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Irradiation-induced amorphization in β-SiC

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Abstract

Single-crystal (001) β-SiC (3C) films have been irradiated with 360 keV Ar^{2+} ions at 175, 310 and 375 K and the damage accumulation after each incremental fluence has been measured in situ by Rutherford backscattering spectroscopy in channeling geometry (RBS/C) along the [011] direction using the dual beam facilities within the Ion Beam Materials Laboratory at Los Alamos National Laboratory. The relative rate of disordering along [011] decreases with increasing temperature. The critical dose for the relative disorder to reach the random level along [011] is 0.35 dpa (17.5 eV/atom), 0.44 dpa (22.0 eV/atom) and 0.61 dpa (30.5 eV/atom) at 175, 310 and 375 K, respectively. The sigmoidal increase in relative disorder with dose is consistent with defect accumulation processes. Although RBS/C along [011] indicates a fully random layer at the highest dose for each temperature, ex situ RBS/C and XTEM along [001] indicate some residual crystallinity remains near the surface and the end of range. © 1998 Elsevier Science B.V.

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1. Introduction

Silicon carbide (SiC) has outstanding physical and nuclear properties that make it an important technological material for electronic devices and nuclear applications. A fundamental understanding of irradiation effects in SiC is required to effectively utilize ion-implantation techniques in device fabrication and to predict performance in nuclear environments. Cubic β-SiC (3C polytype) is one of the SiC polymorphs of interest for these applications. Ion-beam-induced amorphization in β-SiC has been studied by Rutherford backscattering spectroscopy in channeling geometry (RBS/C) [1–3] and cross-sectional transmission electron microscopy (XTEM) [2–5]. For irradiation at low temperatures, damage annealing may occur prior to (or during) transfer, preparation and characterization at room temperature. In situ electron microscopy techniques have been used to study the temperature dependence of amorphization under 1.5 MeV Xe^{+} irradiation in polycrystalline β-SiC [6] and under 2 MeV electron irradiation in single crystal β-SiC thin films [7]; however, quantitative details of the damage accumulation as a function of cumulative dose are not easily determined by these experiments. Recently, in situ RBS/C techniques have been used in the study of amorphization in single crystal α-SiC wafers [8,9]. In the present paper, these in situ RBS/C methods are used to measure the dose and temperature dependence of damage accumulation in single-crystal β-SiC thin films irradiated with 360 keV Ar^{2+} ions. Some preliminary results have been previously reported [10].

2. Experimental procedures

The n-type (001) β-SiC (3C polytype) single-crystal films (5 μm thick) were grown on Si substrates and obtained from Cree Research. The quality of these films is illustrated by the high-resolution XTEM image in Fig. 1, which shows the atomic structure at the β-SiC/Si interface. The irradiation experiments at 175, 310 and 375 K were carried out with 360 keV Ar^{2+} ions at a flux of 2×10^{12} ion/cm^2 s using the dual-beam facilities within the Ion Beam Materials Laboratory at Los Alamos National Laboratory [11,12]. The two beamlines for the ion

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irradiation and the RBS/C analysis are at a relative angle of 60°, which required that both the ion irradiations and the RBS/C measurements be carried out at fairly large angles off the surface normal. The single crystal specimens were oriented such that the irradiation with 360 keV Ar^{2+} ions (10 mm beam spot size) were performed at ~ 15° off the [001] axis. An in situ RBS/C analysis was obtained on each virgin specimen at the irradiation temperature and after each incremental ion dose, using a 2 MeV He^{+} ion beam (2 mm spot size) incident along the [011] channeling direction and a backscattering angle of 165°. Along [011] in these specimens, the measured values of χ_{min} (i.e. ratio of the backscattering yield in the virgin spectrum to the yield in the random spectrum, just below the surface peak) were 0.010, 0.012 and 0.015 at 175, 310 and 375 K, respectively. Although the [011] orientation is not the optimum direction for determining the dose and depth dependence of amorphization in β-SiC by channeling [2], it was necessary in this study given the constraints of the experimental geometry. The irradiations at each temperature were continued until the aligned backscattering yield in the RBS/C spectra reached the random level over the entire irradiated depth, from the surface to the end of range.

After completion of the in situ study, post-irradiation (ex situ) RBS/C along [001] was also carried out on each sample specimen, since this direction provides a more accurate profile of the damage accumulation [2]. In these specimens, the measured value of χ_{min} along [001] was 0.016 at room temperature. The irradiated specimens were also analyzed by XTEM at the High-Resolution Analytical Electron Microscopy Laboratory (University of New Mexico) using a JEOL 2010 microscope operated at 200 kV. The XTEM specimens were prepared by gluing a Si wafer to the surface of the irradiated crystal films, cutting cross-sections of the samples (perpendicular to the irradiated surface), and using a tripod polishing technique to thin the specimens prior to ion milling.

The depth distribution of atomic displacements produced per ion under the irradiation conditions for both the

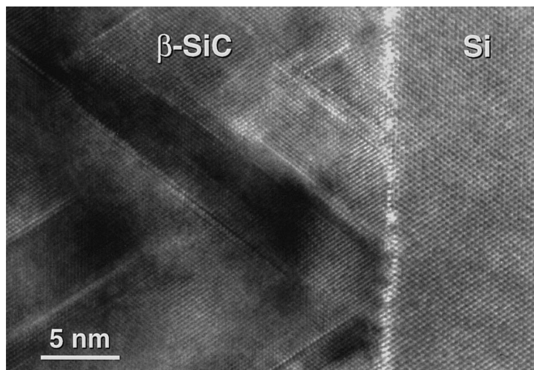

Fig. 1. High resolution XTEM image at β-SiC/Si interface of starting samples.

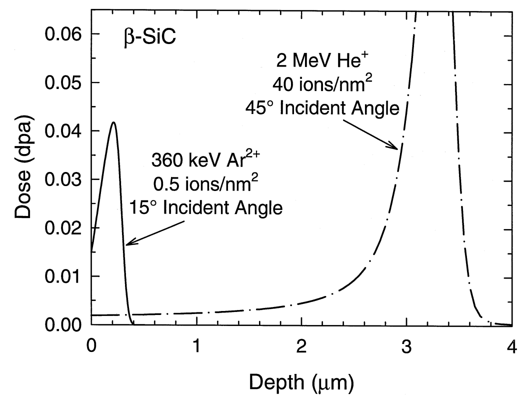

Fig. 2. Damage distributions in β-SiC irradiated to incremental fluences with 360 keV Ar^{2+} ions at 15° incident angle and with 2 MeV He^{+} ions at 45° incident angle (calculated using TRIM-96).

360 keV Ar^{2+} ions and 2 MeV He^{+} ions was determined from full cascade Monte Carlo calculations using TRIM-96 [13] and assuming a displacement energy of 25 eV for both Si and C. The calculated distribution of displaced atoms in β-SiC under the irradiation conditions employed are shown in Fig. 2 for the smallest incremental fluence of 360 keV Ar^{2+} ions and for a typical incremental fluence for each 2 MeV He^{+} (RBS/C) analysis. Each RBS/C spectra contributed 2×10^{-3} dpa to the total incremental dose from the 360 keV Ar^{2+} ions at depths less than 400 nm. Since the 2 MeV He^{+} probe contributes slightly to the overall cumulative damage produced by the 360 keV Ar^{2+} ions during the in situ experiments, the total dose reported below, in units of displacements per atom (dpa), includes the contribution from the RBS/C He^{+} beam. (Note: a previous study of β-SiC by the authors [6] used TRIM-95, which underestimated the displacements produced as a result of an error in the calculation of the stopping powers for SiC.)

3. Results and discussion

3.1. In situ RBS/C along [011]

The irradiation sequence involved irradiating the selected area with an incremental fluence of 360 keV Ar^{2+} ions followed by an RBS/C analysis. The irradiation sequence at 175 K included 13 irradiation steps and 14 RBS/C spectra. At 310 K, 19 irradiation steps and 20 spectra were taken, while at 375 K, 18 irradiation steps and 19 spectra were taken. Several of the sequential RBS/C spectra taken during irradiation at 175, 310 and 375 K are shown in Figs. 3–5, respectively, as a function of ion fluence. Also shown for each specimen are the unirradiated (virgin) aligned spectrum and a random spectrum, which is obtained by rocking the incident He beam a

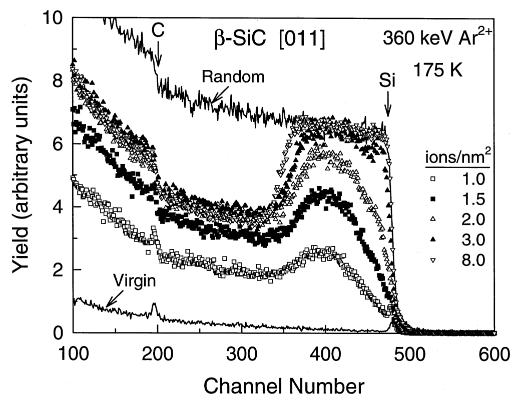

Fig. 3. Several of the aligned backscattering spectra obtained in situ along [011] from β-SiC irradiated sequentially with 360 keV Ar^{2+} ions at 175 K. Also shown are virgin and random spectra.

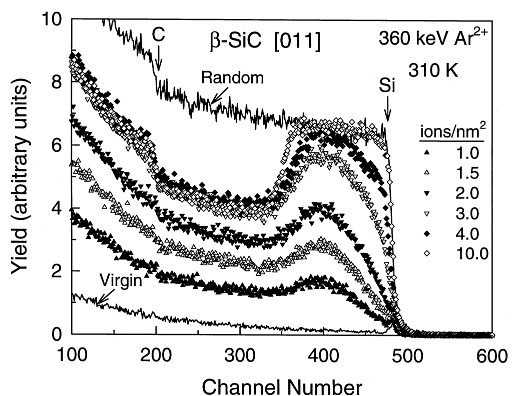

Fig. 4. Several of the aligned backscattering spectra obtained in situ along [011] from β-SiC irradiated sequentially with 360 keV Ar^{2+} ions at 310 K. Also shown are virgin and random spectra.

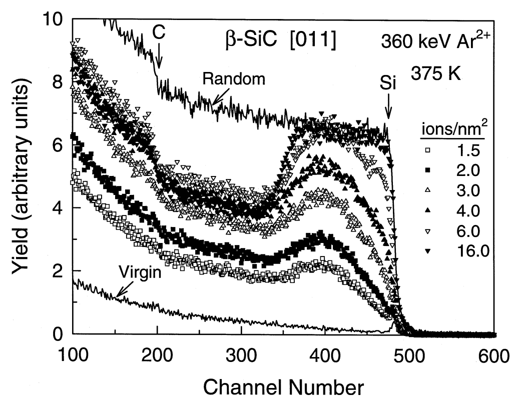

Fig. 5. Several of the aligned backscattering spectra obtained in situ along [011] from β-SiC irradiated sequentially with 360 keV Ar^{2+} ions at 375 K. Also shown are virgin and random spectra.

few degrees off the channeling direction. The increase in the backscattering yield from the Si atoms provides a profile of the distribution of displaced Si atoms on the Si sublattice. At 175 K, the aligned backscattering yield from the Si reaches the random level at a fluence of 3.0 Ar^{2+} ions/nm^2. The fluence at which the aligned backscattering yield from Si first reaches the random level increases with temperature to 4.0 and 6.0 Ar^{2+} ions/nm^2 for the irradiations at 310 and 375 K, respectively. Much higher fluences (nearly a factor of two) are required for the surface regions to reach the random level. At the higher fluences for each temperature, the backscattering yield from the C atoms also noticeably increases and is consistent with a random distribution at the highest fluences. (There is no discernible loss of C in the near-surface spectra.) At intermediate fluences where the peak in the backscattering yield is near or at the random level, a small peak in the backscattering yield was observed just below the surface (e.g. at 3.0 ions/nm^2 in Fig. 3, 6.0 ions/nm^2 in Fig. 5). This peak is similar to that reported by Edmond et al. in β-SiC [1,2], by Weber et al. in α-SiC [9] and by Holland et al. in Si [14] and is evidence for misalignment of the residual crystalline material in front of the buried amorphous layer with respect to the aligned crystalline substrate beyond the amorphous layer. This misalignment is the result of the large volume change (17%) associated with amorphization in β-SiC [4].

Based on the approximately linear relationship between the energy of the backscattered He^+ ions and depth [15] (corrected for the incident angle and assuming a constant specimen density in the damage layer), a profile of the backscattering yield from the Si atoms for each RBS/C spectra has been obtained as a function of depth normal to the surface, which is referenced to the density of the undamaged state of crystalline SiC. (The actual depth profile at each fluence will depend on the specimen density profile in the irradiated layer, which is not easily determined.) The relative Si disorder as a function of depth has been determined from the depth profiles of the backscattering yields of aligned and random spectra, along the [011] direction, using the procedure described by Holland et al. [16]. The depth distributions of the relative Si disorder for several ion fluences at 175, 310 and 375 K are shown in Figs. 6–8, respectively. The relative disorder in the damage layer increases with ion fluence at all temperatures and becomes coincident with the fully disordered or random level (i.e. relative disorder = 1.0) at a depth corresponding to the peak in the ion-damage profile. As the ion fluence increases further, the width of the fully disordered region increases and eventually consumes the entire irradiated layer. As discussed by Edmond et al. [2], reaching the random level for channeling along the [011] direction in β-SiC may not correspond to achieving a fully amorphous state. An anomalous peak in the relative disorder, due to misalignment of the near surface crystalline material, is observed at a depth of about 40 nm once the peak in the

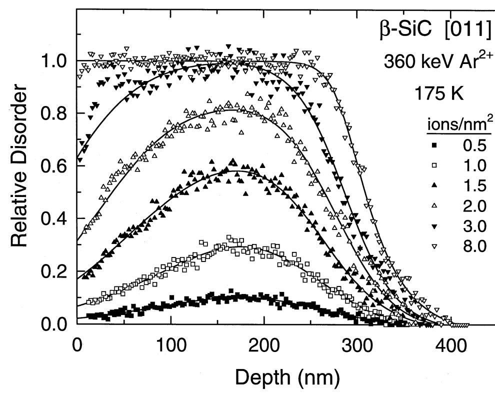

Fig. 6. Profiles of the relative Si disorder along [011] as a function of depth (referenced to undamaged crystalline state) in β-SiC irradiated at 175 K.

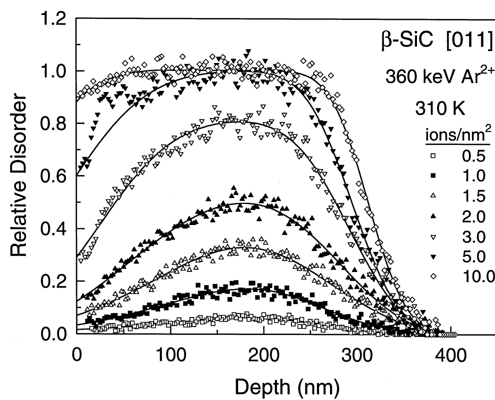

Fig. 7. Profiles of the relative Si disorder along [011] as a function of depth (referenced to undamaged crystalline state) in β-SiC irradiated at 310 K.

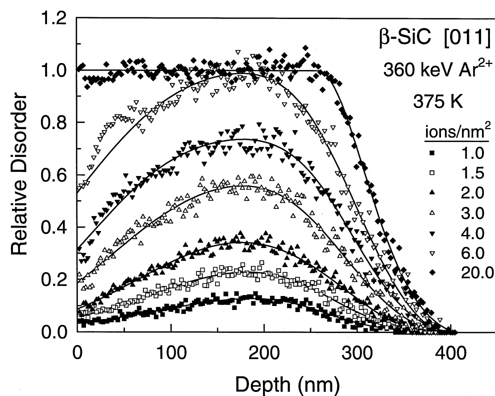

Fig. 8. Profiles of the relative Si disorder along [011] as a function of depth (referenced to undamaged crystalline state) in β-SiC irradiated at 375 K.

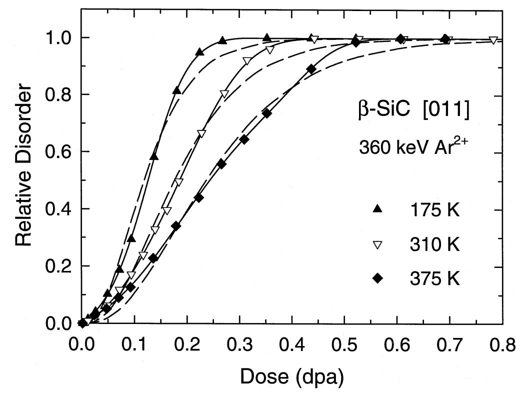

Fig. 9. Relative Si disorder, determined at the peak in the damage profile, as a function of displacement dose for β-SiC irradiated at 175, 310 and 375 K. (Solid curves are polynomial smooth fits to data, while the dashed curves are fits of the double-overlap model [18], Eq. (1), to the data.)

depth profile of the relative disorder reaches 1.0 (the random level), as illustrated by the depth profile for 3.0 ions/nm^2 at 175 K (Fig. 6), 5.0 ions/nm^2 at 310 K (Fig. 7) and 6.0 ions/nm^2 at 375 K (Fig. 8). This near-surface peak was excluded in the curve fitting analysis.

3.2. Temperature dependence of damage accumulation

The ingrowth of the relative Si disorder at both the surface and the depth corresponding to the peak in the damage profile have been determined for each spectrum. When compared on a basis of the calculated displaced atom dose (dpa), the relative disorder along [011] at the surface and at the depth of peak damage (175 nm) exhibited a similar dependence on dose at each temperature, as described previously [10], suggesting a minimal effect of the surface on the disordering process as observed along [011]. The relative Si disorder at the peak in the damage profile is shown in Fig. 9 as a function of displacement dose, at the damage peak, for each irradiation temperature. The solid curves are smooth fits to the data. The relative disorder at all temperatures exhibits a sigmoidal-like dependence on dose that suggests a defect accumulation process, which is consistent with high-resolution transmission electron microscopy observations of the amorphization process in β-SiC [6] and with MD simulations of energetic cascades in β-SiC [17]. As the temperature increases, the rate of disordering decreases. This behavior may be due to increasing defect interactions, recovery and/or migration with increasing temperature.

It has been previously shown [9] that the relative Si disorder in α-SiC irradiated with 360 keV Ar^{2+} ions at 170 K follows the double-cascade overlap model for amorphization [18], which is given by the expression

$$f_a = 1 - \left[\left(1 + BD + B^2D^2/2\right)\exp(-BD)\right], \qquad (1)$$

where f_a is the amorphous fraction, B is related to the effective damage volume per ion and D is the dose. The results in Fig. 9 do not closely follow this model, as illustrated by the nonlinear regression fits of the model (dashed curves) to the data. In particular, as the relative Si disorder approaches the random level, the measured disorder along [011] is larger than that predicted by the model. This difference may be partially due to the orientation effect observed by Edmond et al. [2] in β-SiC, and discussed below, which suggests that near the random level the relative Si disorder measured along [011] in β-SiC overestimates the amorphous fraction. The effect of accumulated damage on this difference has not yet been reported. The values of B determined from the fits of the double overlap model to the data are 22.5, 15.1 and 11.3 dpa^{-1}, respectively. The decrease in B suggests a decrease in the effective damage volume (defect concentration) with increasing temperature.

From the results in Fig. 9, the critical dose for the relative disorder to reach the random level, as measured along the [011] direction, is estimated to be 0.35 dpa (17.5 eV/atom), 0.44 dpa (22.0 eV/atom) and 0.61 dpa (30.5 eV/atom) at 175, 310 and 375 K, respectively. These values are within 10% of the critical amorphization dose values expected in β-SiC at these temperatures, based on the results of Edmond et al. [2] and Weber and Wang [6], as illustrated in Fig. 10. (The dose calculation for the results of Weber and Wang [6] have been corrected using TRIM-96.)

3.3. Ex situ RBS/C along [001]

After completion of the in situ study, the irradiated specimens were analyzed (ex situ) by RBS/C at room temperature along the [001] direction for comparison to the results obtained from the RBS/C along [011] at the highest ion fluence for each specimen (temperature). Virgin and random spectra along [001] were also taken. Based on the results of Edmond et al. [2], the [001] direction should yield a more accurate depth profile of the damage accumulation. Using the same procedure as above, the depth profile of the relative Si disorder was determined from the aligned and random spectra for each specimen. The results, which are shown in Fig. 11, indicate that along [001] the fully random level is not achieved in the near surface region and at the end of range, which is contrary to the results along [011] but in general agreement with the results of Edmond et al. [2]. This behavior is illustrated more clearly in Fig. 12, which compares the depth profiles measured along [001] and [011] in the specimen irradiated at 175 K to 8 ions/nm^2.

3.4. XTEM results

Post-irradiation XTEM was performed on the specimens studied in situ with RBS/C; consequently, the im-

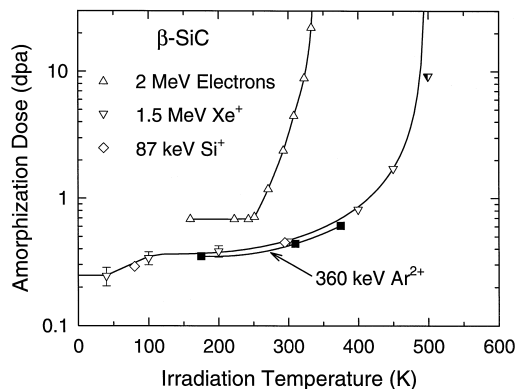

Fig. 10. Temperature dependence of the critical dose to amorphize β-SiC for 360 keV Ar^{2+} (this study), for 1.5 MeV Xe^{+} [6] (dose corrected using TRIM-96) and for 2 MeV electrons [7].

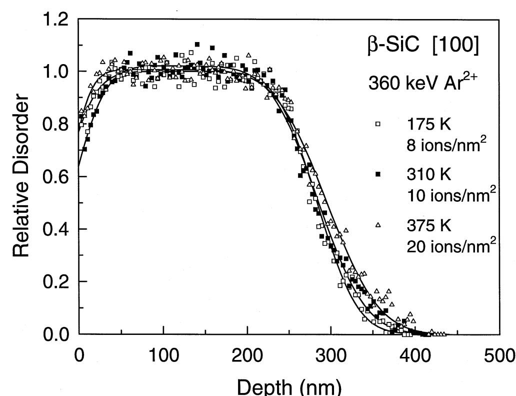

Fig. 11. Post-irradiation profiles of the relative Si disorder along [001] as a function of depth (referenced to undamaged crystalline state) in β-SiC irradiated at 175, 310 and 375 K.

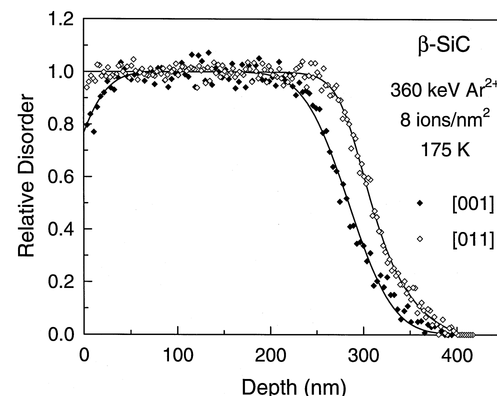

Fig. 12. Comparison of depth; profiles of relative Si disorder determined in situ along [011] and ex situ along [001].

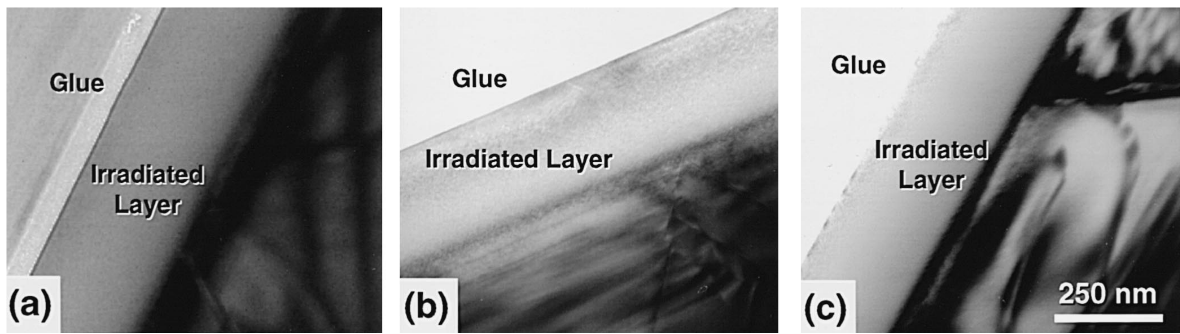

Fig. 13. XTEM images of specimens irradiated with 360 keV Ar^{2+} to (a) 8 ions/nm^2 at 175 K, (b) 10 ions/nm^2 at 310 K and (c) 20 ions/nm^2 at 375 K.

ages correspond to the maximum dose where the RBS/C results along [011] suggest a fully disordered state from the surface to the end of range. XTEM micrographs, parallel to [001], of the specimens irradiated at 175, 310 and 375 K are shown in Fig. 13. The damage layers at this magnification appear amorphous. However, HRTEM indicates that some residual crystallinity is retained at the highest doses in regions near the surface, as illustrated in Fig. 14 for the specimen irradiated at 175 K to 8.0 ions/nm^2. This behavior is consistent with the above RBS/C results along [001] and with the observations of Edmond et al. [2]. Following a procedure described previously [9], the average volume expansion due to radiation-induced disorder has been estimated from the thickness of the damage layer (referenced to the undamaged state) determined from the RBS/C data and the layer thickness measured by XTEM. The average volume expansion at 175 K is estimated to be 20%, which is in reasonably good agreement with the volume change due to amorphization that has been reported for α-SiC [4,9,19].

4. Summary

In situ RBS/C techniques have been used to study the accumulation of ion-beam-induced damage in β-SiC along the [011] direction. The relative disorder along [011], as measured by RBS/C, increases sigmoidally with dose, consistent with disordering occurring by the local accumulation of defects and the rate of disordering decreases with temperature due to defect recovery processes. The critical doses for the relative disorder to reach the random level

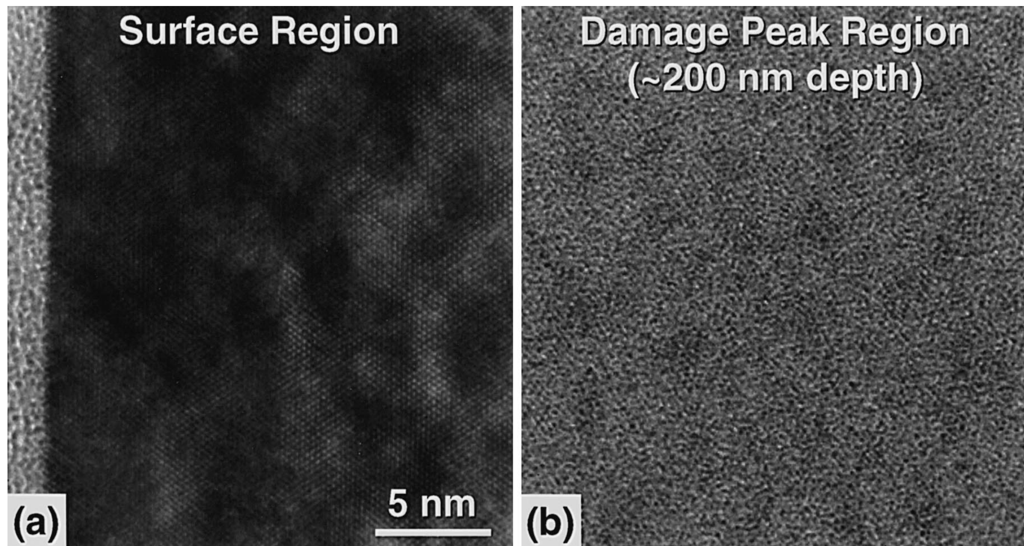

Fig. 14. High-resolution XTEM image of (a) the surface region and (b) the peak damage region in the specimen irradiated with 360 keV Ar^{2+} to 8 ions/nm^2 at 175 K.

along [011] are estimated to be 0.35 dpa (17.5 eV/atom) and 0.61 dpa (~30.5 eV/atom) at temperatures of 175 and 375 K, respectively. These measured critical doses to reach the random level along the [011] direction are within 10% of projected values for amorphization. Although RBS/C along [011] indicates a fully random layer at the highest dose for each temperature, ex situ RBS/C and XTEM along [001] indicate some residual crystallinity remains near the surface and the end of range. The average volume expansion due to the disordering at 175 K is estimated to be 20%.

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References

[1] J.A. Edmond, S.P. Withrow, H.S. Kong, R.F. Davis, in: H. Kurz, G.L. Olson, J.M. Poate (Eds.), Beam Solid Interactions and Phase Transformations, Mater. Res. Soc. Symp. Proc. 51, Materials Research Society, Pittsburgh, PA, 1986, p. 395.
[2] J.A. Edmond, R.F. Davis, S.P. Withrow, K.L. More, J. Mater. Res. 3 (1988) 321.
[3] J.A. Edmond, R.F. Davis, S.P. Withrow, in: J.D. Cawley, C.E. Semler (Eds.), Silicon Carbide '87, Ceramic Transactions, vol. 2, American Ceramic Society, Westerville, OH, 1989, p. 479.
[4] L.L. Snead, S.J. Zinkle, in: I.M. Robertson, L.E. Rehn, S.J. Zinkle, W.J. Phythian (Eds.), Microstructure of Irradiated Materials, Mater. Res. Soc. Symp. Proc. 373, Materials Research Society, Pittsburgh, PA, 1995, p. 377.
[5] S.J. Zinkle, L.L. Snead, Nucl. Instrum. Meth. B 116 (1996) 92.
[6] W.J. Weber, L.M. Wang, Nucl. Instrum. Meth. B 106 (1995) 298.
[7] H. Inui, H. Mori, A. Suzuki, H. Fujita, Philos. Mag. B 65 (1992) 1.
[8] W.J. Weber, L.M. Wang, N. Yu, Nucl. Instrum. Meth. B 116 (1996) 322.
[9] W.J. Weber, N. Yu, L.M. Wang, N.J. Hess, J. Nucl. Mater. 244 (1997) 258.
[10] W.J. Weber, N. Yu, Mater. Sci. Forum 239–241 (1997) 155.
[11] N. Yu, M. Nastasi, T.E. Levine, J.R. Tesmer, M.G. Hollander, C.R. Evans, C.J. Maggiore, Nucl. Instrum. Meth. B 99 (1995) 566.
[12] N. Yu, T.E. Levine, K.E. Sickafus, M. Nastasi, J.N. Mitchell, C.J. Maggiore, C.R. Evans, M.G. Hollander, J.R. Tesmer, W.J. Weber, J.W. Mayer, Nucl. Instrum. Meth. B 118 (1996) 766.
[13] J.F. Ziegler, J.P. Biersack, U. Littmark, The Stopping and Range of Ions in Solids, Pergamon Press, New York, 1985.
[14] O.W. Holland, J.D. Budai, C.W. White, Appl. Phys. Lett. 57 (1990) 243.
[15] L.C. Feldman, J.W. Mayer, Fundamentals of Surface and Thin Film Analysis, North Holland, New York, 1986, p. 48.
[16] O.W. Holland, D. Fathy, J. Narayan, O.S. Oen, Radiat. Eff. 90 (1985) 127.
[17] T. Diaz de la Rubia, M.J. Caturla, M. Tobin, in: I.M. Robertson, L.E. Rehn, S.J. Zinkle, W.J. Phythian (Eds.), Microstructure of Irradiated Materials, Mater. Res. Soc. Symp. Proc. 373, Materials Research Society, Pittsburgh, PA, 1995, p. 555.
[18] J.F. Gibbons, Proc. IEEE 60 (1972) 1062.
[19] C.J. McHargue, J.M. Williams, Nucl. Instrum. Meth. B 80&81 (1993) 889.