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Quantum kinetics of phase transitions at low temperatures

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A theory is developed of quantum decay of a metastable state in a class of problems in which quantum fluctuations governing subbarrier evolution of virtual nuclei of the new phase are related to the local motion of single particles. A model of a crystal with two positions (states) of an atom in a unit cell is considered. The amplitude of the tunnel creation of a critical nucleus is found. It is shown that the associated characteristic sum over various "paths" can be found employing the conventional statistical methods. The discrete nature of the energy structure of the levels makes it necessary to allow for the interaction with phonons, which is done within the framework of the kinetic equation for the density matrix. The method can be applied also to analyze the decay of a metastable state via formation of finite clusters, when the macroscopic description is impossible.

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1. INTRODUCTION

Lifshitz and Kagan[1] (see also Iordanskiĭ and Finkel'shteĭn[2]) developed quantum kinetics of phase transitions at temperatures close to $T = 0$. Decay of a metastable state is found to be associated with subbarrier tunneling of a virtual nuclei of the new phase in the configuration space. This virtual growth of nuclei causes an initially homogeneous system to pass through a sequence of locally inhomogeneous states. Lifshitz and

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Kagan[1] used a macroscopic description of these states presupposing slight changes of all the phenomenological characteristics over interatomic distances and feasibility of describing the kinetic energy in terms of the time dependences of these characteristics. Under certain assumptions and far from the lability point, the problem can be reduced to one-dimensional and quasiclassical in the space of sized of compact nuclei and this problem can be solved directly.

However, there is an extensive class of problems in which quantum fluctuations, which are generally responsible for the evolution of nuclei, are associated with the local motion of single particles and the resultant inhomogeneous states are characterized primarily by the number of particles n in the new-phase nucleus. This situation occurs if a phase transition is accompanied by a change in the state of the particles or, for example, when a unit cell has two nonequivalent positions for an atom separated by a potential barrier, etc. If the specific amplitude of a transition accompanied by a change in the number of particles in a nucleus is small, n is a good quantum number. Then, the part of the Hamiltonian nondiagonal in respect of n acts as the kinetic energy.

The corresponding Schrödinger equation is of the finite-difference type in the particle number space. It is important to note that it is in general impossible to go over to a differential equation for $n_0 \gg 1$ (n_0 is the number of particles in a critical nucleus) and to the usual quasiclassical structure of formulas for subbarrier tunneling. This is due to the fact that the change in the action ΔS corresponding to $\Delta n = 1$ in the particle number space obeys the inequality

$$\Delta S/\hbar \gg 1.$$

In general, n is not the only quantum number. The state of a system with a given value of n depends on the convolution of particles in a nucleus. The subbarrier evolution of a nucleus may pass through various sequences of such configurations. We shall show that the amplitude of decay of a metastable state giving rise to a critical nucleus is expressed in terms of a characteristic sum over "paths" in which the individual path represents a definite sequence of configurations in the virtual growth of a nucleus.

Determination of this tunnel amplitude is effectively the main problem in the kinetics of phase transitions at temperatures close to absolute zero.

We shall consider a very general example of a phase transition which occurs in the light-atom sublattice of a crystal which has two inequivalent positions (states) in each unit cell. The Hamiltonian of the system is then analogous to the Hamiltonian of the Ising model in strong longitudinal and weak transverse fields. The solution of the appropriate Schrödinger equation and determination of the wave functions are given in Sec. 2. The amplitude of the subbarrier creation of a critical nucleus is found and a detailed analysis of the sum of configuration paths is made in Secs. 3 and 4.

The discrete nature of the energy levels makes it necessary to allow for the interaction with the phonon degrees of freedom. This is considered in Secs. 5 and 6 on the basis of solution of the kinetic equation for the density matrix, which makes it possible—in particular—to determine also the preexponential factor in the transition probability. The role of temperature and the change from the quantum to classical transbarrier kinetics is considered in Sec. 7.

2. FORMULATION OF THE PROBLEM. SUBBARRIER WAVE FUNCTIONS

We shall consider a crystal which has a sublattice of light atoms. We shall assume that a unit cell has two inequivalent positions of a given atom separated by a potential barrier. Consequently, the regular sublattice may have one of two configurations, which generally have different energies. We shall assume that both sublattices have the cubic symmetry. For simplicity, we shall confine our attention to the case when only the interaction between the nearest neighbors is important. Then, the Hamiltonian describing the light-atom subsystem can be written in the form

$$H = H_0 + H_1, \tag{2.1}$$

$$H_0 = -\varepsilon \sum_{\mathbf{m}} n_{\mathbf{m}+} + \varkappa \sum_{\mathbf{mg}} n_{\mathbf{m}+} n_{\mathbf{m}+\mathbf{g},-}, \tag{2.2}$$

$$H_1 = -\xi_0 \sum_{\mathbf{m}} (a_{\mathbf{m}+}^{+} a_{\mathbf{m}-} + a_{\mathbf{m}-}^{+} a_{\mathbf{m}+}), \tag{2.3}$$

where

$$\varkappa > 0, \quad n_{\mathbf{m}\pm} = a_{\mathbf{m}\pm}^{+} a_{\mathbf{m}\pm}, \quad n_{\mathbf{m}+} + n_{\mathbf{m}-} = 1.$$

Here, the index "±" refers to the two positions in the unit cell, and their corresponding creation a^+ and absorption a operators introduced in the site (Wannier) representation. The summation over g applies to the nearest neighbors.

The nature of the expressions (2.2) and (2.3) implies that the interaction between atoms is of the pair type and that the amplitude of a transition between two states in a unit cell is independent of the local configuration. The energy in Eq. (2.2) is measured from the value corresponding to the energy of the sublattice with all the atoms in the "−" position. The sign in Eq. (2.3) is selected for reasons of convenience.

We shall assume that a system which is in a stable state with atoms in the "−" sublattice ($\varepsilon < 0$) experiences at $t = 0$ a change in the external parameters which result in $\varepsilon > 0$. Then, the occupied sublattice becomes metastable. In the weak "supersaturation" case corresponding to

$$\varepsilon \ll \varkappa\nu \tag{2.4}$$

(ν is the number of the nearest neighbors) the decay of such a metastable state can occur only as a result of a simultaneous transition of a large number of atoms to the "+" state. On the other hand, in the most typical cases the amplitude of a subbarrier intrasite transition is small and we may assume that

$$|\xi_0| \ll \varepsilon, \varkappa. \tag{2.5}$$

If the condition (2.5) is satisfied, the state of the system can be conveniently represented by the eigenstates of the Hamiltonian H_0. It follows from Eq. (2.2) that the Hamiltonian H_0 commutes with the total number of atoms

in the "+" state, which we shall now denote simply by n. Thus, in this problem, the value of n is a good quantum number. However, the state and energy of the system depend not only on n but also on the configuration of the distribution of atoms in the "+" position. We shall introduce the index α to label the various configurations and we shall denote the eigenstates of the Hamiltonian H_0 by $|n\alpha_n)$ or simply by $|\alpha_n)$ in all those cases when there is no danger of misunderstanding.

We shall expand the complete wave function of the system in terms of these states:

$$\psi=\sum_{n\alpha_n}\psi_{\alpha_n}|\alpha_n). \tag{2.6}$$

Then, the Schrödinger equation corresponding to the Hamiltonian (2.1) can be reduced to the following finite-difference equation:

$$(E_{\alpha_n}-E)\psi_{\alpha_n}=-\sum_{\alpha_{n-1}}(\alpha_n|H_1|\alpha_{n-1})\psi_{\alpha_{n-1}}$$
$$-\sum_{\alpha_{n+1}}(\alpha_n|H_1|\alpha_{n+1})\psi_{\alpha_{n+1}}. \tag{2.7}$$

When n ($n\gg1$) atoms are transferred to the "+" position, the minimum energy corresponds to a compact nucleus with the optimal surface configuration. The energy of a compact nucleus can in general be represented by

$$E_{\alpha_n}=-\varepsilon n+\beta(\alpha_n)\nu\varkappa n^{2/3}. \tag{2.8}$$

We shall use β_n to denote the value of $\beta(\alpha_n)$ corresponding to the configuration with the lowest surface energy for a given value of n. When the number of particles in the "+" state is increased, the energy of a nucleus first becomes less than zero for

$$n_0\approx(\beta_0\nu\varkappa/\varepsilon)^3\quad(\beta_0=\beta_{n_0}). \tag{2.9}$$

This is the critical size of a real nucleus in the particle number space of the new phase. The subsequent fate of this nucleus involves evolution of energy and it is not hindered by barriers. However, the transition from the initial state to the state with a separate critical nucleus is of pure tunnel nature and passes through a sequence of virtual nuclei with particle numbers $n<n_0$.

We note that noncompactness of a nucleus increases greatly its critical size. Let us consider a nucleus consisting of n particles in the "+" state and p ($p\ll n$) holes, i.e., particles in the initial "−" state. Then,

$$E\approx-\varepsilon n+\beta\nu\varkappa(n+p)^{2/3}+\nu\varkappa p.$$

The number of particles in a critical nucleus increased by

$$\Delta n\approx3\nu\varkappa p/\varepsilon\gg p. \tag{2.10}$$

If we allow for the exponential reduction in the transition amplitude on increase of n (this is discussed below), the above result allows us to determine the probability of appearance of a real nucleus by considering only a transition to a state α_{n_0} corresponding to a critical compact nucleus.

It clearly follows from the above and also from Eq. (2.5) that the amplitude of a transition to the state α_{n_0} represents the sum of the amplitudes of formation of a critical nucleus along paths corresponding to transitions to a new state only of the atoms which lie within the region defined by the final configuration of α_{n_0}. Outside this region the atoms are still in the "−" position.

We shall now return to Eq. (2.7). Let us assume that in addition to Eq. (2.5), the following inequality is obeyed:

$$(\xi/\nu\varkappa)^2n_0\ll1. \tag{2.11}$$

We shall consider the solution of Eq. (2.7) which decreases on increase of n and which corresponds to the initial metastable state $E\approx0$. In this case, allowance for Eq. (2.11) and for the right-hand side of Eq. (2.7) makes it possible to neglect the second term of practically all values of n. Then, the solution of Eq. (2.7) is obtained directly in the form

$$\psi_{\alpha_n}=(-1)^n\sum_{\alpha_n\ldots\alpha_1}\frac{(\alpha_n|H_1|\alpha_{n-1})(\alpha_{n-1}|H_1|\alpha_{n-2})\ldots(\alpha_1|H_1|0)}{E_{\alpha_n}E_{\alpha_{n-1}}\ldots E_1}\psi_0. \tag{2.12}$$

It follows from the form of Eq. (2.12) that the configuration α_n is attained as a result of a minimum number of steps, equal to n. Therefore, all the intermediate steps are limited strictly to the configuration of the unit cells corresponding to α_n. Allowance for the population of this configuration with particles in the "+" state gives the number of terms in any intermediate sum

$$\sum_{\alpha_p}=n-p+1.$$

Consequently, the total number of additive terms in Eq. (2.12) is

$$\sum_{\alpha_{n-1}}\ldots\sum_{\alpha_1}=n!. \tag{2.13}$$

Each term effectively represents the product of the reciprocals of the energies which appear in each path as n cells are filled sucesssively with n particles in the "+" state. The filling sequence is random and the number of paths is given exactly by Eq. (2.13).

Bearing these results in mind and using the notation

$$E_{\alpha_p}=\varkappa\varepsilon(\alpha_p), \tag{2.14}$$

we can transform Eq. (2.12) to

$$\psi_{\alpha_n}=\exp\Big[-n\ln\frac{\varkappa}{\xi}\Big]S^{(\alpha_n)}\exp\Big[-\sum_{p=1}^{n}\ln\varepsilon(\alpha_p)\Big]\psi_0. \tag{2.15}$$

Here, $S^{(\alpha_n)}$ denotes functional summation over all $n!$ paths which result in the configuration α_n. The sum in the exponential function represents all the individual paths.

Since the wave function (2.6) is in our case localized in the region of a critical nucleus, we can easily show that normalization of this function to unity subject to the inequality (2.11) corresponds to

$$\psi_0\approx1. \tag{2.16}$$

We can similarly find the solution of Eq. (2.7) corresponding to a level with the index α_{n_0} ($E\approx E_{\alpha_{n_0}}$), which decreases with depth in the barrier on increase in the difference n_0-n. The inequality (2.11) now makes it possible to omit the first term on the right-hand side of Eq. (2.7) and all the comments made in relation to Eq. (2.15) are still applicable.

Adopting $\bar{\psi}$ for the "right-hand" solution, we find that the coefficients of the expansion (2.6) are

$$\bar{\psi}_{\alpha_n} = \exp\left[-(n_0-n)\ln\frac{\varkappa}{\xi}\right]\bar{S}^{(\alpha_n)}\exp\left[-\sum_{p=n}^{n_0-1}\ln[\varepsilon(\alpha_p)-\varepsilon(\alpha_{n_0})]\right]\bar{\psi}_{\alpha_{n_0}}. \tag{2.17}$$

Here, $\bar{S}^{(\alpha_n)}$ denotes summation over the "hole" paths which appear when n_0-n atoms return to the "–" state in a fixed configuration of n_0-n unit cells which are not filled with the new-phase particles in the α_n configuration (compared with the α_{n_0} configuration). Once again the separate paths represent successive random "loss" of n_0-n particles and the total number of the "hole" paths is $(n_0-n)!$.

The normalization of the wave function $\bar{\psi}$ to unity in accordance with Eq. (2.16) corresponds to

$$\bar{\psi}_{\alpha_{n_0}} \cong 1.$$

The wave functions of the states α_m ($E \approx E_{\alpha_m}$) close to α_{n_0} have the same structure as Eq. (2.17) if we make the substitution $n_0 \to m$, $\alpha_{n_0} \to \alpha_m$ and correspondingly allow for the changes in "hole" paths.

3. AMPLITUDE OF A SUBBARRIER TRANSITION

We shall now determine the amplitude of a transition from a metastable state to a state with a critical nucleus of the new phase. We shall introduce localized-state wave functions of the metastable phase ψ_l ("left-hand" state) and of the phase with a critical nucleus ψ_r ("right-hand" state). The function ψ_l is the eigenstate of the Hamiltonian H^l, which differs from Eq. (2.1) because of an additional potential wall for the α_n configurations which are located (on the energy scale) in the immediate vicinity of α_{n_0}. This results in truncation of the wave-function tail precisely in the region adjoining the point of emergence from under the barrier, where the wave function ψ of the Hamiltonian H of Eq. (2.1) has a characteristic peak because of the admixture of the rising solution. Elsewhere throughout the phase space the functions ψ_l and ψ_r are practically identical with one another and with the falling solution (2.15). Similarly, the function ψ_r is the eigenstate of the Hamiltonian (2.1) in which H_0 is replaced with H_0^r, suppressing the peak of the "right-hand" wave function near $n=0$. Over most of the phase space the function ψ_r is practically identical with Eq. (2.17).

We shall determine the matrix element of a transition from "left-hand" to the "right-hand" state employing mutually orthogonal functions $\bar{\psi}_l$ and $\bar{\psi}_r$. Then, in the approximation linear in respect of the overlap, we have

$$\begin{aligned} H_{rl} &= (\bar{\psi}_r|H|\bar{\psi}_l) = (r|H|l) - {}^1/_2(E_r+E_l)(r|l) \\ &= {}^1/_2(r|H-H^l|l) + {}^1/_2(l|H-H^r|r), \end{aligned} \tag{3.1}$$

where $|l)$ and $|r)$ denote the states ψ_l and ψ_r.

We shall use the equations

$$H^l\psi_l = E_l\psi_l, \quad H^r\psi_r = E_r\psi_r,$$

multiplied on the left by ψ_r and ψ_l, respectively, and sum over p and α_p with $n' \leq p \leq n_0$, where n' is displaced considerably to n_0 (but is still to the left of the potential wall). Subtracting the first from the second equation and bearing in mind that in this interval we have $H^r = H$, we obtain

$$(r|H-H^l|l)' = (r|H_1|l)' - (l|H_1|r)' - (E_l-E_r)(r|l)'. \tag{3.2}$$

Here, the primes indicate formally that the summation in the determination of the matrix element is limited to the selected region.

The left-hand side of Eq. (3.2) is clearly identical with the first term in Eq. (3.1) (to within $\frac{1}{2}$), because $H-H^l$ differs from zero only in the interval in question. The expression for the second term in Eq. (3.1) (the corresponding matrix element is denoted by two primes) can be found by following exactly the same procedure and selecting the summation region with $p \leq n''$, where $n'' \ll n_0$. In this way the matrix element of a transition is found to be

$$H_{rl} \cong {}^1/_2[(r|H_1|l)' - (l|H_1|r)' + (l|H_1|r)'' - (r|H_1|l)'']. \tag{3.3}$$

We have omitted above the term

$${}^1/_2(E_r-E_l)[(r|l)'' - (r|l)'], \tag{3.4}$$

because a direct analysis shows that in any case this term is small when n'', $(n_0-n') \ll n_0$ (see also below).

Equation (3.3) is very convenient for calculations. In fact, the differences measured from the kinetic energy, which occur in this equation, give nonzero contributions only at the ends of the selected interval, where ψ_l and ψ_r are practically identical with ψ and $\bar{\psi}$. Bearing this in mind, we find that the first difference in Eq. (3.3) is

$$(r|H_1|l)' - (l|H_1|r)' = -\xi\sum_{\alpha_{n'}\alpha_{n'-1}}[\bar{\psi}_{\alpha_{n'}}\psi_{\alpha_{n'-1}} - \psi_{\alpha_{n'}}\bar{\psi}_{\alpha_{n'-1}}]. \tag{3.5}$$

The second term in the brackets is small compared with the first in respect of the ratio $(\xi/E_{\alpha_{n'}})^2$ and it can be ignored.

The second difference in Eq. (3.3) reduces to the same expression if the substitution $n' \to n''$ is made. We shall now substitute the expressions (2.15) and (2.17) in Eq. (3.5). We shall ignore the small difference $E_0 - E_{\alpha n_0}$ [i.e., the small quantity $\varepsilon(\alpha_{n_0})$] in Eq. (2.17). We can quite easily show directly that Eq. (3.5) gives the following final expression for the matrix element of the transition:

$$\left.\begin{aligned} H_{rl} &\approx -\xi\exp\left[-(n_0-1)\ln\frac{\varkappa}{\xi}\right]S^{(\alpha_{n'})}e^{-\chi}; \\ \chi &= \sum_{p=1}^{n_0-1}\ln\varepsilon(\alpha_p). \end{aligned}\right\} \tag{3.6}$$

We shall check only that the calculation of the path is correct. The formation of $\psi_{\alpha_{n'-1}}$ states requires $(n'-1)!$ paths. The number of paths participating in the formation of states $\bar{\psi}_{\alpha_{n'}}$ is $(n_0-n')!$. The sum over $\alpha_{n'}$ in Eq. (3.5) is equal to the number of different configurations of n' particles in n' particles in n_0 cells, i.e.,

$$\frac{n_0!}{n'!(n_0-n')!}.$$

Then the sum over $\alpha_{n'-1}$ is equal to the number of variants of a transition from any state $\alpha_{n'}$ to a state with $n'-1$ particles, i.e., simply n'. As a result, the total number of paths is

$$(n'-1)!(n_0-n')!\frac{n_0!}{n'!(n_0-n')!}n' = n_0!.$$

Thus, the amplitude of a transition to a state with a critical nucleus of the stable phase is given by Eq. (3.6),

where the summation is carried out over all the $n_0!$ paths representing a random sequence of filling of n_0 cells with particles.

The result (3.6) is independent of the actual solution of n' and n'', because the potential walls truncating the wave-function tails lie within the intervals (n', n_0) and $(0, n'')$. Therefore, the ratios n''/n_0 and $(n_0-n')/n_0$ can be made quite small. It should also be pointed out that if we ignore the difference $E_0 - E_{\alpha_{n0}}$ then the preexponential factor in Eq. (3.6) becomes slightly modified.

4. ANALYSIS OF THE PATH SUM

It follows from the preceding section that determination of the amplitude of a tunnel transition resulting in the formation of a critical nucleus can, in fact, be reduced to the problem in ordinary statistics, with a characteristic summation of the paths in Eq. (3.6), which are sorted at the same time. The transition to the classical case in Eq. (3.6) corresponds to $\xi \to 0$ (and the consequence is $\hbar \to 0$), which reduces the transition amplitude to zero. It is important to note that the path sum in Eq. (3.6) is completely independent of ξ.

Various paths in Eq. (3.6) lie in a fairly wide energy band shown shaded in Fig. 1. The lower edge of this band corresponds to trajectories of growth of a compact nucleus with the optimal surface configuration. It corresponds to Eq. (2.8). The maximum energy on this curve [see Eq. (2.14)] is

$$\varepsilon_m \approx \frac{4}{27}\frac{\varepsilon n_0}{\varkappa}. \tag{4.1}$$

The upper edge of the band is described closely by

$$\varepsilon(p) \approx \nu p, \quad p < {}^1/_2 n_0; \quad \varepsilon(p) \approx \nu(n_0 - p), \quad p > {}^1/_2 n_0. \tag{4.2}$$

The function χ in Eq. (3.6) has its minimum value for the paths corresponding to the evolution of a compact nucleus. We shall consider the behavior of χ in this case by separating the explicit dependence on the number of particles in a nucleus. Then, allowing for Eqs. (2.8), (2.9), and (2.14) we shall obtain the expression for the dimensionless energy $\varepsilon(\alpha_p)$ in the form

$$\varepsilon(\alpha_p) = \beta_0 \nu p^{2/3}\left[\frac{\beta(\alpha_p)}{\beta_0} - \left(\frac{p}{n_0}\right)^{1/3}\right]. \tag{4.3}$$

It follows directly that χ contains the dominant (nonlinear in n_0) term given by

$$\sum_p^{n_0-1} \ln p^{2/3} = \frac{2}{3}\ln(n_0-1)! \approx \frac{2}{3} n_0 \ln\frac{n_0}{e}. \tag{4.4}$$

For arbitrary paths, whose majority corresponds to evolution via noncompact nuclei, the intermediate energies are $\varkappa/\varepsilon \sim n_0^{1/3}$ times greater than for a "compact" path [see Eqs. (4.1) and (4.2)]. In this case, we have

$$\varepsilon(\alpha_p) = p\nu\eta(\alpha_p) \tag{4.5}$$

and the corresponding dominant term of χ is

$$\ln(n_0-1)! \approx n_0 \ln(n_0/e). \tag{4.6}$$

We shall now compare the number of paths in these two cases. For a purely random process of nucleation the number of paths is close to the total number of paths

$$n_0!, \tag{4.7}$$

whereas the number of compact paths is proportional to

$$\prod_p^{n_0} p^{2/3} = (n_0!)^{2/3}. \tag{4.8}$$

The last result is easiest to obtain by proceeding in the opposite direction, i.e., by dismantling a critical nucleus starting from the surface. In each step the number of possible variants is proportional to the surface, i.e., to $p^{2/3}$.

Returning to Eq. (3.6) and comparing Eqs. (4.7) and (4.8) with Eqs. (4.6) and (4.4), we can see that the increase in the number of paths in the first approximation compensates fully the energy "undesirability" of the formation of intermediate noncompact nuclei. The dominant terms in χ cancel out completely in both cases and the argument of the exponential function in (3.6) loses the nonlinear dependence on n_0. This makes it impossible to select *a priori* the optimal trajectories and makes it necessary to determine qualitatively the path sum in Eq. (3.6).

A. Noncompact paths

In general, the energy $\varepsilon(\alpha_p)$ corresponds to the energy of p particles in the "+" state scattered randomly over n_0 cells. Therefore, if $p \gg 1$, this energy is close to the average value

$$\varepsilon(p) = \nu p(1 - p/n_0). \tag{4.9}$$

We shall now modify the expression for χ by replacing the energy $\varepsilon(\alpha_p)$ with the average enery $\varepsilon(p)$ of Eq. (4.9). Changing from summation to integration, we find that

$$\chi = \ln(n_0-1)! + (n_0-1)\ln\nu + \int_1^{n_0-1} dp \ln(1-p/n_0).$$

Bearing in mind that the number of paths in this case is practically equal to (4.7), we find that the matrix element of the transition (3.6) is

$$H_{rl} \approx -\xi \exp\left[-(n_0-1)\ln\frac{\varkappa\nu}{\xi\rho}\right]. \tag{4.10}$$

B. Compact paths

The compact paths lie far from the statistical average paths and their number is negligible compared with that given by Eq. (4.7). There is no contribution from these paths in Eq. (4.10).

We shall use Eq. (4.3) bearing in mind that

$$\beta_0 \nu p^{2/3} = n_S(p)\nu_{S0}, \tag{4.11}$$

where $n_S(p)$ is the number of particles on the surface of a compact nucleus with p particles in the "+" state; ν_{S0}

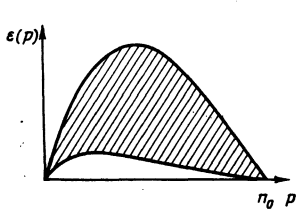

FIG. 1.

is the average number of neighbors in the "−" state for particles lying on the surface of a critical nucleus.

In the calculation of χ for the compact paths we shall be interested mainly in intermediate states with $n_s \gg 1$ and then the energy $\varepsilon(\alpha_p)$ is close to its statistical average, which is obtained by making the following substitution in Eq. (4.3):

$$\beta(\alpha_p)/\beta_0 \to \zeta = \bar{\nu}_s/\nu_{s0}, \tag{4.12}$$

where $\bar{\nu}_s$ is the average number of neighbors in the "−" state on the surface.

We shall substitute Eq. (4.3) in the expression for χ in Eq. (3.6), and we shall allow for Eqs. (4.12) and (4.11). Going over from summation to integration, we obtain

$$\chi = \ln \prod_p^{n_0-1} n_s(p) + (n_0-1)\ln \nu_{s0} + (n_0-1)\varphi(\zeta). \tag{4.13}$$

Here,

$$\varphi(\zeta) = 3\int_0^1 dx\, x^2 \ln(\zeta - x). \tag{4.14}$$

The above integral can be calculated in an elementary manner.

Bearing in mind that the number of the compact paths is

$$\prod_p^{n_0} n_s(p), \tag{4.15}$$

we obtain the transition amplitude (3.6) in the form

$$\left.\begin{aligned} H_{rl} &\approx -\xi n_{s0} \exp\left\{-(n_0-1)\ln\frac{\varkappa c}{\xi}\right\}, \\ c &= \nu_{s0} e^{\varphi(\zeta)}. \end{aligned}\right\} \tag{4.16}$$

The actual value of the argument of the exponential function in Eq. (4.16) depends on the structure of the surface layer (in the direction of its thickness) and also on the lattice structure. The use of a fixed value of ζ given by Eq. (4.12) implicitly presupposes that the distribution in the surface layer remains constant when the radius of a nucleus increases. In the opposite case we have to introduce the dependence of ζ on the number of the surface layer k (or on the radius) and then sum over k. This makes it possible to apply some variational procedure in each specific case.

For simplicity, we shall consider the case when the symmetry of the analyzed sublattice is simple cubic. Then, if we bear in mind that $n_0 \propto \nu_{s0}^3$, we can assume

$$\nu_{s0} \approx 1. \tag{4.17}$$

If the surface structure consists of just one layer, the surface energy is close to the statistical average

$$n_s \varkappa \left[1 + 4\frac{p_s}{n_s}\left(1 - \frac{p_s}{n_s}\right)\right],$$

where p_s is the number of particles in the "+" state on the surface, distributed in n_s cells. Averaging this value over the occupancy of the surface layer, we find that $\zeta = \frac{5}{3}$. Calculation of the quantity in Eq. (4.14) carried out employing this value gives

$$\varphi(^5/_3) = -0.11. \tag{4.18}$$

However, these results postulate implicity that a nucleus grows like a cabbage: a given layer does not begin to fill until all the vacancies are occupied in the layer below. This corresponds to a special set of paths. We can easily see that then the number of paths needed to fill a single layer is $n_s!$ instead of $(n_s)^{n_s}$. Therefore, the total number of paths decreases compared with that given by Eq. (4.15). Allowance for this change has the result that the expression for the transition amplitude (4.16) has

$$c \approx 2.44. \tag{4.19}$$

The values of the arguments of the exponential functions in Eqs. (4.16) and (4.10) are then close although that in Eq. (4.10) is greater than in Eq. (4.16).

The loss in the number of paths in the cabbage-like growth of a nucleus is not compensated by the gain in the energy. The optimal process is the formation of a surface structure from several layers. For example, in the case of a two-layer structure on the assumption that the upper layer begins to fill when the lower is half-filled, we find that direct calculations of the energy and number of paths give

$$c \approx 2.14. \tag{4.20}$$

In this case the coefficient in front of n_0 in the argument of the exponential function occurring in the expression for the transition amplitude is now smaller than in Eq. (4.10) but the values are still close.

In the opposite case of a considerable thickness of the surface layer, we have $\zeta = 3$ and

$$\varphi(3) \approx 0.81. \tag{4.21}$$

Then, assuming that the total number of the compact paths given by Eq. (4.15) is realized [the amplitude of the transition is then given by Eq. (4.16) with the value of φ from Eq. (4.21)], we find that the argument of the exponential function in Eq. (4.16) again has a value slightly greater than in Eq. (4.10).

We may conclude from these results that the optimal intermediate compact nuclei are obtained for limited broadening of the surface layer. The extremal value is most probably close to that given by Eq. (4.21). We shall not go beyond this approximate estimate.

It is clear that the evolution of a nucleus along the compact paths has advantages over the noncompact paths but the difference is not very great. This difference may increase in the case of sublattices with larger numbers of the nearest neighbors.

5. INTERACTION WITH PHONONS

We shall show directly in the next section that the kinetics of a transition depends strongly on the interaction of a nucleus with the phonon subsystem. If allowance is made for the atomic vibrations in a crystal, the original Hamiltonian (2.1) should be supplemented by the terms

$$H_{ph}^0 + V, \quad V = V_1 + V_2 + V_3. \tag{5.1}$$

Here, H_{ph}^0 is the Hamiltonian of the phonon subsystem for a homogeneous metastable phase; $V_{1,2,3}$ are the terms in the Hamiltonian which appear when allowance is made for the dependences of ε, $\varkappa$, and ξ, respectively, on the atomic displacements:

$$V_1=\sum_{\mathbf{m}} B_1(\mathbf{m})n_{\mathbf{m}+},\quad V_2=\sum_{\mathbf{mg}} B_2(\mathbf{mg})n_{\mathbf{m}+}n_{\mathbf{m}+\mathbf{g},-},$$
$$V_3=-\xi\sum_{\mathbf{m}} B_3(\mathbf{m})\,(a_{\mathbf{m}+}{}^{+}a_{\mathbf{m}-}+a_{\mathbf{m}-}{}^{+}a_{\mathbf{m}+}). \qquad (5.2)$$

Here, B_1, B_2, and B_3 are the operators in the phonon space. The operators V_1 and V_2 are diagonal in the $|\alpha_n)$ representation. For each such state the Hamiltonian $H_{ph}^0+V_1+V_2$ governs the renormalized phonon spectrum. The corresponding eigenstates are denoted by $|\gamma\alpha_n)$, where γ represents a set of the occupation numbers. Consequently, the eigenstates of the Hamiltonian

$$\mathscr{H}_0=H_0+H_{ph}{}^0+V_1+V_2 \qquad (5.3)$$

have the form

$$|\alpha_n)\,|\gamma\alpha_n). \qquad (5.4)$$

We shall first ignore V_3. Then, expanding the wave function of the system in terms of the states described by Eq. (5.4) [instead of those described by Eq. (2.6)], we find that the complete Hamiltonian $\mathscr{H}_0+H_1$ is described by the Schrödinger equation in the form similar to that of Eq. (2.7). We can again find the rising and falling solutions corresponding now to a fixed value of γ. The only difference is that the matrix element of H_1 between the states α_n and $\alpha_{n\pm1}$, equal simply to ξ in the earlier treatment, becomes

$$\xi(\gamma\alpha_n|\gamma\alpha_{n\pm1})=\xi f_{\alpha_n}(\gamma), \qquad (5.5)$$

i.e., allowance is made for the "polaron" effect when a single unit cell is considered. For simplicity, we shall assume that the polaron factor is independent of the configuration, i.e., of α_n and also that the local polaron effect is fairly weak. Then, the coefficients in the expansion of the wave function are given by Eqs. (2.15) and (2.17) subject to just one substitution

$$\xi\to\tilde{\xi}=\xi f(\gamma). \qquad (5.6)$$

The phonon-diagonal matrix element of a coherent transition to the state with a critical nucleus is given by the earlier expression (3.6) subject to the substitution (5.6). It is interesting to note that if, as usual, f is represented in the form

$$f=e^{-\Phi_1} \qquad (5.7)$$

($\Phi_1\ll1$), the new matrix element has an additional, compared with Eq. (3.6), factor

$$F=e^{-n_0\Phi_1} \qquad (5.8)$$

typical of the polaron effect in the transition as a whole. Then,

$$\tilde{H}_{rl}=H_{rl}e^{-n_0\Phi_1}. \qquad (5.9)$$

A transition accompanied by creation of a critical nucleus and simultaneous excitation of the phonon subsystem is described by the operator V_3 of Eq. (5.2), which is not diagonal in respect of α_n. Interaction represented by this operator will always be assumed to be small compared with H_1 of Eq. (2.3). In this case the matrix element of the transition is

$$H_{rl}^{\gamma\gamma'}=(V_3)_{rl}^{\gamma\gamma'}=\sum_{n\alpha_n}\sum_{n'\alpha_{n'}}\Psi_{\alpha_n}{}^{r}\Psi_{\alpha_{n'}}^{r'}(V_3)_{\alpha_n\alpha_{n'}}^{\gamma\gamma'}.$$

We shall assume, for simplicity, that the phonon matrix element in this expression is independent of α_n:

$$(\gamma\alpha_n|B_3|\gamma'\alpha_{n'})\approx b_{\gamma\gamma'}\quad(n'=n\pm1). \qquad (5.10)$$

Then, the calculation of the matrix element becomes similar to the calculation of Eq. (3.6) (except that now we have an additional sum over n). In this way we obtain

$$H_{rl}^{\gamma\gamma'}\approx b_{\gamma\gamma'}n_0e^{-n_0\Phi_1}H_{rl}, \qquad (5.11)$$

where H_{rl} has the value given by Eq. (3.6).

The matrix element (5.10) describes the local "shaking" representing the excitation of the phonon subsystem accompanying the transfer of one particle to a different state. In principle, this may occur during any stage of a transition and this is why the factor n_0 appears.

6. EQUATION FOR THE DENSITY MATRIX. TRANSITION PROBABILITY

The discrete structure of the energy levels corresponding to the formation of a nucleus subject to the self-evident condition $|H_{rl}|\ll\varepsilon,\varkappa$ makes it generally necessary to allow for the interaction with the phonon subsystem in transitions of the kind considered here. A consistent description of the transition kinetics in the presence of coherent and noncoherent processes requires solution of the relevant equation for the density matrix.

We shall use $\tilde{H}$ to denote the Hamiltonian whose matrix elements describe a coherent transition diagonal in respect of the phonon variables [see Eq. (5.9)]. In accordance with the nature of selection of the "left-hand" and "right-hand" wave functions (Sec. 3), all the matrix elements in the Hamiltonian $\tilde{H}$ vanish with the exception of that corresponding to a transition to the state α_{n_0} and to the corresponding degenerate states, as well as possibly to a certain number of levels in the immediate vicinity of $E=0$ and directly to the right. All these levels will be denoted by the index r'.

The noncoherent interaction with phonons is characterized by two mechanisms: (1) a transition to a transcritical state accompanied by shaking described by the matrix elements (5.11); (2) fluctuation jitter and decay of a level r' corresponding to an irreversible interaction with the phonon subsystem. The corresponding Hamiltonian of the interaction with phonons will be denoted by H'.

We shall now introduce the density matrix for a nucleus:

$$f=\mathrm{Sp}_{ph}\rho. \qquad (6.1)$$

Here, ρ denotes the total density matrix of the system and the trace (spur) applies to the phonon variables. Then, assuming that the interaction with phonons is weak and retaining only the terms quadratic in respect of this interaction, we obtain (see, for example, Refs. 3 and 4)

$$\partial f/\partial t+i[\tilde{H},f]=-J, \qquad (6.2)$$

$$J=\mathrm{Sp}_{ph}[H',[\underline{H}',f\rho_{ph}]]. \qquad (6.3)$$

Here, the brackets contain a commutator; ρ_{ph} is the equilibrium density matrix of phonons;

$$\underline{H}'=\int_0^\infty dt\exp[-i(\tilde{H}+H_{ph})t]H'\exp[i(\tilde{H}+H_{ph})t]. \qquad (6.4)$$

The characteristic time of the transition kinetics is

governed by the tunnel leakage amplitude. This time is long compared with any other times of the problem and, in particular, with the time taken to drop from any level r' when the process is accompanied by phonon emission. Therefore, we may assume that

$$f_{rr'}\approx 0, \quad f_{r'r'}\approx 0. \tag{6.5}$$

Bearing this in mind, we shall rewrite Eq. (6.2) for the density matrix components in the form

$$\left.\begin{aligned} \frac{\partial f_{lr'}}{\partial t}+i(E_l-E_{r'})f_{lr'}+i\mathcal{H}_{lr'}(f_{r'r'}-f_{ll})=-J_{lr'},\\ \frac{\partial f_{ll}}{\partial t}+i\sum_{r'}\mathcal{H}_{lr'}(f_{r'l}-f_{lr'})=-J_{ll}. \end{aligned}\right\} \tag{6.6}$$

An analysis of Eq. (6.3) shows that the dominant term of the nondiagonal element of the collision integral is

$$J_{lr}=\Omega_{lr}f_{lr}, \tag{6.7}$$

where

$$\Omega_{lr}=\pi\sum_{\gamma\gamma'}\rho_{ph}(\gamma)\delta(E_\gamma-E_{\gamma'})|(H')_{rr}^{\gamma\gamma'}|^2$$
$$+\pi\sum_{\gamma\gamma'}\sum_{r'(\neq r)}\rho_{ph}(\gamma)\delta(E_\gamma-E_{\gamma'}+E_r-E_{r'})|(H')_{rr'}^{\gamma\gamma'}|^2. \tag{6.8}$$

Here, E_γ is the energy of the phonon subsystem.

The first term in Eq. (6.8) describes decay of the coherent phase as a result of scattering of phonons by a nucleus. The second term is associated with transitions of a real nucleus from a level r to lower level accompanied by phonon emission. This term predominates: in the limit $T\to 0$ it remains finite, whereas the first term then vanishes.

Using Eqs. (6.3) and (6.4) and ignoring the contribution due to the nondiagonal elements of the density matrix, we obtain directly the following expression for the diagonal element J_{ll}:

$$J_{ll}=\frac{1}{\tau_l}f_{ll}, \tag{6.9}$$

where

$$\frac{1}{\tau_l}=2\pi\sum_{\gamma\gamma'}\sum_{r'}\rho_{ph}(\gamma)\delta(E_\gamma-E_{\gamma'}+E_l-E_{r'})|(H')_{lr'}^{\gamma\gamma'}|^2. \tag{6.10}$$

Equation (6.9) is derived using Eq. (6.5).

The expression (6.10) describes the noncoherent decay time of a metastable state accompanied by excitation of the phonon subsystem. The matrix element of the transition in the expression (6.10) is given by Eq. (5.11).

We shall substitute Eq. (6.7) on the right-hand side of the first equation in the system (6.6). The expression for Ω_{lr} given by Eq. (6.8) does not contain the overlap integral and it is finite in the limit $T\to 0$. In considering the kinetics of a subbarrier transition we can always ignore the time derivative on the left-hand side of the first equation of (6.6) because the corresponding reciprocal transition time is proportional to the square of the overlap integral. Consequently, we obtain

$$f_{lr'}=\frac{i\mathcal{H}_{lr'}}{i(E_l-E_{r'})+\Omega_{lr'}}(f_{ll}-f_{r'r'})$$

(we are retaining here $f_{r'r'}$ only to demonstrate directly the Hermitian symmetry on transposition $l\rightleftarrows r'$).

Substituting the above relationship into the second equation of the system (6.6), we then obtain with the aid of Eq. (6.9)

$$\partial f_{ll}/\partial t=-Wf_{ll}, \tag{6.11}$$

$$W=\sum_{r'}|\mathcal{H}_{lr'}|^2\frac{2\Omega_{lr'}}{(E_l-E_{r'})^2+\Omega_{lr'}^2}+\frac{1}{\tau_l}. \tag{6.12}$$

As in the problem of quantum subbarrier diffusion (see Refs. 3 and 4), the probability of decay of a metastable state accompanied by creation of a transcritical nucleus W is the sum of the probabilities of the coherent [the first term in Eq. (6.12)] and noncoherent transitions.

If $\Omega_{lr}\ll\varepsilon$, the sum over r' in Eq. (6.12) includes a contribution only from the nearest level, which in fact represents a critical nucleus. In principle, this level is degenerate and the degenerate and the degree of degeneracy g_0 depends on the surface configuration of the critical nucleus. We shall denote Ω_{lr} for this level by Ω_0 and the energy difference E_l-E_r by ΔE. Bearing in mind Eqs. (5.9), (3.6), and (4.16), we obtain

$$W_{coh}=e^{-2n_0\Phi_1}|H_{lr}|^2\frac{2\Omega_0}{\Delta E^2+\Omega_0^2}g_0$$
$$\approx\exp\left\{-2n_0\left(\Phi_1+\ln\frac{\varkappa c}{\xi}\right)\right\}(\varkappa c)^2n_0^{4/3}\frac{\Omega_0}{\Delta E^2+\Omega_0^2}g_0. \tag{6.13}$$

The value of the constant c can be found on the basis of the results in Sec. 4. The preexponential factor in the matrix element corresponds to the evolution of a compact nucleus [see Eq. (4.16)].

If under real conditions the degeneracy is lifted and the spectrum is quasicontinuous, then

$$W_{coh}\approx 2\pi|H_{lr}|^2g(0)\approx\exp\left\{-2n_0\left(\Phi_1+\ln\frac{\varkappa c}{\xi}\right)\right\}\cdot 2\pi(\varkappa c)^2n_0^{4/3}g(0). \tag{6.14}$$

Here, $g(0)$ is the density of states to the left of $E=0$. (It is interesting to note that in this case the expression for W_{coh} has apparently a structure of the kind obtained in the quantum theory of decay accompanied by a transition to a continuous spectrum and the preexponential factor ceases to depend in any way on the phonon subsystem and temperature.)

The explicit form for the probability of a noncoherent transition can be obtained if we use Eq. (5.11) and substitute it in Eq. (6.10). On the basis of Eq. (2.5) the matrix element $H_{lr'}$ decreases considerably for states of lower energy. Therefore, in the sum over r' in Eq. (6.10) we can retain only the level corresponding to a critical nucleus. [If ΔE is found to be very small and there is a correspondingly low density of states in the phonon spectrum at $\omega\sim\Delta E$, it may be necessary to allow for transitions to lower levels in Eq. (6.10).] Then,

$$W_{incoh}\approx e^{-2n_0\Phi_1}|H_{rl}|^2n_0^2\gamma(\Delta E)$$
$$\approx\exp\left\{-2n_0\left(\Phi_1+\ln\frac{\varkappa c}{\xi}\right)\right\}(\varkappa cn_0n_s)^2\gamma(\Delta E), \tag{6.15}$$

where

$$\gamma(\Delta E)=2\pi\sum_{\gamma\gamma'}\rho_{ph}(\gamma)\delta(E_\gamma-E_{\gamma'}+\Delta E)|b_{\gamma\gamma'}|^2. \tag{6.16}$$

Comparing Eqs. (6.15) and (6.13), we can see that the main exponential dependence is the same in both cases. The only difference is in the preexponential factor. Both probabilities remain finite in the limit $T\to 0$. The dependences change with rising temperature. For ΔE

$< \Omega_0$, the two preexponential factors exhibit opposite behavior: in the coherent transition case the factor decreases with rising temperature, whereas in the noncoherent case, it rises.

We have considered so far the probability of creation of a transcritical nucleus in a definite region of a metastable phase. If regions of the order of n_0 are regarded as independent, we find that the probability of creation of a single transcritical nucleus per unit volume is

$$W=\frac{N_0}{n_0}(W_{coh}+W_{incoh})\left[\frac{1}{cm^3\cdot sec}\right], \tag{6.17}$$

where N_0 is the number of unit cells per unit volume.

7. KINETICS AT FINITE TEMPERATURES

The kinetics of a phase transition is considered in the preceding section on the assumption that $T \approx 0$ and that a metastable system is in its lowest energy state. An increase in temperature results in filling of higher energy states which correspond to the appearance of particles or clusters in the "+" position. These levels are characterized by a higher probability of tunnel transitions.

In view of the exponential smallness of the subbarrier transition probability, we may assume that an equilibrium distribution of the "left-hand" states is established in a time which is short compared with that deduced from the decay probability. In this case the total probability of creation of a transcritical nucleus is given by

$$W(T)=\sum_l e^{-E(l)/T}W_l \Big/ \sum_l e^{-E(l)/T}, \tag{7.1}$$

where $E(l)$ is measured from $E(0)$, which is the energy of the lowest state of the metastable phase in question. The index l describes successive states α_m located to the left.

An increase in l causes the probability W_l to rise exponentially because of a reduction in the effective width of the barrier. Then, W_l is described by an expression of the (6.17) type provided n_0 in Eqs. (6.13) and (6.15) is replaced with n_l ($<n_0$). Consequently, we have to consider the optimal path of a transition, i.e., the optimal combination of activated excitation and tunnel leakage. This optimal path corresponds to the minimum value of the function

$$\frac{1}{T}E(l)+A(l)-A(0), \tag{7.2}$$

where $A(l)$ is the argument of the exponential function in W^l_{coh} and W^l_{incoh} given by Eqs. (6.13) and (6.15) for a tunnel transition from a level l. The first excited level has the energy $E_1=\varkappa v$. The value of n_l corresponding to this energy can be found by considering a tunnel transition involving the compact paths (Sec. 4).

It follows from Eq. (2.8) that

$$n_0-n_l \approx 3\varkappa v/\varepsilon.$$

When temperature increases from $T=0$, the first excited level becomes important at a temperature

$$T_1 \approx \frac{E_1}{A(0)-A(1)} \approx \frac{\varepsilon}{3A_0}, \quad A_0=\frac{A(0)}{n_0}=2\left[\ln\frac{\varkappa c}{\xi}+\Phi_1\right]. \tag{7.3}$$

However, since $E''(l)<0$, and the derivative $A'(l)$ is nearly constant, the minimum value of the function (7.2) at $T=T_1$ is displaced and coincides with the values at the top of the barrier. Then, the transition from pure quantum subbarrier leakage from a lower level to the classical transbarrier motion occurs suddenly at $T_0<T_1$, bypassing intermediate levels (a similar result is given in Ref. 1).

The transition temperature is given by the following expression [see Eqs. (4.1), (2.14), and (7.3)]:

$$T_0=\frac{E_m}{A(0)}=\frac{4}{27}\frac{\varepsilon}{A_0}. \tag{7.4}$$

At temperatures $T>T_0$ the transition kinetics is of the classical activated type. The dynamics near the top of the barrier is governed by the operator V_3 of Eq. (5.2), which is responsible for the inelastic (in respect of phonons) rise in the number of particles in a nucleus. Using Eqs. (5.11), (6.15), and (6.17), we find that the probability of creation of a transcritical nucleus per unit volume is

$$\left.\begin{aligned} W &\approx \xi^3 f^2 \gamma(\Delta E_m) g_m e^{-E_m/T}\frac{N_0}{n_0}\left[\frac{1}{cm^3\cdot sec}\right]; \\ E_m &= {}^4/_{27}\varepsilon n_0. \end{aligned}\right\} \tag{7.5}$$

Here, the index m applies to the top of the barrier and g_m is the degree of degeneracy characteristic of the levels which are also near the top of the barrier.

It should be pointed out that in a more rigorous classical analysis of the kinetics one would have to allow for changes in the distribution function compared with the Boltzmann type, which occurs in the direct vicinity of the top of the barrier. This results in some renormalization of the preexponential function in Eq. (7.5). We shall not pursue this point any further.

In conclusion, it should be noted that—according to Eq. (7.4)—the temperature of the transition from the quantum to the classical knietics depends on the scale of the "supersaturation" governed by the parameter ε.

8. CONCLUDING REMARKS

The structure of the expressions (3.6), (4.16), and (4.10) obtained above for the amplitude of subbarrier creation of a critical nucleus of the new phase is fundamentally different from the structure of the expressions obtained in Ref. 1. This is largely due to the discrete nature of the configuration space in which tunneling takes place. The formulas obtained are not quasiclassical and cannot be reduced to the determination of the action. Moreover, an analysis shows that each step in the discrete particle-number space may correspond to the change in the action by an amount $\gg\hbar$. It is then fundamentally impossible to go over from the finite-difference equation (2.7) to an ordinary differential equation in the limit of a large number of particles in a nucleus, i.e., in the limit $n_0 \gg 1$. This very general problem will be analyzed separately in the case of subbarrier relaxation of a large spin, whose kinetics is described by an expression of the (2.7) type but the configuration paths are fixed.

The above results are obtained on the assumption that

the number of particles in a critical nucleus is fairly large. However, it is important to note that the adopted method can be applied effectively to those cases when the size of a critical nucleus is finite and the macroscopic description is difficult. This becomes particularly clear when we turn to the results of Sec. 4, which demonstrate proximity of the subbarrier transition amplitudes for the evolution of a compact nucleus and purely random formation of the interior of a critical nucleus. In particular, the results make it possible to analyze an intrinsic class of problems corresponding to decay of a metastable phase by formation of clusters with very much modified atomic configurations. It should be noted that in the latter case the discrete nature of the energy levels is very pronounced and the inelastic interaction with the phonon subsystem, considered in Sec. 5, may be of fundamental importance.

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Motion of domain walls in an external magnetic field

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It is shown that in a range of magnetic fields exceeding the known value of the Walker limiting field, there can exist stationary-profile waves corresponding to moving domain walls with a definite internal structure.

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1. Investigations of steady-state motions of domain walls[1,2] have shown that with allowance for uniaxial anisotropy of the ferromagnet, dissipation, and an external magnetic field directed along the anisotropy axis, the velocity of a stationary-profile wave is bounded from above by the value

$$U_- = 2|\gamma| \frac{(AK)^{1/2}}{M_s} u_-(\varepsilon). \tag{1.1}$$

Here the following notation is used:

$$u_-(\varepsilon) \equiv (1+\varepsilon)^{1/2} - 1, \quad \varepsilon = 2\pi M_s^2/K \tag{1.2}$$

γ is the gyromagnetic ratio, A and K are the exchange and the uniaxial-anisotropy energy constants, and M_s is the saturation magnetization. Furthermore, as was first mentioned by Walker,[1] the range of existence of stationary motions of domain walls of the Bloch-Landau or Néel type, characterized by a constant orientation of the plane of rotation of the magnetic moment, is bounded from above by a value of the magnetic field equal to

$$H_1 = \frac{2K}{M_s} h_1, \quad h_1 = \frac{1}{2}\alpha\varepsilon, \tag{1.3}$$

α is the damping parameter.

A solution of the Landau-Lifshitz equations that corresponds to a stationary-profile wave with a constant orientation of the plane of rotation of the magnetic moment cannot be continued into the external magnetic field range $H_z > H_1$.

The present paper discusses the possibility of existence of stationary-profile waves in the external-field range $H_z > H_1$. Waves of this type correspond to stationary motions of domain walls that are characterized by a definite internal structure. Specifically, turning of the plane of rotation of the magnetic moment leads to the appearance of a definite number of "internal" domain walls, because of the fact that the projection of the magnetic moment on the direction of the external field changes sign several times during passage from the region of uniform magnetization along the external field to the region of uniform magnetization opposite to the external field.

In the case considered, the system of Landau-Lifshitz equations has the form

$$\begin{gathered}\theta'' - (1+\omega^2+\varepsilon\cos^2\varphi)\sin\theta\cos\theta - h_z\sin\theta = u\omega\sin\theta - \alpha u\theta', \\ (\omega\sin^2\theta)' + \varepsilon\sin^2\theta\cos\varphi\sin\varphi = -u\theta'\sin\theta - \alpha u\omega\sin^2\theta. \\ \omega = \varphi'.\end{gathered} \tag{1.4}$$

Here u is the velocity of the stationary-profile wave divided by the characteristic velocity $2|\gamma|(AK)^{1/2}/M_s$; h_z is the external magnetic field divided by the anisotropy field $2K/M_s$; θ and φ are the polar and azimuthal angles of the vector magnetic moment; the differentiation is with respect to the variable $\xi \equiv x - ut$ (the spatial variable x has been divided by the characteristic thickness $(A/K)^{1/2}$ of a Bloch wall).

For a stationary-profile wave, corresponding to

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