

СЕКЦИЯ 3
ВЛИЯНИЕ ИЗЛУЧЕНИЙ НА СТРУКТУРУ
И СВОЙСТВА МАТЕРИАЛОВ
SECTION 3
RADIATION INFLUENCE ON THE STRUCTURE
AND PROPERTIES OF MATERIALS

**OPTIMISED EXPERIMENTAL WORKFLOW TO MONITOR
RECOVERY OF SWIFT HEAVY ION DAMAGE IN CRYSTALS**

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In this study, the recrystallisation dynamics of swift heavy ion (SHI) tracks have been investigated in single crystal yttrium iron garnet ($\text{Y}_3\text{Fe}_5\text{O}_{12}$ –YIG). The material was irradiated at room temperature with 167 MeV Xe ions to a fluence of $2 \times 10^{11} \text{ cm}^{-2}$. An optimised experimental protocol was developed to monitor the thermal recovery of ion tracks in real time with high temporal and spatial resolution. Plan-view TEM lamellae were prepared with the focused ion beam-scanning electron microscopy (FIB-SEM) and mounted onto the Si_3N_4 MEMS heating chips via FIB lift-out. In-situ TEM annealing was performed by synchronising frame capture from the high-speed camera and heating from the MEMS chip. This procedure was optimised by a custom graphical interface developed in Python to start and stop the camera and heating holder simultaneously, and select the appropriate parameters before the experiment could commence. The results obtained from in-situ BF TEM have been aligned and processed with automated threshold-based procedures that identified individual ion tracks. Other Python modules, such as Trackpy, were also introduced to monitor >150 tracks across the dataset from which measurements, such as equivalent track diameters, were extracted. The tracks are observed to be amorphous due to the uniform contrast and absence of lattice fringes. Large temperature steps have induced shifts and slight bending within the lamella; therefore, a two-stage heating strategy has been introduced to maintain high-contrast imaging, namely pre-heating to 773 K for final zone-axis alignment, then ramping to 823 K. This work demonstrates a streamlined approach to observe ion track recrystallisation on a nanoscale, which could help improve the development of radiation-resistant materials.

Keywords: swift heavy ion irradiation; ion tracks; yttrium iron garnet; recrystallization; in-situ TEM; FIB.

Introduction

Research on improving nuclear reactor materials to solve the radiation stability problems of inert matrix fuel hosts and coated fuel particles has become widely recognised as significant and frequently pursued due to the imminent energy crisis [1]. Fission fragments generated within reactors induce structural defects that result in degradation in material properties and lead to reduced efficiency or failure within nuclear systems. Swift heavy ions (SHIs) can be used to simulate similar conditions found for materials exposed to fission fragments in nuclear reactors [2]. Numerous investigations

have reported that when YIG is irradiated with SHIs; continuous, cylindrical amorphous ion tracks are observed within the material [3]. Due to the properties of YIG, namely its well-known cubic crystal structure and strong resistance to electron beam damage, which is necessary for prolonged in-situ TEM experiments, it was deemed an excellent candidate for studies involving transmission electron microscopy (TEM). Recrystallization dynamics during in-situ TEM annealing have not been reported on to date. Monitoring the crystallisation dynamics in-situ allows for the identification of parameters affecting recovery rate, such as crystallographic

orientation and stress state, which is affected by sample thickness, neighbouring or overlapping tracks, the crystal's anisotropic mechanical properties, and thermally induced bending of the sample.

This work aims to directly monitor and quantify the recrystallisation of ion tracks in YIG through in-situ TEM. As well as determine the main variables affecting track recovery, such as beam exposure and heating conditions, optimising sample preparation and data analysis with Python scripts to create a robust framework for studying defect evolution in real time within irradiated materials and offer insights into the recovery processes within material structures.

Experimental

Single crystal (111) YIG was irradiated at room temperature using 167 MeV Xe ions ($S_e \approx 24.9$ keV/nm near the surface) to a fluence of 2×10^{11} cm⁻² at the Joint Institute for Nuclear Research (JINR) in Dubna, Russia. FIB lift-out was performed with an FEI Helios Nanolab 650 to extract lamellae from ~ 1 μ m below the irradiated surface and place them on Si₃N₄ MEMS heating chips (DENS Solutions Wildfire). Imaging was performed during in-situ heating in BF TEM mode on a JEOL ARM 200F at 200 keV. Frames were captured on a 256x256 pixel Quantum Detectors MerlinEM detector using, 10 ms exposure time.

Results and Discussion

The SHI tracks are observed within the irradiated YIG specimen in the plan-view BF TEM micrograph shown in Figure 1 as bright features, each ~ 10 nm in diameter. Figure 2 represents the same region after in-situ heating up to 550 °C. The amorphous tracks are no longer visible, confirming complete recrystallisation. Bend contours are visible post-annealing due to thermally induced stresses causing slight off-axis tilting in the lamella. BF TEM imaging, in-situ thermal annealing, and feature tracking tools were combined to obtain visual and quantitative information regarding recrystallisation.

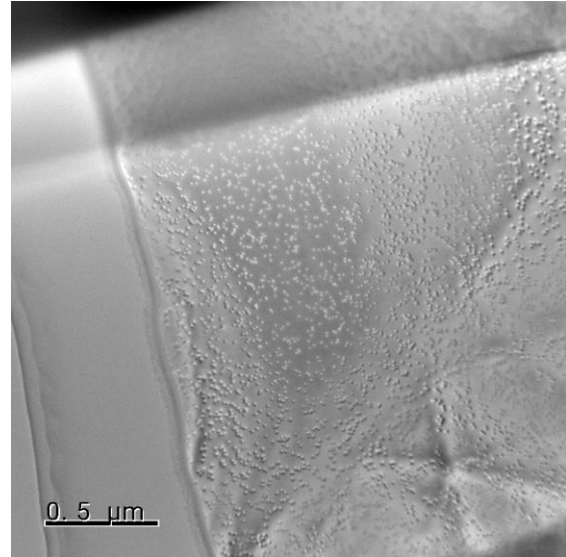


Fig. 1. BF TEM micrograph before heating that shows visible ion tracks as bright dots

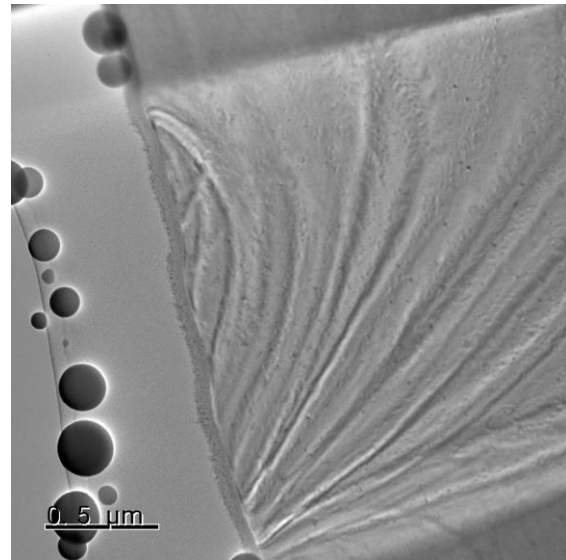


Fig. 2. BF TEM micrograph after heating to 550 °C of the same YIG lamella region as seen in Fig. 1. The SHI tracks are no longer visible, indicating complete recrystallisation. The dark spherical features to the left of the image are gallium droplets segregating from the FIB deposited carbon layer due to increased diffusion at high temperature. Pronounced bend contours are due to thermal stress and slight off-axis alignment due to heating

Image analysis software was used to segment the resulting frames, from which measurements for each individual ion track were obtained, such as the equivalent track diameter. Figure 3 shows the equivalent track diameters for some tracks observed, and Figure 4 illustrates the important time window

between 47 and ~56 s from Figure 3, where the recrystallisation rate rapidly increases.

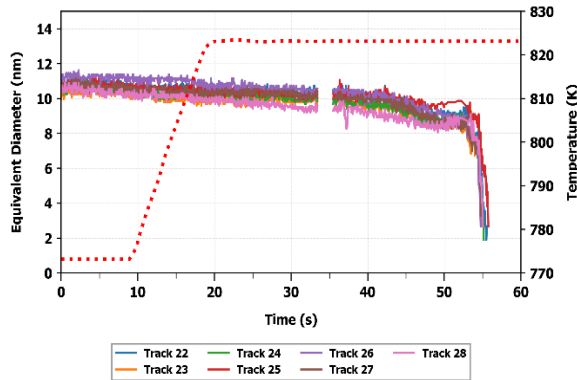


Fig. 3. The relationship between the equivalent track diameters for certain ion tracks, time in seconds and the respective temperatures during the heating of the sample. The curves exhibit a decrease near the end of the period at 823 K, which confirms recrystallisation. The data between 33-35 s was unreliable due to image blur that is attributed to the rapid shift in the field of view of the sample

The sample drifts and bends due to the large temperature adjustments, which result in significant thermal expansion. Therefore, to maintain near zone axis imaging for optimal contrast in BF imaging of the numerous frames, the sample was pre-heated to a specific temperature, which allowed for final adjustments before active recrystallisation could occur; this includes the final zone axis alignment and focusing. As evident from Fig. 3, 773 K was identified as an appropriate intermediate temperature, as insignificant recrystallisation occurs. As seen in both Fig. 3 and Fig. 4, the tracks start to exhibit significant recrystallisation once the temperature is maintained at 823 K.

Conclusions

SHI track recrystallisation in single crystal YIG was observed through in-situ TEM annealing with millisecond temporal material was captured frame-by-frame through the

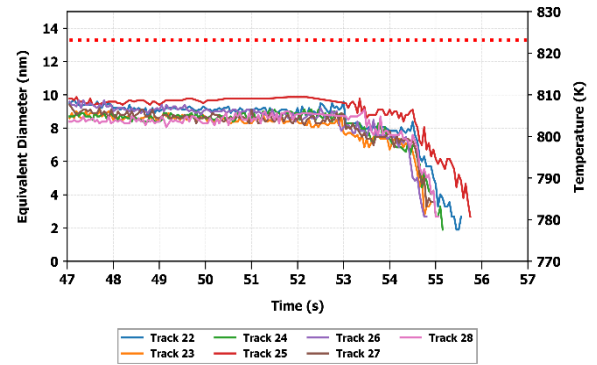


Fig. 4. An emphasis on the specific time window of 47 to ~56 s observed in Fig. 3, which illustrates the rapid increase in the recrystallisation rate

integration of a high-speed resolution. The structural recovery of the camera and precise thermal control, synchronised via a custom Python interface. Recrystallisation was tracked through automated thresholding-based segmentation routines and analysed by custom Python scripts. Limited recrystallisation was observed below 773 K, and only when the temperature has reached 823 K, active recrystallisation was observed within 35 s. Despite challenges from thermal drift and thermal drift induced contrast variations, the results suggest that reliable data on annealing dynamics in irradiated materials can be obtained from these experiments.

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