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Diffuse X-ray scattering measurements of point defects and clusters in iron ☆

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Abstract

The nature of residual damage from atomic displacement cascades in iron has been investigated by diffuse X-ray scattering. A single crystal iron sample was irradiated at ~60 °C in the High Flux Isotope Reactor at the Oak Ridge National Laboratory to a fluence of 1×10^{23} n/m^2 ($E > 0.1$ MeV), or ~0.01 dpa. Diffuse X-ray scattering measurements were carried out at the Advanced Photon Source at the Argonne National Laboratory on three specimens: unirradiated, as-irradiated, and irradiated and annealed for one hour at 450 °C (IA). The specimens were cooled to ~40 K to minimize thermal diffuse scattering. Subsequent measurements of the specimen lattice parameter were completed at ORNL. The diffuse scattering in the as-irradiated specimen was dominated by interstitial defect clusters exhibiting a tetragonal distortion consistent with a $\langle 100 \rangle$ type defect. Substantial recovery of this defect component was observed following the anneal at 450 °C. A lattice parameter increase was observed following irradiation, which is also consistent with a substantial population of interstitial type defects. A net lattice parameter decrease was observed in the IA specimen, indicating loss of the interstitial defects with a residual population of vacancy-type defects.

1. Introduction

In recent years, molecular dynamics (MD) simulations of high energy displacement cascades have provided a detailed picture of primary damage formation in irradiated materials [1–7]. An unanticipated observation in these studies was extensive formation of small interstitial clusters that exhibited very high mobility. Two primary stable interstitial configurations are observed in iron, $\langle 110 \rangle$ and $\langle 111 \rangle$ dumbbells [3,4]. Empirical interatomic potentials predict that the difference in the formation energy between these configurations is ~0.1 eV [1–7]. More recent *ab initio* simulations [9] find the $\langle 110 \rangle$ dumbbell configuration to be lower energy than the $\langle 111 \rangle$ dumbbell configuration, but with a much larger difference of 0.7 eV. There is also significant disagreement between model predictions of mobility. MD simulations find the $\langle 111 \rangle$ dumbbell

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migrates with a low activation energy of ∼0.1 eV, whereas the ⟨1 1 0⟩ dumbbell is essentially immobile near room temperature. However, *ab initio* simulations [9] find a very small energy barrier of about 0.05 eV between the ⟨1 1 1⟩ and ⟨1 1 0⟩ configurations. This suggests that the ⟨1 1 1⟩ dumbbell is not stable and is unlikely to play a significant role in defect migration. In either case, ⟨1 1 0⟩ dumbbells are predicted to dominate.

An understanding of the nature and mobility of these small interstitial clusters is important for predicting microstructural evolution. For example, extensive in-cascade clustering and low cluster mobility would give rise to a high cluster density which would act as sinks for other mobile defects. Conversely, if the clusters are as mobile as they appear in the MD simulations, the defect concentrations would be relatively low since they would easily migrate to fixed sinks such as dislocations and grain boundaries [8]. Similarly, the higher mobility of ⟨1 1 1⟩ defects implies that only the ⟨1 1 0⟩ type should be observed following irradiation. Further, although it is well established that by transmission electron microscopy (TEM) that interstitial loops of both ⟨1 0 0⟩ and ⟨1 1 1⟩ type are observed in iron following neutron and heavy ion irradiation [10,11] the mechanism by which the ⟨1 0 0⟩ loops form is not understood.

During the past two decades, the brilliance of the best X-ray sources has been doubling about every nine months. Recent progress in X-ray optics and commissioning of ultra brilliant third-generation synchrotron X-ray sources now allows for intense X-ray beams with 0.1–100 μm diameter spot size. These intense small beams enable experiments that were previously possible only with large single crystals. Previous X-ray analyses of irradiation-induced point defects, defect clusters, and dislocation loops are described in a number of papers [12–20], although studies in iron are not frequent. Small angle scattering [17] and measurements of electrical resistivity [18] have been used for the analysis of radiation induced defects in iron. Small angle scattering is sensitive to variations in scattering-length density but is not sensitive to local distortions. Since interstitial defects in iron cause strong lattice distortions in the surrounding matrix, diffuse X-ray scattering has been applied to their investigation [20]. The purpose of this work is to use X-ray scattering to investigate the residual damage from a low-dose, low-temperature irradiation of single crystal iron, and to compare the experimental observations with simulated scattering patterns to determine which type of interstitial defect cluster is formed. Lattice parameter measurements and the predictions of a kinetic radiation damage model are also used in the analysis.

2. Experimental: material and irradiation conditions

The material to be irradiated was obtained in the form of 0.999 + purity iron single crystal from Goodfellow Corporation. The disk-shaped sample was 2 mm thick and 1 cm in diameter; with a ⟨1 0 0⟩ specimen surface normal to the circular face. The as-received sample was sliced to provide two ∼0.9 mm thick disk specimens. One of these specimens was irradiated at ∼60 °C in a ‘hydraulic tube’ that passes through the core region of the High Flux Isotope Reactor (HFIR) at the Oak Ridge National Laboratory. The fast ($E > 0.1$ MeV) neutron flux in the HFIR axial location used is 8.9×10^{14} n/m^2/s. The exposure time was 3.12 h leading to a fast fluence of 1×10^{23} n/m^2, which corresponds to a dose of ∼0.01 dpa in iron. Following the irradiation, the disk was cut in half and one of the disk halves was annealed at 450 °C for 1 h. Subsequent measurements were carried out on the single crystal iron in three conditions: as-received (AR), as-irradiated (AI), and post-irradiation annealed (PIA).

3. Diffuse X-Ray scattering measurements and scattering simulations

Diffuse X-ray scattering measurements were carried out at the Advanced Photon Source (APS) at the Argonne National Laboratory. A custom aluminum holder was designed and built for the radioactive AI and PIA specimens to avoid contaminating APS equipment while permitting cryogenic examination. Measurements were carried out in the APS UNICAT beam lines 33BM and 33ID with a beam energy of 20 keV. The small beam spot size obtained in this facility, ∼50 μm × 50 μm, permits essentially single-grain measurements even in polycrystalline samples. The effective beam size was larger for these irradiated single crystal specimens, 1 mm × 2 mm, due to the geometry and constraints imposed by the specimen holder. An energy dispersive detector was employed to measure the Compton scattering to aid in background subtraction and calibrate the incident beam intensity.