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Damage production in silicon carbide by dual ion beams irradiation

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ABSTRACT

Lattice damage and evolution in single crystalline 6H-SiC under Si + He successively dual ion beams irradiation is studied by using Raman spectroscopy, high resolution X-ray diffraction (HRXRD) and nano-indentation tests. Single Si and He ion irradiations are also performed for the comparison. The results of Raman spectra reveal that the damage level increases with the fluence. A normal strain profile along the ion path is generated due to ion irradiation induced dilation of lattices, contributing mainly by interstitial related defects. Moreover, Si and He ion implantation produced different types of defects. The damage and chemical bonding states are significantly changed after He atoms implanted in Si pre-irradiated samples. Si + He dual ion irradiations increase the damage level further, resulting in changes of the damage states because of complex defects interactions. The nano-hardness of irradiated SiC is combined results of hardening effects of some kinds of defects and the breakdown of covalent-bonds. The mechanical properties present significant differences between single Si, He and Si + He successively dual ion beam irradiations, due to defects evolution during the irradiation process.

1. Introduction

Ion irradiation effects in silicon carbide (SiC) have been extensively studied in recent decades for its potential use in the field of nuclear fuel applications, due to its excellent physicochemical properties. Significant results have been obtained concerning damage accumulation, swelling, annealing and microstructural evolutions in SiC [1–8]. However, concomitant irradiations including neutrons and alpha recoils as well as fission fragments will cause complex damage to materials used in nuclear systems. As a consequence, the investigation of multi-ion beam irradiations in materials used in nuclear systems is desired to understand the irradiation effects in harsh and complex environment comprehensively [9]. In view of the fact that cooperative and/or competitive effects may be induced in many practical circumstances concomitant irradiations, two kinds of possible synergistic effects need to be studied. One synergistic effect is between slow and swift particles, and the other one is between H/He and the defects induced by heavy ion irradiation. Actually, the former synergistic effect is about the effect between nuclear collisions (S_n) and electronic excitations and ionizations (S_e) [8,10–14]. Channeling results demonstrate a strong decrease of the damage in MgO and SiC induced by S_n/S_e cooperation in dual-beam irradiations, while no such effect is observed for c-ZrO_2 and $Gd_2Ti_2O_7$ [10,11]. It can be ascribed to inelastic ionization which can effectively anneal pre-existing defects and restore the structural order in SiC [14]. As for the other possible synergistic effect, the combined effect between H/He and the defects induced by heavy ion irradiation is such an issue that needs to be explored.

Most of the work focused on effects of heavy ion (like Au) irradiations or inert gases (like He) effects in SiC [1–8]. However, the interactions between heavy ion irradiations and inert gases have drawn attention [15–17]. For example, the mechanical properties of a polycrystalline β-SiC material irradiated by dual-ion beams have been investigated [16]. The authors found radiation-induced hardening effect is larger by simultaneous irradiation of Si and He ions than that by single Si ion irradiation. The effect is related to temperature and duration of irradiation. In addition, the synergistic effect of H and He gas elements with Si ion implantation on microstructural evolution in SiC/SiC composite is also a scientific

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interest [17].

The aim of this work is to provide demonstration of dual ion beams irradiation effects in 6H-SiC single crystals induced by heavy self-ions (Si) and gas atoms (He) and investigate the damage production mechanism. Synergetic effects of Si/He ion irradiation were studied by comparison of single and dual ion beam irradiations. The damage was evaluated by combining Raman spectroscopy, high resolution X-ray diffraction (HRXRD) and nano-indentation. The complementarity of these techniques used in this work demonstrated that defects induced by heavy ions have strong interactions with He and He related defects.

2. Experimental

350 μm thick n-type 6H-SiC single crystals were implanted with 100 keV He ions at fluences of $1 \times 10^{14}/cm^2$, $1 \times 10^{15}/cm^2$, $1 \times 10^{16}/cm^2$ and $1 \times 10^{17}/cm^2$ and 600 keV Si ions at fluences of $1 \times 10^{14}/cm^2$ and $2 \times 10^{15}/cm^2$, respectively. Si + He successively dual ion irradiations with the fluences corresponding to single ion irradiations were performed. All the irradiations were done at room temperature (RT). The distribution of damage induced by Si implantation and the He ion concentration versus implantation depth according to SRIM 2011 (Stopping and Range of Ions in Matter) full cascade simulations [18] are shown in Fig. 1. The threshold displacement energy for C and Si sub-lattices used are 20 and 35 eV, respectively [19]. The mean projected range of 100 keV He implantation, R_p, is of about 460 nm. Fig. 1 also shows that the damage peak corresponds to 0.06 dpa (displacement per atom) for 600 keV Si implantation at a fluence of $1 \times 10^{14}/cm^2$ (referred to as Si0.06dpa). The peak He concentration is 0.8 at.% for a fluence of $1 \times 10^{16}/cm^2$ (referred to as He1E16). All the tests presented in this work can probe the entire thickness of irradiated layers.

Raman spectra were acquired at RT with a Horiba JY LabRAM ARAMIS apparatus at a spectral resolution of 0.7 cm^{-1}. The excitation laser radiation was a 532 nm line. HRXRD measurements were carried out with a D8 Discovery in the Bragg reflection geometry to study the lattice strain and the evolution of the defects. A Cu $K_{\alpha 1}$ incident beam of wavelength $\lambda = 1.5405$ Å with instrumental resolution 0.005° in 2θ was adopted during the tests. In addition, the ion irradiation induced nano-hardness changes were taken in continuous stiffness measurement mode using a Nano Indenter G200. A diamond Berkovich indenter was adopted in the measurements.

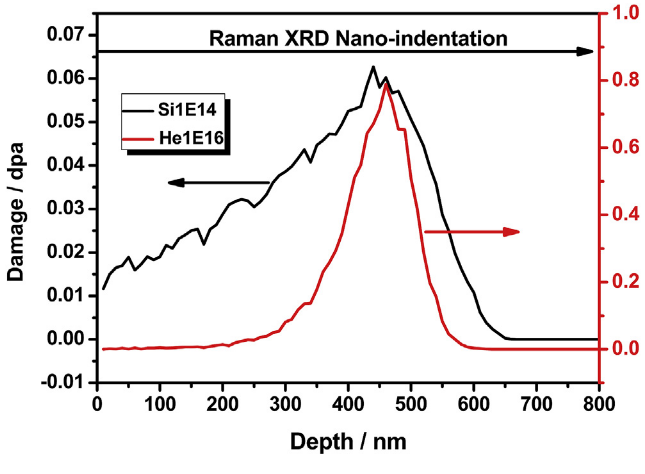

Fig. 1. Depth distributions of He concentration at a fluence of $1 \times 10^{16}/cm^2$ and displacement damage induced by Si ions in SiC at a fluence of $1 \times 10^{14}/cm^2$.

3. Results and discussion

Fig. 2 illustrates the Raman spectra recorded on 6H-SiC irradiated with Si and He ions separately at different fluences. It can be seen that, either for He or Si, the damage degree increases with the fluence till nearly full amorphization is achieved. Si-Si and C-C interactions are clearly seen by He implantation at a fluence of $1 \times 10^{17}/cm^2$ sample (He1E17) and Si implantation at a fluence of $2 \times 10^{15}/cm^2$ sample (Si1.2dpa). With Si-C bond breaking down during irradiation, Si-Si and C-C interactions are formed. Compared He1E16 with He1E17, Si-Si is easier to be formed than C-C bonding, which may be due to the higher cutoff distances for Si-Si bonding [20]. In addition, Si-C disordered/distorted or amorphous signals are also observed, as shown in Fig. 2. The lattice undergoes a distortion during irradiation and the structural relaxation of the lattice occurs [21]. The damage states are dependent on the ion species, fluences and irradiation temperatures.

The Raman spectra of Si + He dual ion beam irradiated samples are presented in Fig. 3. It seems that the He implantation will increase the damage in Si ions pre-implanted SiC. Total disorder is calculated to compare the damage induced by single and dual ion beams for low fluences [22]. It is found that the value of total disorder is 0.417 and 0.073 for Si0.06dpa and He1E14 samples, respectively. And we get 0.573 for Si0.06dpa + H1E14, which is larger than the sum of total disorder values of Si0.06dpa and H1E14, implying dual ion beam irradiation leads to a higher damage level. Though the long range order may be vanished in heavily damaged SiC, some degree of short range order (SRO) may still remain and could continue to decrease during further irradiation. The SRO parameter describes the chemical state corresponding to the local arrangement atoms [21].

The chemical disorder, defined as the ratio of C-C to Si-C bonds, can be used to evaluate the damage state when the damage level is high. Fig. 4 displays the chemical disorder estimated for single and dual ion irradiated SiC by He and Si ions. It can be seen that dual ion irradiations induced more damage than single ions in the present case. The chemical disorder value increases rapidly when the damage level is high, as shown in Fig. 4. The total disorder and chemical disorder are good indications of damage evolution and disorder accumulation. The structural evolution in the present case is in agreement with [21].

In order to fathom the microstructural evolution induced by single and dual ion beam irradiations, HRXRD measurements are performed. Fig. 5 shows X-ray curves obtained after single He and Si ion implantations. It can be seen that satellite diffraction peaks appear at lower angles beside the main Bragg peak θ_{Bragg}, which are corresponding to the disordered part of the implanted samples. The new signals originate at the lower angle side indicate an increase of the interplanar distance in the direction perpendicular to the sample surface. The satellite peaks move to lower angle indicates that more damage is induced by the ion implantation with higher fluence. Note that the interference fringes are observed obviously for the sample He1E16, which is resulted from the same strain area beside the most damaged region. Velisa et al. found that the shape of the strain profile is very similar to the SRIM-predicted He depth distribution in 30 keV He implanted cubic zirconia [23]. On the other hand, Leclerc et al. calculated the strain profile, which is more proportional to the nuclear energy loss profile other than He depth distribution in 160 keV He implanted 4H-SiC [3]. And again, this conclusion is supported by our result shown in Fig. 6, where the strain depth profile is compared to both the nuclear energy loss and the helium concentration in 6H-SiC implanted with 100 keV He ions at $1 \times 10^{16}/cm^2$. These observations demonstrate that a non-homogeneous strain depth profile arises, which is in accordance with the damage profile [3,23]. In addition, a high plateau ascribing