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Single-layer, multi-layer and superlattice chalcogenides for non-volatile memory and artificial intelligence device

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Highlights:

- High-performance phase-change memory using various chalcogenides.
- Potential applications in artificial intelligence devices.

Abstract: Chalcogenide materials such as GeTe, Sb₂Te₃, Ge₂Sb₂Te₅, compounds of sulfur, selenium, and tellurium, are characterized by a rapid and reversible phase transition. They have emerged as versatile candidates for advanced electronic and computing applications due to their unique optical and electrical properties. The potential of single-layer, multi-layer, and superlattice chalcogenides as the active materials in non-volatile memory (NVM) and artificial intelligence (AI) devices is shown in this review. Single-layer chalcogenides often offer exceptional scalability, fast speed, and nonvolatility, enabling them usable for NVM and synaptic devices. Multi-layer chalcogenides with stacked structures exhibit unique properties different from single-layer chalcogenides, allowing high performance such as low power and multilevel storage. Superlattice chalcogenides with alternating chalcogenide layers enable ultra-low power consumption of devices, in which only few of atom moves for the operation of devices. This review also gives some related research details. A comprehensive understanding of these studies provides insights into the design and application of chalcogenide-based devices, offering pathways for future research and innovation in memory and AI hardware.

Keywords: chalcogenide; phase change; non-volatile memory; artificial intelligence

1. Introduction

Chalcogenide materials such as GeSbTe alloys, compounds of sulfur, selenium, and tellurium, have attracted much attention due to their unique optical and electrical properties [1-8]. These chalcogenides are characterized by a rapid and reversible amorphous-crystalline phase transition with a significant change in reflectivity as well as resistivity. The reflectivity change can be as great as 30% while the resistivity change can be as large as six orders of magnitude [9-14]. These properties make them prime candidates for applications in optical disks, non-volatile memories (NVM) and artificial intelligence (AI)



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devices. In order to enhance the performance and extend the functionality of the devices, single-layer, multi-layer, and superlattice configurations are studied in the world. These devices thus can have distinct advantages over other NVM and AI devices using other materials such as oxides. The resistance change is based on phase change between amorphous and crystalline in the single-layer and multiple layer chalcogenides while it is caused by the movement of few atoms in the superlattice [14]. Higher performances such as high speed and low power consumption can be realized by adopting superlattice chalcogenides while low cost can be expected by using simple singe-layer chalcogenides as shown in Figure 1. Generally, the fabrication equipment is complicated and processes require a long time for the multilayer and superlattice chalcogenides.



Figure 1. Schematic of relationship of single-layer, multiple-layer and superlattice chalcogenides.

The origins of Ge-Sb-Te alloys can be traced to the 1960s when the study on amorphous materials by Ovshinsky highlighted the unique switching properties of chalcogenide glasses [2]. Single-layer chalcogenides such as $Te_{48}As_{30}Si_{12}Ge_{10}$ were initially adopted in switching or electrical memory devices. Resistance change was realized by applying a pulse with a duration of 10 µs or more to crystallize the chalcogenide. The discovery of GeSbTe and Ag- and In- doped SbTe (AIST) alloys greatly reduced the switching time of the chalcogenide-based devices to less than 100 ns [8,15]. Due to the high performances of devices, the GeSbTe alloys were intensively and systematically studied from structural, electric and kinetic aspects. In order to further improve the performances such as high speed, low power consumption and high thermal stability, new materials were investigated by doping other elements such as N, O, Ti and Si into conventional chalcogenides [16–21].

Multi-layer chalcogenides are comprised of several stacked layers of different chalcogenide materials, which can be engineered to achieve specific electrical and thermal properties [22–30]. This structural configuration allows for enhanced control over phase transitions, crucial for the high performance of NVM devices. Lai *et al* reported that the multi-level storage capability of a vertical phase change device was confirmed by using multi-layer chalcogenide with a pure 280-nm-thick Ge₂Sb₂Te₅ sublayer and a 200-nm-thick Si-doped Ge₂Sb₂Te₅ sublayer [22]. Pure Ge₂Sb₂Te₅ and Si-doped Ge₂Sb₂Te₅ chalcogenides had much different thermal and electrical properties. For example, the crystallization temperature of these two materials were 163 and 274 °C, respectively. The resistivity of Si-doped Ge₂Sb₂Te₅ at each temperature. Both current-voltage *I-V* and resistance-applied pulse voltage *R-V* exhibited three relatively stable resistance levels. Chong *et al* demonstrated the significant performance improvement

of phase-change device by adopting a superlattice-like GeTe/Sb₂Te₃ multi-layer [27]. Strictly speaking, the multi-layer was not a superlattice structure although it had a similar structure. The RESET current for amorphization of the device with a pure Ge₂Sb₂Te₅ single-layer was 16 mA, much higher that of the device with a superlattice-like GeTe/Sb₂Te₃ multi-layer (6 mA). The speed of SET for crystallization was also increased to twice when the superlattice-like GeTe-Sb₂Te₃ multi-layer was adopted. The difference of the thermal conductivity between the single-layer chalcogenide and superlattice-like multi-layer chalcogenide was thought to be one of the main reasons for the excellent performances.

Two-dimensional (2D) superlattice structures involve alternating layers of different chalcogenide materials, creating periodic potentials that can be tailored to achieve desired electronic properties. These new structures have led to significant advancements in phase-change memory (PCM) technologies [14,31–36]. Simpson *et al* reported an interfacial phase change memory with a superlattice of GeTe/Sb₂Te₃ [14]. The superlattice device consumed an order of magnitude less energy during the SET process with respect to the conventional Ge₂Sb₂Te₅ single-layer device. The reason for this is that the interface between GeTe and Sb₂Te₃ controls the local atomic switching of Ge atoms resulting in a phase transition with substantially reduced entropic losses. Recently, Kang *et al.* demonstrated the possibility of artificial synapse by using a device with a superlattice of GeTe/Sb₂Te₃ [36]. Based on the movement of Ge atoms, a tunable analog weight update of artificial synapses was realized by using the optimal pulse scheme and GeTe/Sb₂Te₃ layer. A nonlinearity of 0.32 and 40 conductance states were achieved for long term potentiation and depression, respectively. The artificial synapse with stable change in the gradual conductance showed the potential for the significant performance improvement of neuromorphic computing.

2. Single-layer chalcogenides

2.1. Ge-Sb-Te chalcogenides

Figure 2(a) shows Ge-Sb-Te chalcogenides such as those along the GeTe- Sb₂Te₃ line, Sb₂Te, Sb₃Te. They are widely used in optical and electrical storage as well as artificial intelligence devices. The origins of Ge-Sb-Te alloys can be traced to the 1960s when the study on amorphous materials by Ovshinsky exhibited the unique switching properties of chalcogenide glasses [2]. These chalcogenides had electrical and optical bistable properties and initiated interest in their use for data storage. GeSbTe, a ternary compound within the Ge-Sb-Te phase diagram as shown in Figure 2, emerged as a particularly promising material due to its remarkable phase transition properties. In the 1980s, GeSbTe alloys were prominently investigated for rewritable optical storage devices such as CDs and DVDs [37–39]. The ability of these chalcogenides to reversibly switch between amorphous and crystalline states with distinct optical reflectivity levels laid the foundation for its application in optical storage media.

Their advanced properties also enabled them to be used in electronic phase-change memories and artificial devices. These devices conventionally utilize a single-layer GeSbTe chalcogenide as the active phase change material sandwiched between two metal electrodes. Figure 2(b) shows the typical *I-V* characteristics of the device with a single-layer chalcogenide. An amorphous phase with a high resistivity can be suddenly changed to a crystalline phase with a low resistivity by applying a voltage higher than a certain voltage (threshold voltage V_t). The two resistance states are distinguished by reading them at a voltage of V_{read} .

GeTe and Sb₂Te₃ are the fundamental binary compounds in the Ge-Sb-Te ternary system and are crucial for understanding the properties of GeSbTe alloys. The combination of GeTe and Sb₂Te₃ in GeSbTe alloys provides a synergistic effect. This balance makes GeSbTe alloys ideal for memory devices.



Figure 2. (a) Typical GeSbTe alloys; (b) Typical *I-V* characteristics of device with a single-layer chalcogenide.

In this review, the fundamental characteristics of GeTe alloys will be discussed as below. X-ray diffraction patterns of GeTe are typically shown in Figure 3(a). As can be seen, there are no peaks for the GeTe annealed at a temperature of 180 and 200 °C. This means that the films were amorphous. Many peaks appeared after annealed at a temperature of 220 °C, implying the films became crystalline. The crystal structure was determined to be face-centered-cubic (FCC). The miller indices are shown in the pattern of the film annealed at 430 °C. GeTe has only one crystalline structure, that is FCC. This is very different from the other chalcogenides such as Sb₂Te₃, and alloys along the line of GeTe and Sb₂Te₃ in the Figure 2(a). As-deposited Sb₂Te₃ is often FCC crystalline because of its very low crystalline temperature while the other widely used chalcogenides are amorphous. Sb₂Te₃ changes to another crystallizes to FCC at about 150 °C and then changes to HEX at about 280 °C. In general, the crystallization temperature

increases from Sb₂Te₃ to GeTe. This means that amorphous phase of GeTe is much stabler than Sb₂Te₃ and thus GeTe device can be used at a high temperature because of its high thermal stability.

Figure 3(b) shows the resistivity change of GeTe as a function of annealing temperature. The resistivity of as-deposited of GeTe was about $70 \Omega \cdot m$ and the high resistivity remained until 200 °C. It suddenly dropped to about $2 \times 10^{-5} \Omega \cdot m$ by more than 6 orders of magnitude after annealed at 220 °C. The decrease in resistivity resulted from the amorphous-to-FCC phase transition as described above. The resistivity almost did not change after annealed at a high temperature. The reason for this should be that there was not the FCC-to-HEX phase transition and obvious crystal growth for the GeTe. On the contrary, compared with GeTe, as-deposited Sb₂Te₃ has a very low resistivity since it has already been crystalline [17]. The resistivity of Sb₂Te₃ decreases gradually as annealing temperature, which is caused by relatively slow crystal growth of FCC and FCC-to-HEX phase transition. As-deposited Ge₂Sb₂Te₅ has an intermediate resistivity, compared with GeTe and Sb₂Te₃. Ge₂Sb₂Te₅ exhibits two sudden decreases in resistivity, corresponding to amorphous-to-FCC and FCC-to-HEX phase transitions.



Figure 3. Typical characteristics of GeSbTe alloys. (a) X-ray diffraction patters of GeTe; (b) Resistivity change of GeTe; (c) Schematic diagram of devices; (d) *I-V* characteristics.

Figure 3(d) shows the *I-V* characteristics of GeTe device with a lateral structure (Figure 3(c)) [40]. The initial device resistance was about 103 M Ω and dropped to 782 Ω by about 5 orders of magnitude. Both high and low resistance states were very stable according to the *I-V* characteristics. These two states

correspond to amorphous and crystalline phases, respectively. The resistance change in device induced by electrical pulse can occur as fast as ns order, even hundreds ps order, depending on chalcogenides.

2.2. Doping for high performance

The Ge-Sb-Te ternary system permits extensive compositional tuning, facilitating the optimization of key parameters such as switching speed, thermal stability, and material endurance. Although a device with a layer of a conventional chalcogenide, for instance, GeTe and Ge₂Sb₂Te₅, exhibits very good performance such as high speed, good stability, it has been demonstrated that doping with elements such as nitrogen, silicon, or carbon into conventional chalcogenides is a good way to refine phase transition kinetics and enhance thermal stability and reliability [41–43]. Doping involves introducing foreign atoms into conventional chalcogenides to modulate their physical, chemical, and electrical properties. The dopant can occupy substitutional or interstitial sites or even form new phases, influencing phase-transition dynamics and stability of the materials.

In this review, the influence of N doping into GeTe will be discussed here. Figure 4(a) shows the X-ray diffraction patterns of the GeTeN1.0 film fabricated by sputtering. The N_2 and Ar flow rates were 1.0 and 9.0 sccm (standard cubic centimeters per minute), respectively. There are no peaks for the film annealed at 220 °C. The crystallization can be almost neglected although very small peaks were observed in the film annealed at 280 °C. This means that nucleation occurred at this temperature. The intensity of XRD diffraction peaks became strong with annealing temperature, implying the crystallization, that is, crystal growth preceded in the film.

Mean crystallite size L can be estimated using the well-known Scherrer's equation

$$L = \frac{0.89\lambda}{\beta \cos \theta} \tag{1}$$

where λ is X-ray wavelength, β is the full-width at half-maximum (FWHM) intensity of the peak in radians and θ is the diffraction angle of the peak. Mean crystallite sizes of undoped and N-doped GeTe films annealed at 360 °C for 3 min are shown in Figure 4(b). The cystallite size of undoped film was 12 nm while the films fabricated at conditions of N₂ flow rates of 1.0 and 2.0 sccm were 7 and 5.1 nm, respectively. As a result, it is very obvious that the crystal size was greatly refined by N-doping.

The crystallization temperature as a function of N_2 flow rate is shown in Figure 4(c). The crystallization temperature was measured by a differential scanning calorimeter and the samples were heated at a rate of 10 °C/min. The crystallization temperature of the undoped GeTe film was about 222 °C while that of the N-doped GeTe film fabricated at 1.0 sccm was even above 295 °C. Doping N into GeTe greatly increased the crystallization temperature, implying the significant improvement of thermal stability.

Figure 4(d) shows the resistivity change with a function of annealing temperature. For the asdeposited films, the resistivity increased by about one order of magnitude after doping N into GeTe. Compared with undoped GeTe, the resistivity of N-doped GeTe suddenly dropped at the high annealing temperature of about 260 °C. The resistivity of N-doped GeTe then decreased gradually while that of undoped GeTe almost did not change. This means that the crystal growth gradually preceded after nucleation in N-doped GeTe while crystal growth should complete very fast in undoped GeTe. The difference in electrical properties between undoped and N-doped GeTe films was in good agreement with the XRD patterns of these films.



Figure 4. Characteristics of doped chalcogenide. (a) X-ray diffraction patterns; (b) Crystallite size as a function of N_2 flow rate; (c) Crystallization temperature as a function of N_2 flow rate; (d) Resistivity as function of annealing temperature (GeTe: blue, N-GeTe: light blue).

3. Multi-layer chalcogenides

Multi-layer chalcogenides involve stacking multiple layers of phase change materials and other functional layers to optimize the performance of phase change devices. For instance, these multi-layer structures can be engineered to enhance thermal confinement, reduce reset current, and improve data retention. Advanced fabrication techniques, such as sputtering and atomic layer deposition, enable precise control over layer thickness and composition. That is crucial for tailoring the electrical and thermal properties of the device. On the other hand, beyond binary storage, phase-change devices with multi-layer chalcogenides can achieve multilevel storage by precisely controlling the degree of crystallization or amorphization and thus enabling intermediate resistance states. In devices, this is accomplished by modulating the amplitude and duration of electrical pulses, allowing each cell to store multiple bits of information. Multilevel storage technology greatly enhances data density without increasing the physical size of the memory array. Figure 5(a) shows the schematic of the multi-layer chalcogenide with two different sublayers of Ch1 and Ch2. By using sublayers with different properties, high performance such as multilevel storage becomes available. Figure 5(b) shows an example of multilevel storage. The first sudden change corresponding to the resistance drop induced by the crystallization of Ch2 occurs at the current I_{th1} or the voltage V_{th1} . The second change corresponding to

the decrease in resistance resulted from the crystallization of Ch1 occurs at the current I_{th2} or voltage V_{th2} . As a result, intermediate resistance levels will be realized if we can control the sequence of crystallization layer by layer in the device with a multilayer chalcogenide.



Figure 5. (a) Schematic of the multiple-layer chalcogenide consisting of two different chalcogenides; (b) High performance by adopting multi-layer and enabling intermediate resistance.

Figure 6(a) shows X-ray diffractions of O-doped GeTe/O-doped GeTe Sb₂Te₃. The multilayer chalcogenide [O-doped GeTe (6.25 nm)/O-doped Sb₂Te₃ (6.25 nm)]₁₆ was composed of 16 cycles of 6.25-nm-thick O-doped GeTe and 6.25-nm-thick O-doped Sb₂Te₃ sublayers. From our analysis, it was clear that O-doped GeTe as the first kind of chalcogenide (Ch1) in the multilayer was amorphous while the O-doped Sb₂Te₃ as the second kind of chalcogenide (Ch2) in the multilayer had already been crystalline and had a face-centered-cubic (FCC) structure when they were as-deposited. The O-doped GeTe (Ch1) sublayers crystallized with an FCC structure while the O-doped Sb₂Te₃ (Ch2) sublayers remained FCC at about 200 °C.

By further increasing annealing temperature to about 260 °C, phase change from FCC to HEX occurred in the O-doped Sb₂Te₃ (Ch2) sublayers. The two subsequent phase transitions are illustrated in Figure 6(a). The sequence of phase change in the multilayer chalcogenide [O-doped GeTe (6.25 nm)/O-doped Sb₂Te₃ (6.25 nm)]₁₆, therefore, can be well controlled by increasing the annealing temperature.

A vertical device with a multilayer [O doped GeTe (6.25 nm)/O doped Sb₂Te₃ (6.25 nm)]₁₆ was fabricated. The multilayer was sandwiched by a bottom TiN electrode and a top Ti electrode. It was demonstrated that the intermediate resistance was realized by adopting the multilayer as shown in Figure 6(b). The high, intermediate and low resistance levels should correspond to the phase states of Ch1: A/ Ch1: FCC, Ch1: FCC, Ch1: FCC, Ch1: FCC / Ch1: HEX, respectively. High performance such as multi-level storage became possible after the multilayer chalcogenide was adopted.



Figure 6. Characteristics of multi-layer chalcogenide. (a) X-ray diffraction patterns; (b) *I-V* characteristics (inserted figure: cross-sectional diagram of devices).

4. Superlattice chalcogenides

As described before, phase change devices utilize chalcogenides such as Ge₂Sb₂Te₅. The transition between amorphous and crystalline phases, representing binary data, can be easily controlled in these materials. However, challenges such as high switching energy and limited endurance persist. Superlattice structures, comprising alternating nanoscale layers of different materials, have emerged as a promising approach to address these challenges, as shown in Figure 7(a). In phase change device applications, it is possible to modulate phase transition dynamics, reduce programming current or power, and thus enhance device performance by adopting superlattice in these devices, as shown in Figure 7(b). Moreover, in neuromorphic computing, biological synapses can be emulated in the superlattice-based devices, offering potential for energy-efficient and high-density neural networks [44–47].





Phase-change memory with a chalcogenide-based superlattice structure such as GeTe/Sb₂Te₃ was demonstrated to exhibit a large resistance change without the conventional phase transition of Ge₂Sb₂Te₅ and significant reduction in power consumption has been proved. GeTe/Sb₂Te₃ superlattice structure can

be fabricated using sputtering technology [48]. Until only recently, this promising GeTe/Sb₂Te₃ superlattice was tried to be applied to the artificial synapse because it is difficult to obtain intermediate states which are usually required for the plasticity of a synapse. However, when the pulse width changed from 1 µs to 70 ns, the synaptic weight varied in a very narrow range of only about 3 when the short pulse width was applied. This implies that the bandgap of GeTe/Sb₂Te₃ superlattice is not wide enough for the application of the high-speed artificial synapse. Doping O into the GeTe/Sb₂Te₃ superlattice has a great influence on the band structure, enabling a wide bandgap. Doping other elements into superlattice structure can be conducted by using doped targets or adding some gases such as O₂ or N₂ [49]. The O content of the superlattice structure can be controlled simply by adjusting the flow rate of O₂.

The primitive cells of the conventional undoped GeTe/Sb₂Te₃ superlattice of Petrov and Inverted Petrov structures are shown in Figures 8(a,c), respectively [50–51]. They consist of atomic layers of -Te-Sb-Te-Ge-Te-Ge-Te-Ge-Te-Sb- and -Te-Sb-Te-Ge-Ge-Te-Te-Sb-. The corresponding band structure of the two superlattice structures are shown in Figure 8(b,d), respectively. Figure 8(e) shows the density of states of undoped superlattice GeTe/Sb₂Te₃. It can be clear that the Petrov structure is metallic while the Inverted Petrov structure exhibits a semiconductor characteristic with a bandgap of 0.22 eV. They correspond to a low resistance state (LRS) and a high resistance (HRS), respectively. The switching between these two states can be done by a slight structural change. This is based on the migration of few Ge and Te atoms with an ultralow energy, instead of the conventional amorphous-crystalline phase transition with movements of a number of atoms requiring a very high energy.



Figure 8. (a) Atomic structure; (b) Band structure of Petrov structure; (c) Atomic structure; (d) Band structure of Inverted Petrov structure; (e) Density of states of undoped superlattice GeTe/Sb₂Te₃.

The primitive cells of the O-doped GeTe/Sb₂Te₃ superlattice of Petrov and Inverted Petrov structures are shown in Figure 9(a,c), respectively. 5 atoms (O: 36%) are incorporated into the primitive cells. The corresponding band structures of the two O-doped superlattice structures are shown in Figures 9(b,d). It was found that the atomic structures are quite different from those of the undoped superlattices after heavily doping O. The bandgap of the doped one is 0.41 eV, much higher than that of conventional superlattice for the Inverted Petrov structure. As a result, by doping O into GeTe/Sb₂Te₃ superlattice, it is possible to increase the bandgap difference to 0.41 eV, which is about twice that of pure GeTe/Sb₂Te₃ superlattice (0.22 eV). It is implied that this can increase the synaptic weight change even when high-speed ns-pulses are applied, indicating the performance improvement of the artificial synapse.



Figure 9. (a) Atomic structure; (b) Band structure of Petrov structure; (c) Atomic structure; (d) Band structure of Inverted Petrov structure; (e) Density of states of O-doped superlattice GeTe/Sb₂Te₃.

5. Conclusion

Chalcogenide materials, in their single-layer, multi-layer, and superlattice forms, have proven to be pivotal in advancing non-volatile memory (NVM) and artificial intelligence (AI) devices. Their characteristics, including fast speed, nonvolatility, and compatibility with modern fabrication techniques, make them indispensable in addressing the evolving requirements of high-performance devices. Single-layer chalcogenides offer significant advantages in terms of energy efficiency, scalability, and functionality, particularly for NVM and AI applications. Multi-layer chalcogenides provide the ability to tune electrical and thermal properties, ensuring improved performance under demanding operational conditions. Superlattice chalcogenides with periodic structures make ultralow power consumption possible with the atom movement during operation of devices. Doping other elements can greatly improve the performances of devices. These chalcogenides have also demonstrated potential in AI systems, enabling the development of innovative hardware such as synaptic devices.

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Authors' contribution

Conceptualization, MM and YY; writing—original draft preparation, MM; writing—review and editing, YY; visualization, YY; supervision, YY; project administration, YY; funding acquisition, YY. All authors have read and agreed to the published version of the manuscript.

Conflicts of interests

The authors declare no conflict of interest.

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