# Radiation-stimulated aggregation of Frenkel defects in solids

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Usp. Fiz. Nauk **160**, 1–33 (October 1990)

Theoretical studies on the effect of generation-recombination aggregation of Frenkel defects created by irradiating solids with radiation are reviewed. We discuss the way in which a substantially non-Poisson spectrum of density fluctuations of the number of defects is formed by the stochastic creation and recombination of immobile and mobile defects. The results obtained by using different approaches are compared—multiparticle densities, diagram technique, scaling, computer simulation. We show that the aggregates created in the course of accumulation of same-type defects (atoms or vacancies) substantially affect the kinetics of defect accumulation and increase severalfold their concentration at saturation at large radiation doses. We examine in detail how the spatial distribution of defects depends on their mobility, on the dimensionality of the space, on correlation in genetic pairs being created, on the mechanism of recombination (annihilation or tunneling charge-transfer), and on the number of crystal-lattice nodes in the sphere of recombination. Experimental data confirming the calculations are presented.

## 1. INTRODUCTION

Irradiation of solids of any type (metals, semiconductors, and insulators) leads to creation of pairs of Frenkel defects–vacancies and interstitial atoms [v, i, which are usually spatially well correlated.[1,2] In most ionic crystals the fundamental Frenkel defects are F- and H-*centers* (an anion vacancy that has captured an electron and an interstitial halogen atom that forms the quasimolecule $X_2^-$ with one of the nearest anions, $X^-$].

At moderate energies the primary event of interaction of particles with a crystal is simple–an elastic pair collision that displaces atoms (or ions) from the nodes into interstices.[3,4] According to the laws of conservation of energy and momentum, the condition for formation of a defect is the transfer to a regular atom (or ion) of the crystal lattice an energy exceeding a certain threshold value[5] (of the order of 10 eV). However, the main fraction of the energy of particles and photons in interaction with a solid is not spent on elastic displacement of atoms, but on exciting the electronic subsystem of the solids. It has been established in recent decades[1–5] that in many broad-gap dielectrics the main mechanism of radiation defect formation is not a *superthreshold mechanism*, as in metals, but a *subthreshold* one arising from the decay of electronic excitations into defects. (The efficiency of this mechanism has been less studied for semiconductors[5]).

The relative spatial distribution function of Frenkel defects of different types depends substantially, not only on which of the two mechanisms takes place, but also on which type of irradiation creates the electronic excitations, e.g., X-rays or photons.[8]

In the creation of a v, i pair, the interstitial atom has an initial kinetic energy, whereby it is displaced from its vacancy, e.g., by a chain of focused collisions. For example, in $\alpha$-Fe the mean free path of a crowdion of energy 50 eV amounts to more than $20d$ ($d$ is the interatomic distance).[6] The stability of the pairs of defects formed by this *primary process* of thermalization depends on their initial distribution. In the simple and pictorial black-sphere model the annihilation of pairs (restoring the perfect lattice) occurs practically instantaneously if the distance in the pair is smaller than a certain critical value $R_0$, whose magnitude is determined by the type of chemical bonding of the given solid, and also by whether the components of the pairs are charged as compared with the nodes of the regular lattice.[4,6–8] Several mechanisms exist of stabilization of unstable pairs of Frenkel defects.[8] In metals the radius $R_0$ of the instability zone usually amounts to several lattice constants, while in ionic crystals it is smaller.[1)] Semiconductors exist with a loose structure and a very large instability region[7] (e.g., $In_2Te_3$).

At temperatures at which the interstitial atoms become mobile (typically $\gtrsim$ 20–30 K in metals and dielectrics), they undergo thermoactivated jumps. In the course of this diffusional wandering they can approach their vacancies within $r < R_0$ and be annihilated with them, or they can move away (with subsequent capture by an impurity or aggregate), which leads to *accumulation* of defects.

In semiconductors and dielectric crystals, in particular alkali halides, an additional recombination mechanism occurs–*tunneling charge transfer*, which depends exponentially on the relative distance $r$ in the pair.[9–12] In the tunneling charge transfer of primary pairs of F- and H-centers, $i^-$ are formed in a crowdion configuration capable of displacement, even at 4.2 K, by several lattice constants, which hinders their recombination with $v^+$.[4,12] Tunneling charge transfer of a nonthermalized pair in the primary stage of their creation assists rapid annihilation owing to Coulomb attraction.[12]

Both the experimental data and the theoretical studies of recent years indicate a *statistical effect of radiation-stimulated aggregation* of same-type defects (vacancies or interstitial atoms) even at low temperatures ($\lesssim$ 30 K) at which the created defects are immobile and the kinetics of their accumulation is restricted only by annihilation of close-lying un-

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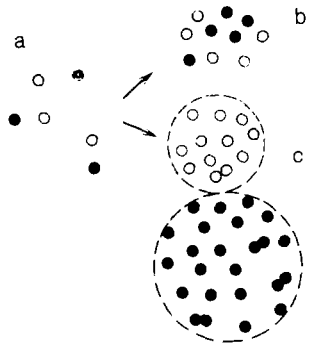


FIG. 1. Spatial distribution of Frenkel defects at low radiation doses (a) and at large doses[9]: with a chaotic distribution of defects (b) and with dynamic aggregation of same-type defects (c) (schematic). In the latter case several dimeric defects can be seen. ○ and ● respectively denote vacancies and interstitial atoms.

stable defects or by tunneling charge-transfer (see Sec. 3.3).

In the course of irradiation the chaotic distribution of defect pairs throughout the volume is replaced by a more ordered one, which amounts to *loose clusters* (aggregates) of noninteracting defects of the same type (Fig. 1). This is manifested experimentally in a substantial deviation in the concentration of pair (dimeric) centers from the value corresponding to a Poisson (chaotic) distribution[13] while at large irradiation doses at low temperatures the clusters of defects are visible in the electron microscope.[14] Owing to aggregate formation, the concentration of defects can exceed severalfold that for a chaotic spatial distribution. Yet the concentration of accumulated defects in turn governs the radiation stability of the material, which is of evident interest for radiation materials science.

Qualitatively the process of radiation-stimulated (or generation-recombination) aggregation is illustrated by Fig. 2. The random creation by radiation of two (or more) close-lying defects of the same type yields a distinctive "nucleus" of accumulations that is *more stable* than isolated defects with regard to the appearance at this same site of defects of the other type. To annihilate a pair of defects of a single type, one must await a corresponding fluctuation–appearance in the given region of two defects of the other type. In other words, in the course of irradiation it is "more favorable" for defects of a single type to form aggregates, since in them the probability of survival is greater than for a chaotic distribution of them, while the effective recombination volume (per defect) in an aggregate of many defects with overlapping recombination spheres is much smaller than for an isolated

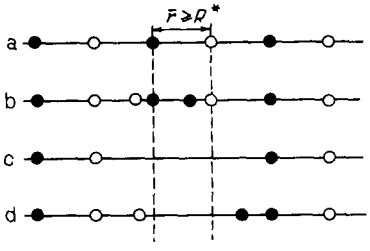


FIG. 2. Qualitative model of the aggregation process.[11] a—Chaotic distribution of defects ($\bar{r}$ is the mean recombination radius). b—Creation of a new pair of defects. c—After recombination. d—Generation of accumulation.

defect[15-17] (the effect of *statistical screening*[18-20]).

Recently reviews have been published[21-23] devoted to a general analysis of the phenomenon of *self-organization* in the most varied physical, chemical, and biological problems. The possibility of effective spatial separation (*segregation*) of i and v defects of different types (up to separate formation of two regions consisting of defects of different types[17]) in this context is important in principle, since it is one of the examples of self-organization of a defect structure, even in the absence of dynamic interaction between localized (immobile) defects of opposite types. This does not contradict the principles of thermodynamics, since the system is far from equilibrium owing to the continual flux of energy caused by the irradiation source that creates the defects.

We recall that in this review, when speaking of diffusion of defects, we are considering only ordinary thermoactivated diffusion, without discussing the problem of radiation-stimulated diffusion at low temperatures as observed in semiconductors under irradiation.[24,25]

While bearing in mind the vast wealth of material on the radiation physics of solids of the most varied nature, we adopt the aim of discussing the general *phenomenological* theory not restricted by assumptions of the concrete chemical nature of the material. We stress that the radiation-stimulated and preferentially *low-temperature* aggregation that we have studied of neutral defects of the same type differs in principle from the thermodynamically equilibrium-type, irreversible coagulation of vacancies (pores) at high temperatures,[26] which sometimes leads to creation of fractal structures.[27] Within the framework of our model the magnitude of the static zone of instability $R_0$ around each vacancy does not depend on the presence of other vacancies nearby, as is correct generally only for neutral defects, and may not be fulfilled for charged defects. Yet the aggregation of the latter is improbable owing to Coulomb repulsion. The model being discussed is valid up to a concentration of defects amounting to $\lesssim 10\%$ of the regular lattice nodes, as is known to be fulfilled, e.g., in dielectric crystals.[4,5] (At larger concentrations we arrive at a problem that has been discussed in disordered semiconductors[28]).

Thus the aim of this study is the detailed analysis of the results of the numerous studies of one of the special cases of self-organization–radiation-stimulated aggregation of defects in solids obtained by the most varied methods–both by analytical (Sec. 2) calculations and by computer simulation (Sec. 3). We study how aggregation depends on the dimensionality of the space, the mobility of defects, the degree of correlation in genetic pairs, the mechanism of recombination (annihilation or tunneling charge transfer), etc. In closing we discuss the description of the experimental kinetics of accumulation of defects (Sec. 4).

## 2. ANALYTICAL THEORIES

In studying the processes of accumulation of point defects one must distinguish the continuum and the discrete models. In the former model the intrinsic volume of the defect is not taken into account and the number of defects of a single type at a given point of space is not bounded. In the discrete model one unit cell can contain no more than one defect (vacancy or interstitial). The results of simulation using the one or the other model lead to substantially different results, as will be seen below. The theoretical results for

TABLE I. Various estimates of the dimensionless concentration of defects $u_0 = c_0 v_0$ at saturation (without taking account of correlation in genetic pairs) in the case of immobile defects in the one-, two- and three-dimensional cases.

| $u_0$ | Notes | References |
|---|---|---|
| 0,5 | Superposition approximation of Kirkwood, 1-D simulation | [30, 31, 32, 33] |
| 0,59 | Neglect of correlation of same-type defects | [30] |
| $\ln 2 \approx 0{,}69$ | Simulation taking account of tunneling recombination (charge transfer) | [11, 12, 29] |
| $\gtrsim 1$ | 1-D simulation with number of nodes from 2 to 700 | [17, 34, 35] |
| 0,46—3,6 | Analytic theory (in the continuum approximation) for the 1-D and 3-D cases without using the Kirk wood approximation | [36, 37, 38] |
| 1,36, 1,08 | Probability estimates for 2–100 nodes in the recombination sphere | [18, 19, 20] |
| 0,33—2,77 | Analogous, for the 1-D and 3-D cases | [39, 40] |
| 3,4, 3,2 | Simulation in the continuum approximation for 1-D, 2-D, and 3-D | [41] |
| 4,2, 2,07, 1,04 | 2-D simulation for 400 nodes in the recombination sphere | [42] |
| 1,36 | Model allowing an infinite local density of defects | [43] |
| $\infty$ | | |

the two models differ to the same extent.

To evaluate the results of analytical calculations using highly different (often implicit) assumptions and to compare them with the results of computer simulation, one must introduce a certain characteristic parameter of the kinetics of accumulation of defects. It is the dimensionless quantity $u_0 = c_0 v_0$, where $c_0 = c(t \to \infty)$ is the steady-state concentration of accumulated defects of the same type (at saturation); $v_0$ is the volume of the sphere of spontaneous recombination [$v_0 = (4/3)\pi R_0^3$, where $R_0$ is the radius of annihilation of defects of opposite types). The probability of recombination of an interstitial atom i in the sphere of spontaneous recombination around a vacancy will be henceforth considered to be uniform within the limits of the sphere and zero outside it]. For the continuum model it is clear from dimensional considerations that its magnitude $u_0$, if it exists, is a *universal* constant of the problem that does not depend on the magnitude of $v_0$. Most of the theoretical constructions have reduced to finding $u_0$. Table I collects the various estimates of this quantity, which will be discussed in greater detail below in Secs. 2.2 and 2.3.

The problem of the kinetics of accumulation is very complex owing to its multiparticle character; clusters of defects of the same type that arise initially owing to statistical fluctuations behave differently with regard to recombination from the way individual defects do. Thus, for example, while the recombination of an isolated vacancy with an individual interstitial diminishes the effective recombination volume by the amount of the volume of spontaneous recombination of the vacancy, the recombination of an interstitial with a cluster consisting of vacancies with overlapping recombination volumes (below, in discussing accumulation, an important role is played precisely by these clusters, rather than simply by the accumulation of vacancies with nonoverlapping recombination spheres), can lead to a quite insignificant change in the effective recombination volume.

### 2.1. Probabilistic models

In studying processes of accumulation of Frenkel defects, one uses three types of simple models: the urn, continuum, and discrete models. In the simplest urn model, which was proposed in Ref. 118, one studies the accumulation of complementary particles in boxes (urns) having a certain capacity, with walls impenetrable for diffusion of particles among the boxes. The continuum model treats a continuous medium; the intrinsic volume of a defect is not taken into account here, and the number of defects of a single type at any point of space is not bounded. In the model of a discrete medium a single cell can contain no more than one defect (v or i).

*The urn model* amounts to a system of $N$ urns, into which one randomly and successively throws white (A) and black (B) balls, with the total numbers of thrown balls $n_A$ and $n_B$ equal to one another at any instant (apart from a single ball). In the absence of annihilation the color of a ball has no meaning and the distribution of balls between the urns is described by the binomial law usual for random events

$$W_k(\nu) = C_\nu^k P^k (1-P)^{\nu-k}. \tag{2.1}$$

Here $W_k(\nu)$ is the probability of filling of an arbitrarily chosen urn with $k$ spheres after $\nu$ throws, $\nu = n_A + n_B$ denotes also the total number of thrown balls, and the $C_\nu^k$ are the binomial coefficients, $P = 1/N$ is the probability that a ball is thrown into an arbitrary urn. As $P \to 0$ and for a finite $s \equiv \nu P$, the distribution of (2.1) goes over into the Poisson formula:

$$W_k(\nu) = s^k \exp(-s)\,(k!)^{-1}, \tag{2.2}$$

while when $k$ is sufficiently close to $s$, into the Gaussian distribution:

$$W_k(\nu) = (2\pi s)^{-1/2} \exp[-(k-s)^2 (2s)^{-1}]. \tag{2.3}$$

Now let us introduce the process of recombination: the entry of a white and a black ball into a single urn leads to their annihilation. When $N = 1$, the number of balls in the urn is determinate, and is equal for alternate throwing to 0 or 1. When $N > 1$ the situation is altered in principle owing to the random character of the entry of A and B into the urns. When recombination is included the distributions (2.1)–(2.3) no longer hold; the probability of finding $m$ balls of a chosen color in an arbitrary urn after $2\nu$ throws ($\nu$ A balls

and $\nu$ B balls) is

$$W_m(2\nu) = \sum_{k=0}^{\nu-m} C_\nu^k P^k (1-P)^{\nu-k} C_\nu^{k+m} P^{k+m} (1-P)^{\nu-k-m}. \quad (2.4)$$

We can also study the case in which the capacity of each urn is restricted to a certain number $M$. Here the pairs of throws in which even one of two balls enters an urn filled to the limit with balls of the same color are not realizable and are not taken into account in calculating the number $\nu$. For small values of $P$, large $M$, and $\nu \to \infty$, Eq. (2.4) is approximately reduced to

$$W_m = \exp(-2s) \lim_{\nu\to\infty} \sum_{k=0}^{\nu-m} s^{2k+m} [k!\,(k+m)!]^{-1}$$
$$= \exp(-2s)\, I_m(2s).$$

Here $I_m$ is the Bessel function of imaginary argument. The asymptotic representation of $I_m$ leads, when $\nu \gg N$, $M$, to a probability $W_m$ that does not depend on $m$: $W_m \sim 1/2(\pi S)^{1/2}$. Here the means are $\bar{m} \approx (M+1)/2$, $m^2 \approx M(M+1)/3$, while we have

$$\delta_m = \frac{(\overline{m^2})^{1/2}}{\bar{m}} \approx 1{,}16. \quad (2.5)$$

Thus the random distribution of the particles among the urns in the generation-recombination process leads to an *equiprobable* distribution, cardinally different from the Poisson distribution, of "boxons" (sets of a given number of particles of the same color) over the urns and to the macroscopic magnitude of the fluctuations of the number of particles in the urn of (2.5).

These characteristic features are manifested also in the generation-recombination processes of Frenkel defects in real crystals. However, in contrast to the urn model, in a crystal a statistical screening of the recombining particles occurs in coordinate space that leads to a complex spatial distribution of vacancies v and interstitial atoms i. This distribution depends on the law whereby the probability of recombination varies with the distance $r$ between complementary particles. Usually one approximates this law by the step distribution $W(r) = 1$ $(r \leqslant R_0)$, $W(r) = 0$ $(r > R_0)$. The quantity $R_0$ is called the recombination radius.

The first attempts to seek a closed expression for $c(t)$ did not include an analysis of pair correlations of defects, even at the level of pair densities, and seem at least to be ambiguous. Such an approach based on simple probabilistic considerations was first used in Ref. 43 (see also Ref. 33). Since here we are not treating explicitly the relationships for two-particle (and higher) particle densities, it is difficult to make a correct estimate of the effect of overlap of the forbidden volumes of several close-lying defects. This leads to the need to introduce some *a priori* assumptions. A characteristic example is Ref. 43, where the implicit assumption of a chaotic distribution of defects through the volume (along with partially taking it into account) led to a physically false result–the *absence* of an effect of saturation of concentration at high doses (see Table I).

In Ref. 39, which assumes the existence of a steady-state concentration of accumulated defects in a one-dimensional model, and which takes the distribution of A and B defects separately in the form of periodically distributed clusters (groups) of identical dimension, it was found from simple probabilistic considerations that the mean number of defects per cluster is

$$\bar{K} = 4e^{R_0/\bar{r}} - 1, \quad (2.6)$$

Here $\bar{r}$ is the mean distance between particles in the cluster (an A-B pair instantaneously recombines if the distance $r$ between A and B is $r \leqslant R_0$). The values were obtained in the model being discussed of $R_0/\bar{r} = 3.43$, $\bar{K} \approx 143$. We can record the results of this study by using the parameter $a_0$. Thus, we easily note that

$$u_0 = \frac{R_0 L/\bar{r}}{L + (ac + cb)}. \quad (2.7)$$

Here we have $L = \bar{K}\bar{r}$, $ac = [2\bar{r}(\bar{r} + R_0)]^{1/2}$, $cb = R_0 - (\bar{r}/2)$. From (2.7) we have ($\bar{K} \gg 1$)

$$u_0 = \frac{R_0}{\bar{r}} = 3{,}43. \quad (2.8)$$

Reference 40 studied a discrete case. Then, in contrast to (2.6), one has

$$\bar{K}_L = 4\left(1 - \frac{c_0}{\nu}\right)^{-\nu R_0} - 1. \quad (2.9)$$

Here $\nu$ is the concentration of traps (localization nodes). The calculation of the dependence of $c_0$ on $\log(2R_0\nu)$ that was performed agrees with the data of computer simulation, with deviation appreciable only in the region of small values of $\log(2R_0\nu)$, where only two lattice nodes exist in the recombination sphere. In the discrete model with the recombination radius $R_0$ any node of the crystal is surrounded by a sphere of radius $R_0/2$ within which only defects of the same type exist or there are none at all. One can call such a region of the crystal a homogeneity interval and characterize each node of the crystal with the number $k$ of same-type defects contained within its homogeneity interval; $n_k$ is the number of such nodes, $M$ is the number of nodes in the homogeneity interval. In a steady state the average over the volume of the crystal of the concentration of defects is

$$\frac{kn_k}{M} = \left(\frac{\partial P_k}{\partial t}\right)_{\text{д}} \tau_k. \quad (2.10)$$

Here $(\partial P_k/\partial t)_{\text{д}}$ is the rate of generation of defects, and $\tau_k$ is the lifetime of a defect with respect to recombination. It was assumed in Ref. 35 as an approximation that $\tau_k$ is determined by the concentration of defects solely within the homogeneity interval (the recombination zone). Here one can derive recursion relationships between the $n_k$, which lead to

$$n_k = n_0 \prod_{s=1}^{k} \left(1 + \frac{1-s}{M}\right), \quad (2.11)$$

and, together with the condition of constancy of the number of nodes

$$N = n_0 + 2\sum_{k=1}^{M} n_k$$

also to the steady-state concentration values and

$$c_0 = N_{\text{v}} = N_{\text{i}} = N(1 + 2q_M)^{-1},$$
$$q_M = \sum_{k=1}^{M} \sum_{s=1}^{k} [1 + (1-s)M^{-1}]. \quad (2.12)$$

At low enough values of $M$ the calculated values of the parameter $u_0$ agree well with the results of computer experiments. For large $M$ the relationships of (2.11) lead to a rapid growth in $u_0(M)$, which qualitatively agrees with experiment. However, the $c_0$ calculated by (2.12) increases with increasing $M$ *more rapidly* than in the computer calculations, as is explained by the mentioned approximation for determining $\tau_k$.

We stress an important feature of the generation-recombination process being discussed. The lifetime of a vacancy, owing to the competition for capture of an i that has fallen at some node $\alpha$ with other vacancies that also lie within the sphere of recombination of i, depends on the concentration of vacancies $N_v(\alpha)$ in the sphere with its center at the node $\alpha$. Yet the quantity $N_v(\alpha)$ depends analogously on $N_v(\alpha')$ for the nodes $\alpha'$ of this sphere; $N_v(\alpha')$ depends on the $N_v(\alpha'')$ for the nodes $\alpha''$ of the recombination sphere with its center at $\alpha'$, etc. Thus an effective interaction arises between immobile point defects caused by the generation-recombination process, which leads to "cleaning out" of the volume of the crystal of single defects and small clusters and their collection into large clusters.

If both components of a Frenkel defect are mobile and no other reactions but annihilation occur, the tendency to generation-recombination clustering is preserved, but the effect of clustering is less sharply marked. However, in real systems usually only capture reactions of mobile v and i by various sinks and by one another are effective; while the complexes that are formed here (e. g., divacancies, tetravacancies, vacancy + impurity atom) lose mobility. The possibility of effective accumulation of clusters of particles of the same type in such systems is determined by the relationships among the capture cross sections of particles of the same type and the recombination of particles of opposite type. There are a number of causes that lead to a systematic difference between the capture cross sections of v and i by various defects (*preference*) that gives rise to an effective separation of them with respect to weak recombination. A consequence of this is a low radiation stability of the materials being irradiated. The effect of statistical screening is manifested also in this case. However, even in the absence of preference a clustering of single-type defects occurs, completely due to statistical screening. The latter statement is well confirmed by the results of computer simulation (see Sec. 3).

### 2.2. Multipoint densities

K. Dettman[30] has created the basis of a rigorous theory of the kinetics of accumulation of Frenkel defects in crystals. He showed that in the absence of diffusion the curve of accumulation $c(t)$ is determined by the infinite sum of the correlation functions $\rho_{n,0}$, $n = 2, \ldots \infty$ that describe the spatial correlations of all orders of defects *of the same type*.

The accumulation equation can be written in the form

$$\frac{dc}{dt'} = p(1 - 2\beta'), \tag{2.13}$$

$$\beta' = \sum_{n=1}^{\infty} \frac{(-1)^{n-1}}{n!} \times \int \theta(\mathbf{r}_1) \ldots \theta(\mathbf{r}_n) \rho_{n,0}(\mathbf{r}_1, \mathbf{r}_2, \ldots, \mathbf{r}_n; t)\, d\mathbf{r}_1 \ldots d\mathbf{r}_n, \tag{2.14}$$

Here $p$ is the rate of creation of stable pairs of defects, while the quantity $\beta'$ is the fraction of the volume overlapped by recombination spheres, i.e., the ratio of the effective recombination volume to the entire volume of the crystal. The problem of constructing the kinetic equation of accumulation for $c(t)$ could be solved if one could obtain $\beta'$ in explicit form, including actually all the information on aggregation. This would mean the solution of the multiparticle problem in accumulation. But in practice one must restrict the treatment to certain approximations. Thus, in the simplest case, upon neglecting the correlation between single-type defects (i. e., assuming that $\rho_{n,0} \approx \rho^n_{1,0}$), the following was obtained:[30]

$$\beta' = 1 - e^{-u_0}. \tag{2.15}$$

The corresponding saturation concentration of defects is $u_0 = \ln 2 \approx 0.69$. When $u_0 \ll 1$, Eq. (2.15) implies that $\beta' = u_0 = 0.5$, i. e., the recombination spheres practically do not overlap, even at saturation.

Recently[32] a more consistent theory was proposed of the accumulation of Frenkel defects that took account of the spatial correlations of defects of the opposite or same types. The cutoff of the infinite chain of linked equations for *multipoint densities* of the number of particles in the superposition approximation (SA) of Kirkwood

$$\rho_{2,1}(\mathbf{r}_1, \mathbf{r}_2; \mathbf{r}_1') \approx \rho_{1,1}(\mathbf{r}_1; \mathbf{r}_1')\, \rho_{1,1}(\mathbf{r}_2; \mathbf{r}_1')\, \rho_{2,0}(\mathbf{r}_1, \mathbf{r}_2)\, c^{-3}, \tag{2.16}$$

which is widely used in studying the kinetics of reactions, leads to a simple equation for the macroscopic concentration:[31,32]

$$c = c_0[1 - \exp(-2pv_0 t)], \quad c_0 = \frac{1}{2}\left(\frac{1}{v_0} - \frac{1}{v_d}\right) \tag{2.17}$$

and to two linked equations for the *pair correlation functions*–of same-type ($X$) and opposite-type defects ($Y$):[32]

$$\frac{\partial X}{\partial t'} = -\frac{2X}{c}\frac{\partial c}{\partial t'} + \frac{1}{v_0 c} - \left(2c + \frac{1}{v_p}\right)\frac{X}{c}(g * Y), \tag{2.18a}$$

$$\frac{\partial Y}{\partial t'} = -\frac{2Y}{c}\frac{\partial c}{\partial t'} + \frac{1}{2v_0 c^2}(2c + f(r)) - \left(2c + \frac{1}{v_d}\right)\frac{Y}{c}(g * X). \tag{2.18b}$$

Here $t' = 2pv_0 t$ is the dimensionless time, and we use the spatial-convolution notation

$$(A * B)_r = \int A(r')\, B(r - r')\, dr' = \frac{2\pi}{r}\int_0^{\infty} dr' r' A(r') \int_{|r-r'|}^{r+r'} d\rho \rho B(\rho) \quad . \tag{2.19}$$

Also we assume that defects are created uniformly in the interval of distances $R_0 \leqslant r \leqslant R_d$, $v_d = (4.3)\pi R_d^3$ and recombine instantaneously when created with $r < R_0$. The pair functions in (2.18) are normalized according to the principle of attenuation of correlation with distance: $X(r \to \infty) = 1$, $Y(r \to \infty) = 1$.

Equation (2.17) implies that the dimensionless concentration at saturation (for uncorrelated creation of defects, $R_d \to \infty$) does not exceed $u_0 = 1/2$. This means that the SA yields a crude result, poorer than the simple neglect of the correlation of single-type defects ($u_0 = \ln 2$). Yet taking account of correlation in genetic pairs has the result that, the greater is the correlation ($R_d \to R_0$), the smaller is the con-

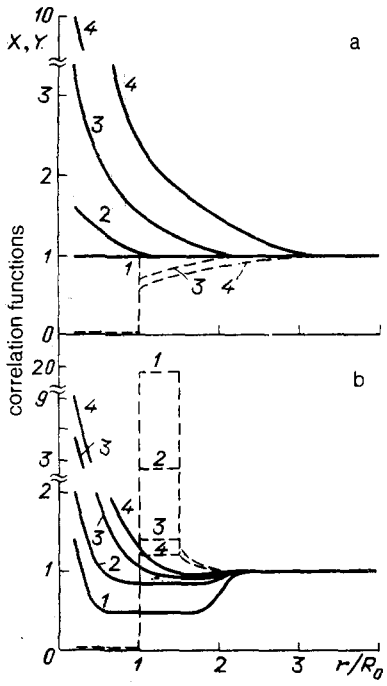


FIG. 3. Dose dependence of the pair correlation functions in the accumulation of immobile defects in three-dimensional space.[32] The solid and dotted lines denote the functions for same-type and opposite-type defects. a and b—Uncorrelated and strongly correlated ($R_d \times R_0^{-1} = 1.5$) generation of defects. $c/c_0$: *1*—0.05, *2*—0.5, *3*—0.9.

centration $c_0$ at saturation.[30,32] An essential point is that the kinetics of accumulation in (2.17) for instantaneous recombination depends on the irradiation *dose* $d = pt$ (the number of created defects) but not on its intensity (power of the dose).

Figure 3 shows the result of calculation for two limiting cases–absence of correlation (a) and strong correlation in genetic pairs (b). We see that, as saturation is approached, the fraction of close same-type defects (A–A, B–B) considerably exceeds the Poisson value ($X = 1$), which confirms their aggregation. The characteristic dimension of the aggregates ($X(\bar{R}) \gtrsim 1$) and the distance between them $\approx 2R_0$ agree with computer simulation in the *three-dimensional* case (Sec. 3). This means that we are dealing with microscopic segregation of defects, since the dimension of the accumulations is small in comparison with the dimensions of the system.

Yet in the case of strong initial correlation at low concentrations, the pair correlation function of opposite-type defects has a sharp peak at $R_0 \leqslant r \leqslant R_d$ that is "accompanied" by a trough at the same $r$ in the correlation function of same-type defects. Again this emphasizes the *interrelation* of the spatial distribution of same- and opposite-type reagents in bimolecular processes, as noted in Refs. 32 and 44. The formation of aggregates with a strong initial concentration is slow and occurs mainly near saturation of the concentration (see Fig. 3b).

Analysis of the SA (2.16) showed[36,45] that it is inapplicable to the region of small relative distances at which $|\mathbf{r}_1 - \mathbf{r}_1'|$ and $|\mathbf{r}_2 - \mathbf{r}_1'| \leqslant R_0$, This leads to a cutoff in the infinite series of correlation functions $\rho_{n,0}$, $n = 2, \ldots, \infty$, and to restriction to the first term, which does not depend on the spatial correlation of defects.

Upon neglecting the initial correlation in genetic pairs in the case of instantaneous annihilation of opposite-type defects created at a distance $r \leqslant R_0$ and in the continuum approximation, it has been possible to overcome the noted defects of the SA and to sum the infinite series of (2.13). The fundamental approximation used was the representation of the multipoint density in the form

$$\rho_{n,0} \approx c^n\Big(1 + \sum_{i<k} \nu(|\mathbf{r}_i - \mathbf{r}_k|)\Big), \tag{2.20}$$

Here $\nu(|\mathbf{r}_i - \mathbf{r}_k|)$ is associated with the pair density of single-type defects $\rho_{2,0}(r)$ and with the relationship $\rho_{2,0} = c^2(1 + \nu(|\mathbf{r}_1 - \mathbf{r}_2|))$, where $\nu(r \to \infty) = 0$. Application of the approximation (2.20) has the consequence that the obtained result is an estimate *from below*, but better than the SA, since here one takes explicit account of the spatial correlation of defects of the same type. The use of (2.18) leads to a system of three linked equations for the dimensionless macroscopic concentration $u(t)$ and the pair correlation functions of same-type $X$ and opposite-type $Y$ defects. We present only the former of them:

$$\frac{\partial u}{\partial t'} = e^{-u}\Big(1 + \frac{u^2}{2}\bar{\nu}\Big) - \frac{1}{2}, \quad u(0) = 0, \quad t' = 2pv_0 t. \tag{2.21}$$

Here the parameter

$$\bar{\nu} = \frac{1}{v_0}\int S(r)\,\nu(r)\,d\mathbf{r}$$

is the fraction of the total volume of two spheres of radius $R_0$, whose centers are separated by the distance $r$. In contrast to the previous approaches, here the equation for the concentration is not closed, and requires one to calculate the correlation function for same-type defects $\nu(r, t)$. It is determined by solving a rather unwieldy system of equations.[36,37]

The solution of these equations on a computer yields the saturation concentration $u_0 = 1.36$ and $1.08$ for $d = 1$ and 3, respectively (Fig. 4a). In the former case this exceeds by approximately twofold the cited value in the absence of aggregation ($u_0 \approx 0.69$). We also see the substantially greater effect in the *one-dimensional* case; however, perhaps also the analytical estimate is substantially low (see Table I and Sec. 3).

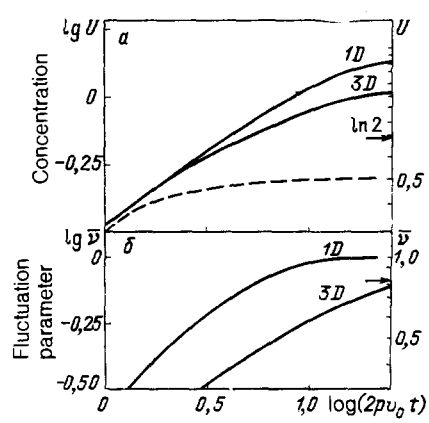


FIG. 4. a—Calculation of the kinetics of accumulation in the three-dimensional case (without using the superposition approximation);[36,37] dotted curve—result of the SA. b—Magnitude of the fluctuation parameter characterizing the deviation of the spectrum of the fluctuations of defect density from the Poisson distribution.

We see from Fig. 4b that a substantially *non-Poisson spectrum* of fluctuation of density of the reagents is formed in the course of accumulation, characterized by the parameter $\bar{\nu}$ and implying *aggregation*. It was shown[36] that

$$\overline{N^2} - \overline{N}^2 = u + u^2\bar{\nu}. \tag{2.22}$$

Here $N$ is the number of defects A (B) in the sphere $v_0$, and $\overline{N} = u$. (For a Poisson spectrum of fluctuation we have $\bar{\nu} \equiv 0$.)

The dynamics of aggregation is shown in Fig. 5. In the course of irradiation the initial chaotic distribution undergoes a substantial change: the fraction of close same-type defects upon prolonged irradiation exceeds approximately *threefold* the Poisson value. This agrees well with the experimental data[13] for dimeric $F_2$ centers in KCl created upon prolonged X-irradiation at 4 K.

An essential result is also the fact that the curve of accumulation $c(t)$ in the large-dose region is not described by the simple formula (2.17), which is often employed in interpreting experimental data (e. g., in Ref. 16), despite the fact that the accumulation process is imitated by a *single* recombination mechanism. Hence we must treat with caution the widespread view (e. g., Refs. 46 and 47) that the successful approximation of $c(t)$ by several exponentials (stages) is unambiguously correlated with several *different* mechanisms of accumulation.

Upon including diffusion of defects the pattern of accumulation becomes substantially complicated. Here it is clear that the effect of aggregation must be weakened. This process has been studied by several methods. Simple approximate answers were obtained by using the SA.[48-54] In this theory the rate coefficient of recombination and the steady-state concentration $c_0$ are determined in a self-consistent fashion. A test of the fundamental equations of accumulation by simulating a two-dimensional model on a computer has been performed in Refs. 49 and 54. The results showed that, for small values of the dimensionless parameter $2pv_0/Dc_0$ (i. e., large diffusion coefficients) ($v_0 = \pi R_0^2$), the following relationship determining $c_0$ is well satisfied:

$$p = \frac{2\pi x K_1(x)}{K_0(x)}, \tag{2.23}$$

Here we have $x = (2pv_0/\pi Dc_0)^{1/2}$, and $K_0$ and $K_1$ are the modified Bessel functions of zero and first order. For $d = 3$ we have

$$p = 4\pi DR_0[1 + (3s)^{1/2}]\, c_0^2, \tag{2.24}$$

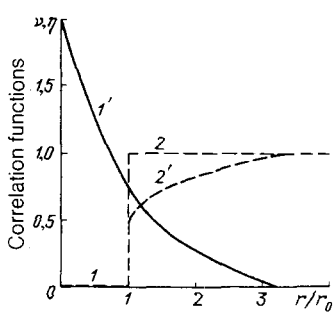


FIG. 5. Pair correlation functions of same-type ($\nu = X - 1$) and opposite-type ($Y$) defects.[36] Curves *1*, *2*—$t = 0$; *1'*, *2'*—$t \to \infty$ ($r/R_0$ is expressed in terms of $r/r_0$).

where we have

$$s = \frac{4}{3}\pi R_0^3 c_0.$$

The problem of taking simultaneous account of aggregation and diffusion (see Sec. 2.4) is most complicated. In this case one can write only an approximate equation:[45,48]

$$\frac{dc}{dt} = p\exp(-2cv_0) - K(\infty)c^2. \tag{2.25}$$

Here the diffusion-controlled recombination constant is

$$K(\infty) = 4\pi DR_0[1 + (3s)^{1/2}]. \tag{2.26}$$

### 2.3. Long-wavelength approximation

One can carry out a systematic treatment of the problem of aggregation of *diffusing* opposite-type defects (while taking account of their interaction) by the *methods of field theory*,[57-59] with account taken of the fact that the problem contains a small gas parameter, $cv_0 \ll 1$. We shall base the presentation of the results of this method on a treatment of the system in the *long-wavelength* approximation, which is most graphic amd which allows one to obtain graphically practically exact answers for the degree of aggregation of defects. Moreover, it has turned out that the field-theory methods and the long-wavelength approximation at low densities of defects yield results that agree. In the stated approximation the density of particles (of types A or B) can be described by the fluctuating function $c(\mathbf{r},t)$. This quantity is determined exactly if one divides the entire volume into microscopic subvolumes containing a large enough number of particles. At equilibrium in the absence of reactions, the probability density of finding the system in a given $c(r)$ (which differs little from its mean value $\bar{c}$) is given by the Gaussian functional

$$P[c(r)] \propto \exp\left[-\frac{\int (c(r) - \bar{c})^2\, dr}{2\bar{c}}\right]. \tag{2.27}$$

In the presence of diffusion $c(\mathbf{r},t)$ obeys the diffusion equation

$$\frac{\partial c(\mathbf{r}, t)}{\partial t} = D\Delta c(\mathbf{r}, t) + i_0(\mathbf{r}, t). \tag{2.28}$$

Here $i_0(\mathbf{r},t)$ is a random Gaussian function with the correlator

$$\langle i_0(\mathbf{r}, t)\, i_0(\mathbf{r}', t')\rangle = \bar{c}(t) D\delta(t - t')\nabla_{\mathbf{r}}\nabla_{\mathbf{r}'}\delta(\mathbf{r} - \mathbf{r}'). \tag{2.29}$$

Equation (2.28) for $t \to \infty$ implies (2.27).

After this remark, let us examine the system of equations controlling the creation and annihilation of A and B particles (to start with, neutral particles), which have equal diffusion coefficients $D_A = D_B = D$.[56] It has the form

$$\begin{aligned} \frac{dc_A(r, t)}{dt} &= D\Delta c_A(r, t) - Kc_A(r, t)c_B(r, t) + i_A(r, t), \\ \frac{dc_B(r, t)}{dt} &= D\Delta c_B(r, t) - Kc_A(r, t)c_B(r, t) + i_B(r, t). \end{aligned} \tag{2.30}$$

Here $K$ is the bimolecular recombination constant (in particular, it can be a diffusion constant, i.e., equal to $8\pi DR_0$ ($R_0$ is the recombination radius of the particles). Also, $i_A(r,t)$ and $i_B(r,t)$ are the densities of creation of A and B

particles by the external source. They have the Poisson statistical properties

$$\langle i_A(r,t)\rangle = \langle i_B(r,t)\rangle = i_0, \tag{2.31}$$

$$\langle (i_A(r,t)-i_0)(i_B(r',t')-i_0)\rangle = 0, \tag{2.32}$$

$$\langle (i_A(r,t)-i_0)(i_A(r',t')-i_0)\rangle = \langle (i_B(r,t)-i_0)(i_B(r',t')-i_0)\rangle = i_0\delta(\mathbf{r}-\mathbf{r}')\delta(t-t'). \tag{2.33}$$

Also $i_A(r,t)$ and $i_B(r,t)$ contain the term with "inner" stochasticity $i_0(r,t)$ of (2.28) and the correlator of (2.29). However, in the long-wavelength approximation and for a small gas parameter, it always proves to be inessential.

The difference $c_A(r,t) - c_B(r,t) \equiv z(r,t)$ obeys the equation

$$\frac{\partial z(r,t)}{\partial t} = D\Delta z(r,t) + \delta i(r,t), \quad \delta i = i_A - i_B. \tag{2.34}$$

The Fourier components $z(r,t) - z_k(r,t)$ are random functions having the correlator

$$g(k,k',t) = \langle z_k(t) z_{k'}(t')\rangle = \frac{2i_0\delta(k+k')}{Dk^2}\exp[-Dk^2(t-t')]. \tag{2.35}$$

For small $k$, $g$ becomes large, which implies a separation of the A and B particles over large scales. In the three-dimensional case ($d = 3$) this can be illustrated as follows.

For completely mixed particles in a large enough volume, the square of the fluctuation of the number of particles $\delta N = N - \bar{N}$ obeys the ordinary relationship

$$\frac{\overline{\delta N^2}}{\bar{N}^2} \propto \frac{1}{\bar{N}}.$$

Yet if the correlator of the number of particles looks like (2.35), we have the stronger level of fluctuations:

$$\frac{\overline{(\delta N)^2}}{\bar{N}^2} \propto \frac{1}{N^{1/3}}. \tag{2.36}$$

In the two-dimensional case the analogous relationship has the form

$$\frac{\overline{\delta N^2}}{\bar{N}^2} \propto \frac{1}{\ln \bar{N}}. \tag{2.37}$$

In the one-dimensional case we have

$$\frac{\overline{\delta N^2}}{\bar{N}^2} \propto 1. \tag{2.38}$$

Equations (2.37) and (2.38) actually imply that the one- and two-dimensional cases actually exhibit already a *macroscopic* separation of the system into regions consisting only of A particles and only of B particles. This is also confirmed by the fact that the integral over the spectrum of spatial fluctuations diverges in these cases at small $k$. To find the aggregation of particles in numerical experiments in the three-dimensional case we must treat the *deviations* from a Poisson distribution in large volumes.

It is interesting to study now in the same approximation the problem of the kinetics of recombination of A and B particles *after* the source has been turned off ($i_A = i_B = 0$). As is known the initial fluctuations in the distribution of the reacting particles in space, and in particular, the thermodynamic fluctuations lead to features in the long-period kinetics of the approach of their mean concentrations to the equilibrium values. Thus, in the case of the irreversible reaction $A + B \to 0$, we have $c_A(t) \propto t^{-3/4}$,[44,59] where the concentrations of A and B particles are equal on the average. This asymptotic behavior is determined by the initial fluctuations of the initial Poisson distribution of particles. If the reaction is reversible, then the approach to the equilibrium values follows the *power-function* asymptotic behavior $t^{-3/2}$ (Ref. 60), which differs sharply from the exponential-in-time law that formal kinetics predicts. A general result of Refs. 59 and 60 was the statement that the long-term dependences are governed by the process of *diffusional* equalization of the fluctuations of the initial distribution of the reacting particles.

When the source is turned off the anticorrelation of the A and B particles of (2.35) is very high, which leads to a *slower* recombination of them for large $t$.

Actually the concentration at a late stage equals the mean value of the modulus $z(r,t)$:

$$c_A = c_B = \frac{1}{2}\langle |z(r,t)|\rangle. \tag{2.39}$$

Calculation of $\langle |z(r,t)|\rangle$ with the spectrum of initial fluctuations of (2.23) (with $t = t' = 0$) is performed analogously[55] and yields the result:

$$c_A = \frac{1}{2}\langle |z(r,t)|\rangle = \frac{1}{2\pi}\left[\frac{i_0}{(2\pi)^3 D}\int \frac{d^3k}{k^2}\exp(-2Dk^2t)\right]^{1/2}. \tag{2.40}$$

For the three-dimensional case we have

$$c_A \propto i_0^{1/2} D^{-3/4} t^{-1/4}. \tag{2.41}$$

For particles that are destroyed according to (2.41), we have $\delta\bar{c}/\bar{c} \approx R_0 c^{1/3}$, where $R_0$ is the recombination radius of the particles. It can be easily observed experimentally. For the two-dimensional case the integral of (2.40) is cut off at $k \sim S^{-1/2}$, where $S$ is the area of the surface, and total disappearance of the particles actually occurs in a time $\sim S/D$. We should note that, in the case of instantaneous generation of particles, faster asymptotic relations hold.[44,59,60]

The case is more interesting of *oppositely charged* particles. The long-wavelength fluctuations of $z(r,t)$ are suppressed by the Coulomb field of the particles. Here, in the initial system of (2.30) a force term appears having the potential $\varphi$, which satisfies the Poisson equation:[61]

$$\Delta\varphi = \frac{4\pi e}{\varepsilon}(c_A(r) - c_B(r)). \tag{2.42}$$

The behavior of charged particles is substantially changed in going from the three- to the two-dimensional case. Let us present the results in three-dimensional space. The spectrum of steady-state fluctuations has the form

$$\langle z_k(t) z_{k'}(t')\rangle = \frac{2i_0\delta(k+k')}{D(k^2+a_D^{-2})}\exp[-D(k^2+a_D^{-2})]. \tag{2.43}$$

Here $a_D = (8\pi c_A e^2/T)^{-1/2}$ is the Debye radius of the system in a steady-state regime. We can easily obtain the spectrum of fluctuations of the potentials and fields. Thus the mean of the modulus of the field in the specimen is

$$|\bar{E}| \approx e\left(\frac{T}{e^2}\right)^{1/8}\left(\frac{i}{D}\right)^{3/8}$$

At large fluxes or small diffusion coefficients it can reach a

considerable magnitude. This phenomenon has been experimentally observed in irradiated dielectrics.[62]

Since the fluctuations of $z(r,t)$ for small $k$ do not have a singularity, the long-term asymptotic behavior of annihilation of particles upon turning off the source does not differ substantially from the laws of formal kinetics.

The singularities are interesting in the two-dimensional case, which arises, e.g., when a flux of particles impinges on a surface. Here the electric fields are not too long-range and the long-wavelength fluctuations are only partly screened.

The spectrum of fluctuations of $z$ has the form

$$\langle z_k(t) z_{k'}(0)\rangle = \frac{2i_0\delta(k+k')}{D(k^2+|k|q)} \exp[-D(k^2+|k|q)t]. \quad (2.44)$$

Here we have $q = 2\pi\sigma_0 e^2/T$ (an analog of the reciprocal Debye radius in two dimensions), and $\sigma_0$ is the two-dimensional particle density. In connection with (2.44) we note the interesting property of the time spectrum of charge fluctuations at a point in the plane

$$\eta_\omega = \int \langle Q(t)Q(0)\rangle \exp(i\omega t)\,dt = 2i_0 \int \frac{d^2k}{D^2(k^2+q|k|^2+\omega^2)}. \quad (2.45)$$

For small $\omega$ and $q=0$ we have $\eta_\omega \sim 1/\omega$, i.e., there is a singularity of the flicker-noise type.

Finally we present without proof the law of decay of the density of particles upon turning off the source:[56] $c_A \sim t^{-1/3}$.

### 2.4. Field-theoretical methods

The *field-theoretical methods* are based on exact formulation of the multiparticle problem of reacting particles. Here we shall give a presentation of these methods as applied to a system of defects of two types A and B in a solid. We shall consider the concentration (on the average) of the two forms of particles, as before, to be the same, $c_A(t) = c_B(t)$. Let us denote by $w_a(\mathbf{y}-\mathbf{x})$ the probability per unit time that an electron (a type A defect) lying at the point $\mathbf{x}$ and a hole (a type B defect) at $\mathbf{y}$ recombine. The inverse process caused by thermal activation and photo-and radiation pair generation is described by the probability $w_b(\mathbf{x}-\mathbf{y})$ that A and B defects are formed per unit time in the vicinity of the points $\mathbf{x}$ and $\mathbf{y}$, respectively. They can be neutral or charged defects or electrons and holes. Thus, in contrast to the preceding section, we can assume that the pairs are created *in correlated fashion* with the distribution function $f(r) = w_b(\mathbf{x}-\mathbf{y})$. For simplicity, we shall not take account of the potential interaction, while considering all the concentrations to be small. That is, the *small gas parameter* exists

$$c_{A(B)} r_{a,b}^d \ll 1, \quad (2.46)$$

where the $r_{a,b}$ are the characteristic lengths of the functions $w_{a,b}(\mathbf{x})$.

We shall characterize the state of the multiparticle system being studied, which includes $m$ electrons (which form the configuration $X^m = \{x_1;\dots;x_m\}$ and $m$ holes $Y^m\{y_1;\dots;y_m\}$ by the distribution function $U^m(X^m,Y^m)$. The evolution of $U_m(X^m,Y^m)$ is described by the corresponding diffusion-balance equations. According to Refs. 60, 63, and 64, the latter can be rewritten in a form analogous to the *Schrödinger equation*:

$$\frac{\partial}{\partial t}|F(t)\rangle = H|F(t)\rangle; \quad (2.47)$$

Here the "wave" function $|F(t)\rangle$ is defined in terms of wave operators by the relationship

$$|F(t)\rangle = \sum_{m=0}^{\infty} \int \frac{dx_1\dots dx_m}{m!} \int \frac{dy_1\dots dy_m}{m!} U_m(X^m, Y^m)\,|X^m, Y^m\rangle, \quad (2.48)$$

$$|X^m, Y^m\rangle = \psi_e^+(\mathbf{x}_1)\dots\psi_e^+(\mathbf{x}_m)\psi_h^+(\mathbf{y}_1)\dots\psi_h^+(\mathbf{y}_m)|0\rangle.$$

Our problem is to calculate the *correlation function* characterizing the spatial distribution of uncompensated charges (particle-number densities)

$$\langle \Delta c(0)\Delta c(\mathbf{x})\rangle = \langle (c_e(0)-c_h(0))(c_e(\mathbf{x})-c_h(\mathbf{x}))\rangle, \quad (2.49)$$

which is expressed in terms of the particle-number density operator

$$c_e(\mathbf{x}) = \psi_e^+(\mathbf{x})\psi_e(\mathbf{x}) \quad (2.50)$$

and the pair densities

$$\langle \Delta c(0)\Delta c(\mathbf{x})\rangle = 2(X-Y) \quad (2.51)$$

of same-type and opposite-type defects. The "Hamiltonian" (2.47) in $\mathbf{k}$-space has the form

$$H = H_0 + H_{int}, \quad (2.52)$$

where $H_0$ corresponds to free diffusion:

$$H_0 = -D_e\sum k^2\alpha_k^+\alpha_k - D_h\sum k^2\beta_k^+\beta_k, \quad (2.53)$$

while $H_{int}$ includes the terms involving reaction:

$$H_{int} = \sum w_b(\mathbf{k})\alpha_k^*\beta_{-k}^+ + \sum w_a(\mathbf{k})\alpha_k\beta_{-k} - Vw_b(0) - V^{-1}\sum w_a(\mathbf{k}-\mathbf{k}_1)\alpha_k^+\alpha_{k_1}\beta_{k_2}^+\beta_{k_3}\Delta(\mathbf{k}+\mathbf{k}_2-\mathbf{k}_1-\mathbf{k}_3); \quad (2.54)$$

Here $\Delta(\mathbf{k})$ is the Kronecker symbol; $\Delta(\mathbf{k}) = \delta_{k_1,0},\dots,\delta_{k_d,0}$. Upon averaging the operator for the total number of particles, we obtain an expression of the concentration in terms of the condensation operators[60] $\alpha_0$ and $\beta_0$ ($\alpha_0 = \beta_0$):

$$c = V^{-1/2}\langle\alpha_0\rangle. \quad (2.55)$$

Further, upon writing the equations of motion for the condensation operators in the Heisenberg representation and replacing them with numbers according to Refs. 57 and 61,

$$\alpha_0^+, \beta_0^+ \to V^{1/2}, \quad \alpha_0, \beta_0 \to cV^{1/2}, \quad (2.56)$$

we obtain the *equation for the concentration*

$$\frac{dc(t)}{dt} = w_b(0) - w_a(0)c^2(t) - V^{-1}\sum{}' w_a(\mathbf{k})\langle\alpha_k\beta_{-k}\rangle, \quad (2.57)$$

Here the prime on the summation sign indicates that the summation index does not take on a zero value. The separation of the condensation operators is performed also in the Hamiltonian and in the correlator being calculated.

Finally, let us go over to the *interaction representation* and introduce the S-matrix in the standard way,[61,62] so that the wave function is given by the equation

$$|F(t)\rangle = S(t;\,-\infty)\,|F(t=-\infty)\rangle, \tag{2.58}$$

Here $|F(t=-\infty)\rangle$ corresponds to randomly distributed particles with the concentration $c$, while we assume the interaction to be introduced adiabatically.

The expression for the correlation function of interest to us acquires the form

$$\langle \Delta c(0)\,\Delta c(\mathbf{x})\rangle = 2c\delta(\mathbf{x}) - 2V^{-1}\sum{}' \cos(\mathbf{kx})\,\langle \alpha_{\mathbf{k}}\beta_{-\mathbf{k}}\rangle + V^{-1}\sum{}' \exp(i\mathbf{kx})\,(\langle \alpha_{\mathbf{k}}\alpha_{-\mathbf{k}}\rangle + \langle \beta_{\mathbf{k}}\beta_{-\mathbf{k}}\rangle). \tag{2.59}$$

To calculate the *correlator* $\Gamma_{\mathbf{k}} = \langle \alpha\beta_{-\mathbf{k}}\rangle$ in the lowest (second) order in the concentration, we must sum the ladder diagrams containing the minimal number of entering condensate lines giving the coefficient $c$ and the vertices with the small interaction $w_b$. They are constructed of the following terms in the Hamiltonian:

$$H_{\text{int}} = \sum{}' w_b(\mathbf{k})\,\alpha^+_{\mathbf{k}}\beta^+_{-\mathbf{k}} - c^2\sum{}' w_a(\mathbf{k})\,\alpha^+_{\mathbf{k}}\beta^+_{-\mathbf{k}} - V^{-1}\sum{}' w_a(\mathbf{k}-\mathbf{k}_1)\,\alpha^+_{\mathbf{k}}\alpha_{\mathbf{k}_1}\beta^+_{\mathbf{k}_2}\beta_{\mathbf{k}_3}\Delta(\mathbf{k}+\mathbf{k}_2-\mathbf{k}_1-\mathbf{k}_3) - \ldots \tag{2.60}$$

As we can easily convince ourselves, the correlators $\langle \alpha_k\alpha_{-k}\rangle$, $\langle \beta_k\beta_{-k}\rangle$, are proportional to $c^3$ and must be omitted.

The free Green's function (GF), as we can easily see from the Heisenberg equations of motion, is determined by the formula

$$G^{\alpha}_{\mathbf{k}}(t) = \exp(-D_e k^2 t)\,\Theta(t). \tag{2.61}$$

The essential point is that $G_{\mathbf{k}}(t) \equiv 0$ when $t<0$.

To avoid the divergences that arise in low-dimensional systems owing to the singularity of the free GF $G_{\mathbf{k}}(\omega)$ as $\mathbf{k}\to 0$, $\omega\to 0$, we shall construct the ladder diagrams from the "clothed" GF:

$$G^{\alpha}_{\mathbf{k}}(\omega) = \left(D_e k^2 - i\omega - \Sigma^{\alpha\alpha}_{\mathbf{k}}(\omega)\right)^{-1}. \tag{2.62}$$

Here the mass operator $\Sigma_{\mathbf{k}}(\omega)$ is also calculated in the ladder approximation. Since it is small in the concentration [see Eq. (2.65)], $\Sigma_{\mathbf{k}}(\omega)$ in (2.62) can be replaced by its value $\Sigma_0$ for $\mathbf{k}=0$, $\omega=0$ when the free GF (2.61) has a singularity.[61]

Let us denote by $\Gamma^{(1)}_{\mathbf{k}}$ the sum of the ladder diagrams beginning with the first term in the interaction Hamiltonian of (2.60), and by $\Gamma^{(2)}_{\mathbf{k}} = \Gamma_{\mathbf{k}} - \Gamma^{(1)}_{\mathbf{k}}$ the sum of the diagrams beginning, correspondingly, with the second term. For $\Gamma^{(1)}_{\mathbf{k}}$ and $\Gamma^{(2)}_{\mathbf{k}}$ we have the integral equations

$$\Gamma^{(1)}_{\mathbf{k}} = (Dk^2 + 2|\Sigma_0|)^{-1}\,(w_b(\mathbf{k}) - V^{-1}\Sigma'\,w_a(\mathbf{k}-\mathbf{k}_1)\,\Gamma^{(1)}_{\mathbf{k}_1}), \tag{2.63}$$

$$\Gamma^{(2)}_{\mathbf{k}} = -(Dk^2 + 2|\Sigma_0|)^{-1}\,(c^2 w_a(\mathbf{k}) + V^{-1}\sum{}' w_a(\mathbf{k}-\mathbf{k}_1)\,\Gamma^{(2)}_{\mathbf{k}_1}). \tag{2.64}$$

Here we denote by $D$ the overall diffusion coefficient $D = D_e + D_h$.

As we can see simply, the *mass operator* $\Sigma_0$ calculated in the ladder approximation is expressed in terms of $\Gamma^{(2)}_{\mathbf{k}}$ as follows:

$$|\Sigma_0| = cw_a(0) + (cV)^{-1}\Sigma' w_a(\mathbf{k})\,\Gamma^{(2)}_{\mathbf{k}}. \tag{2.65}$$

Owing to the presence of the incoming condensate line $\alpha_0$ ($\beta_0$), the mass operator $\Sigma_0$ is a quantity *small* with respect to the concentration.

The relationships (2.57) and (2.63)–(2.65) for a steady-state regime form a *closed* system whose solution determines the concentration of the particles and the correlation function of the excess-charge densities of (2.59). This problem can be easily solved by taking the probabilities $w_a(\mathbf{x})$ and $w_b(\mathbf{x})$ in the form of $\delta$-functions corresponding to the creation and annihilation of pairs at fixed distances. Let us study the case for $d=3$, for which

$$w_b(\mathbf{x}) = p\,(4\pi r_b^2)^{-1}\,\delta(|\mathbf{x}| - r_b), \tag{2.66}$$

and the one-dimensional case, $d=1$, with

$$w_b(x) = \frac{1}{2}\,p\,(\delta(x-r_b) + \delta(x+r_b)), \tag{2.67}$$

while the recombination probabilities $w_a(\mathbf{x})$ are obtained from (2.66) and (2.67) by replacing all indices $b\to a$. (The two-dimensional case entails additional mathematical complications, and we shall not discuss it here.) Here $p$ is the rate of pair creation, $k_a$ is the rate constant for annihilation. The classical *law of mass action*, which is valid for thermodynamic systems, i.e., when the conditions are satisfied of microscopic equilibrium in local form ($r_a = r_b$ in our case), relates the concentration of particles with the constants $p$ and $k_b$ by the simple relationship

$$c^2 = \frac{p}{k_a} \tag{2.68}$$

(dimensionalities $k_a$–$[\mathrm{s}^{-1}\mathrm{m}^{-3}]$, $p$–$[\mathrm{s}^{-1}\mathrm{m}^{-3}]$). We note that in the case in which the local condition of microscopic equilibrium is not fulfilled, e.g., in photogeneration of carriers, the mass-action law (2.68) generally contains substantial corrections (see (2.76)).

Thus, let us examine first the *three-dimensional* case. Here the mass operator $\Sigma'_0$ can be assumed to be zero. That is, the ladder diagrams can be constructed from the free GF; divergences do not arise.

Upon adding the relationships (2.63) and (2.64) and taking account of (2.57), we obtain the system

$$\Gamma_{\mathbf{k}} = (Dk^2)^{-1}\left[w_b(\mathbf{k}) - c^2 w_a(\mathbf{k}) - (2\pi)^{-d}\int w_a(\mathbf{k}-\mathbf{k}_1)\,\Gamma_{\mathbf{k}_1}d\mathbf{k}_1\right], \tag{2.69}$$

$$c^2 w_a(0) = w_b(0) - (2\pi)^{-d}\int w_a(\mathbf{k})\,\Gamma_k d\mathbf{k}. \tag{2.70}$$

The correlator of the densities of excess charges is expressed in terms of the latter as follows:

$$Z(\mathbf{x}) = \langle \Delta c(0)\,\Delta c(\mathbf{x})\rangle = 2c\delta(\mathbf{x}) - \pi^{-2}x^{-1}\int \Gamma_k k\sin(kx)dk. \tag{2.71}$$

In calculating the three-dimensional integrals in the equations of the system and in substituting (2.70) and (2.69) the integral term in the equation for $\Gamma_{\mathbf{k}}$ drops out, and we obtain

$$\Gamma_{\mathbf{k}} = \frac{p}{Dk^2}\left(\frac{\sin kr_b}{kr_b} - \frac{\sin kr_a}{kr_a}\right). \tag{2.72}$$

Finally the expression for the correlation function of (2.71) acquires the form

$$\langle \Delta c(0)\,\Delta c(\mathbf{x})\rangle = 2c\delta(\mathbf{x}) + pk_d^{-1}x^{-1}[x + r_a - |x - r_a| - r_a r_b^{-1}(x + r_b - |x - r_b|)], \tag{2.73}$$

Here the diffusion rate constant $k_d$ in the three-dimensional case is determined by the ordinary relationship:

$$k_d = 4\pi D r_a. \tag{2.74}$$

Figure 6 shows the graph of the corresponding relationship for the case in which the radius of creation of a pair exceeds the annihilation radius. If $r_b > r_a$, then when $x < r_a$ the correlation function consists of a $\delta$-function at zero and a positive constant component $2pk_d^{-1}[1 - (r_a/r_b)]$; when $r_a < x < r_b$, it decreases hyperbolically to zero: $2pk_d^{-1}r_d(x^{-1} - r_b^{-1})$, while when $x > r_b$ it is identically equal to zero. The positive spatial correlation of the density of uncompensated charges describes the phenomenon of *stochastic aggregation* of particles of the same type in the reacting system. It is simple to estimate the mean number of uncompensated charges in such a cluster:

$$N \sim 1 + \frac{4\pi}{3} p^{1/2} k_d^{-1} r_a (r_b^2 - r_a^2)\left[k_a^{-1} + k_d^{-1}\left(1 - \frac{r_a}{r_b}\right)\right]^{-1/2}. \tag{2.75}$$

The magnitude of the excess charge is proportional to the square root of the small probability of pair creation. However, with a small total diffusion coefficient entering as the coefficient $D^{-1/2}$ and with a large radius of creation $r_b$ ($N \propto r_b^2$), it can be not at all small.

According to this pattern, upon radioactive irradiation of a dielectric, regions must spontaneously be formed in the latter that contain preferentially electrons (or holes). Naturally this will be accompanied by the appearance of internal electric fields, and all the more so, the smaller the mobility of the carriers and the greater the intensity of irradiation. This effect has been experimentally observed in Ref. 62, and here the internal fields attained breakdown values. We note that, for a quantitative description of this phenomenon, we must take account of the Coulomb interaction of the electrons and holes.

With the opposite relationship between the radii of creation and annihilation, $r_b$ and $r_a$ change places, and the continuous component in the correlator becomes negative. This corresponds to "screening" of a particle of one type by the particles of the other type.

It is also interesting to study the expression for the mean concentration obtained from (2.70) and (2.72):

$$c^2 = pk_a^{-1} + pk_d^{-1}\left(1 - \frac{r_a}{r_b}\right)\Theta(r_b - r_a). \tag{2.76}$$

The first term on the right-hand side of (2.76) corresponds to the classical mass-action law (2.68), and the second to a correction to it. Such a correction was first found in Ref. 60 in the extreme case of low carrier concentration and low efficiency of annihilation as compared with diffusion; here it was small in the parameter $k_a/k_d \ll 1$.

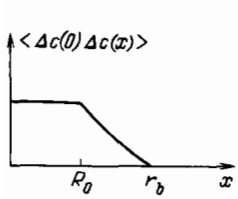


FIG. 6. Graph of the correlation function of the density of uncompensated charges[55] for $r_b > r_a$, $d = 3$, steady-state regime.

We see from (2.76) that, in a system in thermodynamic equilibrium, owing to the condition of microscopic equilibrium $r_a = r_b$, the classical mass-action law is satisfied exactly. If the process of carrier creation is non-equilibrium (although the regime can be steady state), then generally we have $r_a \neq r_b$. If, moreover, an electron-hole pair upon creation separates to distances exceeding its annihilation radius (e.g., upon $\gamma$-irradiation), then the correction to the mass-action law differs from zero, while in the case of slow diffusion and effective annihilation, $k_d \ll k_a$ (the limit of a diffusion-controlled process), *precisely it* becomes decisive.

As we have shown above, in this regime accumulations of single-type particles are formed in space, and they dissipate slowly by diffusion. Evidently the clustering of single-type particles increases the steady-state value of the concentration (the sign of the correction term is strictly positive): if the clumps of particles are uniformly distributed, the annihilation processes become more effective and the equilibrium concentration is lowered. Thus the nature of the correction to the mass-action law lies in *steady-state fluctuation effects*, which are manifested when $r_b > r_a$. The one-dimensional case has been discussed in detail in Ref. 55.

Analogous problems are studied also in Ref. 65 with use of the *scaling method*. The authors confirmed the existence of segregation of defects when $d = 1$ or 2, while denying it for $d = 3$. As we see it, segregation occurs also in the latter case, as is evidently visible from the appearance in the course of defect accumulation of a non-Poisson spectrum of their density fluctuations that does not break down the macroscopic homogeneity of the system (see also Ref. 44). Apparently the scaling method is not sensitive to such a type of phenomena of *microscopic self-organization*[66] having a characteristic radius on the atomic scale.[2)]

## 3. COMPUTER SIMULATION OF DEFECT ACCUMULATION

### 3.1. Quasicontinuum model

To establish the fundamental laws of the process of the kinetics of accumulation, initially the *quasicontinuum* model of a crystal was studied.[17] A one-dimensional "crystal" was represented in the form of a segment of length $L$ of cells with periodic boundary conditions (the ends of the segment are closed). The simulation was conducted for different dimensions $L$ of the crystals and magnitudes $l$ of the recombination region. The fundamental results of the simulation are given in Table II.

The simulation shows that: 1) the curve of the dependence of the number of accumulated defects on the total number of defects created reaches saturation; 2) when $L = 100$–$400$ and $l = 5$–$20$, *complete separation* was attained of the "crystal" into a region of interstitials and a region of accumulation of vacancies; the location of the regions changes upon further generation of defects; 3) the sum of the lengths of the wells filled with vacancies for $L/l > 2$ considerably exceeds the length $L$ (when $L = 2000$, $l = 5$, $u_0 = c_0 l \approx 5$, where $c_0$ is the concentration of *accumulated* defects, in total about $10^6$ pairs of defects was generated).

TABLE II. Results of calculations for a quasi-continuum one-dimensional model.[17]

| $L$ | $l$ | $u_0$ |
|---|---|---|
| 40 | 20 | 1,16 |
| 200 | 100 | 1,16 |
| 100 | 20 | 1,32 |
| 200 | 20 | 2,5 |
| 400 | 20 | 3,5 |
| 2000 | 5 | 5,0 |
| 2000 | 10 | 5,0 |

Let us examine Secs. 1–3 in greater detail. For small values of $L/l$ the magnitude of $u_0$ depends on the ratio $L/l$. However, in all cases one observes the establishment of a steady state (saturation). We note that the existence of a steady-state value was subjected to doubt in a number of studies (e.g., Ref. 43). Therefore one of the purposes of simulation in the quasicontinuum pattern was specifically a rigorous test of this result.

Figure 7 shows a series of successive patterns for $L = 200$ and $l = 20$. We see that in three of the presented patterns for a one-dimensional model, a *complete separation* was established of the "crystal" into regions containing only vacancies or interstitials (cf. Ref. 65). (A number in a square denotes the number of interstitials lying in the corresponding cell, while a number without a square denotes a cell in which vacancies occur, and shows the number of them in the given cell). With time such regions of single-type defects move through the "crystal" and sometimes break up into smaller ones. As a rule, for small values of $L/l$, the regions of single-type defects "crawl" through the "crystal" owing to the fitting at the ends of the regions of defects of the same type. For larger values of $L/l$ regions of single-type defects also exist–*clusters*. However, in this case the separation into two regions was not observed. Evidently, as long as $L/l$ is small, there is not enough room for arrangement of the accumulated defects in the "crystal", and hence they are collected in such a way that the greatest number of them is accommodated. This will happen precisely upon division of the entire "crystal" into only two regions occupied by single-type defects. When $L/l \gtrsim 200$ we no longer find a dependence of $c_0 l$ on $L/l$. We recall that Ref. 39 made estimates on the basis of a simple model of the mean number of defects in a cluster. It was found that $u_0 = R_0/\bar{r} = 3.43$, where $\bar{r}$ is the mean distance between defects. The mean number of particles in a group is $\bar{K} = 120$. These values correlate with the values of $u_0$ from the computer experiment, which obtained $u_0 \simeq 5$ and a mean number of defects in a cluster, respectively, of about 100. (These data were taken from the pattern of accumulation for $L = 2000$ and $l = 5$ with a total number of creation events of $5 \times 10^5$.)

### 3.2. The discrete model

To simulate a real experiment, a simulation was performed for a *discrete* (but one-dimensional) model (described in detail in Refs. 17 and 67). In contrast to the quasicontinuum model, the "crystal" is divided into $2N$ cells of two types: at the initial instant of time the cells with odd numbers are occupied by atoms, and the rest are empty. A vacancy can appear only in a cell with an odd number, and an interstitial atom can lie only in a cell with an even number. Each cell contains no more than one defect.

References 17 and 67 established the existence of saturation of the concentration $c_0$ and a dependence of the number of lattice nodes $v_p$ in the recombination sphere $v_0$ (Table III) ($V = 2M$ in the theory).[18–20]

Reference 35 determined the value of $u_0$ upon approach to the steady state *from above*. One-dimensional crystals were studied of length from $8 \times 10^3$ to $2 \times 10^4$ lattice constants. (Spatial correlation in genetic pairs was absent.) The limiting values $u_0 = 3.5$–$3.6$ for 500 and 700 nodes in the recombination sphere (Table III, third column) are close to the value 3.43 obtained in the continuum approximation by an approximate method[18] and considerably exceed the estimate 1.36 implied by the approach based on multipoint densities.[36,37] Apparently the latter approach takes insufficient account of the specifics of one-dimensional systems.

To determine the existence of a stable steady state, a model was studied[35] of destruction of clusters in the case $v_p = 700$. At the initial instant of time 10 uniformly distrib-

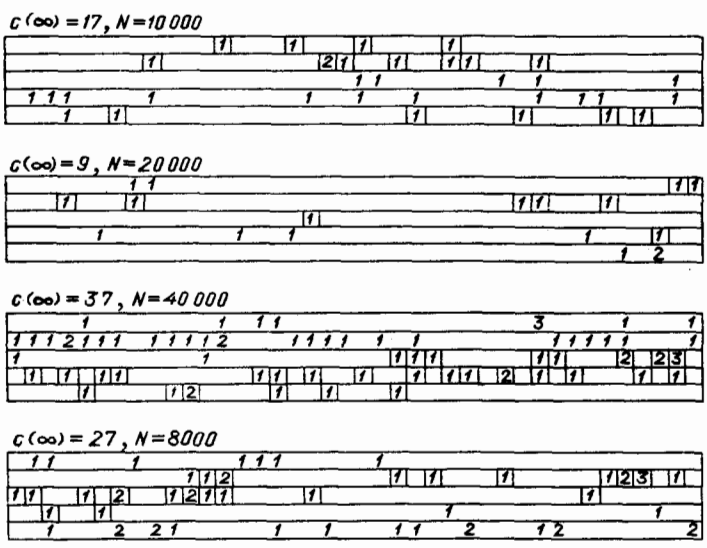


FIG. 7. Sequence of the patterns of accumulation in a quasicontinuum one-dimensional model. $c(\infty)$ is the number of particles in the "crystal" in the saturation region. $N$ is the number of defects that have been created (the dose).[17]

TABLE III. Dependence of the steady-state value of the concentration of defects $u_0$ on the number $v_p$ of nodes in the recombination sphere (one-dimensional case).

| $v_p$ | [35, 67] | [18—20] |
|---|---|---|
| 2 | 0,464 | 0,333 |
| 4 | 0,636 | 0,50 |
| 10 | 0,922 | 0,830 |
| 24 | 1,248 | 1,323 |
| 50 | 1,625 | 1,937 |
| 100 | 2,25 | 2,77 |
| 150 | 2,3±0,2 | |
| 200 | 2,5 | |
| 300 | 3,0 | |
| 400 | 3,2 | |
| 500 | 3,5 | |
| 700 | 3,6 | |
| .... | .... | |
| ∞ | 4,12 | |

uted clusters of 300 vacancies each were put into the "crystal", and interstitial atoms in the intervals between them ($u_0 \approx 10$). Then pairs of randomly distributed defects of differing types were created in the "crystal". The newly generated defects break up the originally existing clusters and the concentration of defects *declines* to a steady-state value. The values of $u_0$ were obtained by averaging a region of the curve of length $2.5 \times 10^4$ events of defect creation. The result unambiguously implies the existence of a *stable steady state* in the problem of accumulation of point defects and in the problem of breakup of clusters.

A connection was established by simulation of accumulation[67] between $c_0$ and the ratio of the number of active interstitials to the total number of them in the crystal

$$u_0 = 1 - 2\beta. \tag{3.1}$$

Here $\beta$ is a function of $v_p$, the number of nodes active toward recombination in the recombination sphere. (In Ref. 67 the concentration and recombination volume were expressed in units of the volume $v_0$ of the unit cell, and $c_0$ coincides with the fraction of nodes or interstitial sites occupied respectively by vacancies or interstitial atoms.) We note that, in the model being discussed, the cell itself in which a vacancy occurs is considered inactive with respect to recombination of an interstitial on it.

For a qualitative study of the clustering effect in accumulation, also *pair correlation functions* have been defined for the same- and opposite-type defects.

We see in Fig. 8 that the distribution function of single-type defects at small values of $r$ has a region of enrichment ($X(r) > 1$) for $r \lesssim 2R_0$, which indicates the clustering of defects. The distribution function $Y$ of opposite-type defects in this same region has a value smaller than unity. (These results agree well with the analytic theory in the continuum approximation.)[36,37] It was established by the simulation that clustering *increases* with increasing number of lattice nodes $v_p$ in the recombination sphere. Thus, when $v_p = 4$, the clustering effect is practically absent: $X \approx 1$ for all $r$. The absence of the clustering effect at low $v_p$ indicates the possibility of applying the ordinary superposition approximation in such cases.

Reference 34 proposed the following accumulation equation on the basis of results of simulating the accumulation of point defects in a one-dimensional crystal in the saturation stage:

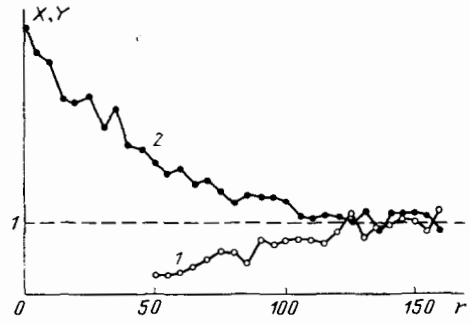


FIG. 8. Pair distribution functions obtained in a computer experiment of opposite-type (*1*) and same-type (*2*) defects[17] in a discrete model with 50 nodes in the recombination sphere. Correlations in genetic pairs are absent.

$$\frac{dc}{dt} = p\left(1 - \frac{2\alpha(t)\,c(t)\,v_0}{1 - c(t)\,v^*}\right), \tag{3.2}$$

(in the notation of Ref. 67 we have $v_0 = v_p v^*$, where $v_p$ is the number of nodes in the recombination sphere), $\alpha(t)$ is the efficiency coefficient of recombination; $\alpha(t=0) = 1$ characterizes the efficiency of recombination of a single defect. With increasing degree of overlap of the recombination sphere, $\alpha(t)$ declines. It was assumed for $\alpha(t)$ in Ref. 34 that

$$\alpha(t) = 1 - (1 - \alpha(\infty))\frac{c(t)}{c_0}. \tag{3.3}$$

Here $\alpha(\infty) = \alpha(t \to \infty)$ is the value in the saturation stage.

To test the hypotheses (3.2) and (3.3), the kinetics of accumulation was simulated on a computer by the method described in Ref. 67. For each of the values $v_p = 10, 16, 24$, and 50, the process of accumulation was performed independently 200 times until the stage of steady-state values of $c_0$ was reached. The relationships $c(N)$, $N = pt$, and $\alpha(c)$ were constructed from the mean values obtained in this series. Figure 9 shows these dependences for the case $v_p = 24$ and $c_0 = 0.052$ as obtained from (3.2) and (3.3). We see that, within the limits of error of experiment ($\approx 5\%$), the slowly varying function $\alpha(c)$ can be well approximated by the linear dependence of (3.3), which confirms the suitability of (3.2) and (3.3) for describing the accumulation of point defects in the discrete model. Analogous results are obtained

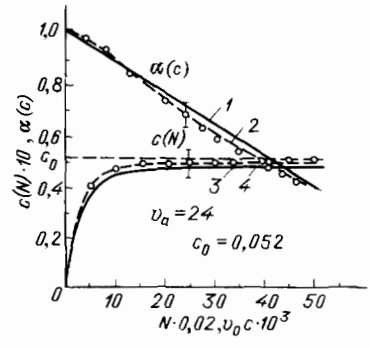


FIG. 9. Theoretical dependences:[17] *1*—$\alpha(c)$, *4*—$c(N)v_p$ calculated by (3.2) and (3.3); the corresponding results of simulation are: *2*—$\alpha(c)$, *3*—$c(N)v_p$.

TABLE IV. Dependence of the concentration of defects at saturation on the degree of correlation in genetic pairs.[17]

| $d/a$ | $u_0$ |
|---|---|
| 1 | 0,75 |
| 2 | 0,81 |
| 3 | 0,85 |
| 5 | 0,89 |
| 30 | 0,92 |

for $v_p = 16$ and 50. For $v_p = 16$ and 50, the values were found by simulation, respectively, of 1.092 and 1.625 for $c_0$ and 0.463 and 0.478 for $\beta(\infty) = \alpha(\infty) v_0 c_0$.

Finally, to estimate the role of spatial correlations in *genetic pairs* of created defects, the case was simulated in which a created interstitial lies at various distances $d$ from the edge of the recombination sphere of its vacancy (see Fig. 2 in Ref. 35). Table IV shows also the values for different $d$ in the case $v_p = 10$. As we see from these results, only when $d/a < 5$ do the correlations substantially alter $u_0$; for small $d$ the magnitude of $u_0$ declines owing to the suppression of the effect of clustering by correlations *within* the pairs being created. The distribution functions of same- and opposite-type defects also have a distinctive form. Thus, in the steady-state distribution function of opposite-type defects one observes a *maximum* at small distances (in the region of interpair correlation). The distribution function of same-type defects in the region of small values takes on smaller values than in the case of absence of interpair correlations. [This agrees well also with the analytic calculations for the continuum model[30,36] (see Fig. 3)].

In Ref. 42 the accumulation was also simulated in a computer of point defects in a *two-dimensional* "crystal" represented by a square lattice with the lattice constant $d$ and dimensions $L \times L$ (in units of $d$). In the initial state $L \times L$ atoms are placed at the lattice nodes, while $L \times L$ interstitial states are free. The recombination region amounts to a square containing $l \times l$ nodes. Otherwise the simulation is analogous to the one-dimensional case. Periodic conditions are imposed on the boundaries of the crystal. The number $N$ of accumulated defects of a single type was determined, together with the dimensionless parameter $u_0$ (the multiplicity of the covering of the crystal by the sum of the areas of the instability zones of the accumulated defects). A steady state is established in the cases studied, as a rule, after $4 \times 10^4$ events of pair creation. To obtain the steady-state values of $u_0$ and $N$, an averaging was performed over the last $5 \times 10^4$ creation events with a total number of them of $10^5$. The results are given in Table V, which shows the results of simulation for a set of parameters $L$, $l$, and $L/l$. In all cases we observe a strongly marked effect of aggregation of same-type defects. Figure 10 shows the spatial distribution of vacancies and interstitial atoms for the case corresponding to the second row of Table V ($10^5$ generation events). The steady-state value of $u_0$ does not depend on $L$ when $L/l \geqslant 10$ (see Table V).

In the two-dimensional case the value of $u_0$ is *smaller* than for the one-dimensional case at the same magnitudes of the region of recombination (thus, when $v_p = 400$, $u_0 = 3.2$ in the one-dimensional case). However, the aggregation effect is expressed rather strongly.

The quantity $u_0$ characterizes the degree of aggregation only on the *average*. Therefore it is important in each concrete case to analyze also the spatial distribution of the defects. Thus, the low-temperature accumulation of Frenkel defects in the two- and three-dimensional cases was simulated in Ref. 68. The obtained values of $u_0$ for $d = 2$ considerably exceed the same in Ref. 42. In contrast to Ref. 42, the authors of Ref. 68 used a *circle* as the recombination region. The values of $u_0$ ($= \lambda_n/2$ in the notation of Ref. 69) obtained in Ref. 69 are considerably larger than the results of Ref. 68. We note that it was assumed in Ref. 69 that, when an interstitial atom occurs at a node where the recombination spheres of several vacancies overlap, it recombines with the *closest* vacancy.

In the three-dimensional case[68] the saturation concentration is *smaller* than for $d = 1$ and 2; the maximum value is $u_0 = 1.02$ (for $v_p = 266$ nodes in the recombination sphere), and its dependence on $v_p$ is weaker.

A two-dimensional simulation has also been performed of the accumulation of defects with an *asymmetric* recombination region[42] chosen in the form of a rectangle with $v_p = a \times b$. The anisotropic case was studied in which the larger sides of all the rectangles are oriented in parallel. For $v_p = 6 \times 16$ and $L \times L = 150 \times 150$, the steady-state value was obtained of $u_0 = 1.35$. The greater lengths of the sides of the rectangle enhance the aggregation effect in this direction. Thus, in a simulation with $v_p = 6 \times 16$, *extended* clusters were observed, which divided the crystals into strips (Fig. 11).

A simulation of accumulation with account taken of *diffusion* was first performed for $d = 2$ in Refs. 49 and 70. Evidently the inclusion of diffusion smears out the clusters—the more efficiently the greater is the mobility of the defects.

TABLE V. Results of computer simulation of accumulation in a two-dimensional crystal of length $L$. The area of the recombination zone is $l \times l$, the number of accumulated defects is N.[42]

| $L \times L$ | $L/l$ | $l \times l$ | $u_0$ | N |
|---|---|---|---|---|
| 50×50 | 5 | 10×10 | 0,89±0,02 | 22,34 |
| 100×100 | 10 | » » | 1,2 | 120,7 |
| 150×150 | 15 | » » | 1,2 | 270,0 |
| 250×250 | 25 | » » | 1,2 | 752,4 |
| 100×100 | 10 | 20×20 | 1,0 | 25,3 |
| 500×500 | 25 | 20×20 | 1,36 | 322,3 |

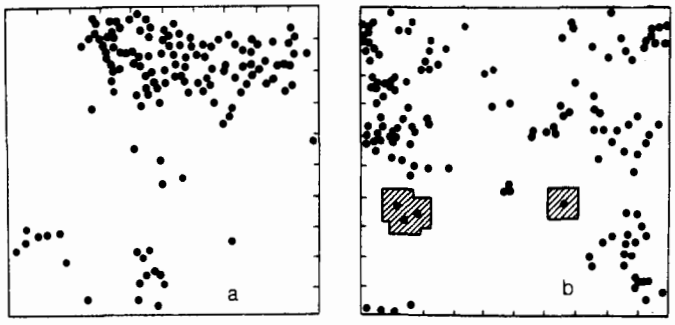


FIG. 10. Distribution of vacancies (a) and interstitial atoms (b) in a two-dimensional computer experiment.[35] The recombination region around a solitary vacancy and an accumulation of three vacancies are cross-hatched.

Recently a simulation was performed on a supercomputer of accumulation with account taken of diffusion—the A and B defects migrate and recombine when they fall into adjacent lattice nodes.[71] They confirm aggregate formation for $d = 1$ and 2. For $d = 3$ the authors interpreted the distribution of defects in the cubic lattice as random—in contrast to the aggregation in the fractal lattice of Serpinskiĭ. (An absence of aggregation in a three-dimensional cubic lattice was also obtained by the scaling method in Ref. 65. However, the authors of Ref. 65 did not calculate the pair correlation functions, while in the case $d = 3$ the aggregation effect, as is known, is relatively weak,[54,68] and hence might remain unnoticed.

Finally, in Ref. 41 the accumulation was simulated of immobile defects for $d = 1, 2$, and 3 in a continuum model. It was easy to show that in these cases $u_0 = 4.2$, 2.07, and 1.04, respectively. This does not contradict the results of simulation in the discrete model[35,42,67]—see Table I, as well as the one-dimensional pattern for the continuum model (see Table III).

The role of correlations in the two- and three-dimensional cases is less important because a genetic pair that has been newly created can overlap in a large number of ways with another pair already existing in the crystal without recombination of the components of different pairs.

### 3.3. Taking account of tunneling charge-transfer of defects

A simulation has been carried out[11,12] of the process of accumulation of immobile Frenkel defects restricted by *tunneling recombination* of defects, as is observed in many solid insulators. The latter consists in the spontaneous transport of an electron between defects of opposite types. In contrast to the ionic process of annihilation of close pairs of the vacancy-atom type, it is characterized by a broad spectrum of recombination times. Thus, the probability for a pair of chosen defects that lie at the relative distance $r$ to survive for $\tau$ seconds is

$$P(\tau) = \exp\left[-W_0\tau \exp\left(-\frac{r}{a}\right)\right]. \tag{3.4}$$

Here $W_0$ and $a$ are constants that depend on the electronic structure of the defects. For small $r$ the lifetime of the pair is minimal: $\tau \gg W_0^{-1}$ (usually $\tau \gtrsim 10^{-5}$–$10^{-8}$ s), and increases *exponentially* with the distance $r$ between the defects.

The simulation was performed for the three-dimensional case with imposition of periodic boundary conditions on the cube in which the defects are being created. The initial distribution function of genetic defects was chosen in the form

$$f(r) = \frac{e^{-r/r_0}}{4\pi r^2 r_0}, \quad \int f(r)\,\mathrm{d}r = 1. \tag{3.5}$$

The results of the simulation confirmed the hypothesis expressed in Refs. 11, 72, and 73 that tunneling recombination is the basis of the secondary reaction that leads to *saturation* of the concentration of defects upon prolonged irradiation of many insulators.

Figure 12 shows the pair correlation function of same-type defects in the region of concentration saturation as calculated from the results of the simulation. We see that the fraction of close-lying Frenkel defects (of the type of paired $F_2$-centers) exceeds by approximately *threefold* the value

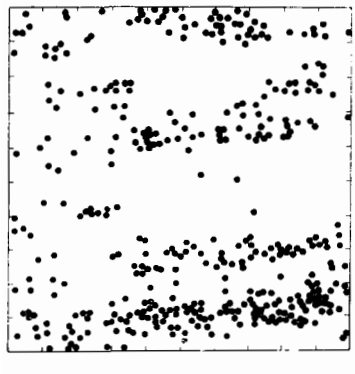

FIG. 11. Spatial distribution of defects in a two-dimensional simulation with rectangular recombination regions.[42]

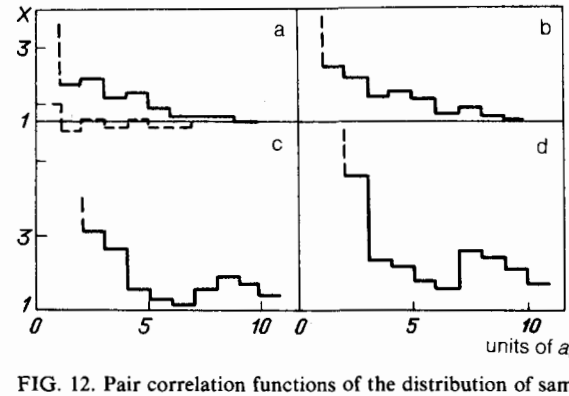


FIG. 12. Pair correlation functions of the distribution of same-type defects obtained in a simulation of accumulation limited by tunneling charge transfer.[12] Intensity of radiation $p$ ($\mathrm{cm}^{-3}\,\mathrm{s}^{-1}$) = $5\times10^{20}$ (a), $6.7\times10^{19}$ (b), $1.2\times10^{18}$ (c), and $2.2\times10^{16}$ (d). The dotted line around the axis of abscissas in Fig. 12a corresponds to absence of recombination.

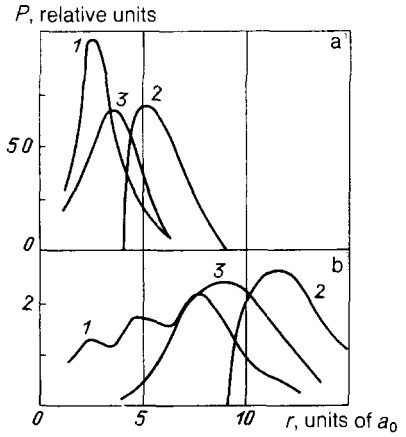


FIG. 13. Probability density of finding a closest defect of the same type (*1*) and of the opposite type (*2*) at the distance *r* from a given defect in the region of concentration saturation.[12] Curve *3*—absence of recombination. Intensity of irradiation $p$ ($cm^{-3}\,s^{-1}$) = $5\times10^{20}$ (a) and $2.2\times10^{16}$ (b).

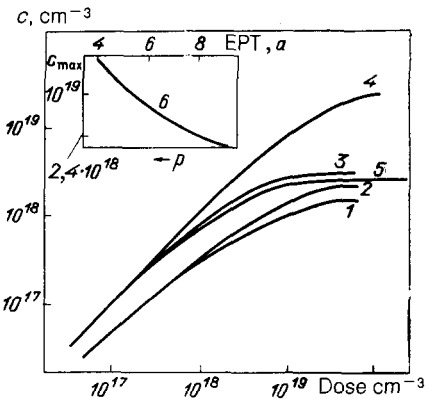


FIG. 14. Curve of the accumulation of defects as a function of the irradiation dose with a tunneling mechanism of recombination.[12] *1*—correlated pairs ($R_g = 10\,a$), $p = 7\times10^{16}\,cm^{-3}\,s^{-1}$, $W_0 = 10^7\,s^{-1}$; *2*—analogous to *1* with $p_2 = 10p_1$; *3*—for an uncorrelated distribution with the same $p_1$, $W_0$ as for *1*; *4*—uncorrelated distribution with $p = 5\times10^{20}\,cm^{-3}\,s^{-1}$; *5*—analogous to *4* with $p = 2.2\times10^{16}\,cm^{-3}\,s^{-1}$; *6*—dependence of the concentration at saturation on the irradiation intensity *p*.

expected in a Poisson distribution, which agrees well with the analytic theory[36,37] and with experiment.[13]

In contrast to the case of annihilation, here the intensity $p$ of irradiation plays a substantial role since, in view of the relatively large lifetimes of close-lying defects, the appearance of a third defect influences their recombination. We see from Fig. 12 that, the smaller is the intensity $p$ of irradiation, the closer the defects of the same type lie, and the better marked is the clustering. (We can interpret the curves of Fig. 13c, d as the creation of periodically arranged accumulations of same-type defects).

Figure 13 demonstrates the form of the obtained probability density of finding *nearest* neighbors of the same or opposite types as a function of the relative distance. We see that a smaller irradiation intensity leads to increasing the minimal relative distance between opposite-type defects; at smaller distances the created pairs efficiently recombine. Owing to the tunneling recombination, the defects of the same type prove to lie closer, and those of opposite types farther than in the case of their equal-probability (Poisson) distribution throughout the volume (curves *3* in Fig. 13).

Since greater irradiation intensities lead to creation of closer opposite-type defects (see Fig. 13), we can expect that this should facilitate attainment also of larger defect concentrations at saturation. The curves and the inset in Fig. 14 illustrate well what we have said.

We can also easily conclude from Fig. 14 that, the stronger is the degree of correlation in genetic pairs ($R_d \to 0$), the smaller is the concentration $c_0$ at saturation. This agrees well also with the analytic theories.[31,32] Figure 14 (curves *1* and *3*) implies that, in the saturation region, the mean distance between defects of the same type $\bar{r} = 0.554/c_0^{1/3}$ amounts to about 50Å, whereas the distance, even in weakly correlated pairs ($R_d/R_0 = 10$) [see (3.5)] is of the order of 25Å. That is, genetic pairs do not mix, even in the saturation region, which leads curves *1* and *3* to reach saturation at *different* concentrations.

The dependence of the low-temperature accumulation curve on the intensity $p$ of irradiation is a characteristic sign of tunneling recombination. It has been observed in the most varied solid matrices: alkali-halide crystals,[14] glasses,[73,74] etc.

## 4. ANALYSIS OF THE KINETICS OF ACCUMULATION

Some analytic expressions are collected in Table VI that have been used in the literature to describe the experimental curve $c(t)$ of accumulation or $dc/dt$, the rate of accumulation. Experimentally the kinetics of accumulation of Frenkel defects in the course of irradiation at liquid-helium temperatures has been studied, both in the alkali-halide crystals[13,14] and in many metals.[75–77] Since often a successful approximation of the accumulation curve is associated with a micromechanism of defect formation (see, e.g., Ref. 47) and with other physical conclusions, it is reasonable to discuss briefly the region of applicability and the substantiation of the expressions that have been used.

Equation (1) of Table VI is the most general and rigorous analytic result. However, it does not take account of the aggregation of same-type defects and hence is applicable only at not very large irradiation doses (up to a concentration of defects $\lesssim 1/2c_0$, where $c_0$ is the concentration at saturation).

In (1) (see Table VI) the existence is allowed of clusters of single-type defects, but here it is actually assumed that these clusters are statistical fluctuations of the Poisson distribution of same-type defects, and do not reflect a real pattern of cluster formation with a substantially non-Poisson spectrum of fluctuations. It is assumed implicitly in (1) that, after each event of creating a new pair of defects, the entire system of defects is shaken up to attain a Poisson distribution. In the case of absence of correlation in genetic pairs we arrive at Eq. (2).

Equation (3) is most widely used in analyzing experimental curves, since its form is intuitively clear: the rate of accumulation is determined by the fraction of the free volume of the crystal not occupied by previously created defects, without taking account of the overlap of the annihilation volumes of same-type defects. Evidently it is applicable only in the initial stage of kinetics at relatively low concentrations of defects, $cv_0 \ll 1$. As was stated (Sec. 2.2), the SA corresponds to the first two terms of the expansion (2) in powers of $cv_0$. The equations (4) and (5) correspond to taking account of two more terms of the expansion. It was erroneously stated in Ref. 31 that the coefficient of the cubic

TABLE VI. Different forms of approximation of the accumulation curve $c(t)$ and the rate of accumulation $dc(t)/dt$.

| Equation | Notes | References |
|---|---|---|
| 1. $\frac{dc}{dt} = p(1-\gamma)(2e^{-cv_0} - 1) - \frac{p\gamma}{2cv_0}[2cv_0 - 1 + 4e^{-v_0c}(1-cv_0) - 3e^{-2cv_0}]$ | Neglecting correlation of same-type defects, $\gamma = \int_0^{R_0} f(r)\,dr$ | [31] |
| 2. $\frac{dc}{dt} = p(2e^{-cv_0} - 1)$, $c = pt - \frac{1}{v_0}\ln(2e^{-cv_0t} - 1)$ | The same, neglecting correlation of genetic defects during creation ($\gamma = 0$), $c_0 = \ln 2/v_0 \approx 0.69/v_0$ | [31, 32] |
| 3. $\frac{dc}{dt} = p(1-2cv_0) = pe^{-2pv_0t}$, $c = c_0[1-\exp(-2pv_0t)]$ | Two terms of the expansion (2) for $cv_0 = 1/2v_0$; coincides with the result of the superposition approximation | [16, 30, 32, 77] |
| 4. $\frac{dc}{dt} = p[1-2v_0c+(cv_0)^2] = p(1-cv_0)^2$ | Three terms of the expansion (2) for $cv_0 \ll 1$ | [75—77] |
| 5. $\frac{dc}{dt} = p\left[1-2cv_0+(cv_0)^2 - \frac{2}{3}(cv_0)^3\right]$ | Four terms of the expansion (2) for $cv_0 \ll 1$. In Ref. 31, the coefficient for the cubical member is pointed out by mistake as $-0.01$, instead of $2/3$. | [31, 75] |
| 6. a) $c = \frac{1}{v_0}\ln(pv_0t+1)$, | At temperatures at which the interstitial atoms are mobile | [43, 47] |
| б) $c = \frac{1}{4v_0}[\ln(4pv_0t-1]+1]$, | For temperatures at which the defects are immobile, and for moderate radiation doses. Owing to incompletely taking account of the correlation of same-type defects, $c_0 \to \infty$ | [43, 47] |
| в) $\frac{dc}{dt} = p(4pv_0t+1)^{-1}$ | (At room temperature the coefficient $1/4v_0$ is replaced by $1/v_0$) | |
| 7. $c = \sum_k A_k(1-e^{-\beta_k t}) + Dt$ | Has been applied preferentially in the room-temperature region | [46] |
| 8. $c = c_0[1-\exp(2Kt)]^{1/2}$ | Empirical equation in which $K$ is not necessarily equal to $pv_0$ | [78] |
| 9. $c = c_0\left[1-\frac{1}{(bt+1)^{1/2}}\right]$ | Empirical relationship with the parameters $b$ and $c_0$ | [79] |
| 10. $\frac{dc}{dt} = p\left(1-\frac{2\alpha cv_p}{1-cv_0}\right)$ | It is assumed that the efficiency of recombination $\alpha(v) = \text{const} = 0.85$ | [33] |
| 11. $\frac{dc}{dt} = p\left\{1-2\left[1-\left(1-\frac{1-c_0v^*}{2c_0v_0}\right)\frac{c(t)}{c_0}\right]\frac{c(t)v_0}{1-c(t)v^*}\right\}$ | Obtained in an approximation of the results of computer simulation, $\alpha(0) = 1$, $\alpha(t) < 1$ | [17, 34] |

term of Eq. (5) equals $-0.01$, which was then copied in all the experimental studies that used this approximation. It is not always acknowledged that these equations also assume the absence of aggregation of defects; the authors themselves of Ref. 31 propose using Eq. (5) up to the saturation concentration. Therefore it is not justified to extrapolate the rate of accumulation of defects to a zero value to obtain $c_0$ from (3)–(5) (cf. Refs. 75 and 76). Actually the saturation concentration can exceed by severalfold the value predicted by these equations.

Equation (6) predicts a logarithmic growth of the concentration of defects without saturation. To obtain the saturation effect experimentally observed by the authors themselves of Ref. 14, a hypothetical secondary reaction was introduced. The defects of this model were discussed in Sec. 3, while we note only that, in the initial stage, the relationships (6) qualitatively resemble the previous ones. They also have been used in interpreting the experimental data obtained for alkali-halide crystals,[47] and they yielded $v_p = 3000$ anion nodes around a vacancy, which explicitly indicates a tunneling mechanism of recombination.

Finally, Eqs. (7)–(9) are approximations of the kinetics of accumulation that are not substantiated theoretically in any way with curves showing saturation, which qualitatively resemble the form of (1)–(5).

The analytic description of the kinetics of accumulation with account taken of the effect of aggregation leads to Eq. (2.9), which contains the fluctuation parameter $\bar{\nu}$, which unfortunately cannot be represented by a simple analytic expression.

Equation (10) was proposed in Ref. 33 (see Table VI), and yields $u_0 = 0.59$. This evidently too low value is obtained owing to the unsubstantiated assumption of independence of the efficiency of recombination $\alpha$ of $v_p$ and of the irradiation dose, as well as the ill-substantiated choice of the value $\alpha = 0.85$.

Finally, Refs. 17 and 34 obtained the simple empirical formula (11) (see Table VI), which describes well the computer results (see Sec. 2.2). It was shown that the efficiency of recombination depends on the time and on the number of lattice nodes in the recombination sphere $v_p$.

It is evident from what we have said that the basis for analyzing the experimental data can be Eq. (11). However, in this case we must convince ourselves by comparing the experimental and theoretical accumulation curves of their qualitative resemblance. Thus we easily note that convex relationships of the type of Fig. 1 in Ref. 75 require the use of more complex models than those discussed above, where the rate of accumulation declines monotonically.

An approximate analysis of (11) for the accumulation curve for Cu (Fig. 7 in Ref. 77) yields the value $c_0 v_0 \approx 1.2$, which clearly indicates an aggregation effect. This result was obtained on the basis of estimates of $p$ and $v_p$ obtained in this same study, and with the assumption of absence of correlation in genetic pairs. (Evidently taking account of correlation would have led to an even larger value of this parameter.) The experimental kinetics of accumulation of Frenkel defects–F-centers in alkali-halide crystals at liquid-helium temperatures–was studied in Refs. 14 and 47 within the framework of a model that yields a logarithmic dependence of the concentration of defects on the irradiation dose (Eq. 6 of Table VI). Although we have criticized this relationship above, at low radiation doses it can be represented as a polynomial in powers of $c_0 v_0$ resembling Eqs. (3)–(5) (see Table VI). At the same time cogent arguments exist favoring the idea that in alkali-halide crystals the accumulation of F-centers is restricted by their tunneling recombination with the complementary H-centers (see Sec. 3.3). This recombination gives rise to its products—pairs of charged $F^+$- and I-centers, which influence the kinetics of accumulation and complicate the very simple model of accumulation being discussed.

We note in conclusion that taking account of correlation of defects in genetic pairs, formation of pairs of new defects (e.g., owing to the tunneling mechanism of recombination), and of dislocation loops, etc., substantially complicate the theory of the kinetics of accumulation. The *temperature dependences* of the efficiency of accumulation contain substantial information on the correlation of defects in genetic pairs and on the nature of their interaction.[38,80]

## 5. CONCLUSION

The analysis conducted in this study of the results of different theoretical approaches to the kinetics of accumulation of Frenkel defects in irradiated solids with account taken of multiparticle effects has shown that all the theories confirm the *effect of low-temperature radiation-stimulated aggregation* of same-type neutral defects and its substantial influence on the spatial distribution of defects and their concentration at saturation in the region of large radiation doses. The aggregation effect must be taken into account in a quantitative analysis of the experimental curves of the low-temperature kinetics of accumulation of Frenkel defects in crystals of the most varied nature—from metals to wide-gap insulators, it is universal, and does not depend on the micromechanism of recombination of opposite-type defects—whether by annihilation of atom-vacancy pairs (in metals) or tunneling recombination (charge transfer) (in insulators).

The kinetics of defect accumulation requires further theoretical studies at temperatures at which they become mobile. The creation under the action of radiation of an ordered structure from the accumulation of radiation defects is an effect akin to those discussed in the theory of self-organization of structures–*synergetics*.

* Deceased

1) A theory of phase transitions to the superionic state was developed[81] on the basis of representations of unstable pairs of Frenkel defects.

2) This same remark also pertains to the recent studies.[82,83]


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Translated by M. V. King