Atomic Resolution Imaging of Black Spot Defects in Ion Irradiated Silicon Carbide

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Silicon carbide is of great interest as a nuclear fuel cladding material. At relatively low irradiation temperatures (< 1000 °C) and doses (< 10 dpa or displacements per atom), the major irradiation induced defects are black spot defects (BSD), which appear as nanometer scale black spots in bright field transmission electron microscopy (TEM) images [1,2]. BSDs are associated with radiation-induced swelling [1]. The detailed internal structure of BSD is unknown. We are working towards understanding the structures and evolution of BSD by combining high resolution scanning transmission electron microscopy (STEM) and defect structure modeling [3].

We have irradiated single-crystal 4H-SiC and polycrystalline 3C-SiC with two ion species. One is 3.15 MeV carbon ions to a dose of 5.14×10^{16} at/cm^2 at 870 K, 1070 K, and 1250 K. The other is 1 MeV krypton ions to a dose of 3×10^{14} at/cm^2 and 6×10^{14} at/cm^2 at 870 K, 1070 K. The corresponding damage levels are 0.4 dpa for carbon irradiation at 1 μm implantation depth, and 0.4 dpa / 0.8 dpa for krypton irradiation at 0.3 μm depth. TEM observation was performed at 300 kV in a FEI Tecnai TF30 and STEM observation was performed at 200 kV in a probe C$_s$-corrected FEI Titan.

Low-angle annular dark field (LAADF) STEM imaging are used to obtain atomic resolution images of BSD strain fields [2]. Figure 1a and 1b are two STEM images of a 4H-SiC sample irradiated with 1 MeV Kr ions at 0.4 dpa, 870 K, with 17.5 mrad semi-convergence angle and 29 – 144 mrad collection angle. They were acquired 24 s apart under 200 keV, 3.0×10^7 electrons/nm^2 continuous irradiation. Non-rigid registration eliminated sample drift and other instabilities, and aligned the images in figures 1a and 1b [4]. Many BSDs moved through the crystal and changed shape in two dimensional projection under 200 keV electrons, as shown by arrows pointing to a BSD in Figures 1a and 1b. Tracking through all the intermediate frames confirms that this is the same defect. Figure 1c shows the mean square displacement of 5 BSDs, each tracked for at least 20 s within a 430 s, 5.56×10^8 e$^-$/nm^2 STEM image series. In general, BSDs cannot be tracked for longer times as they either leave the image field or change in contrast too much, potentially due to motion in the depth of the TEM specimen as well as in the plane.

Irradiation-induced diffusion of clusters of high intrinsic migration barrier, as opposed to irradiation releasing clusters from dislocations or other defect "traps", has not been reported before. We have estimated the migration barrier using density functional theory calculations for a small interstitial cluster in SiC [3]. Figure 1d presents the minimum barrier migration path of carbon tri-interstitials in SiC. Within experimental uncertainty, the predicted beam-induced diffusivity of the BSDs is in reasonable agreement within predictions based on the barrier in Figure 1d. Combining HRSTEM study and theoretical structural models will provide valuable information about the structure of BSD [5].

References:

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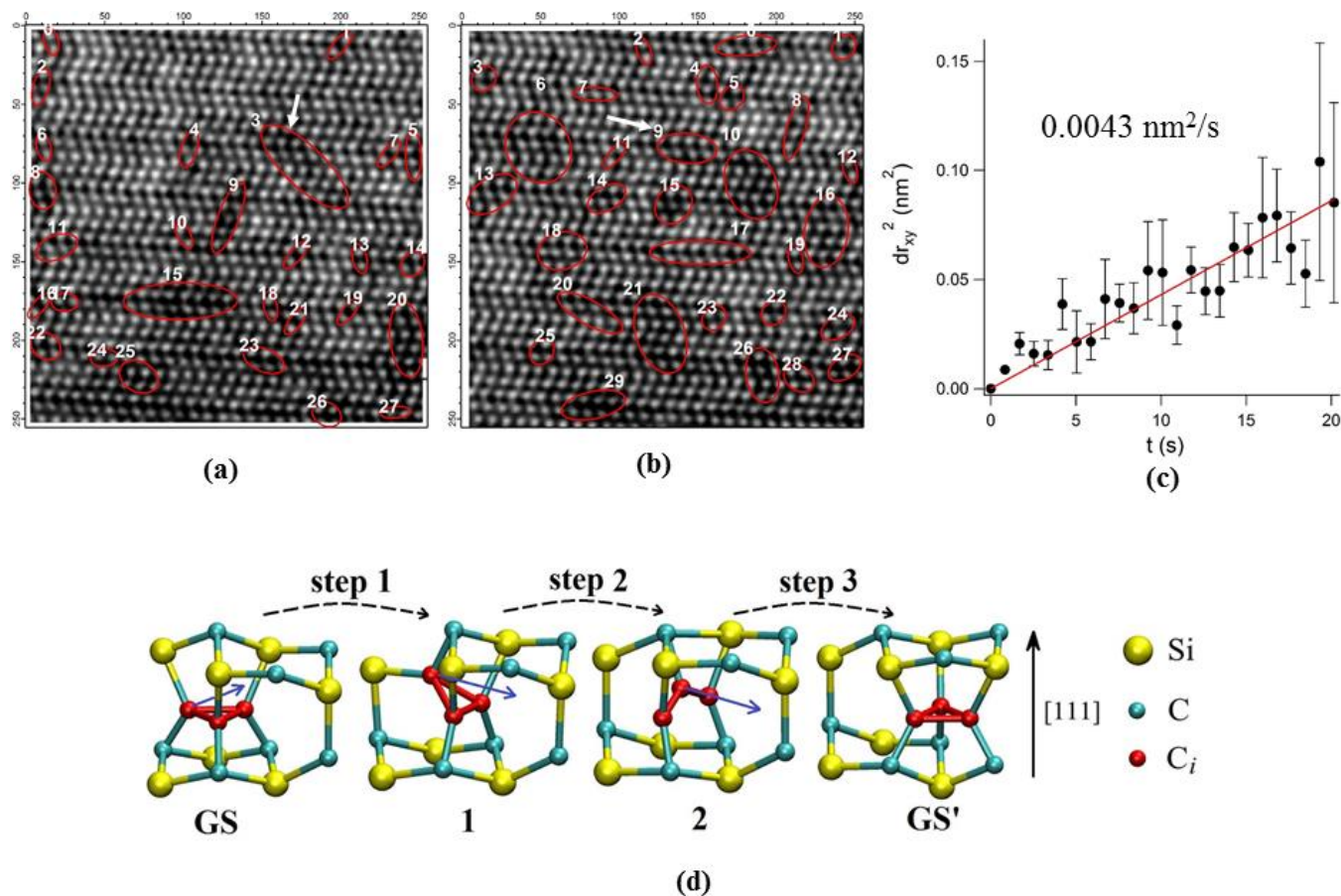Nuclear Energy University Programs under contract number CFP-12-3357.

Figure 1. (a) LAADF STEM image of a 4H-SiC sample irradiated with 1 MeV Kr ions at 0.4 dpa, 870 K. BSDs are circled. The sample is normal to $[11\bar{2}0]$. (b) The same area of (a), after 23.5 s continuous 200 keV, 3.0×10^7 electrons/nm^2 irradiation. (a), (b) are aligned against drifting with non-rigid registration [4]. Arrows indicate displacement of an individual BSD, labeled as # 3 in (a) and # 9 in (b). (a) and (b) have been Gaussian smoothed to reduce noise. (c) Mean square displacement of 5 BSDs. (d) Migration trajectory of a carbon tri-interstitial cluster under electron radiation by ab initio molecular dynamic simulation, assuming 10 eV is transferred to the cluster at each step.