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Effects of swift heavy ion irradiation parameters on optical properties of muscovite mica

S.X. Zhang [a,b,*], J. Liu [a], J. Zeng [b], Y. Song [a], D. Mo [a], H.J. Yao [a], J.L. Duan [a], Y.M. Sun [a], M.D. Hou [a]

[a] Institute of Modern Physics, Chinese Academy of Sciences (CAS), Lanzhou 730000, PR China
[b] University of Chinese Academy of Sciences, Beijing 100049, PR China

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ABSTRACT

Muscovite mica sheets with a thickness of 25 μm were irradiated by various kinds of swift heavy ions (Sn, Xe and Bi) in HIRFL (Heavy Ion Research Facility in Lanzhou). The fluences ranged from 1×10^{10} to 8×10^{11} ions/cm^2 were applied. The electronic energy loss $(dE/dx)_e$ in mica was changed from 14.7 to 31.2 keV/nm. The band gaps and Urbach energy of pristine and irradiated mica were analyzed by ultra-violet–visible spectroscopy (UV–Vis). A red shift of the absorption edge was found in the absorption spectra of muscovite mica irradiated by ions with increasing $(dE/dx)_e$. The results show that the chemical bonds between Tetrahedral–Octahedral–Tetrahedral (TOT) layers of mica were destroyed by ion irradiation. With increasing $(dE/dx)_e$ and fluences, the band gaps became narrow and the Urbach energy increased. It suggests that the amount of defects and the proportion of amorphous structure were increased in mica irradiated with increasing $(dE/dx)_e$ and fluences.

1. Introduction

Muscovite mica is a well-known mineral material with a layered structure. It is sensitive to irradiation damage. The surface of mica is easy to cleave to obtain a smooth plane, and it is widely used as a substrate material [1,2]. A series of studies has been done on the radiation effects of swift heavy ions (SHIs) on mica. Latent tracks on the surface of irradiated mica were firstly observed by Price in 1960s', and the latent tracks have been used to produce the nano-channels after chemical etching [3]. From then on, track etched-pores were applied to prepare a variety of different shapes of nanowires in different diameters [4,5]. Over decades, muscovite mica has played an important role in research of track formations caused by SHIs [6–9], clusters [10,11] or highly charged ions (HCIs) [12].

Latent track was produced by the coupling of several complex processes, including atomic displacement, disturbance and even amorphization. Each of the processes can lead to significant influences on properties of materials, and the crystallographic orientation was changed with respect to the incident ion beam [13]. As a SHI penetrates into the material surface, it transforms its energy first to electrons and then to atoms around its path. Atomic displacements could be generated due to atomic collisions and electronic excitations when $(dE/dx)_e$ was above a certain threshold value. It has been reported in several studies that the track core caused by SHIs irradiation was mainly amorphous structure [8,9]. Several theories have been established to describe the formation and size of latent tracks. The thermal spike model has shown good agreement with experiment data in the track radii of several materials.

Structural modification caused by SHI irradiation such as the formation of defects and amorphous phase was an important factor that affected the materials optical properties. A large number of studies have been focused on the band gap of irradiated materials [14–19]. Sukhnandan Kaur et al. investigated the optical properties of natural phlogopite mica after irradiated by 80 MeV oxygen ions. They found a systematic reduction in both the direct and indirect optical band gap with increasing ion fluences from UV–Vis spectra [18]. Early in 1951, Popper analyzed the absorption coefficient for natural muscovite [19]. Dahr et al. observed transmittance spectral of Indian muscovite mica as the wavelength ranged from 300 to 1000 nm [20]. However, the work on the optical properties of muscovite mica irradiated by SHIs is not available at present. The aim of our work is to get insight into the radiation effects of SHIs on the structural modification and optical properties of muscovite mica. A series of mica sheets were irradiated by various heavy ions with different electronic energy losses $(dE/dx)_e$ and ion fluences.

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[*] Corresponding author at: Institute of Modern Physics, Chinese Academy of Sciences (CAS), Lanzhou 730000, PR China.

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The band gaps and Urbach energy were obtained from the UV–Vis spectroscopy of the irradiated samples. The influences of irradiation parameters on optical properties were discussed.

2. Experimental

Muscovite mica ($KAl_2[AlSi_3O_{10}]\ (OH)_2$) sheets with a thickness of 25 μm were irradiated by SHIs accelerated by HIRFL. Irradiation parameters including ion energy, ion fluences and electronic energy loss $(dE/dx)_e$ and the mean projected range R_p in mica calculated by SRIM-2008 code were shown in Table 1. Prior to irradiation, mica sheets were cut into small pieces about $5 \times 5\ mm^2$. Aluminum foils with different thicknesses were fixed in front of mica sheets to adjust ion energy on the sample's surface.

The UV–Vis transmission spectroscopy (Lambda 900, Perkin–Elmer) was performed on the pristine and the irradiated mica. The analytical wavelength was in the range of 200–1000 nm with a resolution of 1 nm. The direction of the incident light was perpendicular to the surface of the samples. The transmission spectra were transferred into absorption spectra. The direct band gap, indirect band gap and Urbach energy were obtained by analyzing these spectra.

3. Results

Fig. 1 shows the absorption spectra of pristine and irradiated mica. A red shift was observed in the irradiated sample. The absorption edge was shifted from about 305 nm towards higher wavelength.

The UV–Vis absorption spectra were employed to investigate the structure transition and defect information in crystalline and non-crystalline materials. The optical band gap of mica can be calculated using Tauc's expression [21] which relates the absorption coefficient α and the incident photon energy $h\nu$ by the following equation:

$$\alpha(h\nu) = \frac{B(h\nu - E_g)^n}{h\nu} \tag{1}$$

where B is the correction coefficient. E_g is the band gap of the material. When $n = 1/2$ and 2, Eq. (1) stands for the direct band gap and indirect band gap, respectively. The absorbance coefficient $\alpha(h\nu)$ and band gap E_g are calculated by formula (2) and (3) as following:

$$\alpha(h\nu) = \frac{2.303A}{l} \tag{2}$$

$$E_g = \frac{hc}{\lambda} \tag{3}$$

where l is the thickness of the sample. A is the absorbance. h is Planck's constant. c is the speed of light. λ is the related wavelength. Plots of $[\alpha(h\nu)]^{1/2}$ and $[\alpha(h\nu)]^2$ vs. E_g were drawn according to the equations. The values of n give the best linear fits in the lower energy absorption regions. For instance, for a direct band gap, $n = 1/2$ gives the best linear fit. While for indirect band gap, $n = 2$ gives the best linear fit. Once the linear fits were obtained, the band gap energy of the pristine and irradiated mica can be then determined from the intercept of the linear fits in the lower energy absorption region of the plots, as shown in Figs. 2 and 3.

Urbach energy was defined for the irregularities in the band gap level, and the analysis was carried out in the present work to compare the degree of defect generation and amorphization between the pristine and the irradiated mica. It can be calculated according to the Urbach rule [22]:

$$\alpha(h\nu, T) = \alpha_0 \exp\left(\frac{\sigma h\nu - E_0}{kT}\right) \tag{4}$$

where α_0, σ and E_0 are the parameters related to material properties. The logarithm of Eq. (4) is given below:

$$\ln \alpha = \frac{h\nu}{E_{Ur}} + C \tag{5}$$

here, E_{Ur} is the Urbach energy. The dependence of logarithm of α on photon energy of pristine and irradiated mica is depicted in Figs. 2 and 3(c). The Urbach energy E_{Ur} can be calculated by taking the reciprocal of the slopes of the linear portion of the plots of $\ln \alpha$ vs. $h\nu$. The values of the band gap and Urbach energy of the pristine and irradiated mica are shown in Table 2.

Table 1
Irradiation parameters applied in the experiment.

Ions	Energy/(MeV)	$(dE/dx)_e$/(keV/nm)	Fluence/(ions/cm^2)	Projected range/(μm)
Sn	250	14.7	1×10^{11}	12.3
Xe	2020	15.8	1×10^{11}	102.4
	1500	18.0	1×10^{10}	79.5
	1500	18.0	1×10^{11}	79.5
	1500	18.0	8×10^{11}	79.5
Bi	1390	30.9	1×10^{11}	53.6
	1250	31.2	1×10^{11}	49.1

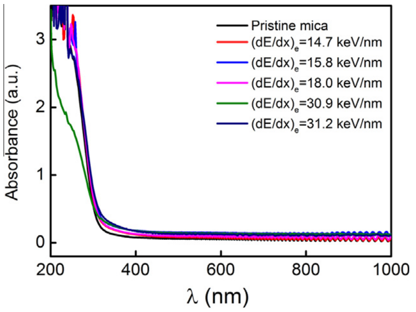

Fig. 1. Ultraviolet absorption spectra of muscovite mica before and after irradiated by SHIs with different $(dE/dx)_e$ where the fluence was 1×10^{11} ions/cm^2.

4. Discussion

Muscovite mica is a two dimensional layered silicate mineral. Concluded from the chemical formula of muscovite, one-fourth of Si atoms are replaced by Al atoms forming the aluminum silicate. Another part of Al atoms are bonded to O atoms or OH forming octahedral structure, and a single octahedral is sandwiched between two identical tetrahedral $(Si, Al)_2O_5$, as shown in Fig. 4. Trilayer aluminosilicate sheets are not electrically neutral, and a layer of K atoms are located between the trilayer aluminosilicate to balance the charge. It is liable to cleave mica as the ionic bonding between the K layers and the trilayer aluminosilicate is weak [23]. As SHIs penetrated into the samples, the energy would convert to atoms around the path via ionization and electronic excitation processes. The chemical bonds between K atoms and the trilayer aluminosilicate sheets were destroyed. The perfect layered structure ceased to exist. The bond cleavage and reconstruction in irradiated mica caused the red shift of the absorption edge.

Concluded from Figs. 2, 3 and Table 2, the direct and indirect band gap of irradiated mica were gradually narrowed down with increasing $(dE/dx)_e$ and ion fluences, while the values of Urbach energy were enlarged in reverse. When $(dE/dx)_e$ was 15.8 keV/nm, the direct band gap and indirect band gap decreased from 4.19 and 3.79 eV to 4.14 and 3.73 eV, respectively. While as $(dE/dx)_e$ increased to 31.2 keV/nm, the direct and indirect band gap were reduced to 3.85 and 3.51 eV, and the values of Urbach energy