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Raman investigation of lattice defects and stress induced in InP and GaN films by swift heavy ion irradiation

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ABSTRACT

InP crystals and GaN films were irradiated by swift heavy ions ^{86}Kr and ^{209}Bi with kinetic energies of 25 and 9.5 MeV per nucleon and ion fluence in the range 5×10^{10} to 3.6×10^{12} ions/cm^2. The characteristic optical bands were studied by Raman spectroscopy to reveal the disorder and defects induced in the samples during the irradiation process. The crystallinity of InP and GaN was found to be deteriorated after irradiation by the swift heavy ions and resulted in the amorphous nature of the samples along the ion tracks. The amorphous tracks observed by transmission electron microscopy (TEM) images confirmed the formation of lattice defects. In typical F_2(LO) mode, in case of InP, the spectra shifted towards the lower wavenumbers with a maximum shift of 7.6 cm^{-1} induced by 1030 MeV Bi ion irradiation. While in case of GaN, the typical E_2(high) mode shifted towards the higher wavenumbers, with maximum shift of 5.4 cm^{-1} induced by 760 MeV Bi ion irradiation at ion fluence of 1×10^{12} ions/cm^2. The observed Raman shifts reveal the presence of lattice defects and disorder induced in the samples after irradiation by the swift heavy ions. This irradiation also generated lattice stress in the samples, which has been investigated and discussed in detail in this work.

1. Introduction

Indium phosphide (InP) and Gallium nitride (GaN) both are direct bandgap semiconductor materials with unique physical and electronic characteristics. They are used in wide range of applications including microwave and short-wavelength optoelectronic devices [1,2]. In 1986, Aksun et al. for the first time reported new type of indium phosphide substrate that successfully produced the micro gate-length high electron mobility transistor (HEMT) [3]. Later on in 1993, the first high-brightness blue light emitting diode (LED) based on GaN epitaxial layer structures was successfully developed [4]. Owing to the high electron mobility of InP, it is considered as one of the most suitable candidates to be used as a substrate material in the field optical fiber communications [5]. Similar to Si-based devices, InP- and GaN-based electronic devices are capable of surviving and working efficiently in radiation environment. High-energy heavy ion irradiation induces defects and stresses in the materials that can alter their structures and modify their physical properties [6–8]. This can severely influence the efficiency and reliability of the devices after irradiation by the heavy ions. Considerable efforts have been made to understand the irradiation effects on these two materials. During the irradiation disordered regions as well as large number of defects are induced in InP and GaN irradiated with Xe and Ni ions [9–11]. The severity and structure of the damage produced in InP by 250 MeV Xe-ion irradiation depend on the ion fluence. At small ion fluences only point defects and their complexes were observed, while with large ion fluences discontinuous tracks were detected [12]. In another work it was found that the roughness of the surface increases from 1.4 to 5.8 nm as GaN films were irradiated by 100 MeV Ni ions when the fluence was increased from 1×10^{12} to 5×10^{13} ions/cm^2 [13]. For InP crystal the value was found to change from 0.33 to 7.49 nm after irradiated with 100 MeV ^{56}Fe ions with increasing ion fluences [14]. Moreover, some investigations were carried out to explore defects and amorphous ion-tracks in InP crystals as a result of the energy deposited along the ion trajectories [15–18]. Wesch et al. observed that 593 MeV Au ion irradiation of InP crystals produced an amorphous layer at the end of the ion trajectory [19]. While Zhang et al. found that with increasing irradiation ion

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fluences, separate amorphous layers develop from the surface of GaN films and near the damaged peak region when irradiated by 2 MeV Au ions [20]. In another work, Kamarou et al. investigated damage produced in InP irradiated with 140 MeV Kr ions by Rutherford backscattering (RBS), which revealed significant defects that are decreased in the pre-damaged InP [21]. Darowski et al. carried out a study analyzing of the surface crystallinity and amorphization of InP irradiated with 390 MeV Xe ions, in which they got information about the structure of large volume of the material by using X-ray diffraction technique [22].

Raman spectroscopy is one of the most powerful techniques to analyze the electronic and dynamic properties of semiconductors, and provides information about the phonon vibrations in the lattices. Due to these vibrations the phonon frequency associated with lattice stresses changes as a result of the irradiation and causes shift in the Raman spectra. The narrow LO mode (the F_2(LO) in InP and A_1(LO) in GaN) shows good crystallinity and the low doping concentration of the materials. Up till now, several reports have focused on the irradiation effects on InP and GaN investigated by Raman spectroscopy [23,24]. One of the study performed on GaN films irradiated with 308 MeV Xe ions showed the blue-shift of E_2(high) mode from 576 to 588 cm^{-1} for the ion fluences in the range 1×10^{13}–3×10^{13} ions/cm^2. While the GaN films irradiated with 100 MeV Ag ions resulted in the red-shift of E_2(high) mode with increasing ion fluences [25,26]. The purpose of this study was to check the Raman shift in GaN and explore the energy and fluence conditions for the red or blue shift in the spectra. To the best of our knowledge, a systematic study of the Raman spectra has not been carried out to study the response of ion irradiation on bandgap of different materials. Another question arises that how the bandgap influences the radiation-resistance in these materials. To answer to these questions, single crystal InP and GaN films were irradiated by swift heavy ions (SHIs) with varying energies and ion fluences in this work. Residual stresses were obtained from the Raman shift in F_2(LO) mode for InP and E_2(high) mode for GaN. The underlying mechanisms for phonon vibration modifications and variation in crystallinity of the materials were analyzed in detail in this work.

Table 1
Basic properties of InP and GaN at 300 K.[16,27]

Material	Nature of energy gap	Energy gap E_g (eV)	Electron mobility [cm^2/(V s)]	Melting point T_M (K)	Thermal conductivity [W/(cm K)]	Bond energy (eV)
GaN	Direct	3.36	900	2790	1.5	2.24
InP	Direct	1.34	4600	1335	0.7	2.07

2. Experimental methods

Single-crystalline (100) InP and (0001) GaN epitaxial layers were irradiated with ^{86}Kr and ^{209}Bi ions of initial kinetic energy 25 and 9.5 MeV/u, respectively. The ion fluences between 5×10^{10} and 3.6×10^{12} ions/cm^2 were applied. The thicknesses of the InP and the GaN films were 500 μm and 30 μm. Some selected basic and physical properties of the investigated materials are presented in Table 1. The table data are mainly from Refs. [16,27]. The wafers were cut into small pieces of 5×5 mm^2. The irradiation experiments were performed at Heavy Ion Research Facility in Lanzhou (HIRFL). The parameters of the irradiation experiments are shown in Table 2. The energy and the energy loss were calculated by SRIM-2010 code [28]. Aluminum foils with different thicknesses were placed in front of the samples in order to adjust the energy of SHIs, and satisfy the needs of electronic energy loss $(dE/dx)_e$. Ion beam scanning was used to obtain homogeneous irradiation over the whole sample under normal incidence at the room temperature.

Micro-Raman spectra were measured using 532 nm laser as an excitation source in backscattering geometry. The measurements carried out on a Horiba JY-HR 800 micro-Raman spectrometer fitted with 1800 lines mm^{-1} grating. The spectral resolution in the Raman spectra is about 0.65 cm^{-1}. Further, some samples selected on the basis of the Raman spectra were additionally studied using transmission electron microscopy (TEM). Samples were prepared for cross-sectional (XTEM) and plane view (PV-TEM) observations performed with electron microscope (FEI Tecnai G2 TF-20) operating at 200 kV. The cross-sectional specimen was prepared using a focused ion beam system (FEI Helios 600).

3. Results and discussions

3.1. InP Raman spectra and TEM images

Fig. 1 shows the Raman spectra of pristine and irradiated InP with Kr and Bi ions of different energies. The pristine Raman spectrum shows a strong longitudinal optical F_2(LO) phonon mode at 350.1 cm^{-1} and a weak transverse optical F_2(TO) phonon mode at 310.5 cm^{-1}. After irradiated by Kr ions, a red-shift in F_2(LO) phonon peak was observed with increasing $(dE/dx)_e$ values as shown in Fig. 1(a). A maximum peak shift of 2.2 cm^{-1} was observed for 12.7 keV/nm $(dE/dx)_e$ value at a fluence of 1×10^{11} ions/cm^2. At the same time, red-shift of F_2(LO) phonon peaks were also observed in samples irradiated by Bi ions with different ion fluences as shown in Fig. 1(b). In the figure, it is seen that the intensity of the peaks decreases gradually as the ion fluence increases.

Table 2
Irradiation parameters in the experiment. $(dE/dx)_e$ and $(dE/dx)_n$ denote the electronic and nuclear energy losses, respectively, as calculated with the SRIM-2010 code [28].

Material	Ion	Energy/(MeV)	$(dE/dx)_e$/(keV/nm)	$(dE/dx)_n$/(10^{-2} keV/nm)	Projected range/(μm)	Fluence/(ions/cm^2)
InP	^{86}Kr	275	12.7	1.9	28.3	1×10^{11}–1×10^{12}
		428	12.1	1.3	40.5	
		585	11.4	1.0	53.9	
	^{209}Bi	1030	31.4	6.6	45.0	5×10^{10}–3.6×10^{12}
		1540	30.4	4.7	61.6	
GaN	^{86}Kr	175	19.0	4.1	13.6	1×10^{11}–1×10^{12}
		488	17.9	1.7	30.4	
		765	15.9	1.1	46.8	
	^{209}Bi	760	46.8	1.2	24.3	5×10^{10}–3.6×10^{12}
		1390	46.4	7.1	37.6	
		1540	45.9	6.5	40.9	