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Radiation damage induced by swift heavy ions and reactor neutrons in $Y_3Al_5O_{12}$ single crystals

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Abstract

The induced damage in single crystals of Yttrium aluminium garnet (Y₃Al₅O₁₂) bombarded at GANIL with 561 MeV ⁵¹Cr, 466 MeV ¹²⁸Te, 880 and 957 MeV ²⁰⁸ Pb and 885 MeV ²³⁸U ion beams, and at Algiers with reactor neutrons has been studied by optical measurements, profilometry and X-ray diffraction techniques (XRD).

The optical absorption spectrum increases with fluence up to saturation level and absorption bands at about 250, 300, and 380 are observed after irradiation with ions and neutrons. X-ray diffraction as well as profilometry measurements show the volume expansion of the sample after irradiation due to the phase transformation from crystalline to amorphous phase. This volume expansion increases as a function of fluence and mean electronic stopping power S_e . It reaches saturation at high fluence, but the evolution with S_e shows a maximum at 17 keV/nm then it decreases significantly at 31 keV/nm. The track radii deduced from these techniques is seen to increase as a function of mean electronic stopping power from 8 to 17 keV/nm. From the analysis of the results, it is clear that the color centres do not contribute significantly to the swelling in the case of heavier ions with S_e larger than 12 keV/nm.

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Keywords: Radiation damage; Swift heavy ions; Neutrons

1. Introduction

Yttrium aluminium garnet $Y_3Al_5O_{12}$ is known for its excellent thermal and opto-mechanical properties (high thermal conductivity, extremely hard and durable, non birefringence, stable mechanical and chemical properties. Furthermore, this garnet is used as new substrate, window and laser host materials [1–5].

Yttrium aluminium garnet $Y_3Al_5O_{12}$ is also a candidate inert matrix fuel material [6]. Therefore, this material has been submitted to several kinds of radiation: alpha-particle, recoil nucleus, fission products, fast neutron and gamma rays. Color centers induced in $Y_3Al_5O_{12}$ crystals, by lowly ionizing radiation (γ rays, electrons, protons and neutrons) have intensively been studied in the past [7–13]. Most of the defects produced by fast neutrons via elastic collision are oxygen vacancies [14,15], and each vacancy can trap one or two electrons to form *F* and *F*⁺ center respectively [15–23]. The latter can be identified by their absorption bands centered at about 243 and 380 nm [17,24] respectively. Whereas, there are no studies on swift heavy ions induced damage in $Y_3Al_5O_{12}$, known to the authors. In this paper, optical properties, volume and structure changes induced by swift heavy ions and reactor neutrons in $Y_3Al_5O_{12}$ are investigated by means

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of optical absorption, profilometry and X-ray diffraction techniques.

2. Experimental

High-quality single crystals of $Y_3Al_5O_{12}$ purchased from "Crismatec" with a thickness of about 0.5 mm have been irradiated [25] with 561 MeV ⁵¹Cr, 466 MeV ¹²⁸Te, 880 and 957 MeV ²⁰⁸Pb and 885 MeV ²³⁸U ion beams. During irradiation, the samples are partially masked in order to analyse the out-of-plane swelling in direct comparison with the virgin crystal.

The irradiations were performed at the GANIL accelerator in Caen, France. In several cases, aluminum degraders of the different thicknesses were used in order to decrease the initial energy of the ions. Table 1 lists the beam parameters of different irradiation experiments. The mean dose Dwas deduced from the fluence using the following formula [26]

$$D = 1.6 \times 10^{-10} \frac{E\phi}{\rho R_{\rm p}},\tag{1}$$

where D is given in Gy, E is the total energy in MeV, ϕ denotes the fluence in ion/cm², $\rho = 4.56$ g/cm³ is the mass density of Y₃Al₅O₁₂, and R_p is the range in cm.

The irradiation with neutrons was performed at NUR research reactor, Algiers, at a position where the maximum temperature and the fast neutron flux were approximately 40 °C and 1.48×10^{13} cm⁻² s⁻¹ respectively. This flux was measured with a Cd-covered Indium foil based on ¹¹⁵In (n,n') In^{115m} reaction with neutron energy threshold of 1.2 MeV. Four samples were separately inserted in a well sealed container made of aluminium and irradiated at different fast neutron fluences up to 2.13×10^{17} , 5.86×10^{17} , 1.33×10^{18} and 4.10×10^{18} cm⁻², respectively. The lower fluence irradiation was performed using the pneumatic tube facility. The sample was inserted in a polyethylene container and irradiated to neutron fluence of $3.39 \times$

Table 1

Irradiation conditions of the samples: The average electronic stopping power is calculated from initial beam energy E_i divided by the projected range R_p . The projected ranges are estimated with SRIM2003 code

Ion species	Energy (MeV/u)	$S_{\rm e} ({\rm E}_i/{\rm R}_{\rm p})$ (keV/nm)	R_p (µm)	Fluence range (cm^{-2})
⁵² Cr	1.7 4.6 6.6 11	8.2 9 8.9 8	10.9 25.7 37.5 69.9	$\begin{array}{c} 4\times0^{11}7\times10^{12}\\ 4\times0^{11}7\times10^{12}\\ 4\times0^{11}7\times10^{12}\\ 4\times0^{11}7\times10^{12} \end{array}$
¹²⁸ Te	1.15 2	12 15	12.56 17.33	$\begin{array}{c} 3 \times 10^{11} 7 \times 10^{11} \\ 3 \times 10^{11} 7 \times 10^{11} \end{array}$
²⁰⁸ Pb	0.93 4.2	17 31	11.6 28.32	$\begin{array}{c} 2 \times 10^{11} 8 \times 10^{11} \\ 2 \times 10^{11} 3 \times 10^{11} \end{array}$
²³⁸ U	3.72	31	28.32	$4 \times 10^{11} 8 \times 10^{11}$

 10^{15} cm⁻². The corresponding doses are deduced using the ICRP51 conversion and quality factor [27].

2.1. Optical measurements

The optical measurements were performed using Cintra 40 UV–Visible spectrometer in the wavelength range 190–900 nm. The absorption coefficients were determined from

$$\mu = \frac{\text{OD}}{e},\tag{2}$$

where OD is the optical density and e is the sample thickness in the case of neutron irradiations and the ion range R_p in the case of ion irradiations.

All the measurements were made in the same conditions at 300 K and the color center concentration N was calculated using Smakula's formula (Eq. (3)) by assuming a Gaussian shape of the absorption band.

$$N(\mathrm{cm}^{-3}) = 0.87 \times 10^{17} \frac{n}{\left(n^2 + 2\right)^2} \frac{w(\mathrm{eV})}{f} \ \mu(\mathrm{cm}^{-1}), \tag{3}$$

where f is the oscillator strength, n is the refractive index, and w is the FWHM of the band.

2.2. Profilometry measurements

The volume expansion of the irradiated sample was measured using DEKTAK³ profilometer. A high precision stage moves the sample beneath a diamond – tipped stylus over the border line between a virgin and an irradiated area. The scans had typically a length of several hundred micrometers. The roughness of the virgin sample does not exceed 5 nm.

2.3. X-ray diffraction analysis (XRD)

XRD was the additional technique used in this work to check the influence of ion and neutron irradiation on the structure form of $Y_3Al_5O_{12}$. The diffractometer is X' PERT PRO MPD Philips using CuK α_1 ($\lambda = 1.5405$ Å). The sample holder is provided with a spinner with adjustable rotation velocity. After data acquisition, the K α_2 stripping was made using High Score Plus software program.

For each acquisition, both virgin and irradiated part of the sample were analysed at the same time. For this reason, two diffraction peaks can be observed in the diffraction pattern. The first one at a known position $2\theta = 52.785^{\circ}$ is the (444) diffraction peak of Y₃Al₅O₁₂ with lattice parameter of $a_0 = 12.003$ Å. The second peak is the shift of the peak (444) induced by irradiation leading to the new lattice parameter *a*. By measuring *a*, the lattice parameter of the irradiated part of the sample, the mean lattice strain can be calculated from

$$\varepsilon = \frac{a - a_0}{a_0}.\tag{4}$$

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