

Many-particle effects in accumulation kinetics of Frenkel defects in crystals

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Abstract. The analytical theory describing accumulation kinetics of immobile Frenkel defects created under irradiation in crystals and based on a hierarchy of equations for many-particle distribution functions of defect densities is presented and developed. Many-particle effects resulting in a radiation-induced aggregation of similar neutral defects (atoms or vacancies) are treated in detail. Their role in accumulation kinetics and the saturation concentration for 1D and 3D cases is discussed. Comparisons with earlier theories and computer simulations are carried out.

1. Introduction

As is well known, irradiation of most crystals (whether metals or insulators) produces Frenkel pairs of defects (vacancies and interstitials are hereafter denoted by A and B respectively). Pairs of dissimilar defects separated by a relative distance r less than a clear-cut recombination radius, r_0 , spontaneously annihilate very rapidly, $A + B \rightarrow 0$, restoring a perfect crystalline lattice. At low temperatures (typically below 30 K) defects are immobile and annihilation of closely created dissimilar defects is the *main mechanism* limiting their accumulation in this regime, and leading to saturation of defect concentration at high irradiation doses. (The same effect is observed in heavily doped crystals at high temperatures when defects become localised by impurities immediately after their production.)

Both the experimental data (Hobbs *et al* 1973, Faraday and Compton 1965) and theory including computer simulations in metals and insulators (Lück and Sizmann 1964, 1966a, b, Tāle *et al* 1975, Kotomin and Valdats 1977, Kalnin 1982, Kalnin and Krikis 1983) as well as analytical estimates (Pooley 1966, Hughes and Pooley 1971, Dettmann 1965, Dettmann *et al* 1967, Vinetsky and Kondrachuk 1982, Vinetsky 1983, Kotomin and Kuzovkov 1981, Antonov-Romanovskii 1981, 1983, 1984) confirm the statistical effect of a *radiation-induced aggregation* of similar Frenkel defects. Under irradiation a random spatial distribution of defects is substituted by a more ordered one consisting of loose similar defect aggregates. This is also supported by the experimental fact that the share of dimer defects (e.g. F₂ centres in KCl) observed after prolonged x-irradiation by Faraday and Compton (1965) considerably *exceeds* the corresponding

Table 1. Different estimates of the dimensionless saturation concentration $u_0 = c_0 v_0$.

u_0	Reference	Notes
0.5	Dettmann <i>et al</i> (1967) Kotomin and Kuzovkov (1981)	Superposition approximation
0.59	van den Bosch (1971)	Monte-Carlo simulations
0.69	Dettmann (1965)	Neglect of similar defect correlation
≈ 1	Kotomin and Valdats (1977)	Computer simulation incorporating tunnelling recombination
1.05	Pirogov (1984)	3D computer simulations
> 1.2	Lück and Sizmann (1966a, b)	Computer simulations of cascades of defects
1.62	Kalnin and Krikis (1983)	1D computer simulations for discrete lattice with fifty sites within v_0
1.08	Present paper	$\left\{ \begin{array}{l} 3D \\ 1D \end{array} \right.$
1.36		
2.77		
2.77	Vinetsky (1983)	An analytical estimate for one hundred sites within v_0
3.2–3.4	Antonov-Romanovskii (1983, 1984)	An analytical estimate for 3D, 1D
∞	Hughes and Pooley (1971)	Model permits infinite local densities of similar defects

value for a Poisson distribution. Aggregation permits us to reach higher defect concentration under saturation as compared to a random defect distribution which may be important for a radiation physics of materials.

A consistent and rigorous theory of defect accumulation kinetics must be based on an infinite *hierarchy of coupled equations* for the many-point defect densities (MPD) $\rho_{n,n'}$ describing spatial correlations of all orders (Dettmann 1965, Dettmann *et al* 1967, Kuzovkov and Kotomin 1981, 1983), the cut-off of which gives an approximate solution of the problem. There is a simple criterion to estimate the approximations involved. In the case of a continuous approximation and uncorrelated defects within Frenkel pairs the saturation concentration c_0 at high irradiation doses is described by the universal parameter $u_0 = c_0 v_0$, $v_0 = \frac{4}{3} \pi r_0^3$, depending only on a space dimensionality (1D–3D). Thus, cutting off the hierarchy of equations for MDP by Kirkwood's superposition approximation results in $u_0 = 0.5$ (Dettmann *et al* 1967, Kotomin and Kuzovkov 1981), whereas summation of an infinite series for the MPD has led Dettmann (1965) to the result $u_0 = \ln 2 \approx 0.69$ †. Both results are independent of the space dimensionality and contradict computer simulations and other analytical estimates where $u_0 > 1$ is obtained as a rule (see table 1). (The shortcomings of these theories are discussed below.)

On the other hand, starting from Pooley (1966) and Hughes and Pooley (1971) several attempts have been made to develop a simple phenomenological theory of radiation-induced defect accumulation based, in fact, purely on combinatorial considerations (also see Antonov-Romanovskii 1981, 1983, Vinetsky and Kondrachuk 1981, Vinetsky 1983).

Although such a semiquantitative approach was useful in understanding the aggregation effect as a whole, it *fails* in fact to describe adequately the spatial defect correlations since one-particle defect densities (concentrations) only are employed here,

† The greater the initial correlation within geminate Frenkel pairs, the less is the saturation concentration c_0 (Dettmann *et al* 1967, Kotomin and Kuzovkov 1981).

which necessitates use of *a priori* assumptions. In particular, an inconsistent incorporation of the aggregation effect has led Hughes and Pooley (1971) to an erroneous conclusion about an *infinite* increase in the defect concentration which would occur unless a back reaction is involved.

The purpose of this paper is the further development of the rigorous analytical approach (Dettmann 1965, Kuzovkov and Kotomin 1981, Kotomin and Kuzovkov 1981, 1983), starting with a hierarchy of equations for the MPD's. The time development of the spatial distributions of defects describing aggregates and their effect on the accumulation curve $c(t)$ are analysed for 1D and 3D cases.

2. The basic equations

Let us introduce the $(n + n')$ -MPDs of defects

$$\rho_{n,n'}(\{\mathbf{r}\}_n; \{\mathbf{r}'\}_{n'}) \quad \{\mathbf{r}\}_n \equiv \mathbf{r}_1, \dots, \mathbf{r}_n$$

yielding a mean number of configurations containing n A defects in volumes $d\mathbf{r}_i$ at \mathbf{r}_i and n' B defects in volumes $d\mathbf{r}_j$ at \mathbf{r}_j ($j = 1, \dots, n'$).

In order to solve the problem analytically, we confine ourselves to the continuous approximation, uncorrelated defects within geminate pairs and instant annihilation. Therefore, a set of equations describing the accumulation kinetics is (Dettmann 1965, Kotomin and Kuzovkov 1983)

$$\begin{aligned} \frac{\partial \rho_{n,n'}}{\partial t} = & p \sum_{i=1}^n \rho_{n-1,n'}(\{\mathbf{r}\}_n^i; \{\mathbf{r}'\}_{n'}) + p \sum_{j=1}^{n'} \rho_{n,n'-1}(\{\mathbf{r}\}_n; \{\mathbf{r}'\}_{n'}^j) \\ & - \sum_{i=1}^n \sum_{j=1}^{n'} \sigma(\mathbf{r}_i - \mathbf{r}'_j) \rho_{n,n'} - \sum_{i=1}^n \int \sigma(\mathbf{r}_i - \mathbf{r}'_{n'+1}) d\mathbf{r}'_{n'+1} \\ & - \sum_{j=1}^{n'} \int \sigma(\mathbf{r}_{n+1} - \mathbf{r}'_j) \rho_{n+1,n'} d\mathbf{r}_{n+1}. \end{aligned} \quad (1)$$

Here $\{\mathbf{r}\}_n^i$ denotes a set of vectors $\{\mathbf{r}\}_n$ with omitted \mathbf{r}_i , p defect production rate and $\sigma(r) = \sigma(|\mathbf{r}|)$ is the annihilation probability of a pair per unit time. We shall take the latter to be in the form of the (Heaviside) step function: $\sigma(r) = \sigma_a \theta(r_0 - r)$ with $\sigma_a \rightarrow \infty$ describing an *instant* annihilation of a vacancy and an interstitial nearby (cf Dettmann 1965). The MPDs reveal the following symmetry property

$$\rho_{n,n'}(\{\mathbf{r}\}_n; \{\mathbf{r}'\}_{n'}) \equiv \rho_{n',n}(\{\mathbf{r}'\}_{n'}; \{\mathbf{r}\}_n) \quad (2)$$

provided defects were absent before irradiation. Due to a spatial homogeneity one of the coordinates in a set of vectors $\{\mathbf{r}\}_n, \{\mathbf{r}'\}_{n'}$ enters only in the form of a difference with the other coordinates, the one-point densities (macroscopic defect concentrations of A and B) $\rho_{1,0} = \rho_{0,1} = c$ (similar for $\rho_{2,0}, \rho_{0,2}$) are independent of \mathbf{r} and the joint densities of dissimilar defects $\rho_{1,1}(\mathbf{r}_1; \mathbf{r}_2)$ (the averaged products of the macroscopic densities taken at \mathbf{r}_1 and \mathbf{r}_2) depend on $|\mathbf{r}_1 - \mathbf{r}_2|$ only.

Let us clarify the disadvantages of the earlier analytical theories. The main approximation made by Dettmann (1965), in an attempt to obtain an analytical solution, neglected the spatial correlation of similar defects $\rho_{n,0} = c^n$. Therefore, the dimensionless saturation concentration obtained $u_0 = \ln 2$ is the *lower* bound estimate since the aggregation effect is neglected here. On the other hand, it contradicts the behaviour of

the joint densities of similar defects, $\rho_{2,0}$, calculated by Kotomin and Kuzovkov (1981), as well as computer simulations (e.g. Kalnin and Krikis 1983) where $\rho_{2,0} \gg c^2$ is observed at $|r_2 - r_1| \approx r_0$ clearly demonstrating the importance of correlation effects ($\rho_{2,0} = c^2$ is the random (Poisson) distribution). The standard Kirkwood superposition approximation for three-point densities

$$\rho_{2,1} \approx \rho_{1,1}(r_1; r'_1) \rho_{1,1}(r_2; r'_1) \rho_{2,0}(r_1, r_2;)/c^3 \tag{3}$$

employed by Dettmann *et al* (1967), Kotomin and Kuzovkov (1981), Kuzovkov and Kotomin (1981) fails at small distances where $|r_1 - r'_1| < r_0, |r_2 - r'_1| < r_0$, since if $\sigma_a^{-1} \rightarrow 0, \sigma_a \rho_{2,1}$ has order of magnitude σ_a^{-1} instead of an exact order $(\sigma_a)^0$ which has been shown quite recently by Kuzovkov and Kotomin (1983). This has led directly to a cut-off in the infinite functional series (derived for the first time by Dettmann (1965)) and a very low estimate $\bar{n}_0 = 0.5$ exhibiting the shortcomings of the standard superposition approximation for accumulation kinetics.

Let us return now to the set (1). For an instant annihilation at $|r_i - r'_i| \leq r_0 \rho_{n,1}$ tends to zero, but

$$\lim_{\sigma_a \rightarrow \infty} \sigma_a \rho_{n,1} = g_n(\{r\}_n; r'_1) \tag{4}$$

remains at a constant value (Kuzovkov and Kotomin 1983). The first equation in (1) can be rewritten as

$$\frac{\partial c}{\partial t} = p - \int \sigma(r_1 - r'_1) \rho_{1,1} dr_1 = p - v_0 \int w(r_1 - r'_1) g_1 dr_1 \tag{5}$$

where $w(r) = \sigma(r)/\sigma_a v_0, v_0$ is the annihilation volume ($= 2v_0, \pi r_0^2, \frac{4}{3}\pi r_0^3$ for 1D, 2D and 3D respectively). To solve equation (5) determining the accumulation curve $c(t)$ the joint density $\rho_{1,1}$ at $|r_1 - r'_1| < r_0$ has obviously to be known.

The second equation of the set (1) for the r_1, r'_1 just mentioned and $n = 1$ is:

$$\lim_{\sigma_a \rightarrow \infty} \sigma_a^{-1} \frac{\partial g_1}{\partial t} = 0 = 2pc - g_1 - 2v_0 \int w(r_2 - r_1) g_2 dr_2. \tag{6}$$

Hereafter time derivatives should be omitted if $\sigma_a \rightarrow \infty$ which results in degenerate equations. The function $g_n, n \geq 2$ can be obtained from the following chain of equations

$$p \rho_{n,0} - n g_n - v_0 \int w(r_{n+1} - r'_1) g_{n+1} dr_{n+1} = 0. \tag{7}$$

The set of equations (5) to (7) is asymptotically exact, since in deriving equation (7) all terms which vanish as $\sigma_a \rightarrow \infty$ were omitted. The formally exact solutions of equations (6) and (7) are:

$$g_1 = 2p \left(c - \sum_{n=2}^{\infty} \frac{(-1)^n v_0^{n-1}}{n!} \int \dots \int \rho_{n,0} \prod_{i=2}^n w(r_i - r'_1) dr_2 \dots dr_n \right) \tag{8}$$

$$g_2 = p \left(\sum_{n=3}^{\infty} \frac{(-1)^n v_0^{n-2}}{n!} \int \dots \int \rho_{n,0} \prod_{i=3}^n w(r_i - r'_1) dr_3 \dots dr_n + \frac{1}{2} \rho_{2,0} \right). \tag{9}$$

In the superposition approximation $g_2 = 0$ and in equation (8) only the first term in the brackets remains. In its turn, substitution of equation (8) into equation (5) leads to the asymptotically exact equation (derived for the first time by Dettmann (1965)).

Note that the MPDS $\rho_{n,0}$ with $n = 1$ to ∞ enter only in equations (8) and (9) which describe similar defect spatial correlations. Let us present these $\rho_{n,0}$ in the form of the expansion in irreducible correlation functions ν_n well known in statistical physics (Balescu 1975)

$$\begin{aligned} \rho_{2,0} &= c^2(1 + \nu_2|r_1 - r_2|) \quad \nu_2(r \rightarrow \infty) \rightarrow 0 \\ \rho_{3,0} &= c^3(1 + \nu_2(|r_1 - r_2|) + \nu_2(|r_2 - r_3|) + \nu_2(|r_1 - r_3|) + \nu_3(r_1, r_2, r_3)) \end{aligned} \quad (10)$$

etc. The result for the lower bound, $u_0 = \ln 2$, found by Dettmann (1965) corresponds to the neglect in equations (5) and (8) of *all* the correlation functions ν_i . A better approximation involves the incorporation of the joint correlation functions $\nu \equiv \nu_2$, i.e. based on the approximation linear in ν :

$$\rho_{n,0} \approx c^n \left(1 + \sum_{i < k} \nu(|r_i - r_k|) \right). \quad (11)$$

When equation (8) is calculated by making use of equation (11) and substituting it into equation (5), we arrive at

$$\frac{\partial u}{\partial t'} = \exp(-u)(1 + \frac{1}{2}u^2\bar{\nu}) - \frac{1}{2}, \quad t' = 2pv_0t, \quad u = cv_0, \quad u(0) = 0 \quad (12)$$

$$\bar{\nu} = \int w(r_1)w(r_2)\nu(|r_1 - r_2|) dr_1 dr_2 = \frac{1}{v_0} \int S(r)\nu(r) dr. \quad (13)$$

$S(r)$ in equation (13) is the share of the common volume of two intersecting spheres of diameter $2r_0$ with centres separated by a distance r . Unlike earlier theories (e.g. Hughes and Pooley 1971), our equation (12) determining the accumulation curve is *not* closed since it is coupled with the correlation function of similar defects ν . At saturation parameter

$$\bar{\nu} = [\exp(-u_0) - 2]/u_0^2 > 0 \quad (14)$$

and $\bar{u}_0 > \ln 2$. This value of $\bar{\nu}$ permits a simple interpretation. As is well known the joint densities $\rho_{2,0}$ describe fluctuations in A(B) defect concentrations (Kotomin and Kuzovkov 1983). It can be shown that the fluctuation of a number N of defects A(B) in volume v_0 can be characterised by a dispersion

$$\overline{N^2} - \bar{N}^2 = u + u^2\bar{\nu}. \quad (15)$$

The second term in equation (15) demonstrates the non-Poisson fluctuation spectrum (for which $\overline{N^2} - \bar{N}^2 = \bar{N}$ holds) with $\bar{\nu}$ as a measure of this deviation.

One can see that $\bar{\nu}$ (equation (13)) and thus the accumulation curve (equation (12)) must be different for the 1D and 3D cases, a situation which has rarely been observed in computer simulations in earlier theories (see, however, Antonov-Romanovskii 1983).

The calculation of $\bar{\nu}$ necessitates knowledge of the joint density $\rho_{2,0}$ (see equation (10)) for which we have

$$\frac{\partial \rho_{2,0}}{\partial t} = 2pc - 2 \int \sigma(r_1 - r'_1)\rho_{2,1} dr'_1. \quad (16)$$

The integration region in equation (16) consists of two domains. For the first one $|r_2 - r'_1| \leq r_0$ and $\sigma_a g_{2,1} \equiv g_2$; the equation (9) for the latter can be solved employing the approximation (11). For the second domain where $|r_2 - r'_1| > r_0$ the superposition approximation can be used directly. Here we also use the definition (4) which permits

us to substitute $g_1(r_1, r_1)$ for $\sigma_a \rho_{1,1}(r_1; r'_1)$. Introducing the correlation function for dissimilar defects

$$\eta(|r_1 - r'_1|) = \lim_{\sigma_a \rightarrow \infty} \rho_{1,1}/c^2 \quad \eta(r \rightarrow \infty) \rightarrow 1 \tag{17}$$

for which $\eta \equiv 0$ if $|r_1 - r'_1| \leq r_0$, we arrive finally at a basic kinetic equation describing approximately correlations of similar defects of all orders

$$\begin{aligned} \partial \nu(r)/\partial t = & Q_1 - Q_2(r) - S(r)(F_1 + F_2 \nu(r)) - F_3 I(r) \\ & - (1 + \nu(r))(G_1 K(r) + G_2 L(r)) \end{aligned} \tag{18}$$

where

$$\begin{aligned} Q_1 &= (2/u)[1 - \exp(-u)(1 + \frac{1}{2}u^2 \bar{\nu})] \\ Q_2 &= (2/u)[\exp(-u)(1 + \frac{1}{2}u^2 \bar{\nu}) - \frac{1}{2}] \\ F_1 &= (1/u^2)[\exp(-u) - 1 + u + 3\bar{\nu}][\exp(-u)(1 + \frac{2}{3}u + \frac{1}{6}u^2) - 1 + \frac{1}{3}u] \\ F_2 &= (1/u^2)[\exp(-u) - 1 + u] \\ F_3 &= (4/u^2)[1 - \frac{1}{2}u - \exp(-u)(1 + \frac{1}{2}u)] \\ G_1 &= (2/u)[1 - \exp(-u) + \bar{\nu}(1 + u + \frac{1}{2}u^2)] \\ G_2 &= (2/u)[\exp(-u)(1 + u) - 1] \end{aligned} \tag{19}$$

and

$$\begin{aligned} I(r) &= \nu_0(\omega^* \omega \mathcal{F}) & \mathcal{F}(r) &= (\omega^* \nu) \\ K(r) &= (\omega^* \eta) & L(r) &= (\omega \mathcal{F}^* \eta) \\ \omega &= \begin{cases} 0, & r > r_0, \\ \nu_0^{-1}, & r \leq r_0. \end{cases} \end{aligned} \tag{20}$$

In equation (20) the definition of the spatial convolution

$$(X^* Y) \equiv \int X(r') Y(r - r') dr' \tag{21}$$

has been used. The initial condition for the equation (18) is $\nu(r, 0) = 0$. To cut off the hierarchy of equation the dissimilar joint density $\rho_{1,1}$ at $|r_1 - r'_1| < r_0$ should be also considered

$$\frac{\partial \rho_{1,1}}{\partial t} = 2pc - 2 \int \alpha(r_2 - r'_1) \rho_{2,1} dr'_1. \tag{22}$$

Since there are no restrictions on the use of the superposition approximation here we obtain easily (for $r \geq r_0$)

$$\frac{\partial \eta(r)}{\partial t} = \frac{1}{u} (1 - \eta(r) - \eta(r'))(G_1 \mathcal{F}(r) + G_2 M(r)) \tag{23}$$

with $M(r) = (\omega \mathcal{F} * \nu)$. A random initial distribution within geminate Frenkel pairs is given by $\eta(r \geq r_0) \equiv 1$.

The conclusion can be drawn from equations (12), (18) and (23) that the accumulation curve $\bar{n}(t)$ is coupled with the radiation-induced fluctuations of *similar* defects (determining their aggregates) which in turn are coupled with the joint correlation functions of dissimilar defects. The spatial correlations of similar and dissimilar defects are *back-coupled* (Kuzovkov and Kotomin 1981, Kotomin and Kuzovkov 1983), since defect annihilation decreases the concentration of dissimilar defects and stimulates simultaneously the aggregation of *similar* defects. These spatial inhomogeneities of defect distribution affect directly the accumulation kinetics. The incorrectness of earlier attempts to guess the closed kinetic equation for $c(t)$ without any analysis of the spatial correlations referred to is quite obvious now. It should be stressed in conclusion of this section that even for a very simplified model we could take into account similar defect correlations of all orders only approximately and thus obtain rather complicated equations which do not permit an analytical solution.

3. Results

Figure 1 obtained by computer calculations using equations (12), (18) and (23) shows the accumulation kinetics of immobile Frenkel defects. One can see that: (i) the concentration saturation is reached at high irradiation doses, (ii) its magnitude is higher for

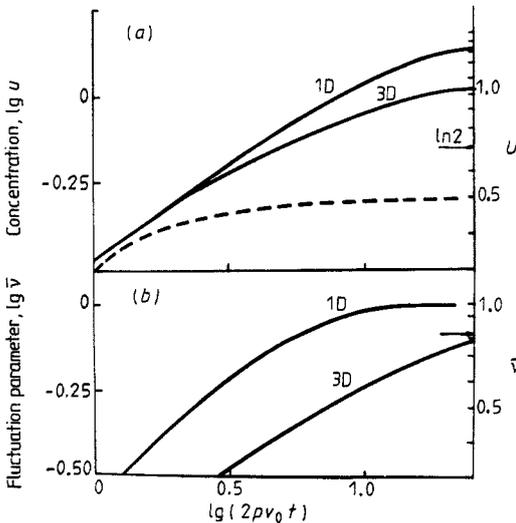


Figure 1. (a) The dimensionless defect concentration plotted against dimensionless irradiation dose. The accumulation kinetics are calculated for the 1D and 3D cases. The broken curve shows the result of the standard superposition approximation (3). An arrow indicates the saturation level obtained by Dettmann (1965) if the aggregation effect is neglected. (b) The fluctuation parameter $\bar{\nu}$ (equation (13)) characterising the deviation from the Poisson (random) distribution. An arrow indicates the saturation level for the 3D case (0.84).

the 1D case ($u_0 = 1.36$) than for the 3D case ($u_0 = 1.084$)†. This is in agreement with computer simulations (see also table 1) and the behaviour of the fluctuation parameter

† Due to the approximation involved (11) these figures are underestimated. At any rate, the aggregation enlarges the saturation concentration by a factor of at least 1.5–2.

$\bar{\nu}$ (equation (13)): in the 1D case it considerably exceeds that for the 3D case. At any rate, the accumulation curve at high irradiation doses deviates greatly from the superposition approximation (see figure 1(a)) and thus can not be described by the corresponding simple relation

$$c(t) = c_0[1 - \exp(-2\rho\nu_0t)]$$

widely used by experimentalists (e.g. Kalnin and Krikis 1983, see also Kalnin and Kotomin 1984), despite this being the *only* recombination mechanism limiting the accumulation process. It demonstrates once more that the successful expansion of the accumulation curve in several exponentials (stages)

$$c(t) = c_0 \left(1 - \sum_i \exp(-a_i t) \right)$$

does *not* necessarily mean that several *mechanisms* are involved.

The joint correlation functions plotted in figure 2 clearly demonstrate how in a course of an irradiation dynamical similar defect aggregates are formed. Their distinctive sizes (about $2r_0$) are determined by an interval at small r values where ν exceeds the Poisson (asymptotic) value. A concentration of dimer defects (e.g. F_2 centres in alkali halides) by a factor $\nu(r \rightarrow 0) + 1 \approx 3$ exceeds the value predicted for the Poisson distribution which is in qualitative agreement with experiments by Faraday and Compton (1965).

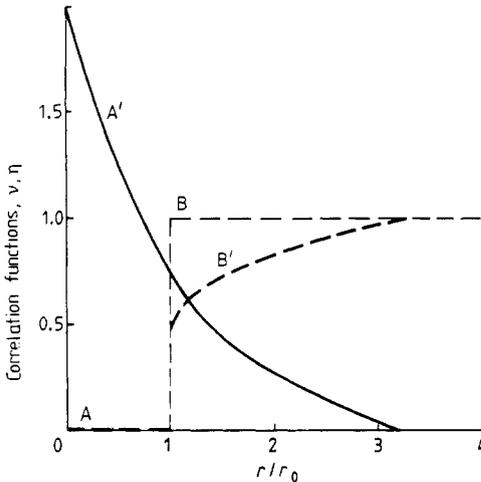


Figure 2. The joint correlation functions for similar (ν) and dissimilar (η) defects. A, $\nu(t = 0)$; B, $\eta(t = 0)$ for 1D saturation level; A', $\nu(t = 0)$; B', $\eta(t = 0)$ for 3D saturation level.

4. Discussion

The theory developed above has demonstrated that the analytical description of the accumulation kinetics and low-temperature radiation-induced aggregation of similar defects requires a *rigorous* analysis of the infinite hierarchy of the equations for the correlation functions for similar and dissimilar defects. The spatial correlations of similar and dissimilar defects are back-coupled which is the common effect in the theory of bimolecular reactions between defects (Kuzovkov and Kotomin 1981, Kotomin and Kuzovkov 1981, 1983).

The aggregation effect has statistical character (see e.g. Tăle *et al* 1975) and is observed at relatively high doses (half of the saturation concentration). Computer simulations by Pirogov and Ekmanis (1981) (see also Lanore 1974) argue that the aggregation also occurs at temperatures when defects are mobile even if similar defects *do not* interact (otherwise their short-range attraction will produce much denser and larger colloids, e.g. Hobbs *et al* (1973), Pirogov and Ekmanis (1981)). However a *correct* analysis of this kinetics is too complicated (a very simple estimate has been made by Hughes and Pooley 1971). This is the more so because the experimental structureless accumulation curves are not sufficiently informative to obtain reliably several parameters (r_0 , p , the initial distribution within Frenkel pairs) (see e.g. Guillot and Nouailhat 1976, Guillot *et al* 1979, 1980, Kalnin and Kotomin 1984). The other point is that in the simple model presented processes such as the formation of other radiation defects (F^+ , I , V_k in alkali halides) or colloids, dislocation loops and cascades of defects etc are neglected. For instance, tunnelling recombination (recharge) of close electron and hole defects is also neglected here. This mechanism is operative in many insulators, e.g. for primary F and H centres in alkali halides (Kondo *et al* 1972, Lushchik *et al* 1982, Shluger *et al* 1982, Tajirov 1983, Gavrilov *et al* 1983). It could be included phenomenologically in equations (1) through $\sigma(r) = \sigma_0 \exp(-r/r_B)$ (Tăle *et al* 1975), σ_0 and r_B are constants exhibiting a wide spectrum of lifetimes $\sigma(r)^{-1}$, instead of the step function with zero lifetime for $r \leq r_0$. Computer simulations incorporating tunnelling recombination of primary Frenkel defects have been undertaken by Tăle *et al* (1975), Kotomin and Valdats (1977), which have also confirmed the aggregation process. The new effect observed is that the accumulation kinetics also depends here on an irradiation *intensity* p : the greater intensity, the greater the saturation concentration[†]. Indeed, this effect, having been observed in alkali halides (Hughes and Pooley 1971), organic compounds (Mikhailov 1971) and glasses (Vojevodsky 1970), serves as a qualitative indication of tunnelling recombination. Another distinctive feature of the tunnelling recombination is a large effective radius (Tăle *et al* 1975) with an order of magnitude exceeding that for annihilation (typically about 200 lattice sites). An estimate by Hughes and Pooley (1971) for an F centre in alkali halides—3000 lattice sites—is in good agreement with the idea that the tunnelling recombination is efficient here.

To our opinion, more valuable information about Frenkel defect interaction, recombination and their initial spatial distribution is available from the *temperature dependence* of the accumulation efficiency (e.g. Kotomin and Fabrikant 1980).

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[†] This is associated with a *reduction* of the aggregation effect (Millers 1976). An aggregation is the more pronounced the greater is the total number of defects produced for a given accumulated concentration.

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