Migration kinetics for interacting atoms in a crystal lattice

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A theory of atomic diffusion in a crystal lattice is developed within the framework of the one-dimensional model with allowance made consistently for the correlation in the distribution of the diffusing particles. Exact solutions to the infinite system of equations determining the probabilities for specific distributions under equilibrium conditions are presented. The properties of the equilibrium system are extended to macroscopically inhomogeneous particle distributions, thereby determining the space-time evolution of these distributions. Expressions are found for the diffusion coefficients for an arbitrary strength of the interaction between the atoms and arbitrary population levels at the lattice sites. It is established that, if the diffusing particles attract each other, then the macroscopic mobility of the ensemble decreases faster than the individual particle mobilities as the temperature is lowered.

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1. INTRODUCTION

The concept of diffusion is used to describe the transport of atomic particles in crystals at most (excluding extremely low) temperatures.[1–4] The fruitfulness of this approach has been demonstrated by numerous investigations. But under certain conditions-correlation of the successive hops, strong inhomogeneity, reciprocal influence of the migrating particles—the applicability of the diffusion description is not obvious. The ideas of the random-walk model,[5,6] which are extremely useful for the justification of the theory of diffusion, need to be modified before they can be applied to such conditions. Extensive work has been done on the description of diffusion under conditions when the population levels at the allowed positions (lattice sites, interstices—hereinafter "sites") are high and when the interaction between the atoms has to be taken into account. Nevertheless, the role of the interaction between the diffusing particles is one of the incompletely elucidated points of the theory of atomic diffusion. This is primarily due to the fact that, in the published papers, the correlation that arises in the distribution of the diffusing atoms as a result of their interaction is either not taken into consideration at all, or is introduced approximately. Below, in application to a model structure, we construct a theory of atomic migration, that takes account of the correlation effects exactly, and can be used in the case of arbitrary population levels at the sites.

The behavior of the system of particles is described by the equations

$$\dot{\varphi}_M=\sum\nu(N')\varphi_{N'}-\sum\nu(N)\varphi_N. \tag{1}$$

Here M denotes any configuration of the particles on some finite set of sites and φ is the probability for this configuration. The summation in the first terms is over the N' different configurations (for the most part on a broader set of sites) that produce the configuration M as a result of one hop, and uniquely determine the probability $\nu(N')$ per unit time of this hop, while the summation in the second term is over a set of N configurations that includes the configuration M, and uniquely determines the probabilities $\nu(N)$ of the exchanges that destroy the configuration M; t denotes the time.

The system of Eqs. (1) is an infinite set of equations, that is similar to Bogolyubov's chain of equations.[7–9] It is of interest to consider those variants of the Eqs. (1) for which the analysis can be carried through. Such is the case of heterodiffusion in a one-dimensional chain in which the atoms at neighboring sites interact with each other. Besides its model import, the one-dimensional case also corresponds to specific conditions that sometimes obtain during the deposition of monatomic layers on a crystal surface. Thus, it has been established[10] that one-dimensional linear chains of atoms and identical interatomic "corridors" form during the condensation of potassium on the [112] face of tungsten. This circumstance allows us to neglect the interaction of the chains,[11] and consider the diffusion on the surface to be one-dimensional.

2. EXACT SOLUTIONS

The one-dimensional form of the Eqs. (1) is as follows:

$$\left.\begin{aligned}
&\dot{\varphi}_1(n)=\nu\varphi_{110}(n-1)+\nu\varphi_{011}(n+1)+\varphi_{010}(n-1)\\
&+\varphi_{010}(n+1)-\nu\varphi_{011}(n)-\nu\varphi_{110}(n)-2\varphi_{010}(n);\\
&\dot{\varphi}_{11}(n+{}^1/_2)=\nu\varphi_{1101}(n-{}^1/_2)+\nu\varphi_{1011}(n+{}^3/_2)\\
&+\varphi_{0101}(n-{}^1/_2)\\
&+\varphi_{1010}(n+{}^3/_2)-\nu\varphi_{011}(n)-\nu\varphi_{110}(n+1);\\
&\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\\
&\dot{\varphi}_{000}(n)=\varphi_{0100}(n-{}^1/_2)+\varphi_{0010}(n+{}^1/_2)-\nu\varphi_{11000}(n-1)\\
&-\nu\varphi_{00011}(n+1)-\varphi_{01000}(n-1)-\varphi_{00010}(n+1);
\end{aligned}\right\} \tag{2}$$

In the Eqs. (2) the order of the numbers in a subscript corresponds to the sequence of occupied and unoccupied sites; φ is the probability for the corresponding sequence; and an argument indicates the number of the central—for the chosen configuration—lattice site or interstice. It is assumed that the equations have been written in dimensionless form, so that ν is the ratio of the probability for a hop of a

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particle having neighbors to the probability for a hop of an isolated particle; the case $\nu < 1$ corresponds to attraction between the migrating particles ($\nu = 0$ corresponds to unbreakable interconnection of the particles); $\nu > 1$, to repulsion.

Notice that the system of Eqs. (2) is not a "chain," i.e., a one-dimensional sequence. The equations constitute a sequence of layers corresponding to the chosen number p of sites with 2^p equations in a layer for each value of n.

Let us write down the n- and t-independent solutions to the Eqs. (2), i.e., the solutions specifying the equilibrium of the system. Detailed balance is guaranteed in the system if, for any pair of configurations that are transformed into each other by one hop, the probabilities per unit time for these hops are equal. This requirement is satisfied if the probability for an arbitrary configuration $a_1 \ldots a_m$ ($m \geqslant 3$) can be expressed in terms of the probabilities for its constituent two-site configurations as follows:

$$\varphi_{a_1 \ldots a_m} = \frac{\varphi_{a_1a_2}\varphi_{a_2a_3}\cdots\varphi_{a_{m-1}a_m}}{\varphi_{a_2}\varphi_{a_3}\cdots\varphi_{a_{m-1}}} \tag{3}$$

under the condition that

$$\varphi_{01}^2 = \varphi_{10}^2 = \nu\varphi_{00}\varphi_{11}. \tag{4}$$

The equalities (3) are reminscent of the Kirkwood approximation, but do not coincide with it, and, what is most important, as will be seen below, they turn out to be exact. Notice further that the equalities (3) automatically guarantee the satisfaction of the obvious homologies

$$\varphi_{M0} + \varphi_{M1} = \varphi_M,$$

where M denotes an arbitrary sequence of occupied and unoccupied sites.

The equation determining the evolution of the arrangement $M\,101$ has the form

$$\begin{aligned}\dot{\varphi}_{M101} = {} & g_0(\varphi) + \nu\varphi_{M10011} + \varphi_{M10010} \\ & + \nu\varphi_{M110} - \nu\varphi_{M1011} - 2\varphi_{M1010},\end{aligned} \tag{5}$$

where $g_0(\varphi)$ is some set of expressions corresponding to the appearance and decompositions of the configuraiton $M\,101$ by all possible migrations within the limits of the section M, i.e. migrations that do not affect the section consisting of the three sites 101.

The corresponding equation for the configuration obtained by adding one occupied site to $M\,101$ is as follows:

$$\begin{aligned}\dot{\varphi}_{M1011} = {} & g_1(\varphi) + \nu\varphi_{M101011} + \varphi_{M101010} \\ & + \nu\varphi_{M1101} - \nu\varphi_{M1011} - \nu\varphi_{M10110},\end{aligned} \tag{6}$$

where $g_1(\varphi)$ is a set of terms similar in meaning to g_0, i.e., corresponding to migrations within the limits of the block M.

Let the substitution of the expressions (3) into the right members of the equalities (5) and (6) make $\dot{\varphi}_{M\,101}$ vanish. Then Eq. (6) with

$$g_1(\varphi) = g_0(\varphi)(\varphi_{11}/\varphi_1),$$

assumes the form

$$\begin{aligned}\dot{\varphi}_{M1011} = {} & (\varphi_{11}/\varphi_1)(\nu\varphi_{M1011} + 2\varphi_{M1010} - \nu\varphi_{M10011} - \varphi_{M10010} \\ & - \nu\varphi_{M110}) + \nu\varphi_{M101011} + \varphi_{M101010} - \nu\varphi_{M10110},\end{aligned}$$

which reduces to the form

$$\begin{aligned}\dot{\varphi}_{M1011} = \varphi_{M1}\Big\{ & \nu\frac{\varphi_{11}^2\varphi_{10}^2}{\varphi_1^2\varphi_0} + 2\frac{\varphi_{11}\varphi_{10}^3}{\varphi_1^2\varphi_0} - \nu\frac{\varphi_{11}^2\varphi_{00}\varphi_{10}^2}{\varphi_0^2\varphi_1^2} \\ & - \frac{\varphi_{11}\varphi_{00}\varphi_{10}^2}{\varphi_1^2\varphi_0^2} - \nu\frac{\varphi_{11}^2\varphi_{10}}{\varphi_1^2} + \nu\frac{\varphi_{11}\varphi_{10}^2}{\varphi_1^2\varphi_0^2} + \frac{\varphi_{10}^3}{\varphi_1^2\varphi_0^2} - \nu\frac{\varphi_{11}\varphi_{10}^3}{\varphi_0\varphi_1^2}\Big\}.\end{aligned} \tag{7}$$

Taking into consideration the fact that

$$\varphi_1 = \varphi_{10} + \varphi_{11}, \quad \varphi_0 = \varphi_{00} + \varphi_{01}, \tag{8}$$

and carrying out obvious transformations of the expression (7), we obtain

$$\dot{\varphi}_{M1011} = (\varphi_{M10}/\varphi_0^2\varphi_1)(\varphi_{10}^2 + 2\varphi_{11}\varphi_{10} + \varphi_{11}\varphi_{00})(\varphi_{10}^2 - \nu\varphi_{11}\varphi_{00}).$$

Thus, $\dot{\varphi}_{M\,101} = 0$ when the condition (4) is fulfilled, which proves that, if the relations (3) and (4) give the steady-state solution of the equation for $\varphi_{M\,101}$, then they also determine the equilibrium conditions for the arrangement with one more occupied site. Using similar constructions, we can show that this assertion is valid for all the eight variants obtained by varying the populations of the three end sites ($M\,000$, $M\,001$, $M\,010$, $M\,100$, $M\,011$, $M\,110$, $M\,101$, $M\,111$), and lengthened by the addition of occupied or empty sites. And since the equalities (3) and (4) guarantee the vanishing of the time derivatives for the single-site (φ_0, φ_1), two-site (φ_{00}, φ_{01}, φ_{11}), and three-site (φ_{000}, φ_{001}, φ_{010}, φ_{110}, φ_{101}, φ_{111}) arrangements, these relations give the exact steady-state solution of the infinite set of Eqs. (2).

3. CHARACTERISTICS OF THE EQUILIBRIUM DISTRIBUTIONS

Taking the homologies (8) into account, we write the relation (4) in the form

$$(\varphi_1 - \varphi_{11})^2 = \nu\varphi_{11}(1 - 2\varphi_1 + \varphi_{11}),$$

from which we obtain the following explicit relation between φ_1 and φ_{11}:

$$\varphi_{11} = \frac{\nu + 2(1-\nu)\varphi_1 - \{\nu[\nu + 4(1-\nu)\varphi_1(1-\varphi_1)]\}^{1/2}}{2(1-\nu)}. \tag{9}$$

For $\nu = 1$, i.e., in the absence of interaction,

$$\varphi_{11} = \varphi_1^2, \tag{10}$$

as it should be.

For a weak interaction, i.e., for $|1 - \nu| \ll 1$, the formula (9) yields the relation

$$\varphi_{11} \approx \varphi_1^2 + (1-\nu)\varphi_1^2(1-\varphi_1)^2. \tag{11}$$

The second term in (11), which characterizes the scale of the deviation of the distribution from the equilibrium distribution (10), has its maximum value at $\varphi_1 = \frac{1}{2}$, and its relative contribution increases as the mean density decreases.

In the case of strong repulsion ($\nu \gg 1$)

$$\varphi_{11} = \begin{cases} \varphi_1^2/\nu(1-2\varphi_1), & \varphi_1 < 1/2; \\ 1/2\sqrt{\nu}, & \varphi_1 = 1/2; \\ 2\varphi_1 - 1 + (1-\varphi_1)^2/\nu(2\varphi_1 - 1), & \varphi_1 > 1/2; \end{cases} \tag{12a}$$

$$\varphi_{00} \approx \begin{cases} 1 - 2\varphi_1 + (1-\varphi_1)^2/\nu(2\varphi_1 - 1), & \varphi_1 < 1/2; \\ 1/2\sqrt{\nu}, & \varphi_1 = 1/2; \\ (1-\varphi_1)^2/\nu(2\varphi_1 - 1), & \varphi_1 > 1/2. \end{cases} \tag{12b}$$

In the limit as $\nu \to \infty$ either of the quantities φ_{11} and φ_{00}, (12), goes to zero, depending on which is greater, the number of sites or the doubled number of particles. The case $\varphi_1 = \frac{1}{2}$ then corresponds to the ordered disposition of the particles ("crystallization" of the diffusing component).

In the case of strong attraction

$$\varphi_{11} \approx \varphi_1 - [\nu\varphi_1(1-\varphi_1)]^{1/2}, \qquad \varphi_{01} \approx [\nu\varphi_1(1-\varphi_1)]^{1/2}. \tag{13}$$

It follows from the formulas (13) that here the migrating particles or the empty sites have a tendency to form long chains (complexes), a fact which corresponds to the smallness of the quantities φ_{01} and φ_{10} and, consequently, the quantities φ_{010} and φ_{101}, which determine the probablity for the isolation of an atom or an empty site.

The formulas (3) and (4) allow us to find the characteristic lengths of the particle and unoccupied-site chains. Thus, the probability for the formation of a chain of N atoms is

$$\psi_N = \nu\varphi_{00}\varphi_{11}{}^N/\varphi_1{}^N.$$

The mean chain length $\bar{N}$, the most probable length N^*, and the variance D are given by the formulas

$$\bar{N} = \frac{\varphi_1+\varphi_{11}}{\varphi_1-\varphi_{11}}, \quad N^* = \ln^{-1}\frac{\varphi_1}{\varphi_{11}}, \quad D = 2\frac{\varphi_1\varphi_{11}}{(\varphi_1-\varphi_{11})^2}. \tag{14}$$

The quantities φ_1 and φ_{11} in these relations are connected by the relation (9).

The formulas (14) give

$$N^* = \ln^{-1}\frac{\bar{N}+1}{\bar{N}-1} \approx \frac{\bar{N}}{2}, \quad D = \frac{\bar{N}^2-1}{2}.$$

4. SPATIALLY-HOMOGENEOUS NONSTATIONARY PROCESSES

The exact solutions given above for the equilibrium state can be used as a basis for the search for various approximate relations for the nonequilibrium regimes. Such, in particular, is the process of redistribution of the atoms in the structure at a fixed mean concentration φ_1. This process corresponds to those reconstructions of the structure of an ensemble of atoms which lead to the appearance of local-equilibrium distributions corresponding to the local level φ_1, and can be regarded as the first stage of the general process of migration of interacting atoms.

It is clear from (2) that $\dot{\varphi}_1$ is zero for any spatially-homogeneous, symmetric—with respect to a change in direction—three-site functions. The situation is somewhat different with respect to the multisite probabilities. Their determination in the nonstationary regime requires uncoupling approximations. The advantage of the representation (3) in application to the nonstationary problem lies in the fact that this form corresponds exactly to both equilibrium distributions and the particularly nonequilibrium, uncorrelated distributions that play the role of the initial state. For example,

$$\varphi_{1101} = \varphi_{11}\varphi_{10}\varphi_{01}/\varphi_1\varphi_0 = (\varphi_1{}^2)(\varphi_1\varphi_0)(\varphi_0\varphi_1)/\varphi_1\varphi_0 = \varphi_1{}^3(1-\varphi_1).$$

Using (3), we obtain for the function φ_{11} the equation

$$\dot{\varphi}_{11} = \frac{2(\varphi_1-\varphi_{11})}{\varphi_1(1-\varphi_1)}\{(\varphi_1-\varphi_{11})^2-\nu(1-2\varphi_1+\varphi_{11})\varphi_{11}\}. \tag{15}$$

If $\varphi_{11}^{(0)} = \varphi_1^2$ at the instant $t = 0$, then (15) describes the interaction-induced variation of φ_{11} provided the diffusing-particle distribution was independent at zero time. This variant corresponds, for example, to diffusion of atoms in a sample that has been subjected to heating followed by rapid cooling. The relative probabilities ν are proportional to

$$\exp\{-(U_1-U_0)/kT\},$$

where U_1 is the potential barrier for the jumps of an atom having a neighbor at the nearest site and U_0 is the corresponding barrier for an isolated atom. Thus, regardless of the values of U_1 and U_0, as $T \to \infty$, the coefficient ν tends to unity, at which value we have, according to (9) and (10), $\varphi_{11} = \varphi_1^2$. But if we rapidly cool the sample, then the deviation of ν from unity increases sharply, and this leads to the redistribution of the atoms in accordance with Eq. (15). In all the cases the quantity φ_{11} tends asymptotically to the vlaue given by the formula (9). In particular, if $U_1 > U_0$, then, for sufficiently low temperatures, $\nu \to 0$ and $\varphi_{11} + \varphi_1$. In the limit $\nu \to 0$, Eq. (15) gives

$$\varphi_{11}(t) = \varphi_1 - [\varphi_1-\varphi_1{}^2][1+4(\varphi_1-\varphi_1{}^2)t]^{-1/2}.$$

From this it follows that the duration of the process of forming a local-equilibrium distribution is of the order of

$$(1-\varphi_1)/4\varphi_1. \tag{16}$$

Let us note that the increase of φ_{11} at a fixed value of the mean density φ_1 corresponds to a situation in which the uniform distribution of the atoms is unstble, and can be interpreted as upward diffusion, leading to the increase in time of the local particle-density inhomogeneity.

5. MACROSCOPIC MIGRATION

The formation of local-equilibrium states is the first stage of the process of particle migration. The second stage is the smoothing out of the macroscopic inhomogeneities. This stage is characterized by the following relation between the characteristic linear dimensions: the correlation length is large compared to the lattice spacing and small compared to the characteristic variation length for the mean density φ_1. The assumption that φ_1 varies slowly in space makes the continuous formulation of the kinetic equation for φ_1, (2), expedient. We obtain in this case the equation

$$\frac{\partial\varphi_1}{\partial t} = \frac{\partial}{\partial n}\left(\lambda\frac{\partial\varphi_1}{\partial n}\right), \tag{17}$$

where

$$\lambda = \nu^2\varphi_{11}{}^2/\varphi_1{}^2[\varphi_1{}^2+(\nu-1)\varphi_{11}{}^2], \tag{18}$$

φ_{11} being given in terms of φ_1 by (9).

The quantity λ is always positive, on account of which the stage in question has the character of macroscopic-density equalization. The quantity $\lambda \to 1$ as $\nu \to 1$, $\varphi_1 \to 0$ and $\lambda \to \nu$ as $\varphi_1 \to 1$. The last case clearly describes vacancy *migration in the diffusing component*. These asymptotic properties of the solution to Eq. (17) agree with the predictions of the theories that use the "method of mean energies" or other simplifying arguments. On the other hand, the formula (18) has some characteristics that qualitatively distinguish it from the corresponding relations obtained on the basis of averaging or approximation procedures.

As can be verified, the function $\lambda(\varphi_1)$ for fixed ν has an extremal point in the region $0.5 < \varphi_1 < 1$, with the case $\nu > 1$ corresponding to a maximum and the case $\nu < 1$, to a minimum. In other words, when $\nu > 1$, we have $\lambda_{\max} > \nu$ and $\lambda_{\min} = 1$, while when $\nu < 1$ we have $\lambda_{\max} = 1$ and $\lambda_{\min} < \nu$. The presence of a sufficiently high peak in the $\lambda(\varphi_1)$ curve at $\nu \gg 1$ will, for high population levels, lead to the decrease of the macroscopic-concentration gradients in the vicinity of this value of φ_1. Thus, the density range adjoining the indicated value turns out to be preferable.

If $\varphi_1 \ll 1$, $\nu \gg 1$, then the coefficient λ is given by

$$\lambda \approx (1-2\varphi_1)^{-2}, \tag{19}$$

i.e., does not depend on ν. This circumstance should be related to the fact that, under conditions of relatively strong repulsion and low mean population level, the diffusing atoms are not, as a rule, in close proximity, which leads to the effective annihilation of their short-range interaction. The growth of φ_1 at $\nu \gg 1$ entails the growth of λ and the relation $\lambda \approx 2\nu$ for $\varphi_1 = \frac{1}{2}$. The position of the maximum of λ approaches the point $\varphi_1 = \frac{1}{2}$ as ν increases. But if $\varphi_1 \to 1$, then

$$\lambda \approx \nu/\varphi_1^2. \tag{20}$$

In the parameter range where the diffusing particles exhibit a strongly pronounced tendency to stick together, i.e., for $\nu \ll 1$, we have

$$\lambda \approx {}^1/_2 (\nu^3/\varphi_1^3 (1-\varphi_1))^{1/2}; \qquad \nu \ll \varphi_1, (1-\varphi_1). \tag{21}$$

It follows from the formula (21) that, in the entire range of variation of φ_1, with the exception of narrow regions around $\varphi_1 = 0$ and $\varphi_1 = 1$,

$$\lambda \ll \nu.$$

Since λ is the ratio of the diffusion coefficient at the density in question to the value of the coefficient for a negligibly small density, attraction between the atoms leads to the decrease in almost the entire density (φ_1) range of the diffusion coefficient down to significantly lower values than even in the case when the allowed positions are practically fully populated. In other words, the macroscopic deceleration proceeds significantly faster at low temperatures that the decrease of the microscopic mobility. Thus, the correlation in the distribution of the diffusing atoms leads to the effective macroscopic freezing of the entire ensemble of particles subject to migrations when their individual mobilities are conserved.

It can be seen that the dynamics of the variation of the macro-distributions in the case $\nu \ll 1$ is such that the smoothing out in the region of low densities proceeds faster than the disintegration of denser condensations. If, on the other hand, $\nu \gg 1$, then the opposite tendencies obtain, so that the substantial macrocondensations disappear relatively rapidly, after which a slower process of "almost linear" diffusion develops.

It is advisable to note that, in all the above-presented particular cases, e.g., (19)–(21), etc., the ν dependence of λ is a power dependence, i.e., the logarithm of the diffusion coefficient is proportional to T^{-1}. But the coefficient of proportionality has different values in different region of φ_1, as a result of which substantial deviations from the Arrhenius law should be expected in the case when the interaction between the atoms is not negligible, and, furthermore, the relative scale of these deviations should increase with decreasing temperature. The fact that deviations from the Arrhenius law are often not observed in experiment should be accounted for in part by the fact that the cases usually studied are the ones in which the diffusion coefficients are fairly large, a situation which corresponds to the case of high temperatures. As T increases, the probability $\nu \to 1$, which results in the loss of the effects determined by the correlation.

Equation (17) determines together with the expression (18) the time scale for the development of the diffusion process. A comparison of this quantity with the characteristic local-equilibrium-establishment time, e.g., (16), yields the conditions for the admissibility of the division of the migration process into two stages. For the case $\nu \ll 1$, to which corresponds the formula (16), we obtain

$$\nu^{3/2} (1-\varphi_1)^{1/2} / 4\varphi_1^{5/2} L^2 \ll 1, \tag{22}$$

where L is the characteristic macroscopic-inhomogeneity length, expressed as a number of lattice spacings. It can be seen that the condition (22) practically does not impose limitations, and can be violated only at extremely low φ_1 values, when the approach to the local-equilibrium states is a macroscopically long process.

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