Effective energy deposition and latent track formation of swift heavy ions in solids

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Abstract In the present paper, latent track formation in yttrium iron garnet (YIG) produced by high energy Ar ions is briefly reported at first. Then, in the framework of thermal spike model, a phenomenological parameter describing the effective energy transfer from excited electrons to lattice atoms, effective energy deposition Q_{eff}, is deduced. Q_{eff} is a function of ion velocity, electronic energy loss (S_e) and mean free path λ of excited electrons in the matter, and is a time moderate term initialized by Waligorski's function of spatial energy deposition of secondary electrons ejected by incident ions. Size of ion latent track is proportional to Q_{eff} value. From Q_{eff} obtained by use of realistic λ values, the sizes of latent tracks in SiO_2, YIG, Ti and Zr produced by given swift heavy ion irradiations are deduced and compared with experimental results. It is found that, from the fits to experimental results, the best λ values for SiO_2, YIG, Ti and Zr are (6±1), (8±2), (6.1±1.0) and (9.6±1.0) nm, respectively. Moreover, the relationship between experimental damage and Q_{eff} is discussed.

Keywords Latent track, Electronic energy loss; Thermal spike; Effective energy deposition

CLC numbers O571.33; $O532^+$.26

1 Introduction

When passing through a matter, an energetic ion loses its energy via collisions with target nuclei and electrons. The energy losses on target nuclei and electrons, named nuclear energy loss S_n and electronic energy loss S_e, will result in defect creations (point defect creation, clustering of defects, phase change of a crystal, and so on) in the matter. In solids, dense defects created in the wake of a single ion is called latent track.

In the case of swift heavy ion irradiations, S_e may be 1000 times higher than S_n, and it will play an important role in the ion latent track formation. Indeed, since the new generation of GeV heavy ion accelerators such as VICKSY at Berlin, UNILAC at Darmstadt, GANIL at Caen and HIRFL at Lanzhou appeared in the 1980's, especially high mass cluster particles such as C_{60} projectile being accelerated to MeV, the range of S_e has been enlarged up to or much higher than that corresponding to Bragg peak of a uranium beam and then the ion latent track formation by S_e was extensively studied. It is now well known that very high S_e can induce latent track formation in solids even if in pure metals.[1-5] Two models, thermal spike model [6-8] and Coulomb explosion spike model,[7,9] have been used to explain defect creation and ion track formation in various condensed materials. In the time scale of a swift heavy ion passing through a metal, the duration of nuclear collision between the incident ion and a target atom is extremely short (of the order of 10^{-16} s) and the electronic excitation induced thermal spike will last long enough (about 10^{-12}—10^{-10}s) compared to the lattice excitations (phonon frequency of the order of 10^{13} s^{-1}). Thus, the electronic process will cover the nuclear process. The purpose of the present paper is, based on the thermal spike model, to give effective energy deposition Q_{eff}, a phenomenological parameter, which can be used to simply evaluate magnitude of the energy of excited electrons converted into kinetic energy of the target atoms and the formation of ion tracks in solid materials.

Supported by NSFC (Projects 10125522, 10175084, 10475102) and the Chinese Academy of Sciences.

Received date: 2004-12-02

In the following, damage formation in yttrium iron garnet YIG ($Y_3Fe_5O_{12}$) produced by high energy argon ions is briefly reported at first. Then, in the framework of thermal spike model, the energy transfer from excited electrons to the target atoms is discussed and the Q_{eff} is deduced. From Q_{eff}, ion track radii in several solid materials are evaluated. Furthermore, the affection of some irradiation parameters to damage level in solids is also discussed.

2 Latent track in YIG produced by high energy argon ion irradiation

Stacked polycrystalline YIG samples with 5 mm in diameter and 60-120 μm in thickness were irradiated with GeV energy ^{40}Ar ions delivered from HIRFL at temperature (195±0.1) K. The irradiation induced amorphisation of YIG was investigated by use of Mössbauer spectra and saturation magnetization (4πMs) analysis. In the analysis the fraction of damaged material F_d is described by Poisson's distribution $F_d=1-\exp(-\sigma\phi)$ where ϕ is the ion irradiation fluence and σ is the damage cross section. If we consider that σ corresponds to the theoretical cross sectional area of the latent track, there is $\sigma = \pi R_e^2$ where R_e is the track radius. Some results of YIG irradiated with swift Ar ions are shown in Table 1, where E_{in} and E_{out} are the energies of ion beam in and out of the sample respectively, and $\langle S_e \rangle$ is the mean S_e value in the sample. From the deduced R_e values, we can see that they are in the region II criticized by Meftah et al.[3] and it implies that though the electronic damage may overcome the nuclear damage, the extended defects are nearly spherical and no continuous track formation could be concluded in the condition of the present work.

Table 1 Damage cross section σ of YIG under high energy argon ion irradiation

E_{in} (MeV)	E_{out} (MeV)	$\langle S_e \rangle$ (keV·nm^{-1})	σ (nm^2)	R_e (nm)
960	724	2.9	0.011±0.004	0.06±0.01
720	434	5.8	0.13±0.05	0.20±0.04
427	45	6.2	0.26±0.10	0.29±0.05

3 Effective energy deposition by S_e and ion track formation

Because the relationship between the energy of the excited electrons and the motion energy of target atom is still an open question, it is difficult to trace the entire details of damage process in solids under irradiations. In order to face the challenge of describing how the energy deposited into the target electrons transfers to atomic motions and induces latent track formation, a phenomenological parameter called effective energy deposition to target atoms from excited electrons, Q_{eff}, is introduced based on the thermal spike model.

In the thermal spike model of the interaction of energetic ion with matter, it is assumed that the energy deposited instantaneously in a very small region, producing a localized increase of temperature which spreads and cools according to the classical laws of heat conduction in a homogeneous continuum. In a cylindrical geometry with the center of ion path as the cylinder axis, the energy transfer process for the atomic system can be expressed as

$$C(T)\frac{\partial T}{\partial t} = \nabla[K(T)\nabla T] + A(r,t) \quad (1)$$

where $C(T)$, $K(T)$ and $T(r,t)$ denote the specific heat, thermal conductivity and absolute temperature, r and t denote the cylinder radius and time, respectively. $A(r,t)$ is the energy density rate converted from excited electrons to target atoms at r and t, and there is

$$A(r,t) = \frac{S_e}{\pi(R_d^2 + 4\lambda^2 t/\tau)\tau}\exp\left(-\frac{r^2}{R_d^2 + 4\lambda^2 t/\tau} - \frac{t}{\tau}\right) \quad (2)$$

with λ and τ are the mean free path and relaxation time of excited electrons in the matter, R_d is a critical radius determined by fitting the original expression given by Waligorski et al.[10] and is a function of the incident ion energy and the target properties. Taking into account two boundary conditions of the energy transfer process for the atomic system:

i) $t=0$, there is $\nabla(K\nabla T)=0$, $C(\partial T/\partial t)_{t=0}=A(r,0)$, and ii) $t=t_0$, there is $\partial T/\partial t=0$, $T=T_{max}$, $\nabla(K\nabla T)_{t=t_0}=-A(r, t_0)$,

the maximum energy transfer from excited electrons to atomic system within a cylinder body with radius r, effective energy deposition $Q_{eff}(r,t_0)$, can be expressed as

$$Q_{\text{eff}}(r,t_0)=\int_0^r\int_0^{t_0} C\frac{\partial T}{\partial t}2\pi r\mathrm{d}r\mathrm{d}t$$

$$=\int_0^r\int_0^{t_0}[\nabla(K\nabla T)+A(r,t)]\times 2\pi r\mathrm{d}r\mathrm{d}t$$

$$\approx\frac{S_e}{2}\left[1-\exp\left(-r^2/R_{\mathrm{d}}^2\right)\right]\left[1-\exp\left(-t_0/\tau\right)\right] \quad (3)$$

If $Q_{eff}(r,t_0)$ is equal to the energy needed to melt the material within the cylinder, we denote that the latent track radius is $r=R_e$. Moreover, we suppose that $t_0/\tau=R_0^2/(4\lambda^2)$. Thus, there is

$$R_e^2=R_0^2\left[1-\exp\left(-\frac{R_0^2}{R_{\mathrm{d}}^2}\right)\right]\left[1-\exp\left(-\frac{R_0^2}{4\lambda^2}\right)\right] \quad (4)$$

with $R_0^2=\frac{S_{\mathrm{e}}}{2\pi\Delta H_{\mathrm{f}}}$ and $\Delta H_{\mathrm{f}}=\int_{T_{\mathrm{irr}}}^{T_{\mathrm{m}}} C\mathrm{d}T+L$, where T_{irr}, T_m and L are irradiation temperature, melting point and latent heat of the target material, respectively. In Eq.(4), λ is the only free parameter. For a material with given energetic ion irradiation, T_{irr}, T_m, L, S_e and R_d are known and thus the ion latent track radius can be deduced by Eq.(4) using realistic λ value.

4 Results and discussion

Sizes of ion tracks in SiO_2, YIG, Ti and Zr under energetic heavy ion irradiations have been evaluated by use of Eq.(4). Table 2 gives the used ΔH_f and λ values in which T_{irr}=300 K is considered.

Table 2 Mean electron free path λ and melting energy ΔH_f values of several selected materials

Parameter	$Y_3Fe_5O_{12}$	SiO_2	Ti	Zr
λ (nm)	8±2	6±1	6.1±1.0	9.6±1.0
ΔH_f (eV·nm^{-3})	37.5	30.4	41.9	30.5

Tables 3-5 show the comparison between the calculated and experimental values of ion tracks, R_{cal} and R_{exp}, in SiO_2, YIG, Ti and Zr. It is found that the best fits to R_{exp} occur when λ=6±1, 8±2, 6.1±1.0 and 9.6±1.0 nm are used for SiO_2, YIG, Ti and Zr, respectively. Considering larger λ values corresponding to smaller R_{exp} values, the size of ion track is mainly affected by the λ value for the condition that R_d is much smaller than R_0. More complete numerical calculations for the determination of the sizes of latent tracks in SiO_2, YIG, Ti and Zr were reported by Meftah et al.[3] and Wang et al.[8], and the corresponding λ values were given as (4.0±0.3), (6.3±0.3), 6.1 and 9.6 nm. The similarity of the λ values obtained in the present work and the previous reports suggest that latent track formation in solids by S_e can be expressed by a transient thermal spike process and characterized by the effective energy transfer from excited electrons to target atoms.

Table 3 Latent track in SiO_2

Ion	E (MeV·amu^{-1})	S_e (keV·nm^{-1})	R_d (nm)	R_{cal} (nm)	R_{exp} (nm)[1]
O	1.88	1.61	4.2	0.42±0.07	0.15±0.02
F	0.79	2.44	2.9	0.94±0.14	0.6±0.1
S	1.56	4.6	3.8	1.7±0.3	2.0±0.3
Cl	4.29	3.82	5.7	1.1±0.2	1.1±0.1
Ni	5.82	7.1	6.5	2.2±0.3	2.6±0.3
	1.9	9.2	4.2	3.6±0.5	3.7±0.4
Cu	0.17	5.2	1.5	2.2±0.4	2.7±0.3
	0.79	9	2.8	3.7±0.5	2.9±0.3
Kr	3.4	12	5.2	4.5±0.6	3.2±0.5
I	1.48	16.4	3.7	6.2±0.7	4.0±0.4
Te	2.1	15.2	4.3	5.8±0.8	5.2±0.8
	1.2	17.4	3.5	6.5±0.8	4.5±0.4
Xe	1.5	16.7	3.7	6.3±0.8	4.0±0.4
Ta	1.1	19.2	3.3	7.1±0.8	5.4±0.5
Pb	1.0	20.9	3.2	7.5±0.8	5.4±0.6
	5.0	27.8	6.2	9.4±0.9	8.2±1.0
	0.30	14	1.9	5.4±0.7	5.5±1.0
U	1.52	27.2	9.5	8.4±0.8	8.2±2.0

Table 4 Latent tracks in Ti and Zr

Sample	Ion	E (MeV·amu^{-1})	S_e (keV·nm^{-1})	R_d (nm)	R_{cal} (nm)	R_{exp} (nm)[5]
Ti	Pb	4.06	36	3.8	9.1±0.8	~2.5
	C_{60}	0.025	43	0.5	10.4±0.9	10.0±2.5
Zr	U	23.3	41	5.1	9.7±0.8	
	C_{60}	0.025	44	0.5	10.3±0.8	7.5±2.5

Table 5 Sizes of ion latent tracks in YIG

Ion	E (MeV·amu^{-1})	S_e (keV·nm^{-1})	R_d (nm)	R_{cal} (nm)	R_{exp}* (nm)
S	1.56	6.9	3.3	1.7±0.4	1.1±0.1
Ar	19.6	2.9	7.7	0.33±0.08	0.06±0.01
	8.1	5.8	5.6	1.1±0.3	0.20±0.04
	5.9	6.2	4.9	1.3±0.4	0.29±0.05
Cu	0.8	13	2.4	3.3±0.8	4.3±0.4
Kr	38.7	6.7	9.7	0.88±0.19	0.26±0.03
	33.3	7.2	9.3	1.0±0.3	0.51±0.05
	29.0	7.8	8.8	1.2±0.3	0.56±0.06
	21.4	9.4	7.9	1.7±0.4	0.86±0.10
	15.7	11	7.1	2.2±0.5	1.3±0.2
	10.7	13	6.3	2.8±0.6	2.0±0.4
	8.75	15	5.8	3.4±0.8	2.87±0.33
	2.8	17	3.9	4.2±1.0	3.30±0.30
	2.8	17	3.9	4.2±1.0	4.1±0.4
	3.28	19.4	4.1	4.7±1.0	3.89±0.41
Te	1.24	22.8	2.9	5.5±1.2	5.92±0.54
	2.17	26	3.6	6.2±1.3	5.97±0.56
Xe	0.42	19	1.8	4.7±1.0	4.6±0.4
	1.74	20	7.3	4.4±1.0	3.7±0.2
	1.36	22	6.7	5.0±1.1	3.4±0.2
	1.4	24.6	3.2	5.9±1.2	6.4±0.5
	1.4	24.6	3.2	5.9±1.2	5.9±1.0
	8.3	25	5.6	5.9±1.2	4.2±0.2
	7.6	25.6	5.4	6.0±1.3	4.5±0.3
	4.9	27.5	4.5	6.5±1.3	5.1±0.6
Mo	8	18	5.6	4.2±1.0	2.7±0.4
Ta	1.3	30	3.0	7.1±1.4	6.4±0.8
	1.6	31.5	3.4	7.4±1.5	6.7±0.8
	3.6	37.5	4.3	8.6±1.6	6.7±0.8
Pb	1.3	31	3.0	7.3±1.4	6.4±0.8
	29	35	8.8	7.5±1.4	5.30±0.30
	19.7	35	7.7	7.8±1.5	4.5±0.4
	16.5	37	7.2	8.2±1.6	5.6±0.3
	12	40	6.5	8.6±1.7	5.1±0.5
	3.6	41	4.3	9.3±1.6	5.9±0.8
	5	43	4.7	9.6±1.6	6.53±0.74
U	0.8	29	2.4	6.9±1.4	6.2±0.8
	1.4	36	3.2	8.3±1.5	6.9±1.5
	2.8	43.5	4.0	9.8±1.7	6.2±0.8
	10.5	47	6.2	10.3±1.9	5.6±0.4
Au_4	25.4	17.6	0.30	4.3±1.0	4.2±0.2
C_{20}	55.8	26.1	0.58	6.2±1.3	5.7±0.4
C_{60}	22.5	47.4	0.51	10.5±1.7	8.4±0.2
	28.1	54.1	0.55	11.7±1.8	9.4±0.4
	41.9	67	0.64	13.9±1.8	10.1±0.4
	55.8	78.3	0.73	15.5±1.8	10.7±0.6

* R_{exp} values of Ar ions are from the present work, while the others are from Refs.[2-4].

In the quasi-free electron model, λ can be expressed as $\lambda=\sqrt{D_e\tau}$ where D_e is the electronic thermal diffusivity, and τ is linked to the electron-phonon coupling factor and specific heat for electron system of the target material. D_e and τ depend on temperature and target electron structure[11] and their determinations are still in question.[3,8,11] Despite of like that, our analyses confirm that, whether in solid insulators or in solid pure metals, the λ value is the main parameter governing the formation of ion latent tracks in the selected materials. From Eq.(4), it is also found that the ion with higher velocity (R_d is larger) and lower temperature irradiation will produce latent track with smaller size, and inversely, produce larger size track. The same energetic ion irradiation will produce smaller size track in the material with larger ΔH_f value. By the way, if we consider a given solid and take $r = \lambda$ in Eq.(3), the Q_{eff} varies only with R_d and S_e, which can be used to explain the damage level in solids where there is no track formation.

Based on the analyses shown above, one point of view can be raised: from the experimental irradiation results for a material we can deduce the λ value, and then can study the mechanism of energy transfer from excited electrons to target atoms.

5 Summary

Latent track formation by S_e in solid materials is closely connected to the mean free path λ of excited electrons and the energy ΔH_f necessary to melt. The track radii increase as λ and ΔH_f decrease. These conclusions have been confirmed by experiments and the analyses in the present work. From the idea of the effective energy transfer from excited electrons to target atoms, the size evaluation of latent tracks was made for SiO_2, YIG, Ti and Zr which were well studied experimentally. It is found that the best fit to experiments is obtained when $\lambda = (6\pm1)$, (8 ± 2), (6.1 ± 1.0) and (9.6 ± 1.0) nm were used for SiO_2, YIG, Ti and Zr, respectively. Furthermore, for the irradiation performed in the present work, argon ions with several hundreds MeV can not produce continuous latent track in YIG.

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