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DIFFUSION IN FLUCTUATIVE MEDIUM

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DIFFUSION IN FLUCTUATIVE MEDIUM

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We consider diffusion in a medium with randomly fluctuating diffusion coefficient. An equation is derived for diffusate concentration averaged over dimensions exceeding those of fluctuations. In the case when fluctuations are weak and their correlation scales small the equation is reduced to a differential one, where the effective diffusion coefficient is always lower than the average one. A stability criterion for uniform distribution of diffusate is derived. Some implications of the theory for the radiation effects in solids are discussed.

Key words: diffusion, solids, fluctuative medium, nonlinear effects, irradiation-induced instability.

1 INTRODUCTION

There is considerable interest in various nonequilibrium phenomena in solids, e.g. phase transitions,[1] development of ordered defect structures,[2] radiation damage,[3] etc. A general mechanism of evolution of nonequilibrium disturbances in solid is the diffusion mechanism. Diffusion fluxes arising in nonequilibrium medium are proportional to the diffusion coefficient D which depends on the medium properties. Usually, it is assumed that D is constant or its dependence of time and space is known. However, in reality the medium properties often undergo numerous random fluctuations from the basic state, e.g. the fluctuations in temperature and concentration of point defects, induced by irradiation. If the fluctuation scales are macroscopic then the diffusion coefficient can be defined locally, but it is a random function of time and position. We are interested in the concentration $\langle C\rangle$ averaged over the scales that are considerably larger than those of fluctuations. We derive an integro-differential equation describing the evolution of the averaged concentration, and show that this equation can be reduced to a differential one in the case of sufficiently small fluctuation amplitudes and correlation scales. The effect of fluctuations results in the appearance of an additional term $\sim\Delta^2\langle C\rangle$ and in that an effective diffusion coefficient is always lower than the average one due to nonlinear interaction between the fluctuations in diffusivity and concentration.

We also derive a criterion of stability of the uniform concentration $\langle C\rangle=\text{const}$ and discuss some fluctuation-induced effects in radiation environment.

2 FORMULATION OF THE PROBLEM

Fluctuations are assumed to be macroscopic, so that a diffusate concentration $C(x, t)$ at point x and time t satisfies the usual diffusion equation (for simplicity we consider one-dimensional case)

$$\frac{\partial C}{\partial t}=\frac{\partial}{\partial x}D(x,t)\frac{\partial C}{\partial x}, \tag{1}$$

where $D(x, t)$ is the random function. Consequently, $C(x, t)$ is also a random function.

We are interested in the evolution of the concentration averaged over scales that are larger than those of fluctuations but less than the dimensions of a whole system under consideration.

To perform the averaging let us describe a random function $D(x, t)$ by its correlation moments:

$$\langle D\rangle = D(x, t) - \tilde{D}(x, t), \tag{2}$$

$$\langle \tilde{D}(x, t)\tilde{D}(x', t')\rangle, \text{ etc.,} \tag{3}$$

where $\langle\ldots\rangle$ is the average over an ensemble of realizations of $D(x, t)$, $\tilde{D}$ is the deviation from the average value $\langle D\rangle$. Note that explicit forms of these moments depend on the origin of the fluctuations, while we are interested here in general properties independent of the specific mechanisms of the fluctuation generation. Accordingly, we assume the moments to be given in the general form.

3 DELTA-CORRELATED IN TIME FLUCTUATIONS

Let us consider first the simplest case assuming $D(x, t)$ to be Gaussian, statistically stationary, spatially homogeneous and, in addition, delta-correlated in time. The first assumption is of a general nature according to a central limit theorem[4] which states that the sum of a large number of sufficiently small independent random functions is Gaussian irrespective of statistical properties of each function. In this case $D(x, t)$ is completely described by its two moments $\langle D\rangle$ and $\langle D(x, t), D(x', t')\rangle$. The next two requirements mean that correlations do not depend on x and t but rather on the differences $x-x'$ and $t-t'$. The delta-correlation in time implies that the characteristic time of fluctuation correlations τ_c is negligibly small as compared to the characteristic time T of $\langle C\rangle$ variation:[5] $\tau_c/T\to 0$. Then we have

$$\langle \tilde{D}(x, t), \tilde{D}(x', t')\rangle = d(x-x')\delta(t-t'), \tag{4}$$

where $d(x-x')$ is the spatial part of the correlation function.

Performing the formal averaging of Eq. (1) we obtain

$$\frac{\partial\langle C\rangle}{\partial t} = \langle D\rangle \frac{\partial^2\langle C\rangle}{\partial x^2} + \frac{\partial}{\partial x}\left\langle \tilde{D}(x, t)\,\frac{\partial C(x, t)}{\partial x}\right\rangle. \tag{5}$$

This equation contains the unknown correlation moment $\langle\tilde{D}(\partial C/\partial x)\rangle$. One can obtain an equation for $\langle\tilde{D}(\partial C/\partial x)\rangle$ multiplying Eq. (1) by $\tilde{D}$ and performing the averaging. Yet, this equation will contain an unknown correlation moment of higher order, etc. Thus, we arrive at the infinite hierarchy of correlation equation that results from nonlinear $C(x, t)$ dependence on $D(x, t)$. This difficulty can be solved using the following relation:[5]

$$\langle Z(x, t)F[Z]\rangle = \iint \langle Z(x, t)Z(x', t')\rangle \left\langle \frac{\delta F[Z]}{\delta Z(x', t')}\right\rangle dt'dx', \tag{6}$$

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where $Z(x, t)$ is the Gaussian random process, $F[Z]$ is the functional of Z, $\delta/\delta Z$ is the variational derivative. The integration is carried out over the whole range of x' and t'.

After substituting $D(x, t)$ for $Z(x, t)$, $\partial C(x, t)/\partial x$ for $F[Z]$ and integrating over t', Eq. (6) acquires the following form:

$$\left\langle \tilde{D}(x,t)\frac{\partial C(x,t)}{\partial x}\right\rangle = \int_{-\infty}^{+\infty} d(x-x')\frac{\partial}{\partial x}\left\langle \frac{\delta C(x,t)}{\delta \tilde{D}(x',t)}\right\rangle dx'. \tag{7}$$

With account of Eq. (1), the variational derivative is given by

$$\frac{\delta C(x,t)}{\delta \tilde{D}(x',t)} = \frac{\partial}{\partial x}\left(\delta(x-x')\frac{\partial C(x,t)}{\partial x}\right). \tag{8}$$

Substituting it in Eq. (5) and integrating by parts we obtain the expression

$$\left\langle \tilde{D}(x,t)\frac{\partial C(x,t)}{\partial x}\right\rangle = d(0)\frac{\partial^3 C}{\partial x^3} + 2\left.\frac{\partial d(x-x')}{\partial x}\right|_{x'=x}\cdot\frac{\partial^2 C}{\partial x^2} + \frac{\partial\langle C\rangle}{\partial x}\left.\frac{\partial^2 d(x-x')}{\partial x^2}\right|_{x'=x}. \tag{9}$$

Note that $d(x-x')$ acquires its maximal value at $x=x'$, which implies that

$$\left.\frac{\partial}{\partial x}d(x-x')\right|_{x'=x} = 0, \quad \left.\frac{\partial^2 d(x-x')}{\partial x^2}\right|_{x'=x} < 0, \tag{10}$$

Therefore, substituting Eq. (9) into Eq. (5) we obtain the following closed equation for $\langle C\rangle$:

$$\frac{\partial C}{\partial t} = D^*\frac{\partial^2\langle C\rangle}{\partial x^2} + d(0)\frac{\partial^4\langle C\rangle}{\partial x^4} \tag{11}$$

$$D^* = \langle D\rangle + \left.\frac{\partial^2 d(x-x')}{\partial x^2}\right|_{x'=x} \tag{12}$$

where D^* is the effective diffusion coefficient which is always lower than the average one $\langle D\rangle$. Furthermore, fluctuations result in the appearance of an additional term $d(0)\ d^4\langle C\rangle/dx^4$. Note that the fourth spatial derivative of $C(x, t)$ arises also in the diffusion equation describing the phase segregation.[1] This implies the possibility of destabilization of a homogeneous distribution ($\langle C\rangle$ = const) due to fluctuations. The instability may arise also when D^* becomes negative, i.e. if

$$\langle D\rangle < -\left.\frac{{}^2 d(x-x')}{x^2}\right|_{x'=x}. \tag{13}$$

This criterion may be expressed in terms of the correlation scales taking into account that $d(x-x') \sim \langle \tilde{D}^2\rangle\tau_c$, $-\partial/\partial x^2 \sim l_c^{-2}$ where l_c and τ_c are the correlation length and time, respectively.

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An upper estimate of the fluctuation amplitude is $\langle\tilde{D}^2\rangle \sim \langle D\rangle^2$ which gives the instability criterion as $\langle D\rangle > l_c^2/\tau_c$.

Note that the above procedure is also applicable when $D(x, t)$ is not Gaussian provided only that it is delta-correlated in time. Then instead of relation (6) one should use its generalized version.[5] As a result, Eq. (11) acquires, in addition to the terms shown above, an infinite series of correlation moments multiplied by the higher order derivatives of $\langle C\rangle$. However, in the case of sufficiently small fluctuations these additional terms are smaller than the shown ones and may be neglected. Moreover, in this case the analysis can be extended beyond the delta-correlated in time approximation. We shall do it in the following section.

4 WEAK FLUCTUATION APPROXIMATION

We assume here that

$$D(x, t) = \langle D\rangle + \tilde{D}, \quad \tilde{D} \ll \langle D\rangle, \tag{14}$$

$$C(x, t) = \langle C\rangle + \tilde{C}, \quad \tilde{C} \ll \langle C\rangle \tag{15}$$

$$l_c, l_f \ll L; \quad \tau_c, \tau_f \ll T; \quad \frac{|\tilde{C}|}{\langle C\rangle} \cdot \frac{L}{l_f} \ll 1 \tag{16}$$

$$\langle\tilde{D}(x, t)\,\tilde{D}(x', t')\rangle = B(x - x', t - t'), \tag{17}$$

where l_f and τ_f are the fluctuation characteristic length and time, while L and T are the characteristic length and time of $\langle C\rangle$ variation, respectively. Substituting Eqs. (14), (15) into Eq. (1) we obtain

$$\frac{\partial\langle C\rangle}{\partial t} + \frac{\partial\tilde{C}}{\partial t} = \frac{\partial}{\partial x}\left[(\langle D\rangle + \tilde{D})\frac{\partial(\langle C\rangle + \tilde{C})}{\partial x}\right]. \tag{18}$$

Averaging of Eq. (1) with account of $\langle\tilde{D}\rangle = \langle\tilde{C}\rangle = 0$ gives

$$\frac{\partial\langle C\rangle}{\partial t} = \langle D\rangle\frac{\partial^2\langle C\rangle}{\partial x^2} + \frac{\partial}{\partial x}\left\langle\tilde{D}\frac{\partial\tilde{C}}{\partial x}\right\rangle. \tag{19}$$

Subtracting Eq. (19) from Eq. (18) and neglecting second-order terms in $\tilde{D}/\langle D\rangle \ll 1$, $|\tilde{C}|L/\langle C\rangle l_f \ll 1$ we obtain the equation for $\tilde{C}$

$$\frac{\partial\tilde{C}}{\partial t} = \langle D\rangle\frac{\partial^2\tilde{C}}{\partial x^2} + \frac{\partial}{\partial x}\tilde{D}\frac{\partial}{\partial x}\langle C\rangle. \tag{20}$$

The standard solution of Eq. (20) is given by

$$\tilde{C}(x, t) = \int_0^t dt' \int_{-\infty}^{+\infty} dx'\, G(x - x', t - t')\frac{\partial}{\partial x'}\tilde{D}(x', t')\frac{\partial\langle C(x', t')\rangle}{\partial x'}, \tag{21}$$

where $G(x-x', t-t')$ is the Green's function of the diffusion equation. Substituting Eq. (21) in Eq. (19) we obtain a closed integro-differential equation for $\langle C\rangle$

$$\frac{\partial\langle C\rangle}{\partial t}-\langle D\rangle\frac{\partial^2\langle C\rangle}{\partial x^2}=\frac{\partial^2}{\partial x^2}\int_0^t dt'\int_{-\infty}^{+\infty}dx'\,B(x-x', t-t')\langle C(x', t')\rangle \times\frac{\partial^2}{\partial x^2}G(x-x', t-t'). \tag{22}$$

Equation (22) coincides with Eq. (11) when $D(x, t)$ is delta-correlated in time. Thus, nonlocal character of Eq. (22) stems from the finitness of the correlation time $\tau_c\neq 0$. However, owing to the presence of small parameters (16) it is possible to reduce Eq. (22) to a differential equation. Indeed, the main contribution to the right-hand side of Eq. (22) results from the integration over the correlation domain $l_c\cdot\tau_c$ since $B(x-x', t-t')$ diminishes sharply beyond these limits. On the other hand. $\langle C(x, t)\rangle$ varies insignificantly in these limits and may be expanded in Taylor series near x, t:

$$\langle C(X-\xi, t-\theta)=\langle C(x, t)\rangle-\xi\frac{\partial\langle C(x, t)\rangle}{\partial x}-\theta\frac{\partial\langle C(x, t)\rangle}{\partial t}+\dots$$

Substituting this expansion into Eq. (22) we obtain, to the second order in l_c L, τ_c T both small, the following differential equation for $\langle C\rangle$:

$$\frac{\partial\langle C\rangle}{\partial\tau}=D^*\frac{\partial^2\langle C\rangle}{\partial x^2}+d\frac{\partial^4\langle C\rangle}{\partial x^4}-\alpha\frac{\partial^3\langle C\rangle}{\partial t\partial x^2}+\beta\frac{\partial^4\langle C\rangle}{\partial x^2\partial t^2}, \tag{23}$$

$$D^*=\langle D\rangle+\int_0^\infty dt\int_{-\infty}^{+\infty}dx\frac{\partial^2 G(x, t)}{\partial x^2}B(x, t), \tag{24}$$

$$d=\frac{1}{2}\int_0^\infty dt\int_{-\infty}^{+\infty}dx\frac{\partial^2 G(x, t)}{\partial x^2}B(x, t)x^2, \tag{25}$$

$$\alpha=\int_0^\infty dt\int_{-\infty}^{+\infty}dx\frac{\partial^2 G(x, t)}{\partial x^2}B(x, t)\cdot t, \tag{26}$$

$$\beta=\frac{1}{2}\int_0^\infty dt\int_{-\infty}^{+\infty}dx\frac{\partial^2 G(x, t)}{\partial x^2}B(x, t)\cdot t^2 \tag{27}$$

Comparison of Eqs. (23) and (11) shows that account of finitness of τ_c results in appearance of additional terms containing derivatives of $\langle C\rangle$ with respect to time while the effective diffusion coefficient D^* remains unaffected. This means that stationary diffusion processes ($\partial\langle C\rangle/\partial t=0$) are described by Eq. (11) irrespective of the τ_c value.

The coefficients D^*, d, α, and β are evaluated in the Appendix for an explicit form of the correlation

$$B(x, t)=B_0\exp\left(-\frac{x^2}{l_c^2}-\frac{t}{\tau_c}\right), \tag{28}$$

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which represents a homogeneous random process.[4] In principle, it is not the explicit form of $B(x, t)$ that matters but rather its sharp dependence on correlation scales l_c and τ_c which allows one to use the Laplace asymptotic method of integration. In this case D^* is given by (see Appendix)

$$D^* \approx \begin{cases} \langle D\rangle - B_0/\langle D\rangle, & \varepsilon \ll 1, \\ \langle D\rangle - 2B_0 \dfrac{\tau_c}{l_c^2}, & \varepsilon \gg 1, \end{cases} \quad \varepsilon = \frac{l_c}{2\sqrt{\langle D\rangle \tau_c}}, \tag{29}$$

whence it follows that D^* is always lower than $\langle D\rangle$ and the difference between them is the largest at $\varepsilon \gg 1$.

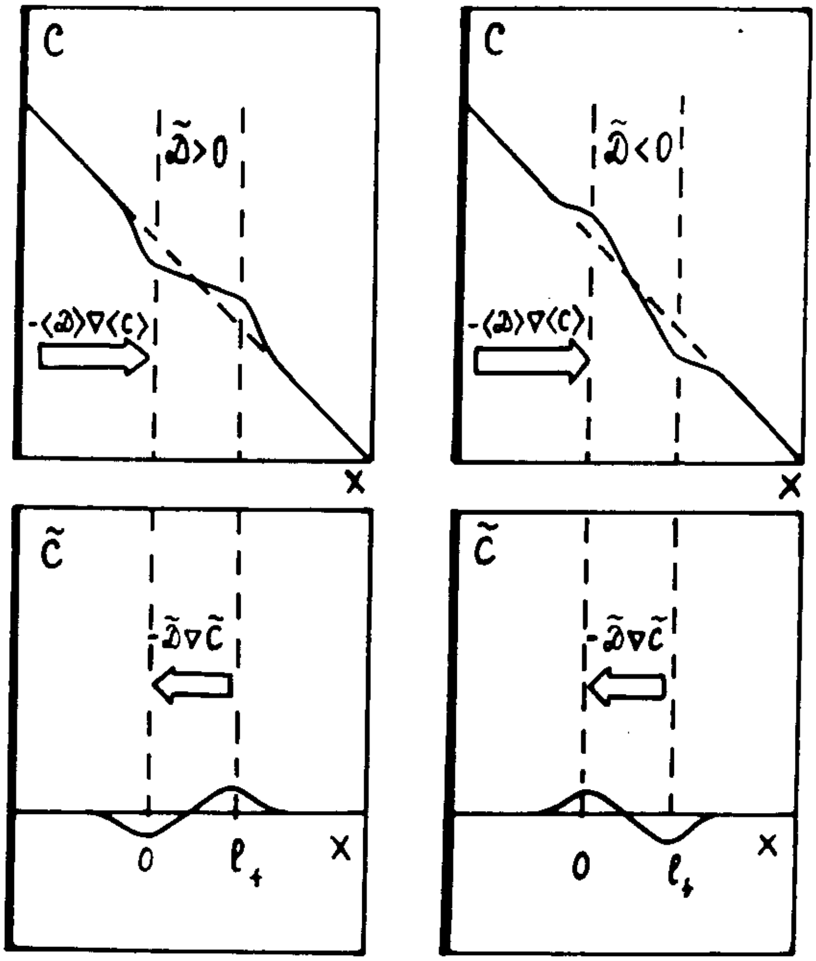

FIGURE 1 Qualitative illustration of the fluctuation-induced diffusivity reduction. In the upper panel the total concentration C (solid lines) deviates from $\langle C\rangle$ (dashed lines) due to the fluctuation of $D=\langle D\rangle+\tilde{D}$, $\tilde{D}\neq 0$ at $0<x<l_f$. The arrows indicate direction of the main flux $-\langle D\rangle\nabla\langle C\rangle$. In the lower panel the fluctuations $\tilde{C}=C-\langle C\rangle$ are depicted and the arrows indicate the direction of additional fluxes $-\tilde{D}\nabla\tilde{C}$.

Let us consider a physical reason of the diffusivity reduction induced by fluctuations. An explicit averaging of the total flux $\mathbf{j} = -D\nabla C$ lead to an obvious expression

$$\langle \mathbf{j} \rangle = -\langle D \rangle \nabla \langle C \rangle - \langle \tilde{D} \nabla \tilde{C} \rangle, \tag{30}$$

since linear fluctuation terms are cancelled by the averaging. Thus, we are interested in the direction of an additional flux $-\langle \tilde{D} \nabla \tilde{C} \rangle$. The direction of this flux becomes clear after inspection of Figure 1, where fluctuations C are shown schematically in two cases $\tilde{D} > 0$ and $\tilde{D} < 0$. Solid lines in the upper panel correspond to the exact concentration C deviating from $\langle C \rangle$ (shown by dashed lines) due to the fluctuations $\tilde{D}$. It can be seen that directions of $\nabla \tilde{C}$ within the fluctuation regions at $\tilde{D} < 0$ and $\tilde{D} > 0$ are different. However, both of the additional fluxes $-\tilde{D} \nabla \tilde{C}$ are directed against the main flux $-\langle D \rangle \nabla \langle C \rangle$ thus reducing the total flux. This is the physical reason of the diffusivity reduction.

5 STABILITY ANALYSIS

Let us consider evolution of the average concentration $\langle C(x, t) \rangle$. The general solution of Eq. (22) may be derived by carrying out the Fourier transformation in space and the Laplace transformation in time

$$C_{kp} = \frac{C_k(0)}{p + \langle D \rangle k^2 - k^2 \int_{-\infty}^{+\infty} k_1^2 \mathscr{L}(G(k_1, t)\, B(k - k_1, t))\, dk_1}, \tag{31}$$

where $C_k(0)$ is the Fourier transform of the initial distribution $\langle C(x, 0) \rangle$, $\mathscr{L}$ is the operator of Laplace transformation in time. Taking into account that $G(k, t) = \exp\{-\langle D \rangle k^2 (t - t')\}$ and $\mathscr{L}(e^{at} f(t)) = f(p - a)$ the solution of Eq. (31) may be written as

$$C_{kp} = \frac{C_k(0)}{p + k^2 \langle D \rangle - k^2 \int_{-\infty}^{+\infty} B(k - k_1; p + \langle D \rangle k_1^2) k_1^2\, dk_1}, \tag{32}$$

where $B(k, p)$ is the spectral density arising as a result of the Fourier transformation in space and Laplace transformation in time of the second correlation moment $\langle \tilde{D}(x, t) \tilde{D}(x', t') \rangle$. Expression (32) completely determines the evolution of $\langle C(x, t) \rangle$ if initial conditions $C_k(0)$ and statistical properties of fluctuations $B(k, p)$ are known.

To analyse the stability of the homogeneous distribution $\langle C(x, t) \rangle = \text{const}$ we need the dispersion equation. This is determined by the poles of Eq. (32):

$$p + k^2 \langle D \rangle - k^2 \int_{-\infty}^{+\infty} B(k_1 - k; p + \langle D \rangle k_1^2) k_1^2\, dk_1 = 0. \tag{33}$$

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Positive values of p following from Eq. (33) correspond to unstable modes. Taking into account that

$$B(k,p)\to 0, \quad \text{if} \quad k,p\to 0,$$

it can be shown that the positive solutions (and hence unstable modes) arise if

$$\int_{-\infty}^{+\infty} k_1^2 B(k_1-k;\, k_1^2\langle D\rangle dk_1 > \langle D\rangle. \tag{34}$$

Thus, the homogeneous distribution of diffusate is unstable when inequality (34) is satisfied.

It is useful to express the criterion (34) in terms of the correlation scales taking an upper estimate of the fluctuation amplitude $\langle \tilde{D}^2\rangle \sim \langle D\rangle^2$, which gives the following instability criterion

$$\langle D\rangle > l_c^2/\tau_c. \tag{35}$$

Note that $D^*<0$ in the delta-correlated in time case when (35) is satisfied (see Eq. (13)). However, (35) is expected to be fulfilled at sufficiently large τ_c when the approximation of delta-correlation in time may be invalid. In addition inequality (34) implies that only short-wave modes are unstable since criterion (34) is satisfied at sufficiently large values of the wave vector k. This may lead to formation of the diffusion profile with irregular, flactal-type geometry.[6]

6 THREE-DIMENSIONAL PROBLEM

Here we extend the derivation of a closed equation for $\langle C\rangle$ to a three-dimensional case, whesre $D=\langle D\rangle+\tilde{D}(\mathbf{r},t)$ with $\mathbf{r}$ the radius vector. Equations (19) and (20) now take an obvious form

$$\frac{\partial\langle C\rangle}{\partial t}=\langle D\rangle\Delta\langle C\rangle+\operatorname{div}\langle\tilde{D}\nabla\tilde{C}\rangle, \tag{36}$$

$$\frac{\partial\tilde{C}}{\partial t}=\langle D\rangle\Delta\tilde{C}+\operatorname{div}\tilde{D}\nabla\langle C\rangle. \tag{37}$$

Standard solution of Eq. (37) is given by

$$\tilde{C}(\mathbf{r},t)=\int_0^t dt'\int d\mathbf{r}'\, G(\mathbf{r}-\mathbf{r}',t-t')\{\operatorname{div}\tilde{D}\nabla\langle C\rangle\}', \tag{38}$$

where the operators in curly brackets $\{\ldots\}'$ act at $\mathbf{r}'$ and t'. Substituting Eq. (38) into Eq. (36) we obtain a closed integro-differential equation for $\langle C\rangle$:

$$\frac{\partial\langle C\rangle}{\partial t}=\langle D\rangle\Delta\langle C\rangle+\operatorname{div}\int_0^t dt'\int d\mathbf{r}'\nabla_{\mathbf{r}}G(\boldsymbol{\rho},\theta)\{\operatorname{div}B(\boldsymbol{\rho},\theta)\nabla\langle C\rangle\}' \tag{39}$$

where $\boldsymbol{\rho}=\mathbf{r}-\mathbf{r}'$, $\theta=t-t'$.

With account of the natural condition

$$B(\rho, \theta) \to 0 \quad \rho \to \infty$$

in a infinite medium, Eq. (39) takes the form similar to Eq. (22):

$$\frac{\partial\langle C\rangle}{\partial t} = \langle D\rangle\Delta\langle C\rangle + \sum_{i,k=1}^{3} \frac{\partial^2}{\partial x_i \partial x_k} \int_0^t dt' \int d\mathbf{r}' \frac{\partial^2 G(\rho, \theta)}{\partial x_i q x_k} B(\rho, \theta)\langle C(\mathbf{r}', t')\rangle. \tag{40}$$

Reduction of Eq. (40) to the differential form is similar to that presented in section 4.

Note that when the medium contains internal sinks, additional terms would arise in Eq. (40) which are proportional to the sink strengths. Thus, the fluctuations may affect the sink strength.

7 SUMMARY AND DISCUSSION

We have demonstrated that in the delta-correlated in time approximation evolution of the averaged concentration under diffusion with random diffusivity is described by a differential equation containing an additional term $\sim \Delta^2\langle C\rangle$ and renormalized diffusion coefficient.

The renormalized effective diffusion coefficient is shown to be always lower than the average one.

An account of the finitness of the correlation time leads to the nonlocal character of the diffusion equation for $\langle C\rangle$. However, the effective diffusion coefficient is again lower than the average one. A physical reason of this diffusivity reduction consists in the nonlinear "interaction" between fluctuations in the diffusion coefficient and concentration fluctuations (see Figure 1).

Let us consider one interesting consequence of diffusion in a fluctuative medium. Above we have derived a criterion of the instability of a homogeneous distribution of diffusate which shows that small-scale pertubations are the most unstable ones. This may lead to development of the diffusion front with complicated irregular geometry. Since any part of the diffusion front may generate a small-scale fluctuation which can grow and generate even shorter fluctuation and so on, such mechanism may be called a cascade. According to Mandelbrot,[6] this may indicate an interesting possibility of formation of diffusion fronts with fractal properties.

In conclusion we discuss some applications of the proposed theory to radiation effects in solids.

First, the fluctuative mechanism may result in radiation-induced segregation since atomic transport in alloys is predominantly caused by motion of point defects (PD), i.e. vacancies and interstitials.[3] During irradiation PD are created by collisions of bombarding particles with lattice atoms. Such collisions occur randomly, and it has been argued that this results in significant fluctuations in PD concentration[7] which consequently leads to fluctuations in diffusivity of alloying elements. As a result, an initially uniform alloy phase may be converted into a nonuniform and even fractal-like distribution of alloying elements when the instability criterion (34) is satisfied. Note that this mechanism requires no preferential coupling of some of the alloy components to PD fluxes, as contrasted with conventional segregation models.[3]

Second, there may be an interesting possibility of fluctuation-induced imbalance between the interstitial and vacancy fluxes to voids and dislocations which is distinct from the conventional elastically induced bias effect.[3] The reason is that collisions of bombarding particles with lattice atoms result not only in PD production, but also in the local enhancements of temperature known as the thermal spikes. This means that PD diffusion coefficient, being a function of temperature, is inhomogeneous together with PD production rate on a local scale. Thus, voids and dislocations are subjected to fluctuating PD fluxes due to the fluctuations in PD production rate and PD diffusivity. The former effect has been shown by Mansur *et al.*[7] to induce no bias factor in the expressions for mean fluxes. It is not surprising since Mansur *et al.*[7] have ignored intrinsic recombination of PD and hence all the nonlinear effects. On the other hand, as we have shown, the diffusivity fluctuations lead to the nonlinear effects irrespective of the intrinsic recombination account. This may lead to the nonlinear response of diffusion fluxes to the type of the sink, and consequently change the sink strengths of voids and dislocations differently which would result in the bias effect. This consideration is, of course, only speculative at the time and needs further development.

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APPENDIX

Let us evaluate the coefficients D^*, d, α and β entering Eq. (23) for the correlation moment having the form (28):

$$B(x, t) = B_0 \exp(-x^2/l_c^2 - t/\tau_c) \tag{A.1}$$

where l_c and τ_c are the correlation length and time, respectively. Substituting Green's function of the one-dimensional diffusion equation

$$G(x, t) = (4\pi\langle D\rangle t)^{-1/2} \exp(-x^2/4\langle D\rangle t) \tag{A.2}$$

together with $B(x, t)$ from (A.1) into Eqs. (24)–(27) and carrying out the integration we obtain the following expressions

$$D^* = \langle D\rangle - \frac{B_0}{\langle D\rangle}(1 - \sqrt{\pi}\,\varepsilon e^{\varepsilon^2}\mathrm{erfc}(\varepsilon)), \tag{A.3}$$

$$d = \sqrt{\pi}\, B_0 \varepsilon^3 \tau_c \left[(1 + 2\varepsilon^2)e^{\varepsilon^2}\mathrm{erfc}(\varepsilon) - \frac{2\varepsilon}{\sqrt{\pi}}\right], \tag{A.4}$$

$$\alpha = \tau_c^2 \frac{\partial}{\partial \tau_c} D^* = -\frac{d}{2\langle D\rangle \varepsilon^2}, \tag{A.5}$$

$$\beta = -\frac{\sqrt{\pi}}{8}\frac{B_0}{\langle D\rangle}\tau_c^2 \varepsilon \left[e^{\varepsilon^2}\mathrm{erfc}(\varepsilon)(1 - 4\varepsilon^2 - 4\varepsilon^4) + \frac{2\varepsilon}{\sqrt{\pi}}(1 + 2\varepsilon^2)\right], \tag{A.6}$$

where $\varepsilon \equiv l_c^2/4\langle D\rangle\tau_c$.

Expression (29) for D^* follows from Eq. (A.3) in two special cases $\varepsilon \ll 1$ and $\varepsilon \gg 1$. Other coefficients in these cases are given by

$$d = \begin{cases} \sqrt{\pi}\, B_0 \varepsilon^3 \tau, & \varepsilon \ll 1 \\ B_0 \tau, & \varepsilon \gg 1 \end{cases} \tag{A.7}$$

$$a = -2\tau d/l_c^2 \tag{A.8}$$

$$\beta = \begin{cases} -\sqrt{\pi}\, B_0 \tau^{3/2} l_c / 16 \langle D \rangle^{3/2} = 1/4 a\tau, & \varepsilon \ll 1 \\ -2\, B_0 \tau^3 / l_c^2 = a\tau_c, & \varepsilon \gg 1 \end{cases} \tag{A.9}$$

Approximate expressions (A.7)–(A.9) are presented here in order to demonstrate the character of their dependence on correlation scales l_c and τ_c. This dependence is expected to be true for a wide classs of correlation functions with sufficiently sharp dependence on l_c and τ_c.

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