Possibility of a diffusion–recombination instability in a solid with two types of defects

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Spatially nonuniform and time-varying distributions of defects are analyzed under the conditions of a diffusion instability. The defects are produced by a steady-state, uniform source. They undergo a mutual recombination. A new instability, which may arise, will increase the total number of defects continuously and exponentially as the result of a spatial redistribution of recombination.

In nonequilibrium systems, a diffusion current of excitations (defects) may be directed opposite the gradient (this is an "ascending" diffusion[1]) and would thus drive a so-called diffusion instability (Ref. 2, for example). In this instability, a spatially uniform distribution becomes unstable, and clusters form. Mechanisms for the diffusion instability have been discussed for the cases of a superconductor (Ref. 2, for example), an excitonic insulator,[3] and a system of vacancies and interstitial atoms.[4]

The time evolution of the diffusion instability and the nonuniform structures which result have received extremely little theoretical study, although a large amount of experimental evidence is available, particularly on clusters of radiation-induced defects (Ref. 5, for example). Analysis of the data shows that, with a steady-state, uniform source, the defects form clusters, whose structure varies continuously in time.

In the present letter we use the example of a system of interstitial atoms and vacancies to study nonuniform steady-state or time-varying solutions of the kinetic equations. We point out the possibility of new instability, which leads to not only a spatial redistribution of defects (as in the diffusion instability) but also a continuous increase in the total number of defects.

We first consider a system which has only a single type of defect. In this case the equation for the defect concentration $n(\mathbf{r}, t)$ is[4]

$$\frac{\partial n}{\partial t}=Q-\alpha n+\operatorname{div}\left[D\frac{\partial n}{\partial r}\left(1-\frac{\Omega^2 Kn}{T}\right)\right], \qquad (1)$$

where Q is the spatially uniform source of defects, the term αn describes recombination, D is the diffusion coefficient, K is the reduced bulk modulus, Ω is the dilatation volume, and T is the temperature. The second term in the derivative in (1), which leads to the anomalous diffusion, describes the motion in the stress field which arises because of the defects. The physical meaning here is that the defects, e.g., interstitial atoms with $\Omega>0$, cause the lattice to stretch out in a certain region.[1] Such a region is attractive for defects with $\Omega>0$ and repulsive for defects with $\Omega<0$.

As has been shown previously,[4] a uniform steady-state solution n_0 of Eq. (1) is unstable with respect to perturbations,

$$n(\mathbf{r},t)=n_0+n_1(\mathbf{r},t), \quad n_0=Q/\alpha, \quad n_1=\tilde{n}\exp(\lambda_\mathbf{q} t+i\mathbf{q}\cdot\mathbf{r}), \tag{2}$$

under the conditions

$$\lambda_\mathbf{q}=-\mathbf{q}^2D(1-n/n_c)-\alpha>0, \quad n_0>n_c=T/\Omega^2K. \tag{3}$$

To study nonuniform states, we put Eq. (1) in dimensionless form and treat the 1D case:

$$\frac{\partial f(xt)}{\partial t}=\frac{\partial}{\partial x}\left[\frac{\partial f}{\partial x}(1-f)\right]-f+\beta, \quad \left[\frac{\partial f}{\partial x}(1-f)\right]_{x=\pm 1}=0, \tag{4}$$

where $f=n/n_c$, $\beta=Q/\alpha n_c$, distances and times are expressed in units of $(D/\alpha)^{1/2}$ and α^{-1}, respectively, and the boundary condition in (4) means that the particle current vanishes at the boundaries of the sample.

A nonuniform steady-state solution of (4) is periodic [with a period of $(24\delta)^{1/2}$]. The behavior over a single period is described by the function

$$f(x)=1+\delta-x^2/6, \quad -1<x<1, \quad \delta=3/2(\beta-1), \tag{5}$$

which has a maximum (at $x=0$) and which reaches the critical value of one at the edges. A stability analysis of distribution (5) leads to the hypergeometric equation. Solutions in the form of the hypergeometric function $\Gamma(\alpha,\beta,\gamma,x)$ and also solutions of the form $(1-x^2)^{-1}$, $(1+x)^{-1}$, and $(1-x)^{-1}$ leave (5) stable.

It is important to note that the total number of particles does not change as the diffusion instability occurs. To demonstrate the point, we integrate (5) over z and impose the boundary conditions. We find

$$\frac{\partial \bar{f}}{\partial t}=-\bar{f}+\beta, \quad \bar{f}(t)=\frac{1}{2}\int_{-1}^{1}dx f(xt)=\beta+(\tilde{n}-\beta)\exp(-t). \tag{6}$$

In other words, the defect concentration quickly reverts to its original value β.

We see a quite different behavior in a system with defects of two types, which is described by the following equations[4] for interstitials, $f_i(xt)$,

$$\frac{\partial f_i}{\partial t}=\frac{\partial}{\partial x}\left[\frac{\partial f_i}{\partial x}(1-f_i)+f_i\frac{\partial f_v}{\partial x}\right]-f_if_v+\beta, \quad \left[\frac{\partial f_i}{\partial x}(1-f_i)+f_i\frac{\partial f_v}{\partial x}\right]_{x=0;1}=0, \tag{7}$$

and for vacancies, $f_v(x,t)$, with the interchange $i \rightleftarrows v$. For simplicity, we assume that all the parameters of the vacancies and the interstitials are identical.

According to Ref. 4, a uniform solution of (7), $f_i=f_v=f_0=\sqrt{\beta}$ is unstable if

$$2f_0-1>0. \tag{8}$$

The recombination contribution has dropped out of (8), and the critical concentration has fallen to half its value [in comparison with (3)]. These results, which stem from the circumstance that the growing perturbations of the vacancies and interstitials are out of phase, play a governing role in the diffusion–recombination instability.

We first note that there is no steady-state, uniform solution of system (7). Subtracting the second term in Eq. (7) from the first, we find the relation $\partial f_i/\partial x=\partial f_v/\partial x$, which shows that there is no longer an anomalous diffusion contribution, and hence no nonuniform solutions with a uniform source. Since it is unlikely that system (7) can be solved exactly, we will use perturbation theory to seek its solutions in the form of the functions

$$f_{i,v}(x,t)=f_0+\sum_{k=1} f_{i,v}^{(k)}(x,t),\quad f_{i,v}^{(k)}(x,0)=0,\quad k\geqslant 2,$$

$$f_{i,v}^{(1)}(x,0)=\pm\tilde{f}\cos\pi x,\quad \tilde{f}\ll 1,\quad f_{i,v}^{(k)}\ll f_0\sqrt{\beta},\tag{9}$$

which satisfy system (7) on the interval $0<x<1$. The reason for the choice $f_i^{(1)}(x,0)=-f_v^{(1)}(x,0)$ is that the instability arises only when there is an out-of-phase situation.

We have derived solutions up to $k=5$, but they are too lengthy to reproduce here. We will simply describe their structure and write some expressions for the total number of particles, which contain the basic results. The functions $f^{(k)}(xt)$ are harmonics $\cos(k\pi x)\exp(k\lambda t)[\lambda=\pi^2(2f_0-1)]$ which grow in time. Harmonics with odd k are out of phase ($f_i^{2n+1}=-f_v^{2n+1}$), while those with even k are in phase ($f_i^{2n}=+f_v^{2n}$, $n=0,1,\ldots$).

Introducing

$$a_k(t)=\int_0^1 [f_i^{(k)}(xt)+f_v^{(k)}(xt)]dx,$$

we can write the rate of change of the total number of particles in the system as

$$\frac{da}{dt}=\frac{d}{dt}\int_0^1 [f_i+f_v]dx=2\beta-2\int_0^1 f_i f_v\,dx=\sum_{k=1}\frac{da_k}{dt}.\tag{10}$$

In the spatially uniform case, we have $da/dt=0$, since we have $f_i=f_v=f_0=\beta^{1/2}$. Let us find da/dt after the beginning of the instability, at $t>0$. Substituting (9) into (7), and integrating along the coordinate, we find equations for $a_k(t)$:

$$\frac{da_1}{dt}=-2f_0a_1,\quad \frac{da_3}{dt}=-2f_0a_3,$$

$$\frac{da_2}{dt}=-2f_0a_2-\int_0^1 f_i^{(1)}f_v^{(1)}dx,\quad a_k(0)=0,$$

$$\frac{da_4}{dt}=-2f_0a_4-\int_0^1 dx[f_i^{(2)}f_v^{(2)}+f_i^{(1)}f_v^{(3)}+f_v^{(1)}f_i^{(3)}].\tag{11}$$

Solutions of (11) can be found exactly. We will reproduce them here for the case in which we are well above the threshold:

$$a_1=a_3=a_5=0,$$

$$a_2(t)=\frac{\tilde{f}^2}{4(\lambda+f_0)}\,[\exp(2\lambda t)-\exp(-2f_0\,t)],$$

$$a_4(t)=\frac{\pi^2\tilde{f}^4}{2(2\lambda+f_0)(\lambda+f_0+2\pi^2)}\,[\exp(4\lambda t)-\exp(-2f_0\,t)]. \qquad (12)$$

The results in (12) can be expected to remain qualitatively correct for all k.

Working from (10), using (12), and omitting terms $\sim\exp(-2f_0\,t)$, we find

$$\frac{da}{dt}\simeq\tilde{f}^2\exp(2\lambda t)\left[\frac{\lambda}{2(\lambda+f_0)}+\frac{\tilde{f}^2\exp(2\lambda t)2\pi^2\lambda}{(2\lambda+f_0)(\lambda+f_0+2\pi^2)}\right]. \qquad (13)$$

We see from (13) that the total number of particles increases exponentially in time. This behavior cannot be explained on the basis of a diffusion instability alone, since the number of particles does not increase when there are particles of only a single species [see (6)]. Accordingly, the reason lies in a separation of the vacancies and interstitials into different regions and a slowing of the mutual recombination as a result. This slowing leads in turn to an increase in the number of particles in a problem with a steady-state source β. Note that (13) is valid for the time interval which satisfies, according to (9), the inequality $\tilde{f}^2\exp(2\lambda t)<\beta$. Since $\tilde{f}^2$ is small, and we have $\beta\sim 1$, Eq. (13) is valid for

$$(2\lambda)^{-1}\ll t<(2\lambda)^{-1}\ln\beta/\tilde{f}^2.$$

This instability, which might be called a "diffusion–recombination" instability, thus yields a qualitative explanation of the continuous change in structure and the increase in the number of radiation-induced defects which are observed experimentally.

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