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Out-of-plane swelling of gadolinium gallium garnet induced by swift heavy ions

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Abstract

Single crystals of gadolinium gallium garnet ($Gd_3Ga_5O_{12}$) have been irradiated with various ions (Cr 10.6 MeV/u, Cu 0.8 MeV/u, Kr 9 MeV/u, and Pb 4 MeV/u) in the electronic stopping power regime. The irradiated areas of the crystals exhibited a pronounced volume expansion. Using a profilometer, the out-of-plane swelling was measured by scanning over the border line between an irradiated and virgin area of the sample surface. The step height varied between 25 and 160 nm depending on the fluence, the electronic stopping power and the total range of the ions. In the high fluence regime, the swelling effect approaches saturation. In order to compare the results obtained for different ion species, the initial swelling per ion was normalised by the length of the damage track. Such an analysis makes evident that swelling occurs only above a critical energy loss of 7 ± 2 keV/nm. The results of $Gd_3Ga_5O_{12}$ will be compared with data obtained earlier in SiO_2 and $LiNbO_3$. © 1998 Elsevier Science B.V. All rights reserved.

1. Introduction

Irradiation of insulators with energetic particles in the electronic stopping power regime (S_e) can lead to amorphisation along the ion path. Although numerous inorganic insulators have been studied during the past decades [1–6], the basic mechanism of ion induced amorphisation is still under discussion. One of the open questions concerns the link between specific properties of a given material and its sensitivity against high electronic excitation. Concerning this problem, it would be very helpful if the damage induced by radiation could directly be related to the change of a specific physical property. Recently, new observations were reported about pronounced volume expansion of various oxides under heavy-ion irradiation [6–9]. Such a change of the sample dimension is possibly well suited to test the radiation sensitivity of materials by an efficient and rather simple profilometer technique. In this paper, we are reporting about such volume changes induced by swift heavy ions in gadolinium gallium garnets ($Gd_3Ga_5O_{12}$). For the analysis, earlier experiments made in various oxides [6–9] were taken into account.

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Volume expansion which results from defects induced by irradiation is known for a long time [10,11]. It is only quite recent, that a huge out-of-plane swelling has been observed and described quantitatively when irradiating Al_2O_3 [7], SiO_2 [9], and $LiNbO_3$ [6,8] with energetic ions. In the two latter cases, it is known from high resolution electron microscopy (HREM), that irradiation induces amorphisation. For SiO_2 quartz, it was demonstrated that swelling occurs only above a critical electronic stopping power threshold of about 1.8 ± 0.6 keV/nm. This threshold is in good agreement with the appearance of topological disorder as measured by channelling Rutherford backscattering (C-RBS) [4].

In the following we are describing systematic measurements of the out-of-plane swelling in $Gd_3Ga_5O_{12}$, using several ion species in the energy regime between 0.2 and 10.6 MeV/u. Earlier experiments have already demonstrated that this material is sensitive to swift heavy ions [12–14]. It was shown, for instance, that the irradiation with Cu and Kr ions [13] induces amorphisation. The diameter of the latent track as measured by HREM is in good agreement with results obtained from C-RBS [14]. Under the same irradiation condition [13], high-resolution X-ray diffraction gave evidence that the lattice of $Gd_3Ga_5O_{12}$ expands normal to the irradiated surface. Furthermore, chemical etching of uranium tracks was reported [12].

2. Experimental conditions and physical characterisation

Slabs of synthetic $Gd_3Ga_5O_{12}$ crystals of optical quality were received from Crismatec. They had a thickness of about 0.5 mm and were covered with a 50 nm carbon layer in order to avoid electrostatic charging during irradiation. The samples were irradiated with Cu ions at the 7 MV tandem Van de Graaff in Bruyères-le-Châtel and with Cr, Kr, Te, Pb ions at the medium energy line of the Grand Accélérateur National d'Ions Lourds (GANIL) in Caen. All irradiations were performed at room temperature and under an angle almost normal to the sample surface. In some cases, thin Al foils were placed in front of the sample for the purpose of varying the energy and thereby the energy loss of the impinging ions. The flux of the beam was of the order of 10×10^8 ions s^{-1} cm^{-2} at the tandem Van de Graaff accelerator and 3×10^8 ions s^{-1} cm^{-2} at the GANIL accelerator. The maximum applied fluence was selected according to the mass and the projected range of the ions and varied between 10^{11} and 10^{13} ions cm^{-2}. During the irradiation, all crystals were partially masked in order to analyse the irradiated surface in direct comparison with a virgin area. A complete list of the irradiation parameters is presented in Table 1. The given energy loss (S_e) and the range (R_p) of the ions was calculated using the TRIM91 code [15].

Due to the fact that the ions were stopped in a thin surface layer of the crystals, the free expansion of the irradiated volume is partially limited by the constraint of the undamaged substrate. As a consequence, the sample bulges outwards mainly normal to the sample surface. The quantitative analysis of this out-of-plane swelling was performed with a profilometer (Dektak 8000) where 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 (Fig. 1). The surface of most of the crystals had a roughness of about 5 nm. Some of the samples exhibited additional corrugations, but even then, the step height could be determined without difficulty (Fig. 1). In some cases, bending of the whole crystal made the profilometer analysis rather difficult. Fig. 1 displays the profilometer scans over the surface of crystals irradiated with Cr ions of fluences between 6.5×10^{11} and 2.7×10^{12} cm^{-2}.

3. Data analysis

The evolution of the step height (δl) as a function of the ion fluence (Φ) is presented in Fig. 2. Each data point represents the mean step height as extracted from several profilometer scans. After an initial linear increase, the swelling approaches saturation at high fluences where the ion tracks begin to overlap. Due to limited beam time, it was

Table 1
Irradiation parameters

Ion	Mass	Energy (MeV/u)	Range (μm)	Mean S_e (keV/nm)	$\delta l / d\Phi$ (cm^3)
Cr	53	10.6	56	10.3	2.8×10^{-18}
Cr	53	6.2	30	11	6.15×10^{-18}
Cr	53	4.5	21.7	11.1	4.65×10^{-18}
Cr	53	1.8	9.6	9.9	2.4×10^{-18}
Cu	63	0.8	6	8.4	1.5×10^{-18}
Cu	63	0.16	2.6	4.2	3.1×10^{-19}
Kr	84	9.0	43	17.5	1.8×10^{-17}
Kr	84	3.4	18	15.7	1.2×10^{-17}
Te	125	2.2	15.7	17.3	1.8×10^{-17}
Te	125	1.2	11.3	13.4	1.2×10^{-17}
Pb	208	4.0	23.2	34.8	5.2×10^{-17}
Pb	208	1.2	10.3	23.2	1.1×10^{-17}

not possible to reach complete saturation as it was found earlier for other solids [6–9].

A first observation is related to the influence of the beam energy and consequently to the range of the incident ion. The step height of Cr ions of 6.2 MeV/u is about twice as large as for Cr ions of 1.8 MeV/u although both ions have approximately the same mean energy loss. A second qualitative observation concerns a comparison of the data of Pb (4 MeV/u) and of Kr (3.4 MeV/u) ions both having approximately the same range. Fig. 2 demonstrates that the out-of-plane swelling is more pronounced for the Pb ions with the higher energy loss. These findings clearly indicate that swelling scales with the range and with the energy loss of the ions.

To confirm the range effect, data of various ion species has been analysed by the following procedure. From electron microscopy (HREM [13]), it is known that in $Gd_3Ga_5O_{12}$, tracks of heavy ions consist of amorphised material. Since the mass density of the amorphous phase is smaller than that of the crystalline phase, it is reasonable to assume that the swelling observed here is based on the volume expansion due to a phase transition. As a consequence, the relevant damage volume should depend on the ion range. In addition, it is expected

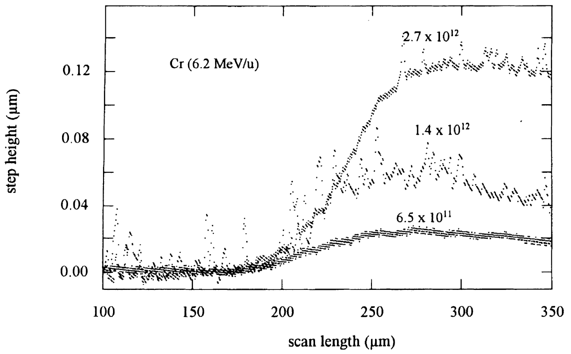

Fig. 1. Profilometer scans from the virgin to the irradiated area of crystals irradiated by Cr ions at three different ion fluences (ions/cm^2).

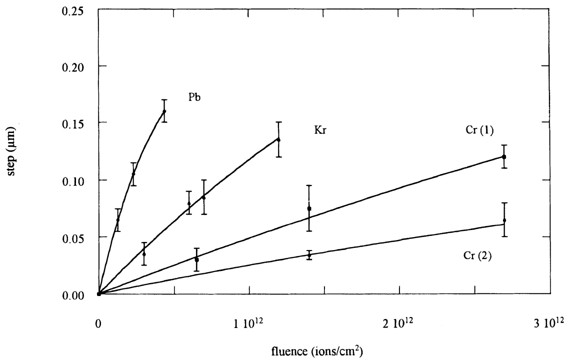

Fig. 2. Step height as a function of the fluence: Pb (4 MeV/u) and Kr (3.4 MeV/u) have approximately the same ion range, while Cr(1) (6.2 MeV/u) and Cr(2) (1.8 MeV/u) have about the same mean energy loss (see Table 1).

that the integral swelling effect for a specific irradiation fluence is a function of the total damage fraction of the irradiated volume. In $Gd_3Ga_5O_{12}$, this damaged fraction (F_d) is known from C-RBS experiments [14]. In Fig. 3, the step height is normalised by F_d and plotted versus the ion range. This presentation clearly demonstrates the linear dependence of swelling on the track length. It also shows that the damage induced by the electronic excitation is not efficient along the total range of the incident ion.

In order to test the correlation between swelling and the energy loss, we determined the initial swelling rate in the low fluence regime ($\delta l/\delta \Phi$ is given in Table 1) and normalised this parameter by the damage length of the given ion. In the beginning, it is assumed that the total length of the damage is equal to the projected ion range (R). This relative dimensional change per ion ($\delta l/\delta \Phi/R$) plotted versus the mean energy loss (initial beam energy E_i dividing by the TRIM range) is presented in Fig. 4 (circular symbols). The error of the energy loss takes into account that in this energy regime, the uncertainty of the values given by the TRIM code is between 10% and 20%. For further analysis, only data of the energy regime between 0.8 and 4.5 MeV/u were considered (full circles in Fig. 4) in order to avoid velocity effects [3]. If we fit a straight line to the restricted experimental data, it intercepts the abscissa at an energy loss $S_c = 5.5$ keV/nm. This value can be regarded as a threshold which has to be surpassed before the dimensional change can take place. It also tells us that the stopping end of the ion path where S_c is not reached does probably not contribute to swelling. We tried to take this effect into account by an iterative correction of the threshold. For each ion, the inefficient part ($R_o = R(S_c)$) of the trajectory was calculated and subtracted from the projected range ($R - R_o$). The value for swelling and for the mean energy loss was adjusted. This procedure converges already after two iterations (diamonds in Fig. 4) leading to a slightly higher critical threshold of $S_c = 7 \pm 2$ keV/nm. The error given here includes the threshold as determined from the unprocessed data. It also should be mentioned that possible variations of track parameters such as discontinuity, inhomogeneity of the damage or change of the track radius along the ion path does not influence this result significantly since the volume swelling is an effect integrated along the full length of the damage.

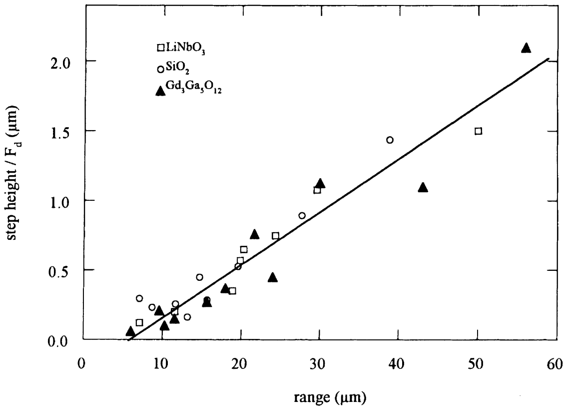

Fig. 3. Step height normalised by the damaged fraction F_d as a function of the ion range for $Gd_3Ga_5O_{12}$ (present results), SiO_2 [9] and $LiNbO_3$ [6,8].

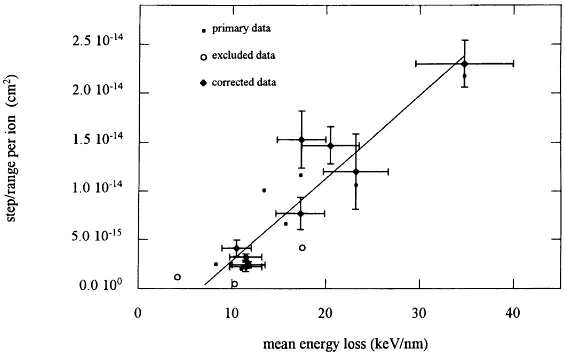

Fig. 4. The initial swelling per ion normalised by the range versus the mean energy loss (the exclusion of the some data points is discussed in the text).

Another interesting aspect is the comparison of our results with data of SiO_2 [9] and $LiNbO_3$ [6,8] shown in Fig. 3. A linear fit to the data gives for all three oxides a relative change of the linear dimension of the order of 0.04, indicating that the ion induced swelling is probably based on the same effect namely on the transition from the crystalline to the amorphous state.

4. Conclusion

Pronounced swelling effects have been found when irradiating $Gd_3Ga_5O_{12}$ with various swift heavy ions. To induce swelling, the ions have to surpass a critical electronic stopping power of 7 ± 2 keV/nm. This threshold is in agreement with the result from channelling Rutherford backscattering performed in the same energy range [14]. It is interesting to note that the relative dimensional change of 0.04 is the same as in SiO_2 [9] and $LiNbO_3$ [6,8] indicating that the effect is related to ion induced amorphisation.

The experiments described demonstrate that profilometry can be applied to probe the sensitivity of various material. Using this simple and effective technique will improve our understanding of the damage creation and the underlying basic mechanisms. It is planned to extend such measurements to non-amorphisable materials like alkali-halides in order to see if there exists a direct correlation between defect formation and volume increase.

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