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Nuclear Instruments and Methods in Physics Research B 146 (1998) 420–425

NIM B
Beam Interactions with Materials & Atoms

Monoatomic and cluster ion irradiation induced amorphous tracks in yttrium iron garnet

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Abstract

Track evolution in $Y_3Fe_5O_{12}$ is analyzed in the range 0.02 MeV/nucleon $< E <$ 30 MeV/nucleon. By applying the phenomenological thermal spike model the track sizes in cluster beam experiments can be predicted within experimental error from studies with monoatomic ion beams. It is concluded that the various energy density parameters are not useful in the description of the track evolution in the full range of track sizes. A quantitative analysis indicates that the Waligorski distribution of δ-electron dose is not directly related to the velocity effect. The contributions of the electron–lattice ion and the ion–ion interactions to the energy transfer are discussed. © 1998 Published by Elsevier Science B.V. All rights reserved.

PACS: 61.80; 61.82M
Keywords: Latent tracks; Electronic energy loss; Cluster irradiation; Velocity effect; δ-electrons

1. Introduction

The formation of amorphous tracks in insulators by swift heavy ions has been studied for over 40 years. The growth of our knowledge in this field was rather slow compared to other fields in physics. At present, there is no general agreement even on the basic mechanism of track formation: a Coulomb explosion or thermal spike mechanism. Recently, thermal spike models were suggested which were suitable for a quantitative comparison with experimental data [1,2]. This does not hold for the ionic spike mechanism: no appropriate model has been reported which would be suitable to predict quantitatively the evolution of amorphous track sizes in insulators.

Previously, the phenomenological thermal spike model was successfully applied to predict the sensitivity of dielectric materials to heavy ion irradiation and also the analytical form of the evolution of the effective track radii R_e with the electronic stopping power S_e. The model has the advantage that no adjustable parameters are used and the predictions are based on the macroscopic thermal properties of the targets: average specific heat c and melting point T_m. A detailed comparison with experimental data has been published in previous reports [3].

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0168-583X/98/$ – see front matter
PII: S0168-583X(98)00519-9

In this paper the model is applied to irradiation experiments performed on $Y_3Fe_5O_{12}$ (YIG) crystals by cluster ion beams [4]. YIG crystals have been most extensively studied among insulators. The cluster beam experiments extended the range of specific ion energies down to $E = 0.02$ MeV/nucleon. Thus at present, track data have been reported in the range 0.02–30 MeV/nucleon. It will be shown that these new data are in good agreement with the predictions based on the analysis of irradiation experiments with monoatomic beams.

2. Analysis of track data and discussion

2.1. Linear regime of track evolution

YIG crystals were irradiated at room temperature with C60 and C20 cluster beams with specific ion energies 0.022–0.056 MeV/nucleon [4]. The effective track radii R_e were measured by high resolution electron microscopy. Compared to monoatomic beams, the application of cluster ions extended the range of S_e values by a factor of two. This offered a possibility to check the predictions of our model with respect to the variation of the damage cross sections and the analytical form of the track evolution.

Previously, we analyzed amorphous tracks induced by monoatomic beams in YIG. We found that the track formation proceeds in two stages: an initial $R_e^2 \sim \ln S_e$ relation is followed by a linear one $R_e^2 \sim S_e$. Because of the velocity effect, low velocity ions induce tracks more effectively [5]. We observed that a high efficiency $g = 0.36$ is stabilized for low velocity ions with $E < 2$ MeV/nucleon. We showed that $R_e^2 \sim S_e$ is valid for 14 keV/nm $< S_e <$ 36 keV/nm and a further linear increase is expected at higher S_e values according to our model. In Fig. 1 we completed the figure which we used in our previous analysis [6] with the new experimental data [4]. Our model predicts the following function for the linear range of track formation for insulators

$$R_e^2 = \frac{gS_e}{2.7\pi\rho c T_o} \quad \text{for } R_e > 4.5 \text{ nm}, \tag{1}$$

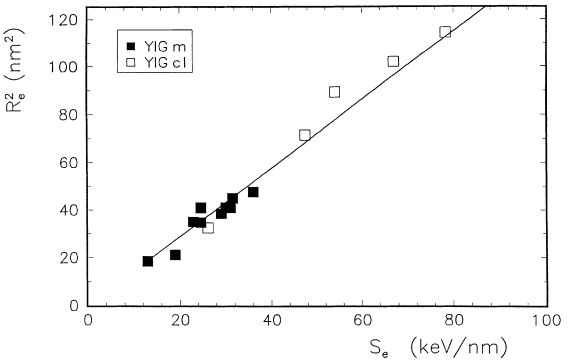

Fig. 1. Tracks with $R_e > 4.5$ nm induced in YIG by monoatomic (m) and cluster (cl) ion beams ($E < 2$ MeV/nucleon).

where ρ and c are the density and the average specific heat, respectively, and $T_o = T_m - T_{ir}$ (T_{ir} – irradiation temperature). We note that for $E < 2$ MeV/nucleon, g has the same value for YIG, $LiNbO_3$ and SiO_2 quartz [6]. Thus, the efficiency is not an adjustable parameter but a constant in Eq. (1). Therefore, Eq. (1) completely determines R_e. This is a very strict condition for a model. The slope of the solid line in Fig. 1 is equal to our previous estimate [6] within experimental error. The plot in the figure clearly shows that the predictions of the model are fulfilled both for the analytical form of the track formation and regarding the size of the tracks. This is an important evidence which proves that the basic idea of the model is correct.

On the other hand, the evolution of tracks according to Eq. (1) in the full range of S_e indicates that the mechanism of track formation is the same for monoatomic and cluster ion bombardment. For cluster beam irradiation the initial energy density in the thermal spike can be very high. Therefore, it is not obvious that the mechanism is unchanged. At about $S_e = 14$ keV/nm the peak temperature is $T_p \approx 2.7 T_o$. Thus $T_p \gtrapprox 15 T_o \approx 23\,000$ K is expected at $S_e = 80$ keV/nm.

We note that S_e for cluster ions is estimated assuming that the electronic stopping power of the individual carbon atoms can be linearly superimposed. This method is reasonable but there is little experimental evidence to support it [4]. The track data for $S_e < 40$ keV/nm and $S_e > 40$ keV/nm were obtained by monoatomic and cluster ion irradia-

tions, respectively. In the two ranges, there is no change in the slope of the solid line in Fig. 1 within experimental error. This indicates, that the rule of the linear superposition is correct in the evaluation of S_e for cluster ions.

There is a general agreement that S_e alone is not sufficient to describe track formation since at S_e = constant larger tracks are induced by low velocity than by high velocity ions [5]. Dunlop et al. claimed that the volumic energy density $P = S_e/\pi R_e^2$ is a more relevant parameter for track formation than S_e [4]. They found that $P = 235$ eV/nm^3 for cluster ion irradiation, and Fig. 1 shows that this is also valid for irradiation by monoatomic ions up to 2 MeV/nucleon. We remind that according to Eq. (1) $R_e^2 \sim S_e$ only for $R_e > 4.5$ nm. When P is calculated for smaller tracks, one finds that $\infty > P > 235$ eV/nm^3 for $0 < R_e < 4.5$ nm ($R_e = 0$ for $S_e = S_{et}$; S_{et} – threshold electronic stopping power). Obviously, this strong variation of P is not related to the velocity effect, it is a direct consequence of its definition. Therefore, it is doubtful that P is the relevant parameter. Moreover, $R_e < 4.5$ nm for the majority of amorphous tracks which have been measured in various materials till now. Thus, this feature of the parameter P is not a purely theoretical problem.

Previously, we found that at low ion velocities a higher fraction of S_e is converted into the energy of the thermal spike than at high ion velocities [7]. Thus, the efficiency g varies with E and it has high value at low ion velocities. According to Eq. (1) $P \sim 1/g$ for $R_e > 4.5$ nm. However, as opposed to the behavior of the parameter P, the efficiency of energy deposition does not depend on whether $R_e > 4.5$ nm or $R_e < 4.5$ nm [7]. Therefore, in experiments with E = constant, g has the same value for any track radii, and it varies only when the ion velocity is changed. This is a very important feature of the efficiency g. Our opinion is that g is the most important parameter in the track formation in insulators, therefore, it is reasonable to follow the variation of g with E in greater detail.

2.2. Velocity dependence

We plotted in Fig. 2 the variation of g in YIG in the range 0.02 MeV/nucleon $< E <$ 30 MeV/nucleon. For the calculation of the efficiency Eq. (1) was applied to track data with $R_e > 4.5$ nm, taken from Refs. [4,5]. The full arrow shows the magnitude of the efficiency g that was determined by another method from tracks with $R_e < 4.5$ nm [3]. This is a reliable value because g was not estimated from individual tracks, as in Fig. 2 but from many data. The full lines are to guide the eye where some very large deviations were not taken into account. The figure shows the well-known damage cross section velocity effect: lower efficiency at high velocities. The efficiency g is constant within experimental error in the ranges 0.02 MeV/nucleon $< E <$ 2 MeV/nucleon and for $E > 8$ MeV/nucleon. The variation of the efficiency in $LiNbO_3$ is also shown in the figure making use of data in Refs. [8,9].

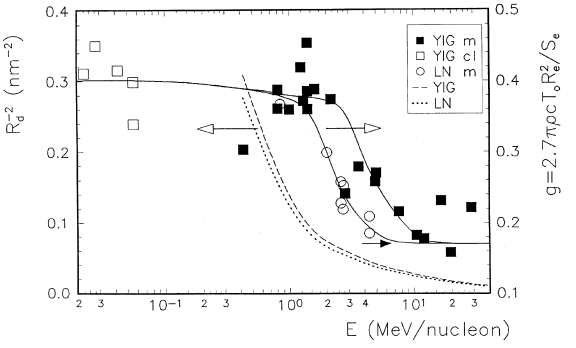

Fig. 2. Velocity dependence of the efficiency g and of R_d^{-2} (R_d – Waligorski radius) for YIG and $LiNbO_3$ (LN). The efficiency g was calculated from tracks with $R_e > 4.5$ nm induced by monoatomic (m) and cluster (cl) ions [4,5,8,9]. The full arrow shows the g value obtained for $E > 7.6$ MeV/nucleon from track data with $R_e < 4.5$ nm [3]. The full lines are to guide the eye.

We note that an estimate of the efficiency as $g \sim R_e^2/S_e$ is reasonable for any model. Obviously, the factor of proportionality may depend on the model. However, the shape of the variation of g with E and the information that can be extracted from it has a general meaning, the validity of which is beyond the actual model.

In the considerations in Refs. [4,5] the energy density has an important role. Meftah et al. claim [5] that the reduction of the track size with increasing ion velocity is the consequence of the broadening of the spatial distribution of δ-electrons. Thus, at higher ion velocities the initial en-

ergy density of the δ-electrons is lower and this is the origin of the damage cross section velocity effect. Based on the calculation of Waligorski et al. [10] they introduced the so-called Waligorski radius R_d which is the radius of a cylinder in which 66% of S_e is deposited. R_d increases with increasing E, therefore, the initial energy density D_e is reduced leading to smaller tracks. Meftah et al. claim that by applying the Waligorski distribution in their model they can quantitatively account for the velocity effect.

In another paper we already disputed this statement, because Meftah et al. could achieve an agreement with the experimental data only when the fitting parameter λ was different (6.6 and 6 nm) at various E values [11]. The fit was performed for $R_e > 3$ nm in three intervals 0.4–2, 2–8 and 8–30 MeV/nucleon. We note, that the maximum reduction of R_e due to the velocity effect is about 33% [5]. This means, that in average an 11% reduction is expected in each of the three selected velocity intervals. According to Figs. 2–4 in Ref. [11], 10% increase of λ at $E =$ constant induces about 7–8% average reduction in the calculated value of R_e. However, the agreement is not satisfactory for a number of points in each velocity interval (see Figs. 2–4 in Ref. [11]) and significantly larger $\Delta\lambda$ values would be necessary to cover the full range of the variation of R_e. The above estimate shows that in the model of Meftah et al. the variation of the fitting parameter accounts for a considerable fraction of the velocity effect and the energy density D_e is less important in this respect.

The plot in Fig. 2 makes it clear why the velocity effect cannot be directly related to the δ-electron distribution. We also show in the figure the variation of R_d^{-2} which is proportional to D_e. The R_d values for YIG were taken from Ref. [5] and the curve for $LiNbO_3$ was calculated from these data by scaling. Though R_d and g are parameters of different models, the comparison shown in Fig. 2 is reasonable, because according to Eq. (1) g is proportional to the damage cross section at $S_e =$ constant. If there were no velocity effect the full curve would be a line parallel to the E axis. In reality, the $g(E)$ curves in Fig. 2 directly show the sensitivity of the track size to the variation of E, i.e. the velocity effect, and according to Ref. [5] R_d^{-2} is a key parameter of this effect.

It is obvious from the figure that the variation of R_d^{-2} is small for E values where $g(E)$ strongly varies. On the other hand, $g(E)$ is constant below 2 MeV/nucleon where the variation of R_d^{-2} is sharp. We also showed in the figure that there is a shift of about 2 MeV/nucleon between the curves characterizing the velocity effect in YIG and $LiNbO_3$. The efficiency g of YIG is systematically higher than that of $LiNbO_3$ in the range of 2–5 MeV/nucleon. Such a dependence on the composition of the target can be highly useful in the experimental and theoretical studies of the velocity effect. This shift cannot be explained by the δ-electron distribution either, because the R_d^{-2}–E curve for $LiNbO_3$ is not shifted along the E axis, but is slightly reduced compared to YIG along the other axis. We also recall here the observation of Dunlop et al. that the track size is constant along a certain part of the trajectory of a fullerene ion while the energy density of the δ-electron distribution varies by about two orders of magnitude [4]. The same observation was used in Ref. [4] to show that the initial energy density of δ-electrons is not directly related to the track size as proposed by Tombrello [12]. Thus, it is hardly believable, that there is a direct correlation between the velocity dependence of track sizes and R_d.

In Ref. [13] we discussed the possible contributions of the electron–ion and ion–ion interactions to the velocity effect. We also analyzed the consequences of the ion–ion interaction model of Seiberling et al. [14]. They assumed that the ionized lattice atoms can gain considerable kinetic energy as a result of their electrostatic interaction. They suggested that this process can lead to the formation of a thermal spike after thermalization. However, it was not estimated whether the effective lifetime of the multiply charged ions is sufficiently long to acquire considerable energy. In Table 1 the results of the calculation for UO_2 lattice are given. We assumed that the ions are triply ionized and estimated the time necessary to increase the kinetic energy of various ion pairs by 0.1, 1 and 10 eV. The results show that the shortest time, about 4 fs are sufficient for the U–O (heavy–

Table 1
Ion interactions in UO_2

Ion pair	Ion separation	Parameters	$\varepsilon = 0.1$ eV	$\varepsilon = 1$ eV	$\varepsilon = 10$ eV
O^{3+}–O^{3+}	0.27 nm	t (fs)	0.72	2.32	8.48
		Δs (nm)	0.001	0.01	0.13
U^{3+}–O^{3+}	0.167 nm	t (fs)	0.39	1.25	4.29
		Δs (nm)	0.001	0.01	0.15
U^{3+}–U^{3+}	0.386 nm	t (fs)	5.73	18	73
		Δs (nm)	0.0015	0.015	0.21

ε – kinetic energy of the ion pair; t – time necessary to acquire ε; $2\Delta s$ – increase of the ion separation.

light) pair to get 10 eV. This is shorter than the screening time in insulators which is about 10 fs. As the number density of highly charged ions is rather high in the track region, the estimates seem to be realistic. Thus, the thermalized Coulomb explosion mechanism can be one of the efficient direct channels of the energy transfer to the matrix.

A high density of highly charged ions is excited in the track region by the projectile [15]. In a considerable number of lattice points multiply charged ions can be found. It is a ''drastic approximation'' [11] to calculate the energy transfer in this region as a simple electron–phonon interaction process in an unperturbed lattice. Our opinion is that the electron scattering by target ions has to be taken into account [12].

Both the ion–ion and the electron–ion cross sections are sensitive to the spatial distribution of the ion number density. The δ-electron distribution and the ion distribution around the trajectory must be in strong correlation to minimize the electrostatic energy. The initial δ-electron distribution is narrower compared to the Waligorski distribution. The initial number density of ions can be very high [15] and it is also lower at high ion velocities. Thus, at low ion velocities we expect a higher contribution to the energy transfer from the ion–ion and the electron–ion interactions. We assume that the electron–ion and ion–ion interactions are closely related to the velocity effect. For a quantitative comparison, the average ion charge and the mean lifetime of ions must be taken into account as well.

3. Conclusions

No difference is observed between track formation induced by cluster and monoatomic ion irradiations for $R_e > 4.5$ nm. The volumic energy density P applied in Ref. [4] depends both on E and R_e while the efficiency g varies only with E. Therefore, the efficiency g is the more suitable parameter to characterize the velocity effect. The evolution of the velocity dependence of track formation is quantitatively analyzed and a shift is observed between the onsets of the velocity dependence for YIG and $LiNbO_3$. The variation of the target composition in the velocity effect can be very useful tool in the investigation of the phenomenon. The comparison of the variations of the efficiency g and of the Waligorski radii R_d shows that the velocity dependence of track formation and the variation of R_d are not directly related. The contributions of the electron–ion and ion–ion interactions to the energy transfer are briefly discussed.

Acknowledgements

This work was accomplished with the partial support of the National Science Research Fund (OTKA, Hungary) under contracts T014987 and T025805.

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