Neutron Radiation-induced Damage to the Local Structure and Optical Properties of Amorphous Phase-Change Materials

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Abstract. Phase change materials are being developed for aerospace applications due to their unique storage mechanisms and excellent optical storage performance, where radiation damage is an issue that needs to be addressed. This paper used a 300 KeV neutron beam to irradiate amorphous $Ge_2Sb_2Te_5$ (a-GST) films, the radiation damage to the local structure and optical properties of a-GST were studied. The results showed that neutron radiation partially damaged the Ge-Te and Sb-Te, causing changes in the values and distribution of the refractive index, but had a lesser impact on reflectivity. Good neutron radiation tolerance was exhibited by a-GST.

Keywords: Neutron irradiation; Optical properties; Radiation damage; $Ge_2Sb_2Te_5$.

1. Introduction

The irradiation environment in space not only poses a threat to human health but also equally endangers the stable operation of electronic devices. In addition to constantly maintaining the stability of the cabin's internal conditions, aerospace equipment also requires monitoring of the surrounding environment. Therefore, a multitude of information needs to be collected, stored, and transmitted, making data storage a critical component within the system. The widely used storage medium is FLASH. However, in space, numerous radiation particles such as protons, neutrons, and heavy ions induce ionization and displacement effects, leading to data errors or permanent functional failures in FLASH [1, 2]. Therefore, there is an urgent need for a storage medium with better radiation resistance to meet the future demands of space exploration.

Phase-change materials are considered a viable storage medium for space environments. There are many types of phase-change materials, among which Ge2Sb2Te5 has been extensively researched and is more readily available. Different from the charge storage mechanism, phase-change materials are rapidly and reversibly switched through laser excitation [3]. Their amorphous and crystalline states exhibit significant optical differences, allowing them to be defined as "1" and "0" in a binary system. In addition, its storage speed [4], capacity [5], and power consumption [6] also surpass those of the currently used storage devices. To harness the full potential of phase-change memory, it is essential to first investigate the behavior of this material in a radiation environment. Ionization effects alter the carrier concentration within the material and also generate some thermal effects. However, phase-change storage does not rely on the quantity of charge and exhibits excellent thermal stability. Displacement effects occur when radiation particles collide with atoms, causing a cascade reaction that generates vacancies and interstitial atoms, disrupting the local structure of the material. The structure and performance of phase-change materials are closely interconnected. This study utilized neutron beam irradiation at various doses on a-GST to investigate the changes in local structure and optical properties.

2. Materials and Methods

The 200 nm thick GST films were deposited on Si substrates using magnetron sputtering. The neutron beam had an energy of 300 KeV, with doses of 2.1×105 p/cm2, 1.0×106 p/cm2, 1.1×107

p/cm2, and 5.2×108 p/cm2, ensuring complete irradiation of each sample. Samples before and after irradiation were subjected to phase identification using an X-ray diffractometer (Shimadzu). Raman spectra (Renishaw) were collected to characterize the local structure of the samples. The bonding characteristics were detected using an X-ray photoelectron spectrometer (Thermo Fisher). In the wavelength range of 400-800 nm, the reflectivity was measured using a UV-Vis-NIR spectrophotometer (Hitachi UH-4150), and the refractive index was measured using an ellipsometer.

3. Results and Discussion

The XRD spectra of the deposited GST films before and after irradiation are shown in Fig. 1(a). The results indicate the absence of any diffraction peaks, suggesting that the films remain in an amorphous structure. After irradiation with varying doses of neutrons, no new diffraction peaks emerged, indicating that the irradiated GST films still maintain an amorphous state. The Raman spectra of the a-GST film before and after irradiation are shown in Fig. 1(b). A strong peak is at 150.7 cm-1, named the T-peak, originating from the vibration of the pyramid-shaped SbTe3 units [7]. The peak position and integrated intensity of the T-peak under different doses are shown in Fig. 1(c) and Fig. 1(d), respectively. After neutron irradiation, the T-peak exhibited a blue shift, which can be attributed to increased compressive stress [8]. The atomic arrangement is random in the amorphous structure. Neutrons entering the films collide with atoms, causing some of the impacted atoms to move toward the interior of the films and come to a halt at a new position. This process will lead to a certain degree of volume expansion. Neutron irradiation enhances the vibration of the SbTe3 units, indicating an increase in disorder within the films. The atoms become highly active, and the pyramid-shaped structure is in an unstable state.

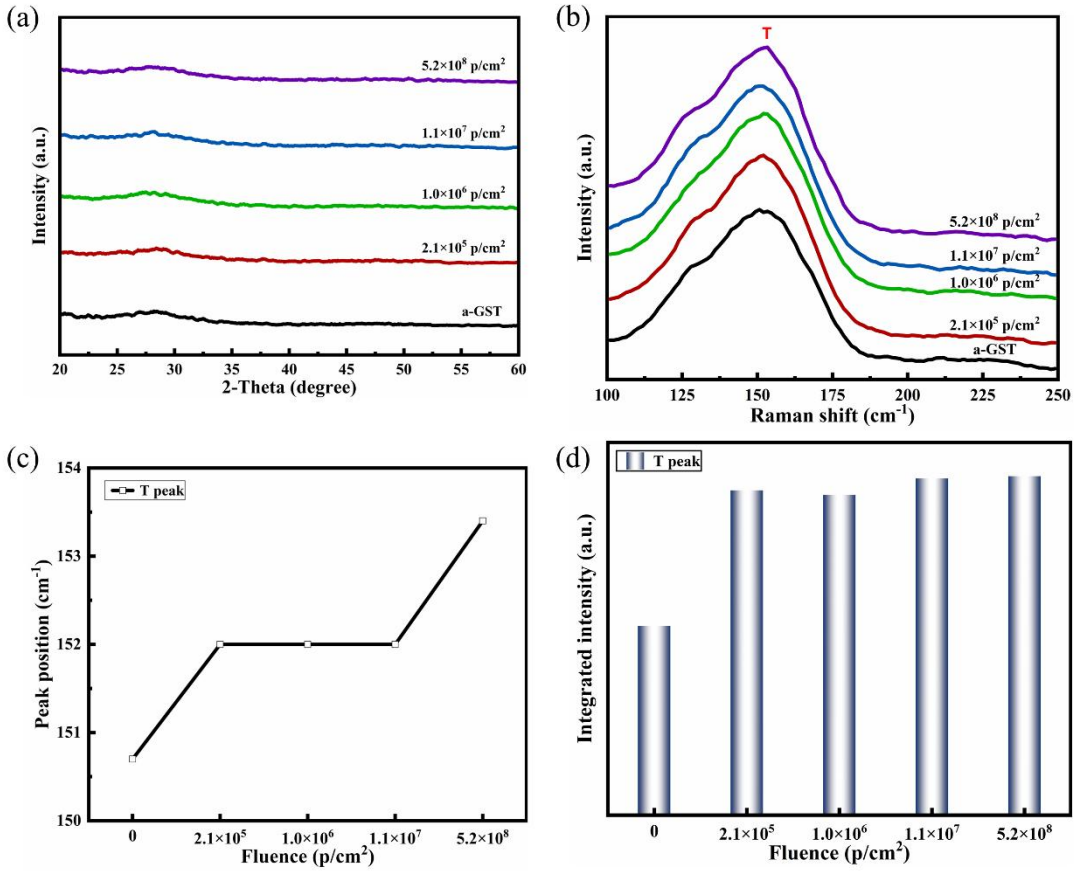

Fig. 1 (a) XRD pattern of a-GST films before and after neutron irradiation. (b) Raman spectra of a-GST films before and after neutron irradiation. (c) The position of T peak at different irradiation doses. (d) T peak integrated intensity at different irradiation doses.

The energy spectra of Ge 2p3/2, Sb 3d5/2, and Te 3d5/2 in the a-GST films are shown in Fig. 2. At 1217.4 eV in Ge 2p3/2, the spectrum corresponds to the Ge-Te bond [9]. At 529 eV in Sb 3d5/2 and 573.15 eV in Te 3d5/2, they both correspond to the Sb-Te bond [10]. The binding energy of the Ge-Te bond increases after radiation, while the binding energy of the Ge-O bond is approximately 1220 eV. The irradiation causes the breaking of Ge-Te bonds, attracting oxygen atoms to form Ge-O bonds by binding with Ge atoms. The decrease in the binding energy of the Sb-Te bond indicates that some of the Sb-Te bonds within the pyramid-shaped SbTe3 units are breaking, leading to the disruption of the local structure. The absence of a trend towards oxide bonding can be explained by the relatively weak nature of the Sb-Te bond, making it more prone to breakage under neutron radiation. Even if oxidation bonds are formed, they may be subsequently disrupted. Furthermore, once the oxide of Ge is formed, it creates a protective layer that hinders the further ingress of oxygen. To mitigate damage, a protective layer on the surface of the GST films is necessary. Additionally, doping can be considered to alter the bonding structure and form more stable chemical bonds

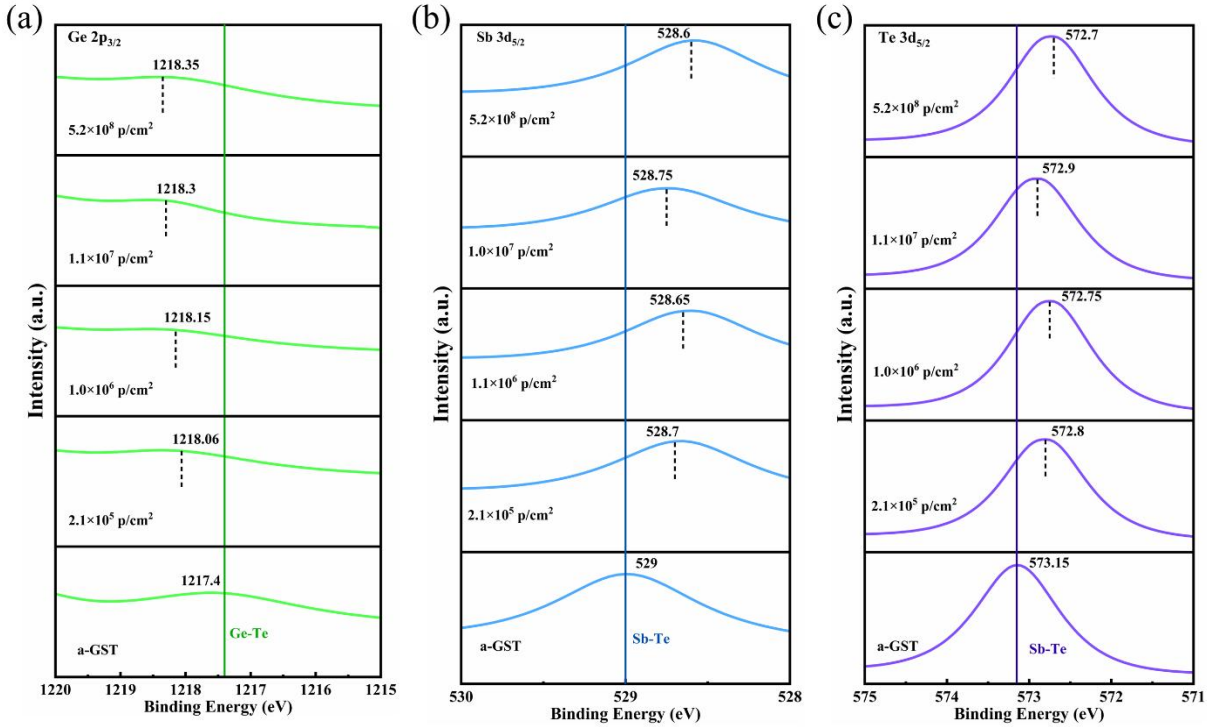

Fig. 2 XPS spectra of a-GST films before and after neutron irradiation. (a)Ge $2p_{3/2}$. (b)Sb $3d_{5/2}$. (c)Te $3d_{5/2}$.

The reflectivity and refractive index of the irradiated a-GST films within the wavelength range of 400-800 nm are shown in Fig. 3. The reflectivity of the a-GST films exhibits an increasing trend followed by a decrease within the wavelength range of 400-800 nm, reaching its maximum reflectivity of 20.789% at 632 nm. At a radiation dose of 2.1×105 p/cm2, the reflectivity is nearly identical to that of the unirradiated films. At radiation doses exceeding 2.1×105 p/cm2, the reflectivity of the films decreases slightly by approximately 1%. However, radiation did not alter the overall distribution trend of reflectivity or the wavelength at which maximum reflectivity occurs. The refractive index of the a-GST is around 1.55. It can be observed that although the refractive index values change very slightly after irradiation, the distribution differs noticeably at different doses. The reflectivity is primarily correlated with surface roughness. When neutrons first penetrate the film surface, their cumulative doses are relatively low, resulting in a minimal impact on surface properties. In contrast, the refractive index is closely associated with the bonding characteristics of the films. Under neutron radiation, there is a phenomenon of bond breaking and formation within the films, which has a more significant impact on the refractive index. Additionally, the changes in

binding energy at different doses are distinct. When constructing optical storage functionalities, it is crucial to consider the influence of the external environment on phase-change materials, particularly in terms of their structure and optical properties.

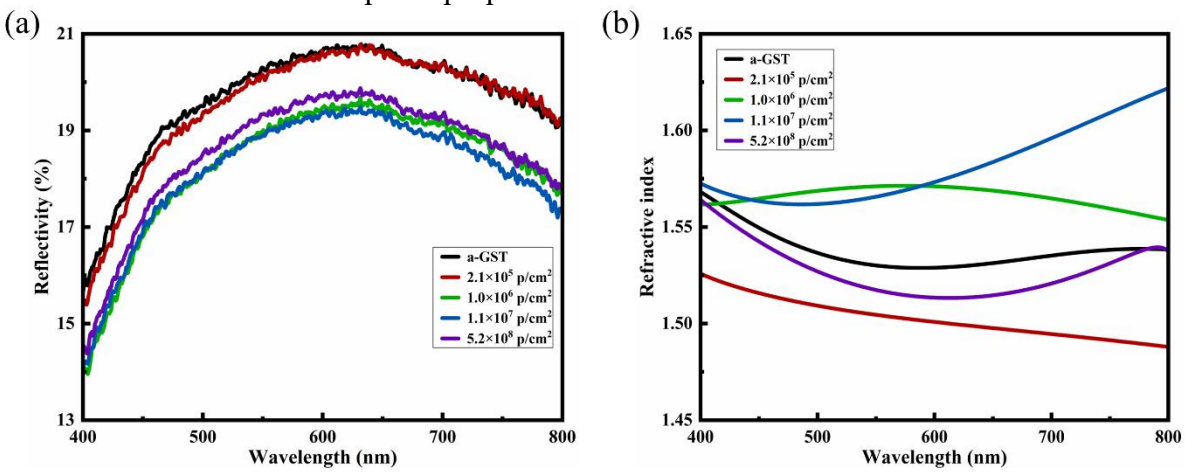

Fig. 3 (a)The reflectivity of a-GST films before and after neutron irradiation. (b) The refractive index of a-GST films before and after neutron irradiation.

4. Summary

In summary, 300 KeV neutrons radiation primarily led to the breaking of Ge-Te and Sb-Te bonds in a-GST films, resulting in changes in reflectivity and refractive index. Moreover, the refractive index was more influenced by the bonding state. The recognition of storage states relies on optical properties, but the radiation-induced changes are not significant enough to trigger a “1” state error.

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