Modeling of primary defect aggregation in tracks of swift heavy ions in LiF

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(Received 27 April 2001; revised manuscript received 12 July 2001; published 24 September 2001)

To simulate aggregation of primary *F* centers created along the path of swift heavy ions in LiF, Monte Carlo simulations were developed. Parameters relevant for defect aggregation as a result of their random hopping, such as the migration energy, temperature in the track, initial defect concentration, and diffusion time, were estimated from available experimental data. It is estimated that in the electronically excited state and under temperature locally increased up to 1200 K *F* centers are mobile enough to make several tens of hops. Most of the *F* aggregates formed are extremely small and consist only of two or three *F* centers. The fraction of larger *F* clusters (with more than 10 defects) is negligibly small, at least for defect concentrations reasonable for ion tracks. Even at the largest initial defect densities, the aggregates are isolated from each other and do not form a percolating trail of defects. Such a track morphology is in good agreement with various experimental results.

DOI: 10.1103/PhysRevB.64.144108 PACS number(s): 61.72.Ji, 61.80.Az, 61.80.Hg, 61.82.Ms

I. INTRODUCTION

When passing through dielectric materials, swift heavy ions induce a trail of electronic excitations and ionizations of the target atoms. In subsequent processes, the excitations can decay nonradiatively creating various types of damage structures in the lattice such as single point defects, defect clusters, local phase transitions, or even decomposition of the solid by radiolysis.[1–5] The peculiarities of the damage process due to energetic heavy ion is mainly determined by the two following facts: (1) the excitation density is very high, reaching up to several keV nm^3 close to the ion path, and decreasing with the radial distance approximately as $1/r^2$, and (2) the energy deposition to the target electrons is extremely fast occurring on the time scale 10^{-16}–10^{-14} s (Refs. 6–10). Compared to this scale, defects in the lattice are created at a much later stage, around 10^{-12}–10^{-11} s. In addition to these specific projectile characteristics, damage creation in a given material depends on many other aspects such as material properties (e.g., the nature of chemical bonding) and irradiation conditions (e.g., temperature).

Although several attempts have been made to give a kind of general description for track formation,[11,12] at present the interpretation of damage processes under heavy ion irradiation is difficult and still leaves many open questions. This is also true for track formation in ionic crystals, for which—in contrast to many other insulators—amorphization is not expected. Detailed knowledge exists concerning defect creation upon excitation of the electron subsystem by low-energy ionizing radiation (e.g., gamma rays, electrons, or neutrons), in particular for alkali and alkali-earth halides. Their damage mechanism is based on self-trapping of excitons (either by relaxation of free excitons or by electron-hole recombination) and their subsequent nonradiative decay into Frenkel defects. In this paper, we focus on lithium fluoride (LiF) in which the most significant stable defects at room temperature are electronic *F* centers (a halogen-ion vacancy with one trapped electron) and complex electron centers. The creation of complex color centers and aggregation of neighboring defects can lead to F_2, F_3, or F_n centers, eventually even to metallic alkali colloids, and to the aggregates of the complementary hole centers (V_n- centers and molecular fluorine aggregates). This *scenario* probably also holds for the irradiation with energetic heavy ions, but the high dose and thus the high density of Frenkel defects close to the ion path, certainly plays a crucial role in defect diffusion and aggregation processes and therefore in the final damage structure in the ion tracks.

In this paper, we present model calculations concerning the aggregation of such primary defects along the trajectory of heavy ions of several hundred MeV. As material, we used LiF crystals mainly due to the fact that a large body of experimental track data exists[4–8,11] thus providing various parameters relevant for our calculations.[2,13]

II. EXPERIMENTAL RESULTS OF ION-INDUCED DAMAGE IN LiF CRYSTALS

Ion tracks in LiF have been studied by various techniques such as optical absorption spectroscopy, small-angle x-ray scattering (SAXS), chemical etching, and scanning force microscopy.[2,4,5,14,15] Combining the different results, the following track description can be given.

(1) In a large halo region around the ion trajectory, the dominant type of defects are single *F* and dimer F_2 centers identified by their absorption bands in the ultraviolet and visible spectral range between 200 and 900 nm. The complementary hole centers are absorbing at room temperature in the vacuum ultraviolet region. They were not studied here but are known to coexist. Figure 1 shows a typical absorption spectra of crystals irradiated with U ions (1.4 GeV) and Ni ions (580 MeV) of 5×10^{10} ions cm^2. The efficiency of the creation of single defects is approximately the same as under conventional irradiation. At higher fluences, the spectra become more complex due to track overlapping. Although the position of some F_n bands is known [F_3 centers (318, 380 nm) and F_4 centers (518, 540 nm)], the analysis of such aggregates is not straightforward. Finally, it should be men-

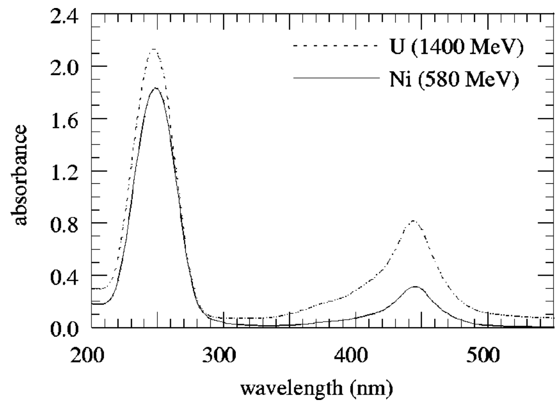

FIG. 1. Absorption spectra of LiF crystals irradiated at room temperature with U ions (1400 MeV; $dE/dx=28$ keV/nm) and Ni (580 MeV; $dE/dx=10$ keV/nm) of about 5×10^{10} ions/cm^2. The bands at 245 and 450 nm belong to single F centers and F_2 centers, respectively. The complementary hole centers absorb in the vacuum ultraviolet region [$\sim$113 nm (Ref. 16)], and cannot be seen here.

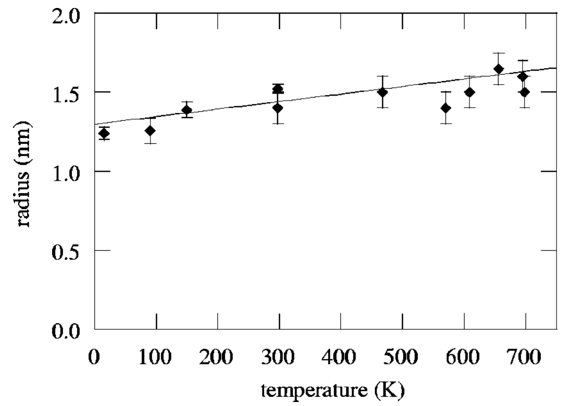

FIG. 2. Track radii deduced from SAXS measurements as a function of the irradiation temperature. The LiF crystals were irradiated with Pb ions of 730 MeV (Ref. 17) or 1230 MeV (Ref. 18).

tioned that the absorption bands of Li colloids are expected to be close to the F_n bands, but they are not so well known and it is difficult to identify them in complex optical spectra. From the evolution of the F-center concentration as a function of the ion fluence, the radius of the halo surrounding the track core can be deduced. It varies from 5 nm for light ions (e.g., S, 1.6 MeV u, 4.3 keV nm) up to 40 nm for heavier ions (e.g., Au, 11.4 MeV u, 24 keV nm).[2,4]

(2) If the ions surpass a critical energy loss (dE/dx) of about 10 keV nm, a new effect occurs, namely the creation of complex defect clusters in a narrow cylindrical core with a radius between 1–1.5 nm (Refs. 2 and 14). The size of this track core has been determined from analyzing the highly anisotropic SAXS pattern which is due to a modified electronic density $\Delta\rho$ in the tracks (the scattering is proportional to $\Delta\rho^2$). Note that the single F centers do not contribute to the SAXS contrast. Above this threshold, tracks can be attacked by a suitable etchant. Both phenomena are stable up to much higher temperatures than single F and hole centers exist. Although the nature of the specific damage in the core is not studied so far, several observations indicate that the track consists of a quasicylindrical, discontinuous array of defect aggregates.

Track formation in LiF crystals was also tested in a wide temperature range from 15 to 750 K (Refs. 17 and 18). The most remarkable fact is that the track core is created even when the irradiation is performed at as low a temperature as 15 K. This is in clear contrast to conventional irradiations (x rays or fast electrons), where aggregation processes at such a low temperature are strongly suppressed because primary Frenkel defects are not mobile. Based on the temperature dependence of the SAXS radius (Fig. 2), we assume that during irradiation, the local temperature around the ion path increases by ΔT.

III. DEFECT AGGREGATION

From a general point of view, aggregation of single F centers to more complex clusters is possible only under the following specific conditions:[4,19] (1) F centers and hole centers must be separated in space, (2) the concentration of F centers surviving recombination with complementary hole (H) centers must be sufficiently high ($C_F\geqslant10^{21}$ cm^{-3}), and (3) diffusion of F centers must be possible.

A. Estimate of defect concentration in the track

The concentration of primary F defects in the track core can be estimated from optical measurements where we find a typical value of about $C_F\approx2\times10^6$ F centers in the halo region of a single track.[4] Monte Carlo calculations of the lateral energy distribution[4,20,21] demonstrate that about 30% of the total ion energy is deposited in the track core and about 70% in the halo. Assuming similar defect creation efficiencies in both regions, the number of primary point defects in the track core should be around 10^6 (Ref. 4). The volume of the track core is given by the cylinder radius (as deduced from the SAXS experiments) and the length, corresponding to the ion range. Taking a projectile of 10 MeV per nucleon with a typically range of 80 μm (Refs. 14 and 22) and a radius of 1.5 nm, the track volume is 5.7 $\times10^{-16}$ cm^3 [corresponding to about 3.4×10^7 (Li^+-F^-) ion pairs]. It follows that the concentration of primary Frenkel pairs in the track core is about 2×10^{21} cm^{-3} which is about 3% of all lattice sites. As mentioned above, this number is certainly high enough for efficient aggregation of F centers.[19]

B. Estimate of diffusion parameters and local heating in the track

For a reasonable simulation of the aggregation process, various parameters such as diffusion length, activation energy, and local temperature have to be known. The mean square diffusion length $\langle x^2\rangle$ of defects is given by[23]

$$\langle x^2\rangle=\lambda^2M=6\tau D, \tag{1}$$

where $\lambda=\sqrt{2}d=0.28$ nm is the hopping (F-F ion) distance to the nearest lattice site (Li-F distance is $d=0.2$ nm), M is the number of hops of a single defect, τ is the diffusion time, and D is the diffusion coefficient determined by

$$D=\frac{1}{6}\nu\lambda^2\exp(-Q/kT^*), \tag{2}$$

where $\nu\approx 10^{13}\ \mathrm{s}^{-1}$ is the frequency (attempt) factor, Q is the hopping activation energy and T^* denotes the effective temperature during defect diffusion given by the irradiation temperature T_{irr} plus the temperature increase in the track region ΔT $(T^*=T_{\mathrm{irr}}+\Delta T)$.

In order to estimate ΔT, the activation energy for the diffusion process has to be known. Unfortunately, for heavy ions it is impossible to monitore experimentally the diffusion kinetics in the track. From conventional irradiations it is known that defect aggregation takes place via the *radiation-enhanced diffusion* (RED),[19,24] i.e., F-centers migrate in the electronically excited state and thus, with a much lower activation energy than under conventional thermal activation.[24,25] In alkali halides, a typical activation energy for F-center diffusion in the excited state is about $Q_F\approx 0.2$ eV (Refs. 25 and 26) with the lifetime of the order of nanoseconds.[8,11] Combining Eqs. (1) and (2), and supposing that the mean diffusion length $\langle x_F\rangle$ of F centers corresponds to the SAXS radius of the core r_c, we obtain the following relation:

$$r_c^2\sim\exp[-Q_F/k(T_{\mathrm{irr}}+\Delta T)]. \tag{3}$$

Inserting the SAXS radius r_c from irradiations at two different temperatures T_{irr} (Fig. 2), we obtain $\Delta T=1200\pm 100$ K (Ref. 18). The effective local temperature for irradiation at 15 K is therefore $T^*=1215$ K and close to the melting point (1115 K). Notice that this estimate comes directly from the activation energy $Q_F\approx 0.2$ eV. For higher values, e.g., $Q_F=1$ eV, the temperature increase would be unreasonable large, reaching $\Delta T\geqslant 2000$ K, i.e., above the boiling point of 1949 K in LiF. It should be emphasized that point defects and their aggregates do not exist in a melt phase. For our simulations, we therefore assume that track formation occurs in the solid phase under a moderate increase of the temperature (a local heating).[13,27]

We can also estimate the diffusion length $\langle x_H\rangle$ of primary H centers. In contrast to the excited F centers, the activation energy for H-center diffusion in the ground state is well known and has a smaller value of $Q_H\approx 0.1$ eV (Refs. 11 and 28). At $T^*=1215$ K, the diffusion coefficient $D_H\approx 5\times 10^{-4}$ cm^2 s and thus the diffusion length for the H centers becomes $\langle x_H\rangle\approx 2$ nm, larger than the core radius (typically 1.5 nm), $\langle x_F\rangle$. Considering that RED is also effective for hole centers, $\langle x_H\rangle$ would be even larger.[11,29] We thus conclude, that under given conditions, F centers and H centers are obviously separated in space and thus efficient aggregation of F centers can take place.

IV. COMPUTER MODELING OF DEFECT AGGREGATION

A. Model description and simulation parameters

Based on the above-estimated experimental parameters, we performed Monte Carlo computer simulations of the F-center aggregation kinetics. The basic physical model has the following assumptions.[13,27]

(i) Defects 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.)

(ii) The initial distribution of defects is described by the exponential function $c(r)=c_0\exp(-r/a)$, where r is the distance from the ion path, and the initial density c_0 at the track center and the radius a can be varied. The total number of F centers in a whole track is given by

$$N(\infty)=\int_0^\infty c(r)L2\pi r dr=2\pi a^2Lc_0 \tag{4}$$

and the fraction of defects inside a cylinder of some arbitrary radius r is

$$N(r)/N(\infty)=1-\left(1+\frac{r}{a}\right)e^{-r/a}. \tag{5}$$

According to Eq. (5), the fraction of defects created inside a cylinder with radius $r=a$ equals $N(a)/N(\infty)=26\%$, in agreement with the estimated lateral energy distribution for LiF irradiated with heavy ions above the dE/dx threshold of 10 keV/nm (Ref. 4).

(iii) Hole H centers are more mobile than F centers and are separated from the F centers before these latter start to migrate. Note that in LiF, H centers are known to aggregate to fluorine (F_2) molecules as point defects or as molecular gas bubbles. In this way, their recombination with F centers is effectively prevented.[16,30,31] Based on this, our model calculations on F-center aggregation do not take into account H centers.

(iv) Single F defects start to perform random hops at $t=0$ and come to rest as soon as they meet another defect at one of the nearest lattice sites. As estimated above, the typical diffusion time is $\tau_F=10^{-11}$ s which corresponds to a number of F-center hops $M\approx 25$. Periodic boundary conditions have been applied on the z axis.

A series of simulations has been performed using the following dimensionless parameters. The length of the ion track was fixed as $L=100\lambda$, which is sufficiently large to exclude finite-size effects along the z axis and restrict the calculations to a reasonable computing time (proportional to L^2). All distances are given in $F-F$ separation units of $\lambda=0.28$ nm. The initial dimensionless defect density c_0 was varied from 0.025 to 0.6 in steps of 0.025, including the estimated experimental value ($c_0=0.03$). This is a fraction of anion sites occupied by F centers.

The distribution of the F clusters, the fraction of surviving single defects and their radial distribution was recorded during random walks where the number of hops was varied between $M=0$ and 100. We define as a *cluster* F_n aggregate center with $n\geqslant 2$, i.e., containing two or more single F centers. With this assumption, we do not included structural properties of smaller (F_2 centers) or larger aggregate centers (Li colloids) in the lattice. Our simulations show that the results depend only weakly on the initial core radius a, therefore only data for $a=5.0$ are presented.

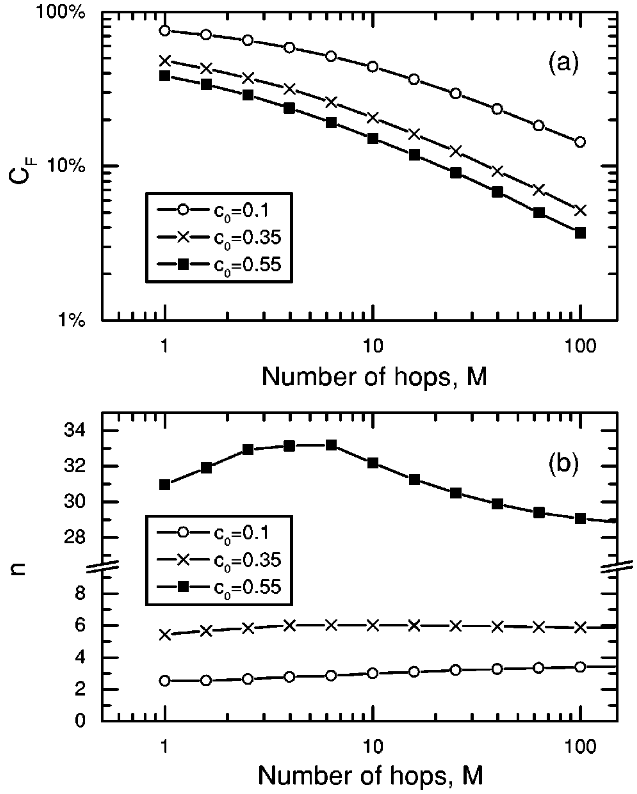

FIG. 3. (a) The fraction of single F centers (C_F) with respect to all F centers, and (b) mean number of F centers in clusters (n) as a function of the number of hops for different initial defect densities c_0.

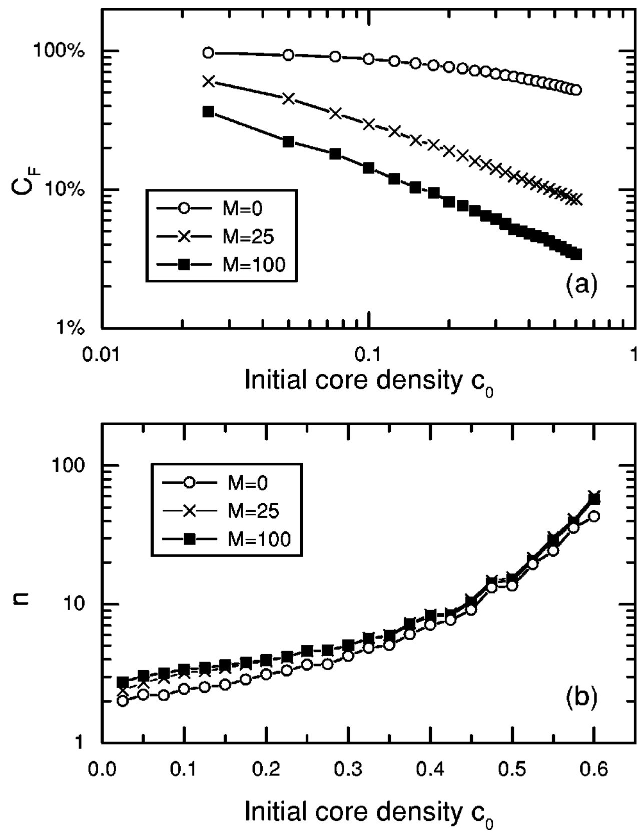

FIG. 4. (a) The fraction of single F centers (C_F) and (b) a mean number of F centers in clusters (n) as a function of the initial defect densities c_0 for various numbers of random hops.

V. SIMULATION RESULTS

Figure 3(a) shows the fraction of single F centers (C_F) as a function of the number of defect hops. Depending on the initial defect densities c_0, some of the F centers are already statistically aggregated even before starting random walks: 13% at $c_0=0.1$, 36% at $c_0=0.35$, and 46% at $c_0=0.55$. In the course of diffusion, the number of single defects decreases significantly, reaching after 25 hops the value of $C_F=29\%$, 12%, and 9%, respectively. The evolution of C_F versus the number of hops, proportional to the diffusion time t, follows a simple asymptotic power-law $C_F \propto t^{-0.6}$. This asymptotic dependence is the same for all initial defect densities tested here.

The average number of F centers per cluster (n) as a function of the hop number is presented in Fig. 3(b). For a realistic initial defect density ($c_0=0.1$), n is extremely small (2–3) and remains nearly constant during the entire hopping process (up to $M=100$). In other words, the average cluster size varies very slowly with time. For a large defect density ($c_0=0.55$), n slightly increases in the initial stage $M=6$ and then returns back to the initial value. Obviously, growth of existing large and formation of new small clusters are the two competing processes, reaching a state of quasiequilibrium after a certain number of hops.

Figure 4 shows again the fraction of single F centers (C_F) and the average cluster size (n), this time as a function of the initial defect density c_0. C_F can be asymptotically fitted by a power law $C_F \propto c_0^{-\alpha}$ with the exponent $\alpha=-0.84\pm0.01$. The mean number of F centers in clusters (2–3) is almost independent of the aggregation time but strongly increases for initial densities $c_0 \geqslant 0.4$.

Figure 5 shows the fraction of F centers (ν_F) in relatively big aggregates ($n \geqslant 10$) with respect to all F centers (single and aggregated defects). For intermediate and high initial densities ($c_0 \geqslant 0.2$), defect hopping obviously results in a significant increase of big aggregates. This observation

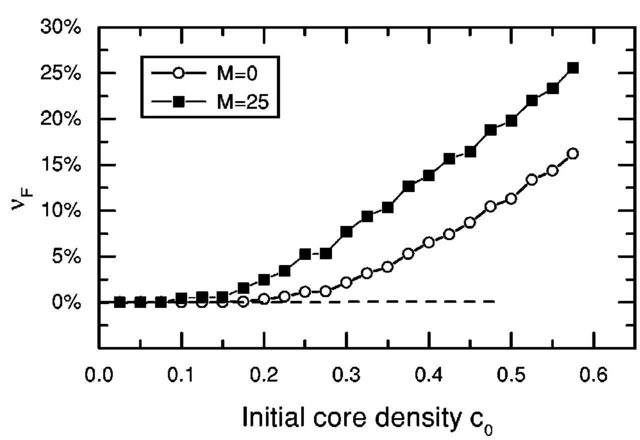

FIG. 5. The fraction of F centers in big clusters ($n>10$) with respect to all F centers (single and aggregated) versus the initial defect densities c_0.

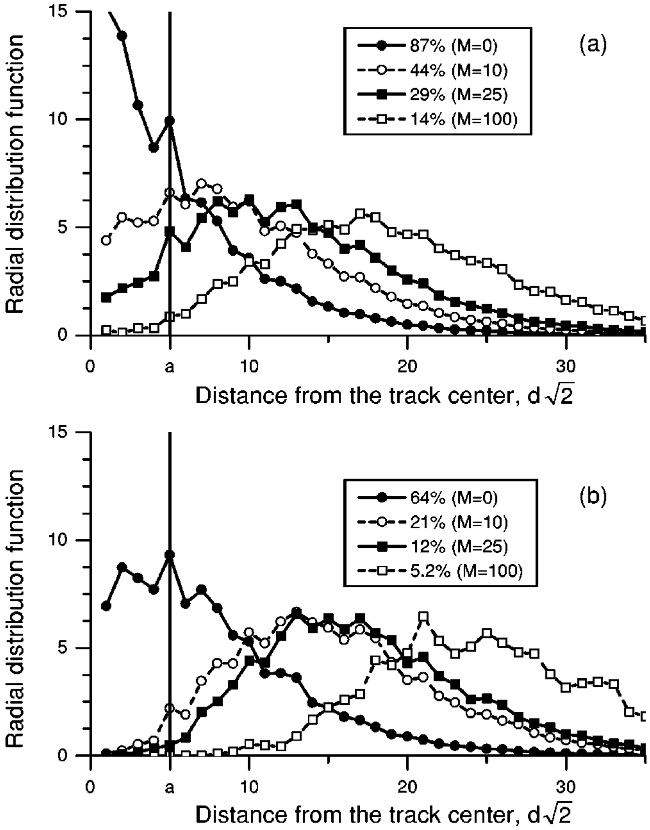

FIG. 6. The radial distribution of isolated defects for defect density (a) $c_0=0.1$ and (b) $c_0=0.35$, for various hop numbers M. The percentage in the legend gives the fraction of single F centers survived after the given number of hops [see also Fig. 3(a)]. The core radius $a=5\lambda$ (~ 1.4 nm) is marked by the vertical line. All distribution curves are normalized to unity.

agrees with the results of a previous study[13] in which a step-like initial defect distribution was used. Note however, that for the initial core density of $c_0=0.1$ (most realistic case), the fraction of F centers in big aggregates is only 0.005, i.e., extremely small.

The radial distributions of single F centers for two different initial defect concentrations is presented in Fig. 6(a) ($c_0=0.10$) and Fig. 6(b) ($c_0=0.35$). As mentioned above, the fraction of F centers which aggregated before the hopping process is 13% ($c_0=0.1$) and 36% ($c_0=0.35$) (cf. Fig. 3). With increasing number of hops, the radial distribution of the single F centers changes, moving out from the track axis to the larger distances. After 100 hops, the distribution maximum has shifted to (15–20) λ length units (corresponding to $\approx$4.2–5.6 nm) for $c_0=0.1$ and even 25λ ($\approx$7 nm) for $c_0=0.35$. For the small initial defects densities [Fig. 6(a)], there is still a considerable amount (14%) of single F defects close to the ion path, while for large densities [Fig. 6(b)], most single defects close to the ion path are aggregated even before the hopping process. In the legend of Fig. 6, the fractions of single defects surviving M hops are indicated. After 25 hops and for $c_0=0.1$, which we consider more realistic estimate, C_F decreased from the initial 87% down to 29% [see also Fig. 3(a)]. This is in qualitative agreement with the experimentally observed F-center concentration in the track halo [see Sec. II(1)].

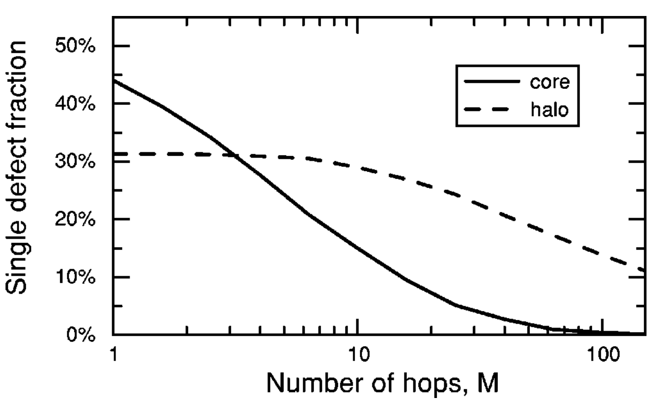

FIG. 7. The fraction of single F centers for the initial defect density $c_0=0.1$ as a function of number of hops M. The solid and dashed curves show the concentration kinetics of single F centers inside core and in a halo, respectively. Their sum is C_F in Fig. 3(a).

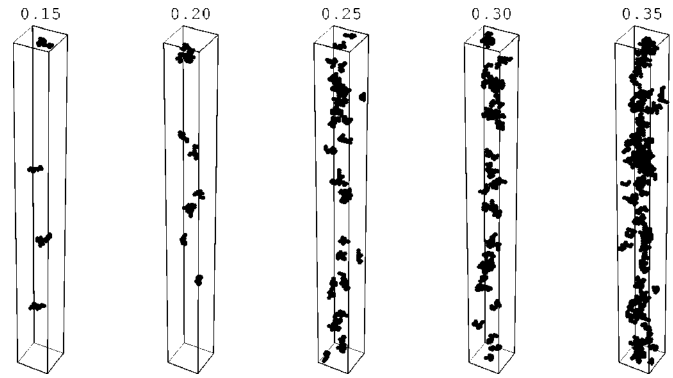

FIG. 8. Visualization of the F-center aggregates formed along the ion trajectory for various initial defect densities c_0.

The dynamics of F-center aggregation, illustrated in Fig. 7, demonstrates in more details quite different situations inside (core) and outside (halo) of a fixed track radius $a=5\lambda$. In the initial stage, concentration of single F centers within the core region (solid curve) exceeds that in the halo (dashed curve). However, after 25 hops, C_F in a core decreases from initial 44% down to 5%. That is, single F centers survive mainly in the halo (decrease from 32% down to 12%). This can be explained by the obvious fact that at larger distance from the ion path, the density of F centers is much smaller and the probability for two F centers to meet and thus to aggregate becomes less and less likely.

Finally, in Fig. 8, we visualize the formation of large F-center aggregates ($n>10$) in ion tracks for various initial defect densities. For $c_0\leqslant 0.25$, the aggregates are well separated from each other and do not form a continuous trail. Therefore, an electrical conductivity based for example on percolating small metallic Li clusters should not be expected, in agreement with test experiments.[32]

VI. DISCUSSION AND CONCLUSIONS

Monte Carlo computer simulations were performed for aggregation kinetics of F centers created along the trajectory of swift heavy ions. Main parameters, such as the migration energy, temperature in the track core, initial defect concentration, or diffusion time were estimated from the experimental data available for LiF. From the optical absorption measurements in LiF crystals irradiated with heavy ions (having the stopping power above the threshold of 10 keV/nm), we can conclude that in the core after irradiation about 3% of the lattice sites are occupied by the *F* centers or their aggregates. In the initial stage of damage creation, the number of defects is obviously larger. We therefore assumed that $c_0 \approx 0.1$ is a realistic value. In order to explain the efficient aggregation of the *F* centers, we have to assume that they diffuse in the electronically excited state. In addition, *F*-center aggregation requires a moderate local heating within the track. Based on experimental SAXS (Fig. 2) results for low and high temperature irradiations, we propose a local temperature increase of $\approx$1200 K. The short lifetime ($\tau \approx 10^{-11}$ s) of the excited *F* centers corresponds to about 25 random hops on the lattice. For a larger number of hops, the simulation results show no qualitative changes, i.e., within this time limit a quasistationary state of aggregation is already reached.

According to the calculations, most defect clusters are very small and typically consist of 2–3 *F* centers. The fraction of single *F* centers decreases with the aggregation time reaching a quasiequilibrium of about 30% after 25 hops. They occupy mainly lattice sites in the track halo, i.e., a few nanometers away from the original ion path. Larger *F*-aggregates (e.g., $n > 10$) are in negligible concentration, even for moderate initial defect concentrations ($c_0 < 0.3$). They could play a role for much larger concentrations, that are probably not realistic. The formation of extremely small defect aggregates along the track is in agreement with experimental results from small angle x-ray scattering.[2,4,5] It also explains the absence of any electron spin resonance[33] as expected for metallic aggregates that are larger than 10 nm (Refs. 34–36). Due to the small size of the aggregates and the absence of larger clusters, a discontinuous defect trail is formed. Such a track morphology also explains why tracks in LiF did not show any increase of electrical conductivity.[32]

Although our model calculations gave good agreement with the experimental situation, several questions remain open. In particular, the diffusion of *F* center in the electronically excited state and the efficient initial separation of the electron and hole centers have to be further investigated, at least theoretically. It should be mentioned that at present, there is no direct experimental technique suitable for a study of the microstructure of such small *F*-center aggregates. Also, the local temperature increase on the time scale of 10^{-11} s is very difficult to access.

ACKNOWLEDGMENTS

V.K. and V.N.K. were partly supported by the European Center of Excellence in Advanced Materials Research and Technology (Contract No. ICA-1-CT-2000-7007).

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