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SAXS and TEM investigation of ion tracks in neodymium-doped yttrium aluminium garnet

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ABSTRACT

Neodymium-doped yttrium aluminium garnet ($Nd:Y_3Al_5O_{12}$ or Nd:YAG) crystals were irradiated with 2.2 GeV ^{197}Au ions. Ion track formation was investigated using synchrotron based small angle X-ray scattering (SAXS) and transmission electron microscopy (TEM). Cylindrical ion tracks consisted of an amorphous core with sharp boundaries within the crystalline matrix. The SAXS results, modelled as long cylindrical tracks, provided a constant track density that is 0.6 ± 0.3% different to that of the surrounding matrix. The average track radii determined by both techniques were in excellent agreement, 4.4 ± 0.1 nm from SAXS and 4.4 ± 0.5 nm from TEM. A comparison with previous results of ion tracks in YAG indicates that the track radius is not affected by the presence of 1 mol% Nd dopant.

1. Introduction

In many materials, in particular in insulators, the high electronic excitations generated by swift heavy ion irradiation produce long columnar defects, so called ion tracks. These tracks create nanoscale domains that can have very different chemical, structural, and optical properties as compared with the unirradiated matrix. Recently, the irradiation with swift heavy ions (SHIs) has emerged as a promising technique to modify the refractive index of optical materials [1], making it suitable for the fabrication of optical waveguide devices [2,3].

For photonic applications [4], the precise control of the dimensions of the waveguides is essential and this requires a detailed knowledge of the ion track morphology in order to effectively utilize SHI irradiation for waveguide fabrication. Isometric Nd:YAG crystals are widely used as a gain medium for high-power solid state laser systems. The SHI-irradiated Nd:YAG waveguide lasers could be used as new miniature light sources for integrated photonic chips as reported by Ren et al. [5,6]. SHI induced modifications of YAG was studied previously utilizing Rutherford backscattering spectroscopy in channelling geometry (RBS/C) providing information of defect cross sections and average track sizes [7]. In addition, beam-induced volume expansion and hillock formation on the sample surface were analysed by means of X-ray diffraction (XRD) and atomic force microscopy (AFM), analysing the volume expansion in the bulk sample and hillock formation on the sample surface, respectively [8].

In the past, we have demonstrated that small-angle X-ray scattering (SAXS) is a powerful, non-destructive technique to investigate the size of ion tracks [9–13]. This work presents new results on the morphology of swift heavy ion tracks in 1 mol%-doped Nd:YAG crystals using synchrotron SAXS and transmission electron microscopy (TEM).

2. Experimental

As samples we used single crystals of Nd:YAG doped with 1 mol% Nd^{3+} ions and polished to a thickness of 110 μm. Ion tracks were produced by irradiating the crystals with 2.2 GeV (11.1 MeV

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per nucleon MeV/u) ^{197}Au ions to a fluence of 1×10^{11} ions/cm^2 at the UNILAC accelerator at GSI in Darmstadt, Germany. At this fluence, the ion tracks are well separated and track overlap can be considered negligible (later confirmed by TEM). Ion irradiation was performed at room temperature under normal beam incidence. The irradiation direction was perpendicular to a [0 1 2] plane of the crystal. The projected range of the ions and the electronic energy loss were calculated with the SRIM-2011 code [14], considering a mass density of 4.55 g/cm^3. The estimated projected range is ~69 μm and the energy loss averaged along the full path length is ~33 keV/nm. Based on previous reports [7], a track formation threshold in YAG of approximately 6 keV/nm was determined by extrapolation of the thermal spike calculations for high velocity ions. This yields an estimated track length of ~65 μm under the given irradiation conditions.

The irradiated samples were first analysed by SAXS to reveal the track structure. Subsequently the same samples were thinned down for TEM studies by polishing and subsequent ion milling from the surface opposite to the ion irradiation. The microscopy information thus originates from the zone close to the irradiated surface. The TEM images were recorded at 300 keV with a JEOL 3011 electron microscope at the University of Michigan. The tracks in this material are very stable under the electron beam even at high resolution TEM (HRTEM) mode during extended imaging, thus sample modification due to the electron beam (if there is any) can be neglected.

All SAXS measurements were performed in transmission geometry at the SAXS/WAXS beamline at the Australian Synchrotron. The X-ray energy was 12 keV (wavelength $\lambda = 1.0332$ Å) and the distance between the sample and the Pilatus 1 M detector was 968 mm. Silver behenate and glassy carbon standard samples were used for q-space calibration and normalization of the absolute scattering intensity, respectively. The samples were mounted on a three-axis goniometer for precision alignment. SAXS data were taken at room temperature with the ion tracks tilted between 0° and 10° with respect to the X-ray beam. To test the thermal stability of tracks, a small piece of the sample was used for SAXS measurements combined with *ex situ* annealing (30 min, temperature range from 200 to 500 °C, step width 50 °C).

3. Results and discussion

3.1. Small angle X-ray scattering analysis

Fig. 1 shows SAXS images of the ion tracks recorded on the 2D detector under different angles of the X-ray incidence (0°, 5° and 10°) with respect to the direction of the aligned tracks. When the tracks and X-ray beam are collinear, the SAXS pattern shows concentric scattering rings (Fig. 1a). Under tilted beam incidence, the scattered intensity is highly anisotropic and concentrates in curved streaks (Fig. 1b and c). The anisotropy originates from the high aspect ratio of the ion tracks (few nanometres wide and tens of micrometres long). The SAXS patterns typically contain many intense straight diffraction lines that originate from the single crystalline substrate. Data reduction was performed by excluding the parasitic scattering lines.

For analysis of the SAXS data, the scattering intensities corresponding to the tracks were extracted from isotropic images (tracks collinear to X-rays, Fig. 1a). Alternatively, we analysed streaks (tilt between tracks and X-ray beam, Fig. 1b and c). Both methods yield identical results. The background produced by parasitic scattering from the crystalline matrix was removed by measuring scattering intensities from radial sectors excluding the streaks produced by the tracks.

As the energy loss is almost constant over the first 58 μm of the ions trajectory in the material (varies less that 15% respect the value on the surface), and SAXS provides a volume weighed average track radius, the ion tracks were modelled as parallel oriented, well-separated cylindrical scattering objects. The simplest model that adequately reproduces the experimental data consists of a cylinder with a constant electron density different from that of the surrounding matrix [11]. The form factor of this model can be expressed as

$$f(q) = 2\pi LR\Delta\rho\frac{J_1(Rq)}{q} \quad (1)$$

where q is the scattering vector, L is the length of the track, R is the track radius, $\Delta\rho$ is the density difference between track and matrix, and J_1 denotes the first order Bessel function. The change in electron density between the track and matrix is described by a step function. In order to account for deviations from identical cylindrical and perfectly parallel tracks, largely caused by the above mentioned variation of the energy loss, polydispersity of the cylinder radius was considered in form of a narrow Gaussian distribution.

The measured scattering intensity of tracks (2.2-GeV Au tracks) and the corresponding analytical fit to the hard cylinder model are shown in Fig. 2. The model assumption of sharp boundaries is consistent with the formation of amorphous tracks in a crystalline matrix. The fit yields an average track radius of 4.4 ± 0.1 nm and a radius polydispersity (corresponding to the width of the distribution) of 0.4 ± 0.1 nm. The density change between track and matrix, deduced with the procedure detailed in Ref. [11], is 0.6 ± 0.3%.

The model assumption of sharp boundaries is consistent with the formation of amorphous tracks in a crystalline matrix. From the irradiated sample, a small piece was utilized for a series of SAXS *ex situ* annealing measurements. The SAXS signal from

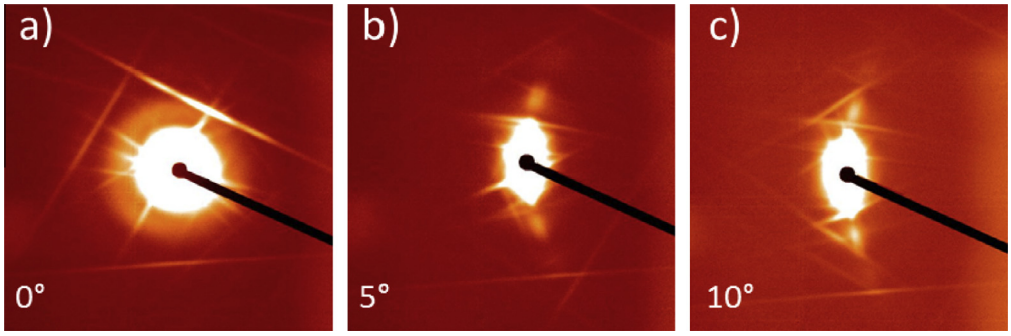

Fig. 1. SAXS images of Nd:YAG irradiated with 2.2 GeV Au ions to a fluence of 1×10^{11} ions/cm^2. (a) Tracks collinear with the X-ray beam, providing an isotropic scattering pattern (the halo corresponds to the second maximum of intensity). (b) Tilted by 5° and (c) tilted by 10° with respect to the X-ray beam. Under tilted X-ray geometry, the track pattern becomes highly anisotropic due to the large aspect ratio of the tracks. The intense straight lines are ascribed to parasitic X-ray scattering from the crystalline matrix.