Electron diffraction study on radiation-induced chemical disorder in covalent materials

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Amorphous and polycrystalline materials can be analyzed by radial distribution functions in which their atomistic structures are characterized by the probability of finding another atom at a distance between *r* and *r*+d*r* from a specific atom. Radial distribution functions can be obtained by x-ray, neutron, and electron diffraction techniques. Among the diffraction techniques, electron diffraction has the following merits: (1) strong interaction between the material and electrons has the advantage of detecting light atoms; (2) thanks to short wavelength of high-energy electrons, an intensity profile up to high scattering angles is easily obtained; (3) local structures on the nano-scale can be obtained by a combination of other techniques, such as high-resolution electron microscopy and nano-beam spectroscopy analysis. In the present study, we examined radiation-induced chemical disorder in SiC [1] and GaN [2] via radial distribution functions.

4H-SiC single crystals were irradiated at room temperature with 10 MeV Au^{3+} ions to a fluence of 10^{15} Au/cm^2, while GaN thin films were irradiated at ~200 K with 2 MeV Au^{2+} ions to a fluence of 7.35×10^{15} Au/cm^2. The specimens were observed using a JEOL JEM-3000F transmission electron microscope (TEM) at an electron energy of 300 kV. TEM images and electron diffraction patterns were recorded on an imaging plate (Eu^{2+}-doped BaFBr). The intensities of the electron diffraction patterns were analyzed quantitatively using a Digital Micro-Luminography FDL 5000.

Figure 1 shows high-resolution TEM images of ion-beam-induced structures in (a) SiC and (b) GaN. The high-resolution image in Fig. 1(a) shows homogeneous fine 'salt and pepper' contrasts, and no detectable clusters nor phase-separated regions exist. The electron diffraction patterns obtained from the damage region also show typical halo rings of amorphous materials. These results suggest that the constitute elements mix on the atomic-scale in amorphous SiC. On the other hand, both halo rings and Debye-Scherrer rings are observed in the diffraction patterns of ion-irradiated GaN, suggesting that amorphous and poly-nanocrystalline phases are intermixed. In fact, ordered atomic arrangements and 'salt and pepper' contrast regions coexist in the high-resolution TEM image of Fig. 1(b). In addition, the round-shaped contrast due to bubbles can be seen in ion irradiated GaN.

Figure 2 shows reduced radial distribution functions of (a) amorphous SiC and (b) amorphous/nanocrystalline GaN, obtained by a quantitative analysis of electron diffraction intensities. For comparison, the interatomic distances and coordination numbers of crystalline SiC and GaN are also indicated on the abscissa. In Fig. 2(a), prominent peaks are apparent at ~0.19 and ~0.31 nm, corresponding to the first (Si-C: 0.188 nm) and second (Si-C-Si and C-Si-C: 0.307 nm) nearest neighbors in crystalline SiC, respectively. In addition, there exist two subpeaks located at ~0.15 and ~0.23 nm which are comparable to the bond length of C-C and Si-Si atomic pairs, respectively. These results suggest that amorphous SiC networks contain not only heteronuclear Si-C bonds but also homonuclear Si-Si and C-C bonds within the first coordination shell. Like SiC, the amorphous/nanocrystalline GaN contains both the homonuclear and heteronuclear bonds [Fig. 2(b)]. There is a peak at ~0.25 nm between the first and second prominent peaks, which is assigned to Ga-Ga bonds. Although there is a tail on the small *r* side of the first peak, no apparent evidence of N-N bonds is obtained.

We found the existence of the homonuclear bonds in ion-irradiated SiC and GaN, but their origin is essentially different between the two materials. As described above, the constitute elements in amorphous SiC networks are intermixed on the atomic scale, and no prominent phase separation is observed. On the other hand, an inhomogeneous damage layer, including N_2 gas bubbles, is formed in ion-irradiated GaN. Due to the formation of N_2 gas, the number of the Ga-N bonds decreases, and the excess Ga atoms form self-bonded atomic pairs. This means that phase

segregation induced by irradiation plays an important role for the formation of the homonuclear Ga-Ga atomic bonds in ion-irradiated GaN.

References

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[2] M Ishimaru *et al.*, J. Appl. Phys. **106** (2009) 053513: 109 (2011) 043512.
[3] The author would like to thank Prof. Emeritus Hirotsu (Osaka University) for his establishment of a precise quantitative analytical technique of electron diffraction intensities for radial distribution function analysis. Ion irradiated specimens were provided by Profs. Weber and Zhang (University of Tennessee, Oak Ridge National Laboratory).

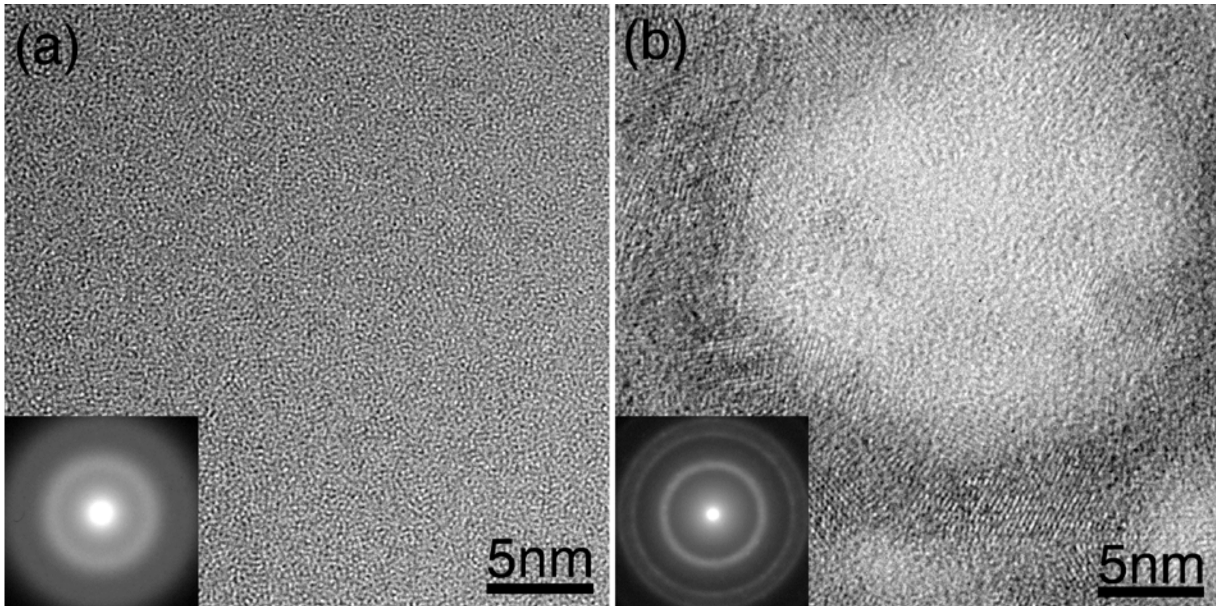

Figure 1. *High-resolution TEM images and electron diffraction patterns of ion irradiated (a) SiC and (b) GaN. Typical 'salt and pepper' contrast due to amorphous phase is observed in ion-irradiated SiC (a), while inhomogeneous contrast consisting of amorphous, nanocrystalline phases, and nitrogen bubbles are apparent in ion-irradiated GaN (b). The appearance of halo rings (a) and Debye-Scherrer rings (b) in the diffraction patterns is consistent with the high-resolution images.*

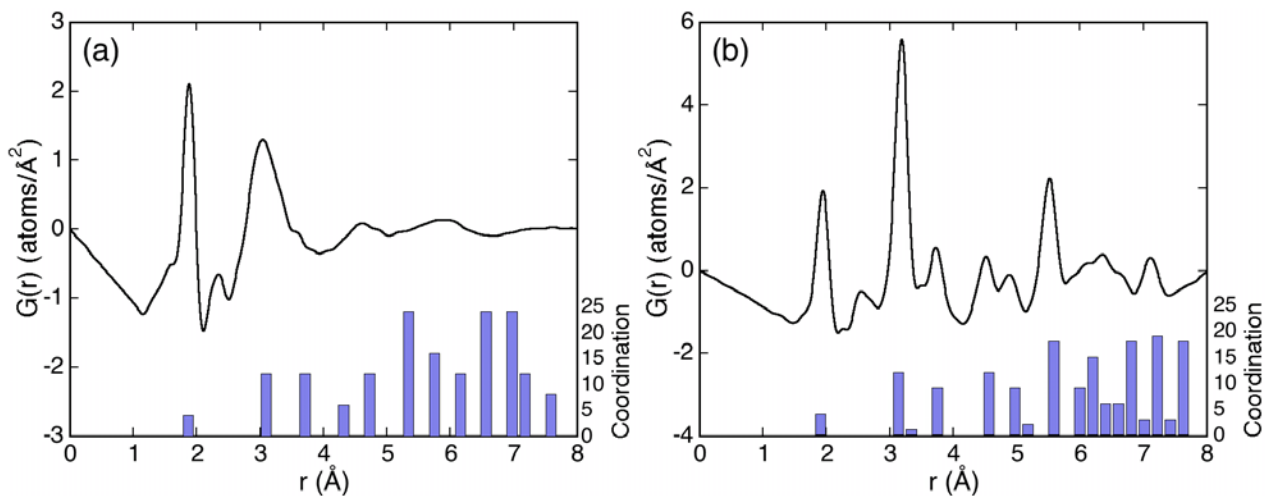

Figure 2. *Reduced radial distribution functions of ion-irradiated (a) SiC and (b) GaN. The vertical bars reveal the coordination number at the distance of r (every 0.02 nm) in crystal SiC and GaN. Additional peaks corresponding to the atomic distance of homunuclear bonds are apparent in both ion-irradiated SiC (~0.15 and ~0.23 nm) and GaN (~0.25 nm).*