Modeling of primary defect aggregation in tracks of swift heavy ions in alkali halides

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Abstract

Dynamic Monte Carlo simulations of primary defect aggregation in tracks of swift heavy ions in alkali halides have been performed. The study was motivated by recent experimental findings on dense F-center clusters observed even at 15 K in LiF irradiated by GeV heavy ions. We relied on a recently suggested model, which assumes local heating and diffusion of single defects in their excited electronic state. The main parameters (migration energy, initial defect concentration, diffusion time) are estimated from available experimental data. Simulation results confirm the formation of a core with large F-aggregates and a broad halo of single and dimer defects around it. The fraction of large (more than 10 defects) aggregates in the core is negligible, making ESR detection impossible. The low sensitivity of the model to changes in parameters can be explained by its universal features, which we first report in this paper. Unexpected power laws in the kinetics of single defect concentrations are also emphasized.
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1. Introduction

Alkali halides as a target material remain a focus of radiation damage studies over the past decades. This interest is mainly due to the simple structure of their ionic lattice and easiness of defect creation. While the damage structure and mechanisms for low ionizing radiation (e.g. electrons, photons, neutrons) are relatively well understood [1,2], the effects of irradiation by energetic (GeV range) heavy ions leave many open questions.

A large bulk of experimental data [3–6] on the irradiation of alkali halides (mainly LiF) with swift heavy ions shows the formation of ion tracks with complex structures. One of the peculiarities in comparison to conventional low ionizing radiation is the formation of a dense core with modified electronic density along the path of the projectile [3,5]. For LiF, this effect is observed if the energy loss dE/dx exceeds a critical value of approximately 10 keV/nm. The most remarkable fact is that the track core is created even at temperatures as low as 15 K [7], at which primary Frenkel defects are believed not to diffuse and aggregate. The narrow (1–1.5 nm) core of defect aggregates is surrounded by a much broader (20–40 nm) halo containing mainly single and double Frenkel defects [3–5].

Recently, a microscopic model of track formation, which takes into account local heating and defect diffusion in their excited electronic state has been suggested [8,9]. In the present paper, we perform additional simulations of this model and study its applicability limits.

2. The model

The physical basics and parameter estimation for the model to be studied have been already presented in detail [9]. In this section, we outline the basic physical assumptions and give typical values of the simulation parameters.

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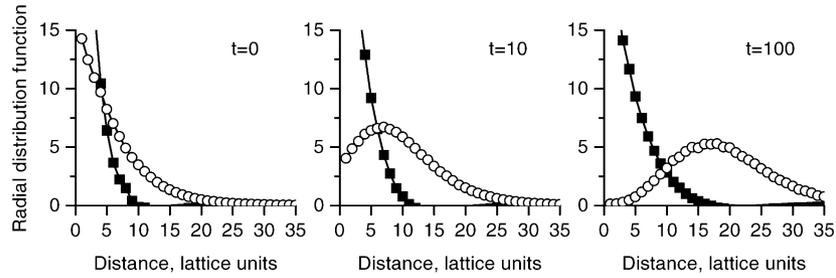*E-mail address:* slava@latnet.lv (V. Kashcheyevs).

Fig. 1. Radial distribution of single F-centers (○) and defect aggregates (■) in the course of the aggregation process. Corresponding fractions of single defects (aggregates) are 87% (13%), 44% (56%) and 14% (86%) for time instants $t=0$, 10 and 100 hops per defect, respectively. Model parameters: $a=5$ (lattice units), $L=100$ (lattice units), $c_0=0.1$ (dimensionless fraction). All distributions are normalized to unity.

1. Primary Frenkel defects, F- and H-centers, are created in a simple cubic lattice of size L in the z direction and of infinite size in the x and y directions. (The z-axis is parallel to the trajectory of the projectile.) Periodic boundary conditions are applied along z-direction.
2. The mobility of defects is ensured by the local temperature increase and the radiation enhanced diffusion [1–10]. H-centers are more mobile than F-centers and separate from the F-centers in space before the latter start to migrate. Only the diffusion and aggregation of the remaining F-centers is simulated. The estimated time scale of the diffusion process is $\tau_F=10^{-11}$ s [9].
3. At the starting point of a simulation, the F-centers are placed randomly in the lattice according to the exponential density distribution $c(r)=c_0\exp(-r/a)$ where r is the distance from the track axis. The track radius a and the initial density c_0 can be varied as the parameters. Estimated realistic values [9] for LiF and 10 MeV/u projectiles vary with $c_0=0.01–0.1$ defects per site [$(0.5–5)\times10^{21}$ cm^{-3}] and $a=3–10$ lattice constants (1–3 nm).
4. Single F defects start to perform random hops at $t=0$ and become immobile as soon as they meet another defect at one of the nearest lattice sites. The simulation is stopped when the total number of hop attempts per defect t reaches M, which is the natural measure for the lifetime of the local temperature increase. From the experimentally measured diffusion distances, M is estimated as $M=25–100$ [9].

3. Simulation results

3.1. Comparison with the experiment

The key results of physical significance that can be extracted from the Monte Carlo simulations of the model are as follows:

The experimentally observed track structure [3,4] is qualitatively reproduced by the model. Fig. 1 shows the time evolution of the radial distribution of single defects (open circles) and defect aggregates containing two and more F-centers (solid squares). (Analogous graphs in Kotomin et al. [9] suffer from uncorrected distortions due to defect counting on a discrete lattice.) As the aggregation process evolves, more and more defects stick in the core, but single F-centers separate, thus forming a halo.

The fraction of large aggregates (containing 10 and more defects) which could transform into metallic colloids is negligible for realistic values of initial density c_0 [9]. This explains the absence of any spin electron resonance signal from the microscopic aggregates in the core [4].

The defect clusters are well separated from each other and do not form a continuous metallization region along the track axis. Therefore, metallic conductivity is not expected in this system.

3.2. Universal features of the model

We have performed a thorough analysis of Monte Carlo data which has revealed a number of universal relations emphasizing general robustness of the model.

According to the model, even at the initial moment $t=0$, some of the F-centers will be already aggregated due to obvious statistical reasons. A simple argument enables us to calculate the fraction of free F-centers among all defects at $t=0$ (denoted C_{F0}) as a function of the initial concentration c_0. Let us consider a site at distance r from the track axis. Since the defects are distributed independently in the lattice, the (not-normalized) probability that the selected site will be occupied and the neighboring six sites will be empty is roughly $p(r)=c(r)[1-c(r)]^6$. Integrating this probability over the whole space and normalizing gives:

$$C_{F0}=\left(\int_0^\infty 2\pi r\, c(r)\mathrm{d}r\right)^{-1}\int_0^\infty 2\pi r\, p(r)\mathrm{d}r$$
$$=1-\frac{3c_0}{2}+\frac{5c_0^2}{3}-\frac{5c_0^3}{4}+\frac{3c_0^4}{5}-\frac{c_0^5}{6}+\frac{c_0^6}{49} \qquad (1)$$
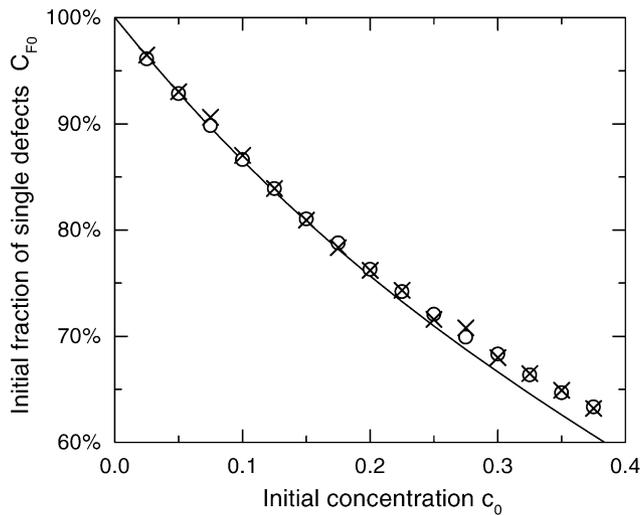
This result is compared to simulations in Fig. 2. Devi-

Fig. 2. The fraction of single F-centers at $t=0$ according to the analytical result [Eq. (1)] (solid line) and the simulations with $a=5$ ($\times$) and $a=10$ ($\bigcirc$) (both in lattice units).

ations appear only for high values of c_0 (which are physically not reasonable) due to the ignored radial gradient of $c(r)$ over distances of one lattice spacing.

A remarkable feature of the polynomial [Eq. (1)] is its independence of the track radius a. This confirms the observation [9] that simulations give the same results for different a-values (see also Fig. 2).

Another intriguing relation has been discovered after the kinetics of the fraction of single defects that survive the aggregation process (C_F) have been carefully re-examined. The functional dependence of C_F (c_0, t) on
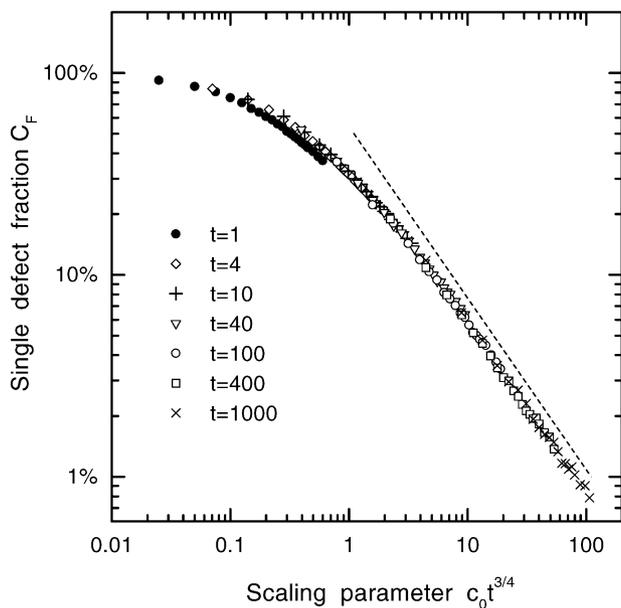

Fig. 3. Scaling function for the fraction of single F-centers $C_F(c_0, t)$. The straight line guiding the eye has slope -0.85.

its two arguments—initial concentration and time—appears to be very well described by a single variable $\xi = c_0 t^\nu$ with the exponent $\nu = 3/4$ in a wide range of concentrations and times. Fig. 3 shows C_F as a function of ξ for c_0 between 0.025 and 0.6 and t going from 1 to 1000. Within the statistical errors, the data fall on a unified scaling curve for $t \geq 4$, which means that a universal regime is attained already after each defect has performed just few hops. The long-time/high-concentration tail of the dependence $C_F(\xi)$ obeys an asymptotic power law $C_F \propto \xi^{-\alpha}$ with $\alpha = 0.85 \pm 0.01$.

Analysis of the kinetic equations for the defect concentration could reveal the nature of the observed power law relations. However, such an approach was not attempted in the present work.

4. Conclusions

A model for the track structure formation in alkali halides irradiated with swift heavy ions has been discussed. The kinetics of F-center aggregation is studied by means of Monte Carlo simulation using the experimental estimates of the model parameters obtained earlier [9].

Although some of the physical assumptions (initial separation of F- and H-centers, F-center diffusion in the excited electronic state) should be further investigated and justified, the model reproduces well the main features of the track structure: a dense core of defect aggregates surrounded by a broad halo of single defects; absence of metallic conductivity along the track; and negligible concentration of metallic colloids.

Despite the low precision or even the lack of experimental data for some of the model parameters, the physical conclusions appear to be quite robust. We reported additional evidence of the universality of the model, namely, tolerance to changes in track radius and data collapse for aggregation kinetics at different initial densities. The observed power-law behavior needs additional theoretical investigation, which could improve our understanding of the aggregation process.

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