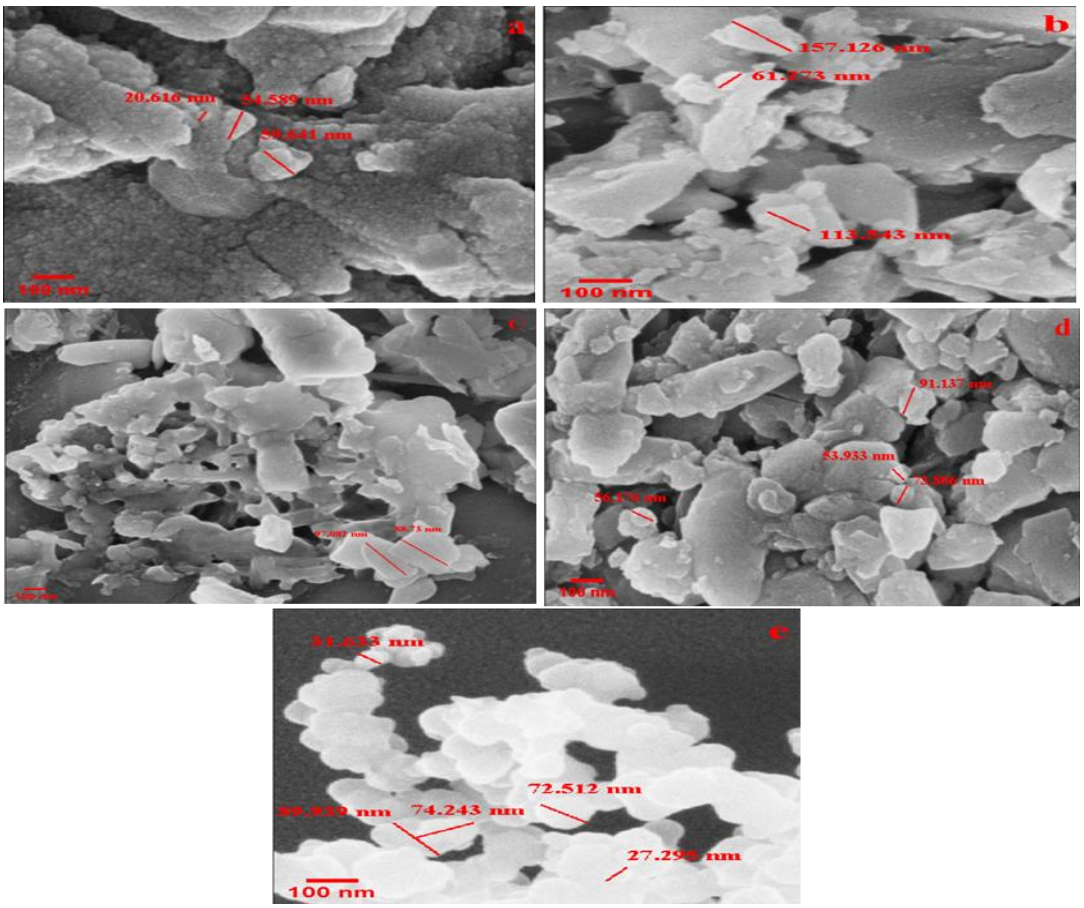

Figure 4: FESEM of (a)as-deposited, and annealed Bi-doped GeTe thin films at (b) 100°C, (c) 150°C,(d) 200°C,(e) 250°C**.**

The crystallite GeTe compound's structure is amorphous and lacks grain, as seen by the FESEM image of Figure (4-a). On the other hand, Figure (4-b) of the thin films that were heated at 100°C for an hour demonstrates that the GeTe crystallites have well-appeared, with an average size of 110.64 nm. This is consistent with the XRD results that indicate a crystalline rhombohedral structure (Figure 3). The annealed thin films at 150°C, 200°C, and 250°C are shown in Figures (4-c, d-e). These figures show that the crystallites have an even distribution throughout all of the thin films. From the aforementioned Figures, it can be inferred that, as indicated in Table (1), the crystallite size decreases as you raise the annealing temperature.

Table 1: The Effect of annealing temperature on grain size.

Temp(°C)	**Average grain Size(nm)**
27	44.948
100	110.647
150	73.718
200	69.592
250	64.772

This compound's aberrant behavior could be caused by uneven crystallite growth throughout the thin films and insufficient annealing their time [20]. When compared to undoped GeTe, the transformation temperature that occurred from the amorphous to the crystalline state decreased when just a little of bismuth was added to the material. With the addition of the Bi dopant, the transformation temperature can drop along with the binding energy across the atoms in the substance [1]. Additionally, because bi atoms encompass the lattice locations in GeTe thin films, as the annealing temperature rises, the film becomes denser, as shown in Figure (4). Furthermore, every single one of these numbers matched the XRD patterns.

3.4. Optical transmission and energy gap

To understand how thermal annealing affects transitions between phases, it is essential to investigate the optical characteristics of bi-doped GeTe thin films, such as transmission and optical band gap [36][37]. The transmittance of the heated and as-deposited Bi-doped GeTe thin films is shown in Figure (5). As the annealing temperature increased, a shift in the absorption edge to a longer wavelength in addition to a decline in transmittance was noted. At a wavelength of 1100 nm and 250 °C, the transmittance of as-deposited thin films dropped to 65.25 %, while that of heated thin films boosted to 32.57%. The optical spectra of the bi-doped GeTe thin films exhibit redshifts following annealing.

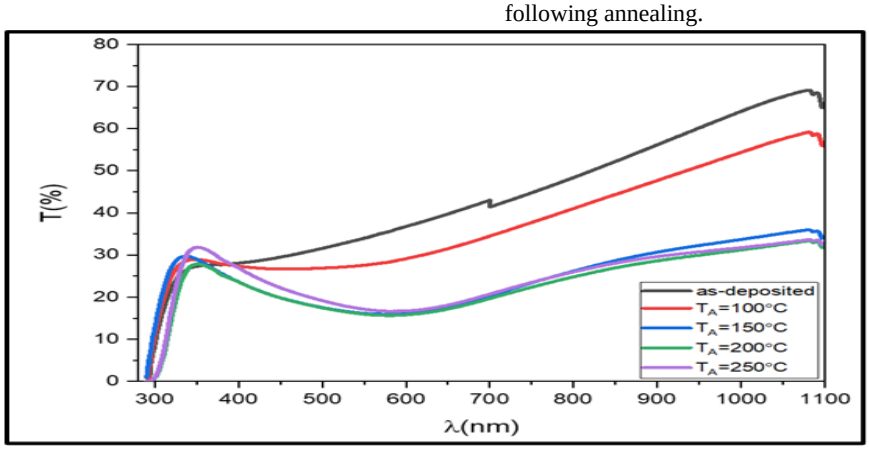

Figure 5: The transmittance for as-deposited and annealed Bi-doped GeTe films versus wavelength.

Figure (6) shows the plot of $(\alpha h\upsilon)^2$ against $h\upsilon$ for the as-deposited Bi-doped GeTe thin films and annealed ones. At the higher absorption region ($\alpha \geq 10^4$), the dependence of photon energy on the absorption coefficient was calculated using (Eq.2) [29, 30, 33].

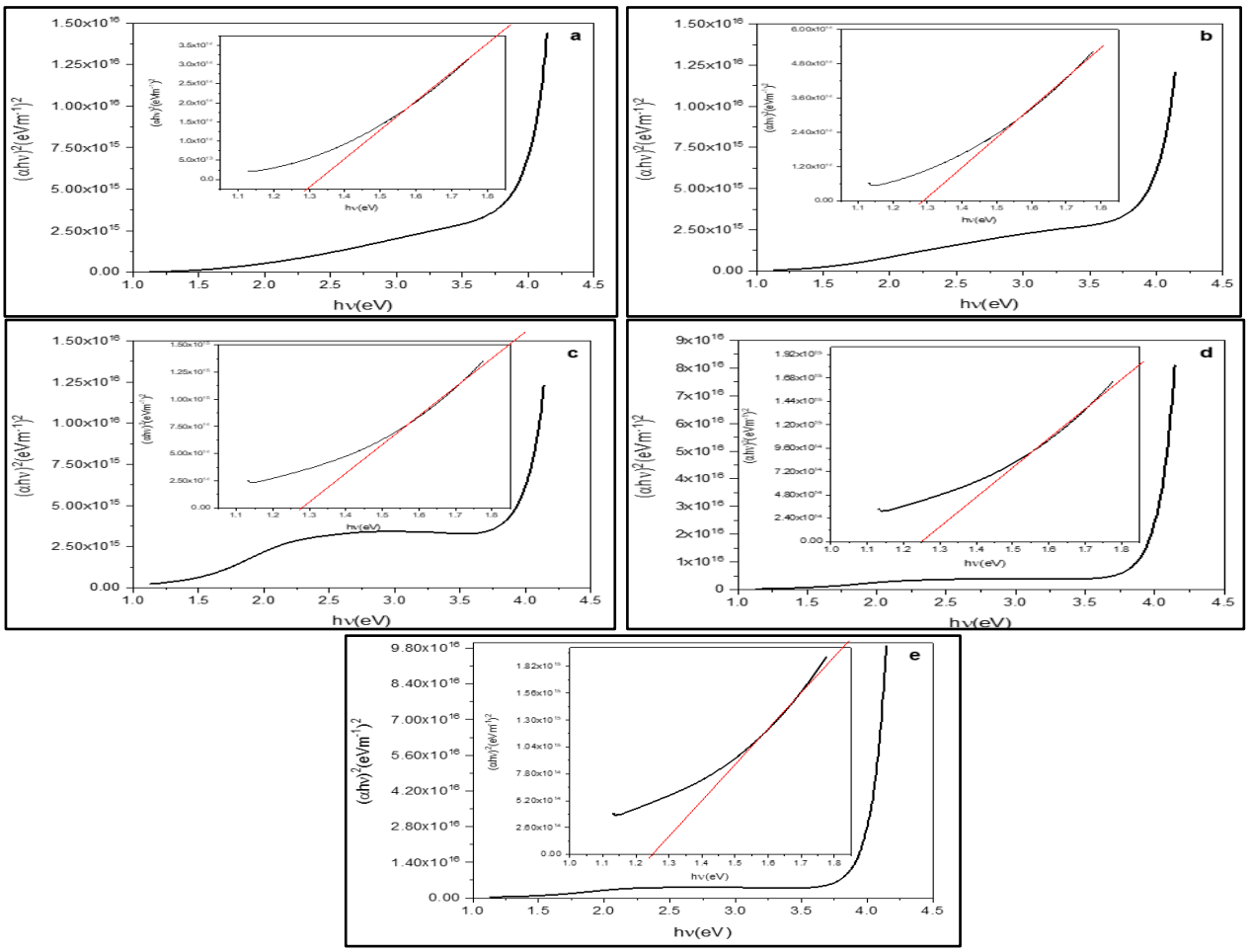

Figure 6: The variant of $(\alpha h\upsilon)^2$ versus photon energy($h\upsilon$) for as-deposited and annealed Bi-doped GeTe against ($h\upsilon$).

Figure (7) illustrates the manner in which the optical gap changes with the annealing temperature. As the annealing temperature rises from 100°C to 250°C, Figure (7) illustrates a corresponding reduction in the optical band gap. One would anticipate denser films and smaller energy gaps as a result of stuffed in gaps that exist in the film, which could be the result of structural a state of relaxation [38]. Furthermore, Bi-doped GeTe shows an increased bandgap regard (0.3-0.8 eV) [39] than common GeTe, which is explained by the amorphous phase. Electron transitions in the amorphous phase can occur from extended states in the conduction band to localized states in the valance band, or vice versa [40]. Furthermore, the size of GeTe's quantization impact, which results in a number of discrete states in the valence and conduction bands, could be the cause of the broad bandgap energy value [41]. Table (2) presents a comparison between the current work alongside additional works.

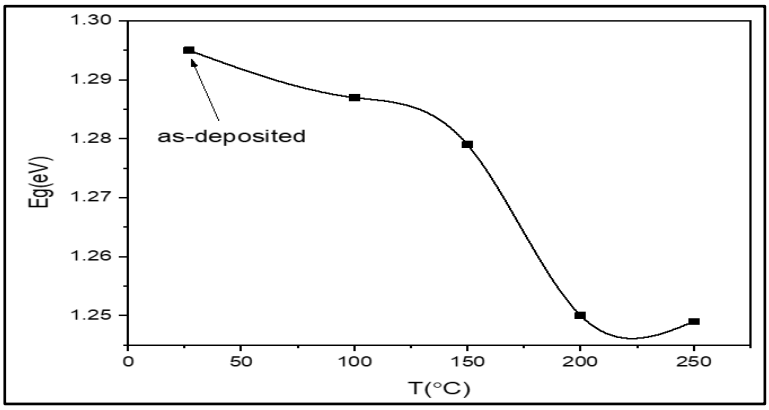

figure 7: Optical band gap (Eg) for as-deposited and Bi-doped GeTe films as a function of annealed temperature.

Table 2: A comparison between the present work and other**s.**

Author	**Bi/percentage**	**Eg (eV)**	**Technique**	**Annealing Temperature**
Our Work	**0.05**	**0.42**	**PVD**	**100℃ to 250℃**
Zihang Liu et al.[9]	0.04, 0.08	-	Alloying	300 to 773 K
B. Ben Yahia et al[32]	-	-	Magnetron sputtering	235℃
M. Shpotyuket al[42]	-	-	Melt–quench	203, 233, and 273 K
C M I Okoy et al[15]	-	0.399	Implementation of the FP-LAPW method	
Xia Xiao Qiu [43]	$0 \leq Bi \leq 0.08$		Melting-quenching-annealing process followed by spark plasma sintering	300–600 K
Thi Hoa Vu[44]	0.05	0.06	Température gradient technique - Bridgman technique	300k
Suresh Perumal[45]	0.06	0.08	Vacuum-sealed tube melting reaction	300–723 K
P. Lazarenko et al[26]	0.5, 1 and 3 wt.%		Quenching	350℃
Ki-Hong Kim et al[46]	GeBi (6 at.%)Te		Sputtering	100–400 °C

CONCLUSIONS

Rapid and reversible transitions between the amorphous (a) and crystalline (c) indicate a characteristic of phase-change materials (PCMs). Bi-doped GeTe thin films were successfully manufactured using the PVD method, allowing phase-change thin films to be properly doped at the right levels to modulate phase-change behaviors. The dopant process modifies the structural characteristics of semiconductor materials and is a highly effective control method to achieve changes in the optical properties. The structure that is annealed at 100°C is crystalline; whereas, the one that gets deposited is amorphous. The crystallite GeTe compound's arrangement is amorphous, with no grain visible, according to the FESEM image. On the other hand, the GeTe thin films that have been annealed at 100°C are attractive crystallites with an average size of 110.64 nm. After annealing the thin films at 150°C, 200°C, and 25 0°C, it is evident that the crystallite is evenly dispersed throughout each thin film. The crystallite is dispersed throughout all of the thin films at annealing temperatures greater than 100°C. A rise in the annealing temperature results in both a decrease in transmittance and a significant shift in the absorption edge with respect to a broad wavelength. during which the transmittance dropped for annealed thin films coming from (65.25%) as-deposited thin films to (32.57%).

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