Nuclear Instruments and Methods in Physics Research B 579 (2026) 166225

Contents lists available at ScienceDirect

Nuclear Inst. and Methods in Physics Research, B

journal homepage: www.elsevier.com/locate/nimb

A model reasoning low-energy track-formation and enhanced nuclear energy loss in Si irradiated with C_{60} ions☆

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ARTICLE INFO

Keywords:
Ion track
C_{60} ion
Silicon
Synergy effect
Cluster effect

ABSTRACT

Ion energy dependence of track radius in Si was investigated under C_{60} ion irradiation between 30 keV and 9 MeV. The tracks were observed from 9 MeV down to 60 keV but not at 30 keV. The track radius gradually decreased with decreasing the energy from 9 MeV to 500 keV, showing a tentative increase around 300 keV followed by further decrease. Both (i) the track formation at exceptionally low energies and (ii) the non-monotonic energy dependence of the track radius, are explained by the ion energy dependence of electronic and nuclear energy losses of C_{60} ions in Si, i.e., the cluster-ion energy loss (CIEL) model.

1. Introduction

Since ion implantation/irradiation to Si is recognized as one of the most important processes in fabricating micro-/nano-integrated circuits for information technology, the ion–solid interactions in Si have been extensively studied particularly in the energy range from sub-keV to several MeV [1]. While the ion track formation has been observed in many materials under heavy-ion irradiation in the energy range of tens MeV or higher, which are often called swift heavy ion (SHI) irradiation, the track formation in crystalline Si has, however, remained controversial. To extend the applicability of high energy heavy ions in the range of tens MeV and higher to the Si micro-/nano-technology, the understanding of the ion–solid interaction between Si and ions in this energy region, particularly concerning the ion track formation, is significant.

Although various SHI irradiations have already been attempted to Si [2,3], no ion tracks have ever been formed under up to high-energy 3.6-GeV U ion irradiation (electronic energy loss $S_e = 24$ keV/nm) [3]. These ions correspond to the Bragg peak region, in which the highest S_e attainable by monatomic SHIs (m-SHIs) is induced. These observations indicate that any of m-SHIs cannot form ion tracks in Si. However, it does not mean that ions other than m-SHIs cannot form ion tracks in Si. In fact, 30- and 40-MeV fullerene C_{60} cluster-ions (S_e of 43 and 50 keV/nm in Si, respectively) succeeded in forming ion tracks in Si [4,5]. After then, many researchers believed that the failures of the track formation in Si under m-SHI irradiation were ascribed to the limitation of S_e available against the quite high S_e threshold in Si, which cannot be overcome by any of m-SHIs but C_{60} ions with delivering extremely high S_e. Since sixty carbon atoms from a C_{60} molecule were injected into a solid at almost the same time and the same nanometric region, an extremely higher S_e was provided even with C_{60} ions of only tens MeV [6–8].

It should be, however, noted that both Canut et al. [4] and Dunlop et al. [5], who independently observed the first track formation in Si under C_{60} irradiation, have also pointed the importance of the velocity effect. Much slower velocity of C_{60} ions compared to the m-SHIs may induce much higher excitation density for the track formation at the expense of more localized excitation volume.

Chettah et al. carried out the inelastic thermal spike (i-TS) calculations [9] in Si irradiated with C_{60} ions with including the velocity effect [10]. However, they suggested that the calculated velocity effect was not high enough to explain the completely different track formation behaviors between C_{60} ions and m-SHIs, if the tracks were formed by melting [10]. According to their calculations [10], the S_e threshold was estimated as low as ~ 4 keV/nm for C_{60} ions under the melting criterion of the track formation. This quite low threshold looks inconsistent with all the past experiments where the tracks were not formed at least less than 30 keV/nm. These two thresholds, 4 and 30 keV/nm, which are

☆ This article is part of a special issue entitled: ‘2025 REI-22 Conference’ published in Nuclear Inst. and Methods in Physics Research, B.

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https://doi.org/10.1016/j.nimb.2026.166225
Received 14 February 2026; Received in revised form 24 May 2026; Accepted 12 June 2026
Available online 17 June 2026
0168-583X/

quite different from each other, were assumed as the thresholds at low and high velocity limits. However, it is quite difficult to reproduce these two widely different thresholds from the i-TS calculations [10]. In fact, $S_{e,th}$ of 6 keV/nm was estimated for the high velocity of 5 MeV/u (i.e., 3.6 GeV U ion) [10], which is comparable to the low velocity value of ~4 keV/nm. Therefore, Chettah et al. suggested the boiling criterion for the track formation in Si under m-SHI irradiation, instead of the melting criterion [10]. Another origin of the inconsistency on the $S_{e,th}$ of Si could be ascribed to the recrystallization of the ion tracks [11]: In this model, the $S_{e,th}$ of Si is assumed to be not so high. Therefore, the tracks are easily created in Si. However, because of the highly enhanced recrystallization, the created tracks are rapidly annihilated. Consequently, no tracks are observed in Si after irradiation by any m-SHIs with any energy.

However, the recrystallization model has not been broadly accepted yet, but still under debate. In fact, Länger et al. reported negative evidence for the recrystallization model in Si [12]. Further studies are necessary to clarify why no tracks are formed in Si under m-SHI irradiation.

Contrary, our experimental results show the ion tracks are formed and survived in Si under C_{60} ion irradiation *without the annihilation due to the recrystallization*. We tentatively ascribed the track survival without the annihilation due to the recrystallization in Si under C_{60} irradiation to relatively high nuclear energy deposition S_n [13]. While S_n is negligibly low under m-SHI irradiation, it is relatively high under C_{60} irradiation. Damage generated by S_n of C_{60} ions could disturb the recrystallization of the tracks [13].

It seems that the melting criterion describes better the track formation in Si under C_{60} ion irradiation less than 9 MeV [13–15]. It should be noted that the threshold $S_{e,\ th}$ of ~ 4 keV/nm is consistent with our recent study [15], where the ion tracks were traced with decreasing the C_{60} ion energy from 9 MeV to 30 keV. Cylindrical damage zones with high aspect ratios (ARs) formed under 9 MeV irradiation, decrease their diameters, lengths and ARs, with decreasing the ion energy [15]. While something like tracks were observed down to 60 keV irradiation, no discernible localized structures like tracks were observed under 30 keV irradiation [15].

In this paper, some experimental results are examined from the energy dependence of the electronic and nuclear energy losses (S_e and S_n) of C_{60} ions. Hereafter, it is called "the cluster-ion energy loss (CIEL) model" and is applied to; (i) why the ion tracks are formed in low energy down to 60 keV under C_{60} ion irradiation, and (ii) the inconsistency of the track formation threshold energy between the prediction of the i-TS model (300 keV) and the experimental observation (60 keV). The CIEL model also predicts that S_n is enhanced around the track threshold. While the track formation down to 300 keV is ascribed to the purely S_e processes, the formation below 300 keV cannot be explained except the synergy effect of S_e and S_n.

2. Experimental

The experimental conditions were the same as the previous paper [15]. Samples of single crystalline Si (boron-doped p-type) were cut from commercially available wafers with a resistivity of ~1 Ω cm. The samples were immersed in hydrofluoric acid before the irradiation to remove surface oxide. The irradiation of C_{60} ions was conducted at the Takasaki Institute for Advanced Quantum Science, of the National Institutes for Quantum Science and Technology (QST). C_{60} ions between 30 and 750 keV were accelerated using a 400-kV single-ended ion implanter with the different charge states of C_{60}^{+}, C_{60}^{2+}, and C_{60}^{3+}. The C_{60} ions between 1 and 9 MeV were accelerated by the 3 MV tandem accelerator. To avoid the overlap of the track, the ion fluence was set to 5×10^{10} or 1×10^{11} C_{60}/cm^2. The ion tracks were evaluated by transmission electron microscopy (TEM) with an operating voltage of 200 kV (JEOL JEM-2100). Focused ion beam (FIB) milling with 30 keV Ga ions were applied for the thinning of the TEM samples.

3. The cluster-ion energy loss (CIEL) model

Fig. 1 shows calculated energy dependences of S_e and S_n in Si irradiated with (a) C_{60} ions and (b) monatomic Xe ions. The energy losses of Xe ion in Si were evaluated from SRIM 2013 code [16] and plotted in Fig. 1(b). Those of C_{60} ions, S_e (E, C_{60}) and S_n (E, C_{60}), were evaluated and plotted in Fig. 1(a) under the approximation that the energy losses of a C_{60} ion with the energy E are comparable to 60 times of those of a monatomic carbon ion with the same velocity, i.e., E/60,

$$S_e\ (E,\ C_{60}) = 60\ S_e\ (E/60,\ C_1), \quad (1)$$

$$S_n\ (E,\ C_{60}) = 60\ S_n\ (E/60,\ C_1). \quad (2)$$

These relationships were proposed for the electronic ones from Ref. [6] and the nuclear ones from Ref. [7], respectively. S_e (Xe) is plotted again in Fig. 1(a) by a broken curve for comparison with that of C_{60} ion, S_e (C_{60}), which clearly indicates that S_e (C_{60}) is much higher than S_e (Xe) due to the enhancement described by eq. (1).

The horizontal broken lines in Fig. 1 indicate $S_{e,th}$, i.e., the threshold value of S_e, which corresponds with the lowest S_e value to induce thermal melting for track formation. Because of the velocity effect [17], the $S_{e,th}$ value could weakly depend on the energy. However, according to Chettah et al. [10], $S_{e,th}$ calculated at the low energy 0.07 MeV/u and at the high energy 5 MeV/u were ~4 keV/nm and ~ 6 keV/nm, respectively. The energy dependence of $S_{e,th}$ in Si can be approximated by the horizontal line.

In the case of Xe irradiation to Si, as shown in Fig. 1(b), the S_e curve crosses with $S_{e,th}$ line at the threshold energy E_{th} of ~15 MeV. This is inconsistent with the experimental observations, because ion tracks

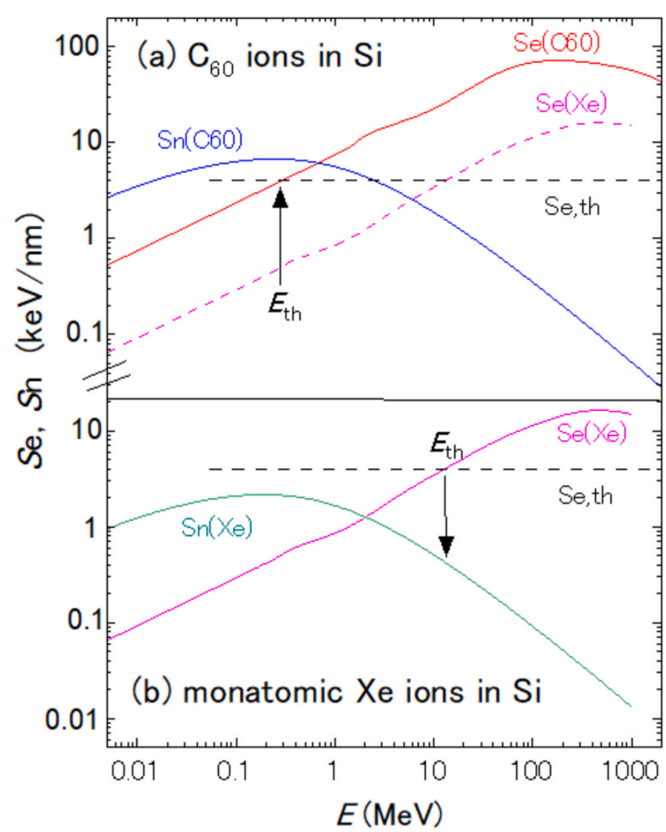

Fig. 1. Ion energy dependence of electronic energy loss S_e and nuclear energy loss S_n in Si, irradiated with (a) C_{60} ions and (b) monatomic Xe ions. Horizontal broken lines indicate $S_{e,th}$, i.e., the lowest energy loss required to induce the thermal melting for track formation. For comparison, S_e of Xe ion is also plotted in (a) by a broken curve. Arrows indicate the electronic track formation thresholds.

have never been observed in Si under m-SHI irradiation up to 3.6 GeV U ions. However, as already described in Introduction, this inconsistency is reasonable and can be ascribed to the recrystallization of the tracks under m-SHI irradiation [11], because the track recrystallization is not included in the CIEL model. Rather, Fig. 1(b) could be intuitive to understand similar situations that happened in many materials where ion tracks are formed under m-SHIs irradiation, i.e., much lower S_n than S_e at the threshold energy E_{th}.

Tracks are formed in the energy region where S_e is higher than $S_{e,th}$. In the case of monatomic ions (Fig. 1(b)), the S_n value is much lower than S_e at the threshold energy E_{th}, which is indicated by a downward arrow at $S_e = S_{e,th}$. Consequently, the behaviors around the (electronic) track formation threshold under m-SHI irradiation are determined by S_e-related processes only. The monotonic decrease in the track radius with decreasing the S_e is a typical consequence in many materials under m-SHI irradiation.

As shown in Fig. 1(a), S_e (C_{60}) is much higher than S_e (Xe). Consequently, the energy at the cross-point between $S_e(C_{60})$ and $S_{e,th}$, i.e., the threshold energy $E_{th,}$ considerably shifts to low energy side. Using the $S_{e,th}$ of ~ 4 keV/nm, which was the value derived in the previous paper, E_{th} was estimated to be 300 keV. It means that ion track formation, which was known as a high energy phenomenon, is realized at low energies under C_{60} irradiation.

In the case of the monatomic ions, S_n was negligible to S_e at E_{th}, as shown in Fig. 1(b). However, S_n overcomes S_e at E_{th} in the case of C_{60} irradiation as shown by an upward arrow in Fig. 1(a). The behaviors of tracks around the S_e threshold may strongly be influenced by the cooperation of high S_n under C_{60} irradiation.

It has been often suggested that the existence of the non-negligible S_n when comparing the irradiation effects of C_{60} ions and m-SHIs, both of which provide comparable S_e. The origins of the non-negligible S_n are considered as following: (a) originally low energies of constituent C atoms of C_{60} ions and (b) the S_n enhancement due to the simultaneous injection of 60 carbon atoms as described as eq. (2). However, we here propose the third one: (c) S_n enhancement due to the huge low-energy shift of the threshold energy E_{th}, which is induced via eq. (1).

4. Experimental results

Fig. 2 shows the electronic energy loss S_e dependence of the experimental mean-track-raii of Si irradiated with C_{60} ions. The corresponding C_{60} ion energy is shown close to each data point. Two significant behaviors are observed: (1) Even at the low C_{60} ion energy of 60 keV, i.e., $S_e = 1.8$ keV/nm, ion tracks of 1.6 nm in the mean radius are formed. (2) With decreasing the ion energy from 9 MeV, the mean radius monotonically decreases down to 500 keV. However, with further decrease, the radius increases once and decreases again. The non-monotonic decrease of the track radius with S_e is exceptional. In many materials, monotonic changes in the radii with S_e have been reported [18].

Regarding (1), ion track formation was known as one of the high energy phenomena, but the tracks were observed down to extremely low energy of 60 keV. The track formation at low energy is well supported by the CIEL model as shown in Fig. 1.

Regarding (2), similar behaviors, i.e., non-monotonic S_e dependence of the track radius was reported by Toulemonde, et al., in amorphous SiO_2 irradiated with Au ions ranging from 300 keV to 185 MeV ($S_e = 0.71 - 16.2$ keV/nm and $S_n = 3.2 - 0.16$ keV/nm) [19]. According to them, the track radius of SiO_2 decreased with decreasing the Au energy from 185 MeV to 10 MeV, but increased from 10 MeV to 300 keV. They suggested the track formation in SiO_2 via the synergy effect of S_e and S_n, since S_n increases with decreasing the energy. As shown in Fig. 1, S_n is enhanced around the track formation threshold in Si irradiated with C_{60} ions. The synergy effect between S_e and S_n is expected for the track formation also in Si irradiated with C_{60} ions.

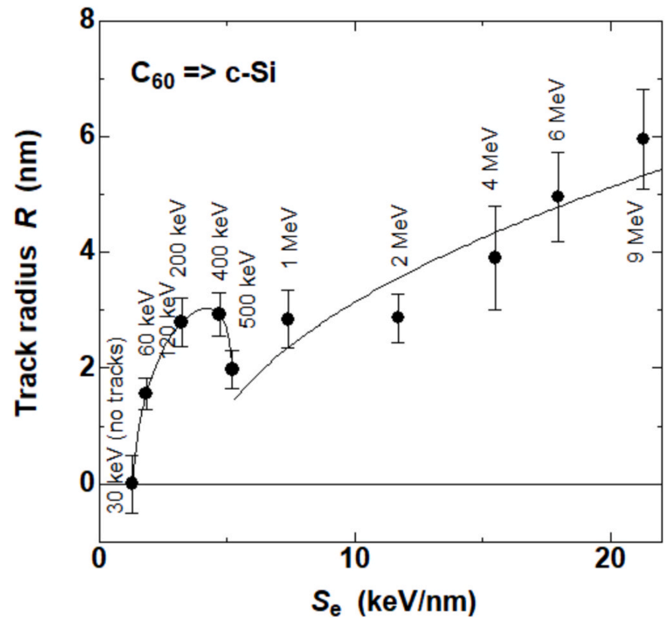

Fig. 2. Electronic energy loss S_e dependence of the experimental mean-track-radii in Si irradiated with C_{60} ions. Corresponding ion energies are shown in the figure. Solid curves are guides to the eye, indicating the qualitative consequences from the CIEL model.

5. Discussion

While the predicted threshold energy E_{th} was 300 keV from the relationship $S_e(E) = S_{e,th}$, the tracks were observed down to 60 keV irradiation. The difference between 60 and 300 keV could be ascribed to the enhanced S_n around the S_e threshold. The S_e dependence of the track radius R of semiconductors under C_{60} ion irradiation is described by an empirical rule [20],

$R^2 = C\,(S_e - S_{e,th})$, (3).

where C denotes a proportional factor. The data points shown in Fig. 2 were replotted in Fig. 3 with S_e versus squared R, i.e., R^2. When the data points follow the relationship indicated by eq. (3), they fall on a straight line in the plot of Fig. 3. In fact, the data points between 500 keV and 9 MeV are well fitted by a straight broken line as shown in Fig. 3. This observation indicates that the track formation between 500 keV and 9 MeV is mainly due to the S_e-related process. The fitted value of $S_{e,th,}$ was 4.2 keV/nm.

A solid curve shows calculated results from the i-TS model [10]. While the i-TS model slightly overestimate the track radii, the extrapolated threshold $S_{e,th}$ reached to ~3.5 keV/nm, in good agreement with the value extrapolated by eq. (3). In this paper, $S_{e,th}$ of ~ 4 keV/nm is used. It should be noted that squared track radius R^2 increased again below 500 keV and then decreased and disappeared at 30 keV. The behaviors below 500 keV irradiation cannot be explained by the monotonic decay of the electronic stopping S_e only. Rather, the deviation of the data points from eq. (3) below 500 keV probably indicates a change in the track formation mechanism from higher to lower than 500 keV. Since S_n increases in this energy region, the peak in R^2 at 300 keV could be ascribed to the synergy effect of S_e and S_n.

While we have assumed the constant $S_{e,th}$ independent of the ion energy in Si in this discussion, which is a good approximation as discussed in section 3, the model can be extended for variable $S_{e,th}$ including the large velocity effect. The $S_{e,th}$ increases mostly with the ion energy. Consequently, the two effects discussed here become more significant.

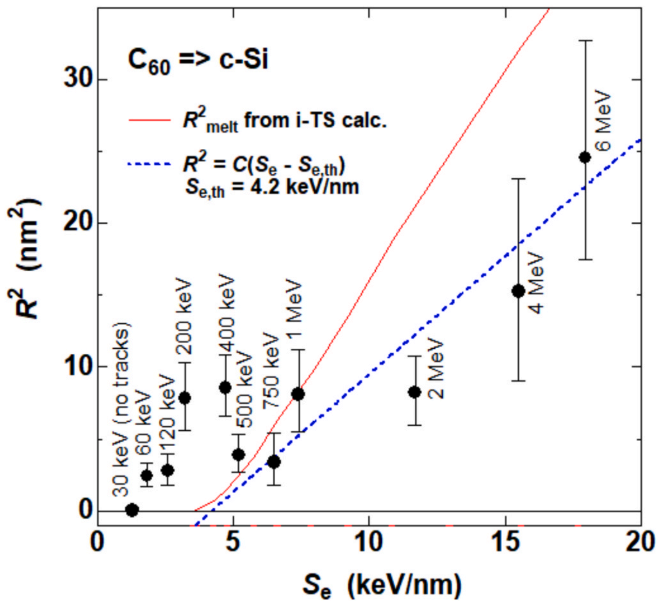

Fig. 3. Data shown in Fig. 2 were plotted with S_e versus R^2, where R^2 denotes the squared mean track radius. The data following with the eq. (3) are observed on a straight line in the figure. The broken line indicates a linear fitting of the data higher than 500 keV. The solid curve indicates calculated results from the i-TS model by Chettah et al [10]. Reproduced from Ref. [15] by the Creative Commons CC-BY license.

6. Conclusions

The mean track radii of Si were evaluated by TEM by changing the ion energy of C_{60} ions from 30 keV to 9 MeV. The tracks were observed down to 60 keV but not at 30 keV irradiation. In many materials under monatomic-ion irradiation, tracks are formed in higher energy than tens MeV or more. The track formation under tens keV irradiation is unusual. However, this phenomenon is described by the large low energy shift of the threshold energy E_{th}, which is ascribed to much higher S_e of C_{60} ions. Furthermore, the low energy shift of E_{th} also results in a shift of the threshold to the enhanced S_n peak. The track formation around the threshold could be largely modified by the enhanced S_n. In the case of Si, the track radius monotonically decreased with decreasing the ion energy from 9 MeV. However, the tracks turned to an increase below 500 keV but soon decreased and disappeared. From the empirical rule, eq. (3), the track formation higher than ~500 keV is explained by the S_e-related processes. However, the track formation below ~500 keV cannot be explained by the purely S_e-related processes alone. The synergy effect between S_e and S_n is suggested, which is favorable since the S_n is largely enhanced around the threshold under C_{60} ion irradiation.

CRediT authorship contribution statement

H. Amekura: Writing – review & editing, Writing – original draft, Investigation, Funding acquisition, Formal analysis, Data curation, Conceptualization. **K. Narumi:** Writing – review & editing, Methodology, Investigation, Conceptualization. **A. Chiba:** Methodology, Investigation. **Y. Hirano:** Methodology. **K. Yamada:** Methodology. **S. Yamamoto:** Methodology, Investigation. **Y. Saitoh:** Supervision, Methodology. **H. Segawa:** Supervision, Project administration, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgments

A part of the study was supported by the Inter-organizational Atomic Energy Research Program through an academic collaborative agreement among JAEA, QST, and the Univ. of Tokyo. The authors are grateful to the crew of the accelerator facilities at QST-Takasaki for their help. HA was supported by JSPS-KAKENHI Grant number 25K08531. This work was supported by "Advanced Research Infrastructure for Materials and Nanotechnology in Japan (ARIM)" of the Ministry of Education, Culture, Sports, Science and Technology (MEXT). Proposal Numbers JPMXP1225NM5095 and JPMXP1226NM5009.

Declaration of generative AI in scientific writing.

No generative AI has been used in the preparation of this paper.

Preprints.

No preprints have been shared.

Funding

This work was supported by JSPS-KAKENHI Grant number 25K08531.

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