

Damage to nonmetallic materials due to braking of high-energy ions

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The paper considers the defect formation in solids due to ionization of inner shells of atoms (ions) located at neighboring lattice sites with the subsequent Coulomb expulsion of atoms from their sites. This mechanism is effective only for high-energy bombarding ions. The effectiveness of the mechanism is analyzed versus target properties and parameters of bombarding ions. We demonstrate that in some cases more defects can be generated by this mechanism than through the elastic slowing down of high-energy ions. The distribution of primary displacement defects caused by elastic and nonelastic slowing down over the projected range is numerically calculated. The calculations correlate with the published data on the distribution of point defects in diamond and boron nitride bombarded with high-energy ions ($E=1-8$ MeV/a.m.u.) © 1995 American Institute of Physics.

1. INTRODUCTION

High-energy charged particles primarily lose energy in solids due to excitation and ionization of atoms and generation of plasmons (inelastic losses) and due to collisions with nuclei of the target (elastic losses). A new ionization mechanism for the damage to the target caused by inelastic slowing down of charged particles in crystals¹ is considered. In some cases this mechanism may be the major contributor to the damage produced in solids by high-energy ions.

Varley² was the first to propose an ionization mechanism of damage for binary compounds. According to Varley, an inner anion shell is first ionized by a high-energy particle. The resulting Auger cascade³ produces a multiply charged ion. The particle thus creates an unstable group of positive ions. The Coulomb repulsion of these can then generate a lattice defect. Later, Karpov and Klinger⁴ suggested a new mechanism of damage by ionizing radiation in nonmetallic crystals. The participation of an impurity atom in inelastic scattering is essential for this mechanism. Incident radiation ionizes an inner shell of a host atom located next to a charged impurity. Then, by Varley's mechanism,² the host atom is multiply ionized through the Auger cascade. A couple of positive ions is thus created, and the Coulomb repulsion transfers one member of the couple to an interstitial position. This mechanism has been used to interpret some experiments on defect generation in semiconductors irradiated with electrons and gammas with energy below a threshold.⁵⁻⁷

The damage mechanism proposed here can be applied to crystals with either ion or covalent binding. In terms of this mechanism, a defect is generated through ionization by a high-energy particle of the inner shells of two host atoms located at neighboring lattice sites. The mechanism is only effective when the solid is exposed to high-energy ions. Specifically, the probability for inner shells of neighboring atoms to be ionized is controlled by the mean free path L of a high-energy particle between two subsequent ionizations of inner shells

$$L = (\sigma N_e)^{-1}, \quad (1)$$

where σ is the inner-shell impact ionization cross section and N_e is the number of electrons in this shell per unit volume. For high-energy electrons, the impact ionization cross section σ can be estimated in the sudden approximation³ as

$$\sigma = \frac{\pi e^4}{E_e} \left(\frac{1}{E_b} - \frac{1}{E_e} \right), \quad (2)$$

where E_e is the fast electron energy and E_b is the inner-shell electron binding energy. For estimates we assume $E_b=300$ eV because this value is usually less than the binding energy of K -electrons in most atoms and slightly higher than that of L - and M -electrons in medium and heavy atoms, respectively (hereinafter we shall discuss only K -electrons because all the results can be easily generalized for other inner shells of target atoms which can be ionized with subsequent Auger cascades).

For incident electron energy $E_e=2E_b$, which corresponds to the maximum in $\sigma(E_e)$, the cross section for the inner-shell ionization is $\sigma \leq 10^{-19}$ cm², which is equivalent to $L \geq 10^3$ Å. (An important point is that this estimate of L neglects the braking of electrons, and the estimated mean free path therefore exceeds not only the interatomic distance, but also the entire electron range (at $E_e \sim 2E_b$), so the probability of the sequential ionization of inner shells of neighboring atoms by a fast electron is practically zero). The sequential ionization of K -shells of atoms located at neighboring lattice sites by a γ -quantum due to the Compton scattering is theoretically feasible, but the probability of this process is even smaller than that of the electron ionization because the Compton scattering cross section is on the order of $\sim 10^{-26}$ cm², which corresponds to a mean free path $L \sim 10^2$ cm.⁸ This probability can be also assumed zero, since $W \propto L^{-2}$ (the formula for $W(L)$ will be derived below).

Two cases are possible when a solid is bombarded with ions.

1. For ion energy ~ 10 keV/a.m.u., using the sudden approximation to estimate the impact ionization cross section does not make any sense because the energy an ion transfers to a K -electron in a head-on collision is smaller than electron's binding energy. However, more accurate calculations and experimental data⁹ indicate that K -shells can still be ionized by a medium-energy ion (this discrepancy is caused by the fact that classical impact parameters cannot be applied here); the K -shell ionization cross section is less than 10^{-20} cm² (free path $L \geq 10^4$ Å) for almost all targets. Hence in this case the probability for K -shells of two atoms located at neighboring crystal sites to be ionized sequentially is also negligible.

2. If the ion velocity is high ($v_i \gg Z_2 e^2/h$, which is the case in high-energy ion implantation), the cross section of host atom K -shell ionization can be roughly estimated in the sudden approximation as

$$\sigma_k = \frac{M_i \pi e^4}{m_e E_i} = Z_{\text{eff}}^2 \left(\frac{1}{E_k} - \frac{M_i}{4m_e E_i} \right), \quad (3)$$

where M_i and m_e are the ion and electron masses, respectively, E_i is the ion energy, and Z_{eff} is the effective ion charge determined by its velocity¹⁰ as

$$Z_{\text{eff}} = Z_i [1 + (v_i/vZ_i^\alpha)]^{-1/k} \quad (4)$$

(here $\alpha=0.45$, $v=3.6 \cdot 10^8$ cm/s, $k=0.6$, and Z_i is the ion atomic number). At typical velocities of the high-energy ions ($v_i \sim (1-5) \cdot 10^7$ m/s), we have $Z_{\text{eff}} \geq 10$. Assuming, as previously, $E_k=300$ eV, we obtain an estimate of the impact ionization cross section $\leq 10^{-16}$ cm² which corresponds to a mean free path $L \geq 1$ Å, i.e. the path length is around the interatomic distance. Hence in high-energy ion implantation, the probability of K -shell ionization of two atoms at neighboring lattice sites can be close to unity in some cases. (The high-energy ion slowing-down over a portion of the range $dx \sim 10^3$ Å may be ignored because $\Delta E/E \ll 1$.)

Thus the mode of defect formation discussed in this paper cannot be realized using traditional sources of radiation. This mechanism is effective only when a target is exposed to a beam of high-energy ions. The aim of this study was to assess the effectiveness of this mechanism of defect formation to calculate the distribution of defects over the projected range in a target, and to compare the calculations with experimental data.

2. PARAMETERS WHICH DETERMINE THE EFFECTIVENESS OF THE MECHANISM OF DEFECT FORMATION

The effectiveness of the mechanism of defect formation is largely controlled by two parameters:

1. The probability of generating a pair of close atoms with ionized K -shells.

2. The probability that one ionized (multiply ionized) atom of the pair leaves its site.

Let us consider these two factors in detail.

1. The probability for a K -shell to be ionized by a high-energy ion as a function of the depth inside the target may be considered in terms of a sequence of identical independent

events. Hence the probability that the K -shells of n atoms on a section Δx of the projected range are ionized is derived from the Poisson distribution:

$$\Psi(n, \Delta x) = \exp(-\Delta x/L) (\Delta x/L)^n / n!, \quad (5)$$

where $L = (\sigma_k N_e)^{-1}$. Then the probability that K -shells of two atoms at neighboring lattice sites are ionized is defined by the formula

$$\psi = 1 - \Psi(0, a) - \Psi(1, a), \quad (6)$$

where a is the average interatomic distance, $\Psi(0, a)$ is the probability that none of the two atoms at neighboring lattice sites is ionized, and $\Psi(1, a)$ is the probability for one of the two atoms located at neighboring lattice sites to be ionized. Taking into account Eq. (5), we obtain

$$\psi = 1 - \exp(-a/L) - (a/L) \exp(-a/L). \quad (7)$$

In the limit $L \gg a$ we have

$$\psi = a^2 N_e^2 \sigma_k^2(x). \quad (8)$$

The number of pairs of atoms with ionized K -shells located at neighboring lattice sites on a section Δx of the projected path length is given by

$$dN_{2i} = \psi dx / a. \quad (9)$$

Hence the distribution of pairs of atoms with ionized K -shells at neighboring lattice sites is

$$\frac{dN_{2i}}{dx} = a^{-1} \psi. \quad (10)$$

A vacancy in the K -shell is filled as a result of either radiative or nonradiative transition of an electron from one of the outer shells of the host atom. If the transition is nonradiative, the probability of multiple ionization is controlled by the atomic number Z of the host atom. According to a semi-empirical formula,³ the probability that the K -shell ionization is followed by an Auger cascade is

$$w(Z) = \frac{1}{1 + (-k + bz - cz^3)^4}, \quad (11)$$

where the coefficients k , b , and c are equal to 0.064, 0.034, and $1.03 \cdot 10^{-6}$, respectively. According to Eq. (11), we have $w=1$ for atoms with $Z \leq 20$. [The probability of a radiative transition is, accordingly, $1-w(Z)$.] The charge acquired by the atom after the Auger cascade increases as a function of atomic number Z . For example, when the K -shell of an argon atom ($Z=18$) is ionized, the average charge after the Auger cascade is ~ 4 , and whereas it is 2–3 when the L -shell is ionized; for K - and L -shells of krypton ($Z=36$), the ionization multiplicity is 5–6.⁸

2. For a defect to be generated from a pair of close ionized (multiply ionized) atoms, two conditions—one involving time and one involving energy—have to be met. The energy condition is that the Coulomb repulsion energy should be higher than the energy needed to displace one atom of the pair from its site:

$$\frac{q_1 q_2}{a} > E_{\text{dis}}, \quad (12)$$

where a is the average interatomic distance ($a \approx C^{-1/3}$), and C is the concentration of atoms in the target. According to the generally accepted concept, the displacement energy E_{dis} in this case is considerably lower than that of the impact displacement because an atom is removed from its site by the Coulomb force adiabatically, i.e. surrounding atoms have enough time to relax when the atom is being displaced. The displacement energy E_{dis} is of the same order as the diffusion activation energy,^{1,8} and equals several electron volts in most materials. Hence the energy condition can be satisfied in a wide range of materials (at $q_1 = q_2 = 1e$ and $a \sim 1 \text{ \AA}$, the left-hand side of (12) is $\sim 15 \text{ eV}$ and at $q_1 = q_2 \geq 2e$ it is over 60 eV ; at a distance $a \sim 1 \text{ \AA}$ the dielectric constant of the material can be taken equal to unity).

The time condition is that the lifetime τ_d of a charge in an ionized atom should be longer than the time τ_{dis} needed to impart to the atom an energy E_{dis} required to shift it from its site. The defect formation probability W_d and τ_{dis} are determined by^{4,8}

$$W_d = \exp(-\tau_{\text{dis}}/\tau_d), \quad (13)$$

$$\tau_{\text{dis}} = \frac{(2ME_{\text{dis}})^{1/2}}{F}, \quad (14)$$

where M is the atomic mass and F is the Coulomb repulsion force between ionized atoms. For interatomic distance $\sim 1 \text{ \AA}$, displacement energy $\sim 10 \text{ eV}$, and $M \sim 10 \text{ a.m.u.}$, the time needed to generate a defect through repulsion of atoms with a unit charge is $\geq (1-3) \cdot 10^{-14} \text{ s}$, and for atomic charge $q \geq +2e$ the time is $\tau_{\text{dis}} \leq 2 \cdot 10^{-15} \text{ s}$. The neutralization time for an atom with ionized K -shell is determined by the lifetime of a vacancy in the K -shell, i.e. by the rate of nonradiative (radiative) transitions³ and the time in which the charge ($q \geq 2e$) is localized in this atom after the transition is completed:

$$\tau_d = \tau_a + \tau_l. \quad (15)$$

The rate of the nonradiative (radiative) transition ν_a in an isolated atom ($\nu_a = 1/\tau_a$) increases with the atomic number; τ_a is fairly long only in light atoms ($Z \leq 10$). For example, in a carbon atom we have $\tau_a \sim 2 \cdot 10^{-14} \text{ s}$ (nonradiative transition). But in crystals with a wide valence band ($D_v \geq 5 \text{ eV}$), the probability of electron transition from the valence band to an atomic shell with a vacancy may be smaller due to the delocalization of valence electrons,⁴ and this time may be considerably longer (up to $\sim 10^{-13} \text{ s}$). The lifetime τ_l of a charge in an atom after the end of the Auger cascade is generally controlled by the chemical bond type.^{1,4,8} In most dielectrics with strong electron-phonon coupling, τ_l can be an order of magnitude longer than the period of lattice oscillations.³ In covalent crystals, additional factors have to be taken into account in order to correctly estimate the neutralization time of a multiple charge, e.g. the presence of two multiply ionized atoms at two neighboring lattice sites (i.e., a fairly deep potential well for valence electrons) or a high level of ionization on a high-energy ion track. Without discussing this topic in detail, we may assume that this time is at any rate longer than the hole delocalization time ($\tau_l \sim h/D_v$), which is $\leq 10^{-15} \text{ s}$ for most materials. Thus our approximate estimates

indicate that in some cases the time needed to produce a defect is longer or at least of the same order as the charge neutralization time. Hence the defect generation probability W_d can be close to unity in some cases.

Thus we obtain the following equation for the distribution of primary defects over the projected ion bath generated by the mechanism considered in this paper:

$$\frac{dN_d(x)}{dx} = W_d \frac{dN_{2i}(x)}{dx} = \sigma_d(x)C, \quad (16)$$

where σ_d is the cross section of inelastic generation of primary defect and C is the atomic density in the target. At $W_d \sim 1$, $a \sim 1 \text{ \AA}$ and $\sigma_k(E_k) \sim 10^{-16} \text{ cm}^{-2}$ we have $\sigma_d \sim 10^{-16} \text{ cm}^{-2}$, which is more than one order of magnitude larger than the cross section for primary defect generation caused by elastic scattering of high-energy ions at a depth much smaller than their projected range in a target.¹²

3. CONCENTRATION OF DEFECTS VERSUS DEPTH: CALCULATIONS AND COMPARISON TO EXPERIMENTAL DATA

Thus, the effectiveness of the damage mechanism depends on the properties of the material, as well as on the parameters of the ion beam. (In metals this mechanism is hardly feasible because the screening radius is $\sim 1 \text{ \AA}$.) Our estimates indicate that under realistic experimental conditions the effectiveness may be fairly high. A clear indicator of this damage mechanism being involved might be a change in the depth distribution of defects on the projected path of a high-energy particle. In fact, the distribution of defects generated by the elastic slowing down of an ion follows that of elastic energy losses, i.e. the concentration of defects increases monotonically with the distance from the target surface and is largest at the end of the projected ion path in the material.

The defect formation mechanism discussed in this paper should, on the contrary, generate more defects on the initial portion of the ion path, on which the ion charge and energy are still high and, hence, the cross section of K -shell ionization is also high. At some critical depth, the ion energy drops to a value at which the cross section for ionization of the inner shell sharply drops, and defects are not generated by this mechanism. On the remaining portion of the ion path defects are produced only through the elastic slowing down of ions. Hence, if the cross section for inelastic defect generation is higher near the target surface than that of elastic defect generation, the defect distribution curve should have a clearly expressed minimum between the surface and the end of the ion path.

The defect distribution curves which can be interpreted in terms of the ionization damage mechanism were recorded in experiments on implantation of high-energy ions ($E \geq 1 \text{ MeV/a.m.u.}$) in diamond and cubic boron nitride^{13,14} (Figs. 1-4). Note that the authors of Refs. 13,15 indicated that such distribution curves can be accounted for only in terms of an ionization mechanism, but they did not propose any specific defect-generation model due to high-energy ions. These curves cannot be interpreted in terms of nuclear ionization losses of ions, because their shapes cannot be described even

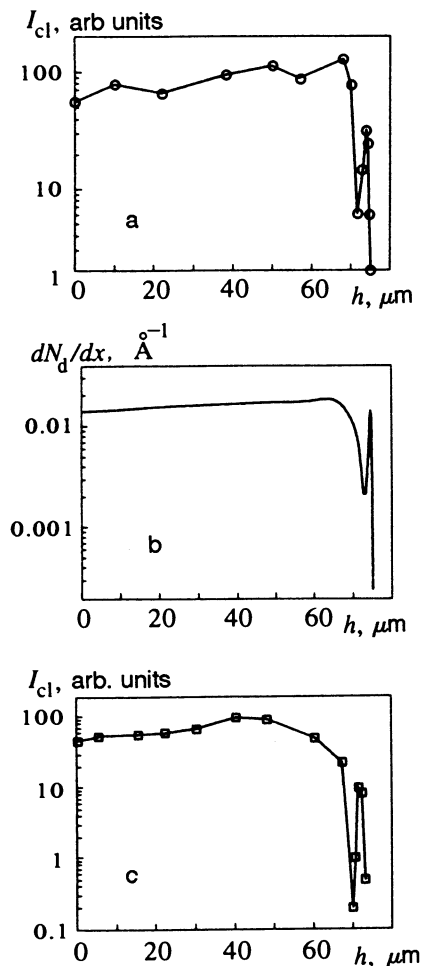


FIG. 1. a) Concentration of isolated vacancies (GR1-centers) in 2a diamond (nitrogen concentration $\leq 10^{18} \text{ cm}^{-3}$) irradiated with carbon ions ($E=81 \text{ MeV}$, $D=1 \cdot 10^{13} \text{ cm}^{-2}$, $T_{\text{impl}} \leq 400 \text{ K}$) versus depth¹³. b) Calculation of primary defect concentration produced by elastic and inelastic (the mechanism discussed in the paper) slowing of carbon ions as a function of depth. c) Concentration of TR12 nitrogen-centered defects versus depth in 1a diamond (a nitrogen concentration of $\sim 10^{19} - 10^{20} \text{ cm}^{-3}$), bombarded with carbon ions ($E=82 \text{ MeV}$, $D=3.3 \cdot 10^{15} \text{ cm}^{-2}$, $T_{\text{impl}} \leq 400 \text{ K}$).¹⁴

qualitatively by these models.¹² In our opinion, these curves also cannot be interpreted in terms of diffusion of primary defects because the diffusion activation energies of interstitial defects in these materials are about 3 eV, and the activation energy of vacancy migration in diamond is 2.3 eV,^{16,17} whereas primary defects (such as an isolated vacancy of an interacting Frenkel pair) in these crystals are annealed only at a temperature of about 1000 K^{18,19} (the temperature of the exposed targets was less than 400 K). On the other hand, diamond and boron nitride are materials in which their damage mechanism should be efficient. This statement is derived from the following properties of these materials.

1. Small binding energy of *K*-electrons (284 eV in carbon atoms, and 188 and 399 eV in nitrogen and boron, respectively).

2. Small atomic masses resulting, on the one hand, in short displacement times τ_{dis} needed to shift an atom from its site ($\tau_{\text{dis}} \sim M^{1/2}$), and, on the other hand, in a low rate of the

Auger cascade, i.e. a long lifetime of a vacancy in the *K*-shell.^{3,9}

3. Short interatomic distance ($a \sim 1 \text{ \AA}$) also leads to small displacement time τ_{dis} ($\tau_{\text{dis}} \sim a^2$).

4. The small annihilation probability of Frenkel pair components, which can be deduced, in particular, from the existence and high annealing resistance of interacting Frenkel pairs and isolated vacancies.^{13,19}

5. The small distance an atom should pass to form a defect (the distance between Frenkel pair components in diamond and boron nitride is $\sim 0.1 \text{ \AA}$).^{14,19}

In order to prove the feasibility of the proposed mechanism and to interpret the distribution of defects in diamond and boron nitride irradiated with high-energy atoms, we have calculated the distributions of primary defects generated by both elastic and inelastic ion braking. The distribution of primary defects due to the elastic slowing and ion energy and charge versus the ion penetration depth were calculated using the TRIM software package.¹² The energy and ion-charge dependence of the *K*-shell ionization cross section were taken from the data quoted by Garcia *et al.*⁹ The distribution of primary defects generated by inelastic ion slowing was calculated using Eq. (16). The probability of generating a primary defect through Coulomb repulsion of ions with like charges was assumed to be unity. The effect of recoil atoms on the generation of defects in this treatment was neglected because an ion is unlikely to impart much energy to a host atom in the initial portion of its range.¹² The calculated concentrations of defects due to elastic and inelastic braking have been summarized.

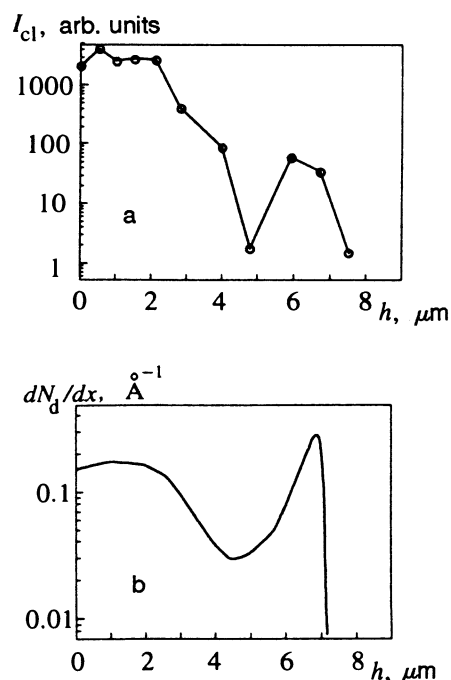


FIG. 2. a) Distribution of unidentified centers (tentatively Frenkel pairs¹⁴) in 2a diamond (nitrogen density of $\leq 10^{18} \text{ cm}^{-3}$), exposed to cobalt ions ($E=64 \text{ MeV}$, $D=6 \cdot 10^{14} \text{ cm}^{-2}$, $T_{\text{impl}} \leq 400 \text{ K}$).¹⁴ b) Calculated concentration of primary defects produced by both elastic and inelastic (mechanism discussed in the paper) slowing of cobalt ions.

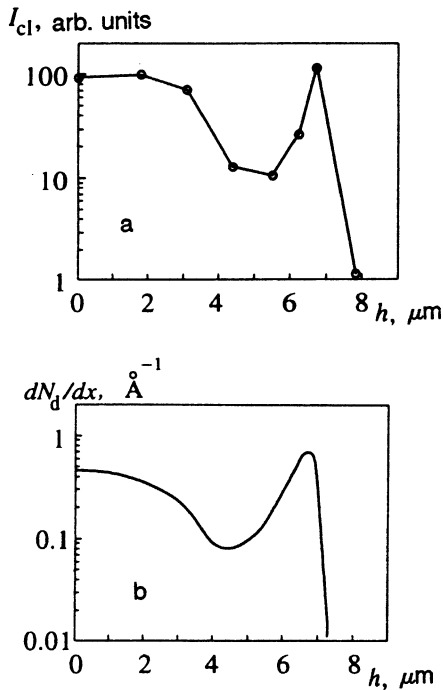


FIG. 3. a) Distribution of Frenkel pairs (RC2-centers) in boron nitride exposed to copper ions ($E=64$ MeV, $D=5 \cdot 10^{14}$ cm $^{-2}$, $T_{\text{impl}} \leq 400$ K).¹³ b) Calculated distribution of primary defects produced by both elastic and inelastic (the present mechanism) slowing of copper ions.

The calculations for various ion-implantation parameters in diamond and boron nitride are compared to experimental data^{13,14} in Figs. 1–4. It is clearly seen in the graphs that the calculated curves are in good agreement with the experimental data on the distribution of isolated vacancies in diamond (GR1-centers^{13,18}), vacancy centers around a nitrogen atom (TR12-center^{14,18}) (Fig. 1), the distribution of centers detected at a wavelength $\lambda=511$ nm¹⁴ (tentatively Frenkel pairs¹⁴) (Fig. 2), and the distribution of Frenkel pairs in boron nitride, which are detected through cathode-ray-induced luminescence (CR2-centers,^{13,19} Figs. 3, 4). The intensity of luminescence lines is proportional to the defect concentration.¹⁵ In terms of R_{min} (the position corresponding to the minimum defect concentration for a penetration depth in the range of $0 < x < R_p$), the calculations agree with the experimental data to within 10–15% (Figs. 1–4).

In our opinion, this fairly good agreement between calculations and experimental data over wide ranges of both energy and the mass of the ions implanted in two different materials is an important indication that defects are generated by the proposed mechanism near surfaces of diamond and boron nitride bombarded with high-energy atoms. Note that the calculated ratio of concentrations of primary defects generated by inelastic (near the surface) and elastic slowing of ions deviates from experimental data. It is evidently caused by the difference in the effectiveness of secondary defect formation, experiment deduced, from primary atomic displacements due to elastic and inelastic ion slowing.

It is also noteworthy that the lattice structure was not taken into account in the calculations, i.e., the target was treated as amorphous. Generally speaking, the probability of

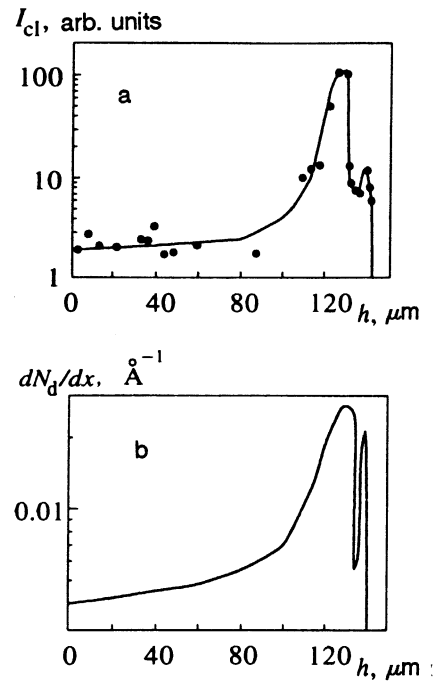


FIG. 4. a) Distribution of Frenkel pairs (RC2-centers) in boron nitride irradiated with boron ions ($E=92$ MeV, $D=3 \cdot 10^{13}$ cm $^{-2}$, $T_{\text{impl}} \leq 400$ K).¹³ b) Calculated distribution of primary defects generated by both elastic and inelastic braking of boron ions.

simultaneous ionization of the inner shells of two atoms located at neighboring lattice sites may depend on the beam alignment with respect to crystal axes. The probability of generating an ion pair should be maximum when the beam is aligned with the [111] axis (if the crystal structure is diamond or zinc-blend). In our case this simplification should not result in major errors because, first, the maximum impact parameter in most cases ranged from 0.1–1 Å, which should result in a very weak anisotropy, and, second, in all the experiments cited the beams of implanted ions were directed along the [111] axis.

4. CONCLUSION

In this paper, we have discussed a defect generation mechanism in nonmetallic crystals. According to this mechanism, pairs of atoms with ionized inner shells are produced at neighboring lattice sites by high-energy ions and one component of the pair is then displaced from its site. We have demonstrated that this damage mechanism is effective for high-energy ions and can produce more primary defects than elastic slowing of ions. Primary defect distributions as functions of depth path calculated with allowance for defects generated due to the elastic slowing of ions are in good agreement with experimental distribution curves of point defects detected through electron-induced luminescence in diamond and boron nitride exposed to beams of high-energy ions, which confirms the proposed model of damage.

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