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In situ X-ray diffraction study of irradiation-induced lattice expansion in Al foils by MeV-energy heavy ions

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ABSTRACT

Using in situ X-ray diffraction measurements, we investigate lattice deformations of a free-standing aluminum foil induced by irradiation with MeV-energy heavy projectiles (C, O, and Si ions). The dependence of the ion-beam flux on the lattice expansion is analyzed in terms of two types of irradiation effects: (i) electronic excitation collision-induced lattice heating and (ii) elastic collision-induced displacement damage. We observe that the change in the lattice parameter is proportional to the energy in lattice heating, irrespective of projectile species. This result is in good agreement with a model calculation for thermal lattice expansion caused by beam heating. Moreover, with the correlation between lattice expansion and displacement damage, we consider a simple model for lattice expansion originating from the accumulation of Frenkel defects. From the model, we obtained the relationship between the relative changes in lattice parameter and the value of displacement per atom (dpa) rate. A comparison of the results from model calculations and experiments shows that the dpa rate calculated from the model, which takes account of athermal defect-recombination, is strongly correlated with the change in lattice parameter. This result suggests that the concentration of surviving defects under irradiation diminishes because of spontaneous recombination of defects produced.

1. Introduction

The effects of energetic-particle irradiation on metallic materials have been extensively studied in nuclear and radiation materials science [1]. Fast heavy-ion irradiation imparts its kinetic energy into lattice atoms of a material via two energy dissipation processes, electronic excitation and nuclear collisions, resulting in lattice heating and displacement defects. This irradiation effect leads to lattice expansion or compaction following radiation disordering or irradiation deformation of materials. Irradiation-induced microstructural strains associated with a change in lattice parameter can be characterized by X-ray diffraction (XRD) analysis [2]. Recently, an ion accelerator-based irradiation apparatus combined with an X-ray diffractometer as an end-station has been developed for investigating the evolutionary dynamics of microstructures under irradiation [3,4], enabling real-time observations of defect accumulation and its dependence on ion fluence. Progress in these investigations makes possible the control of microstructural modifications induced by irradiation with energetic particles.

Ion irradiation induces crystal lattice disorder following various dynamic relaxation processes including defect recombination and aggregation. During irradiation, damage evolution proceeds by the reciprocal processes of defect production and relaxation, and then reaches a certain equilibrium damage state. To date, experimental and computer simulation studies have focused on an understanding of a transient damage state under irradiation [5], and a new model calculation relevant to dynamic annealing has recently been reported [6].

The evolution and relaxation of transient defects that vanish after irradiation play a key role in the dynamic response of a material under irradiation. In our previous studies, we have investigated the deformations of a free-standing metallic foil that occur under irradiation with MeV-energy projectile beams, from shape measurements using a laser displacement meter [7,8]. These results indicated that macroscopically visible foil deformation appears in the beam-spot area under irradiation, and vanishes on timescales of several tens of milliseconds after irradiation ceases; i.e., the deformation is reversible under irradiation and non-irradiation conditions. A length change in the deformed area linearly increases

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with beam flux, suggesting that the driving force is mainly thermal energy induced by beam heating. For high flux irradiation, in contrast, non-linear flux dependence is observed, implying that the length change is caused by not only thermal energy, but also a defect production (synergy effect).

To obtain information on microscopic behavior in reversible foil-deformation, we focused in this study on the change in lattice parameter during deformation using our developed in situ XRD measurements. A high-purity well-annealed aluminum foil with a thickness of 3 μm was used as a target specimen. The target foil was irradiated with three different projectiles (C, O, and Si ions) at MeV energies. We investigated the dependence of the energy flux of a projectile beam on the change in lattice parameter, and a correlation between lattice expansion and energy deposition. We analyze here the results obtained in terms of the deposited energy flux, and discuss a mechanism of lattice expansion originating from thermal stress resulting from the beam-heating effect and the volume changes arising from radiation-defect production.

2. Experimental method

The experiments were performed using a 1.7-MV Cockcroft–Walton-type tandem accelerator of Quantum Science and Engineering Center, Kyoto University. Fig. 1 shows the experimental apparatus for in situ XRD analysis of materials under ion irradiation. This apparatus enables measurements of the expanding lattice during deformations of materials induced by ion irradiation. We chose aluminum foil as the specimen because its thermal conductivity is insensitive to temperature. Rolled aluminum foil has a high purity of 99.999% and a thickness of 3 μm. To remove residual defects or strains originating from rolling, the target foil was annealed at 598 K under vacuum for 10 min. The target was mounted on a sample holder equipped with a device enabling wrinkles or flexures in the foil to be removed (details are described in our previous study [7]). The holder was placed on a goniometer in a vacuum chamber.

A free-standing aluminum foil was irradiated with three different projectiles, specifically 2.5-MeV C, 3.0-MeV O, and 4.3-MeV Si ions. The ion beams were carefully collimated to have a uniform intensity distribution and a spot size of diameter 1.5 mm on the target. The range of these projectiles was almost the same as the foil thickness, achieving then an ion-irradiation region extending over the target depth. Irradiation conditions for each projectile are summarized in Table 1. In XRD measurements, we employed a conventional point-focusing diffractometer (XRD; RINT 2000; Rigaku Co. Japan) operated at 5 kW (50 kV, 100 mA) for Cu K radiation to obtain a high intense X-ray microprobe. The X-ray beam

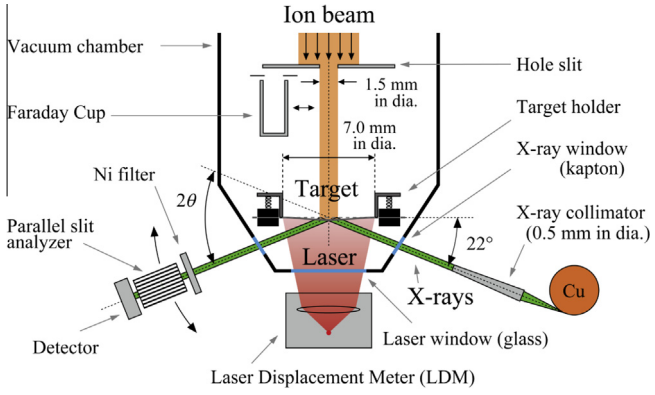

Fig. 1. Schematic of the experimental setup for in situ measurement of X-ray diffraction under ion irradiation.

Table 1
Irradiation parameters: E_0 is the incident projectile energy, E_T/E_0 is the ratio for the energy converted from E_0 as thermal energy E_T, and ϕ is the ion-beam flux.

Projectile	E_0 (MeV)	E_T/E_0 (%)	ϕ (ions cm^{-2} s^{-1})
C	2.5	99.9	3.8×10^{12}–7.0×10^{13}
O	3.0	99.8	2.6×10^{13}–4.9×10^{13}
Si	4.3	99.7	2.8×10^{12}–5.6×10^{13}

was collimated to a spot size of diameter 0.5 mm, and entered the target at a 22° angle of incidence to a target surface in vacuum. Therefore, the beam spot on the target has an elliptical shape (the major and minor radii of the ellipse being 1.3 mm and 0.5 mm). Thus, the X-ray beam was incident within the ion-irradiation area. Diffracted X-ray beam optics consists of a Ni filter for suppressing the Cu K_β X-rays (monochromatization), a parallel slit analyzer, and a scintillation counter detector. This optical configuration of X-ray beam enables the prevention of a shift or broadening of the diffraction peak resulting from a rough sample surface or a displacement of sample position. XRD spectra were taken at 2θ diffraction angles from 43.9° to 45.3° with a step interval of 0.01° and a scanning rate of 4 s per step. XRD scanning was performed in the 2θ scan mode, i.e., the X-ray incident angle was fixed at 22° with respect to the target surface during the diffraction-angle scan. In each measurement of the diffraction spectrum, the total time to collect data was 560 s, according to scanning rate and scanning range. To keep good counting statistics, the total counts in the diffraction peak area measured before irradiation were $\geqslant 10^5$. The overall experimental uncertainty for lattice parameter measurements arising from the limitation of the parallel slit analyzer opening angle (0.01°) was ±0.02% or less.

3. Results and discussion

Fig. 2 shows a typical example for a change in diffraction peak from Al (200) plane observed before, during, and after irradiation from 4.3-MeV Si ions at a flux of 3.3×10^{13} ions cm^{-2} s^{-1}. In terms of the diffraction peak position, intensity, and width, one can see the following characteristic features. First, peak position observed for un-irradiated specimen was around 2θ = 44.71°, which corresponds to a lattice parameter (a_0) of 4.049 Å calculated from Bragg's law. This result is in good agreement with the standard value (4.050 Å) taken from Ref. [9] and within experimental uncertainty. In calculating the lattice parameter using Bragg's law, the wavelength for the incident Cu Kα radiation, λ, was taken as

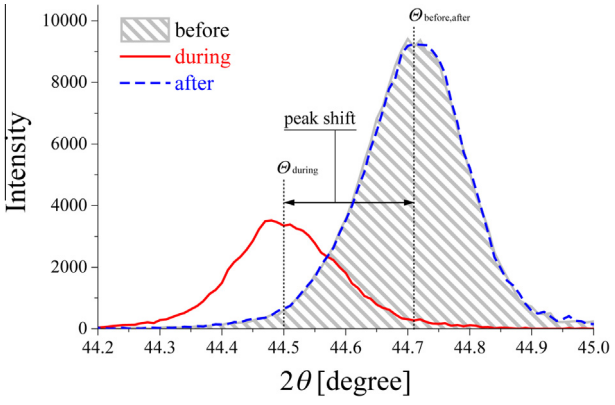

Fig. 2. Typical example of Al (200) diffraction spectra observed during irradiation of 4.3-MeV Si ions at flux of 3.3×10^{13} ions cm^{-2} s^{-1} (solid curves), and after irradiation (dashed curves). The peak of shaded area indicates the spectrum for non-irradiated specimens (before irradiation).