

# Dissipative nucleation and phase-separation kinetics of $^3\text{He}$ - $^4\text{He}$ liquid solutions at low temperatures

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Nucleation in two-component metastable systems as compared with single-component systems is analyzed. In this case the main dissipative mechanism becomes diffusion, which predominates over viscosity near the phase equilibrium curve or when the critical nuclei are sufficiently large. This results in new temperature dependences in the exponential characterizing the probability of quantum tunneling. A characteristic of nucleation in a superfluid is that the lifetime of the metastable phase is very sensitive to the presence of a low concentration of impurities.

## 1. INTRODUCTION

Investigation of the kinetics of nucleation with phase transformations in a metastable system at low temperatures is undoubtedly of interest as a method for observing quantum phenomena at the macroscopic level. The point is that at sufficiently low temperatures, instead of the standard activation mechanism caused only by thermal fluctuations, the transition from the metastable state into a stable state also occurs with the help of quantum-mechanical fluctuations corresponding to subbarrier tunneling.<sup>1,2</sup> One metastable system where observation of macroscopic tunneling is possible is the helium isotopes, in which phase transitions exist right down to absolute zero. Thus in superfluid helium  $^4\text{He}$  supercooling of the liquid was observed experimentally with crystallization at temperatures below 2 K. The average lifetime of the metastable liquid increases as the temperature decreases.<sup>3,4</sup>

Supersaturated liquid or solid  $^3\text{He}$ - $^4\text{He}$  solutions, in which a phase transition also exists down to absolute zero, can be used as a possible metastable system for investigating macroscopic tunneling. Phase-separation processes in solid  $^3\text{He}$ - $^4\text{He}$  solutions at low temperatures have been systematically investigated in recent years with the help of the NMR method by V. A. Mikheev *et al.*<sup>5</sup> Phase separation of liquid solutions  $^3\text{He}$ - $^4\text{He}$  near the tricritical point was studied in Ref. 6. However the temperature corresponding to the tricritical point is still too high for a phase transition to occur through the mechanism of subbarrier quantum tunneling. In this paper we shall study the kinetics of nucleation and phase separation at low temperatures only of the liquid  $^3\text{He}$ - $^4\text{He}$  solutions.

The phase diagram of a liquid  $^3\text{He}$ - $^4\text{He}$  solution is well known (Fig. 1).<sup>7</sup> At temperatures  $T < T_t = 0.87$  K the solubility of  $^3\text{He}$  in liquid  $^4\text{He}$  is limited,<sup>8</sup> and solutions with a molar concentration of  $^3\text{He}$  corresponding to the concentration range  $x_1 < x < x_u$  are unstable and decompose into two phases with temperature and pressure dependent concentrations  $X_1(T, P)$  and  $X_u(T, P)$ . The lower phase  $x < x_1$  is a solution of  $^3\text{He}$  in superfluid  $^4\text{He}$ . As  $T \rightarrow 0$ , depending on the pressure, the  $^3\text{He}$  content lies in the range  $0.065 < x_1 < 0.094$ . The upper phase  $x > x_u$ , conversely, is enriched with  $^3\text{He}$  and is a normal (not superfluid) solution.

The quantum kinetics of nucleation and phase separation of dilute liquid solutions  $^3\text{He}$ - $^4\text{He}$  at low temperatures were first studied in Ref. 9. The analysis in Ref. 9 was per-

formed under the assumption that no dissipative processes occur in the medium and referred only to separation of a supersaturated superfluid solution, which refers to the region of the lower phase. Below we shall study the quantum kinetics of phase separation in the upper phase, corresponding to a supersaturated solution of the normal liquid, and in both types of solutions we shall also study the effect of dissipative processes occurring in the liquid solution on the quantum kinetics of nucleation at low temperatures. Nucleation processes in the upper and lower phases will evidently occur differently as a result of the difference in the flow of dissipative processes in the normal and superfluid solutions. It is well known that dissipative processes play an important role in the kinetics of decomposition of the metastable state. These processes lead to a decrease of the rate of decomposition at zero temperature<sup>10</sup> and sharply change the temperature dependence of the average lifetime of the metastable state.<sup>11</sup> The effect of dissipative processes on the quantum kinetics of phase transitions occurring in a medium consisting of a one-component liquid was studied in Ref. 12.

The formation and growth of a nucleus of a stable phase

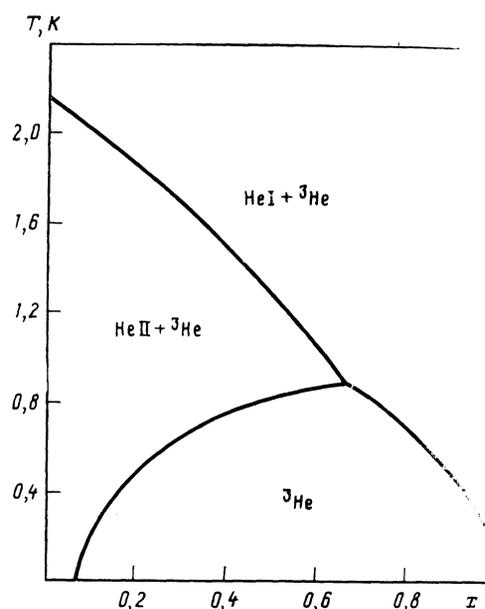


FIG. 1. Phase diagram of a liquid  $^3\text{He}$ - $^4\text{He}$  solution.

in a medium is unavoidably accompanied by dissipation of the total energy of the nucleus. Two types of dissipative processes occur in both one- and two-component media: heat conduction, governed by the presence of a temperature gradient in different sections of the medium, and viscosity (internal friction), which is manifested in irreversible momentum transfer from sections of the medium which move with high velocity into sections of the medium which move with low velocity. The presence of a second component in the solution leads to an additional mechanism of dissipation of energy of the nucleus as compared with a one-component system. The point is that as a nucleus of the stable phase forms and grows the distribution of the concentration of the solution changes. The concentration of the solution can change by two methods. The first method involves purely mechanical mixing of the liquid, when each element of the liquid moves together with the liquid as a whole with constant composition. If the dissipative processes of heat conduction and viscosity (internal friction) can be neglected with such mechanical motion, then such a change of the concentration occurs in a thermodynamically reversible manner without dissipation of energy by the nucleus. The second method is diffusion, when the concentration of each element of the liquid is equalized by means of molecular transfer of the solution components. Diffusion is an irreversible process and together with heat conduction and viscosity is an additional mechanism of energy dissipation in the solution.

The formation and growth of nuclei of a stable phase in a metastable medium involves the overcoming of an energy barrier, whose magnitude depends on the degree of metastability of the system, and this is accompanied by dissipation of the total energy of the nucleus. This motion can be described by adding to the standard potential forces the forces of friction, which prevent growth of the nucleus. With the help of the approach developed in Refs. 10 and 11 we shall find the probability of subbarrier formation of the critical nucleus. As in Ref. 12, the description of the action of dissipation on the probability of decomposition of the metastable phase reduces to finding the effective action that is nonlocal in the imaginary time. In what follows we shall study separately the case of phase separation of a normal solution and the case of phase separation of a superfluid solution.

## 2. QUANTUM FORMATION OF NUCLEI IN A NORMAL SOLUTION

We study phase separation of a liquid supersaturated two-component solution, which is a nonsuperfluid (normal) liquid, for example, phase separation of the solution  ${}^3\text{He}-{}^4\text{He}$  in the region of the upper phase. We designate by  $\rho$  the density of the solution and we characterize the composition of the solution by the concentration  $c$ , which we define as the ratio of the mass of one component of the solution to the total mass of the liquid in a given volume. For simplicity we assume that the thickness of the transitional layer between the phases is small compared with the size of a nucleus. Then the optimal distribution of density and concentration corresponds to a spherical fluctuation of the stable phase with radius  $R$  and potential energy of the nucleus  $U(R)$ , which has the standard form

$$U(R) = 4\pi\sigma R^2 (1 - R/R_c). \quad (1)$$

Here  $\sigma$  is the surface tension of the interphase boundary and  $R_c$  is the critical size of the nucleus, determined from the condition  $U(R_c) = 0$ . The critical size  $R_c$  characterizes the degree of departure of the system from equilibrium and is inversely proportional to  $\Delta x$ , where  $\Delta x$  is the supersaturation of the solution. The relation between the molar concentration  $x = N_3/(N_3 + N_4)$  and the concentration  $c$  is given by the simple formula

$$x = m_4 c / [m_3 + (m_4 - m_3) c], \quad (2)$$

where  $m_3$ ,  $N_3$  and  $m_4$ ,  $N_4$  are the mass and the number of  ${}^3\text{He}$  and  ${}^4\text{He}$  particles.

The formation and growth of a nucleus of a new phase is unavoidably accompanied by the motion of the liquid. This motion is connected with the transport of the components of the solution to the nucleus or away from it. In the description of the dynamics of a macroscopic nucleus of a new phase several macroscopic variables are usually distinguished: the volume, density, pressure, concentration, velocity, etc. In so doing, it is assumed that all other "unimportant" variables are uniquely determined by the macroscopic variables that are singled out. This means that the relaxation times for establishing an incomplete equilibrium, i.e., equilibrium with respect to the "unimportant" variables with fixed macrovariables, are short compared with the relaxation times necessary for establishing the equilibrium values of the macrovariables. This dynamics of the "unimportant" variables has the character of adiabatic adjustment and occurs reversibly and without dissipation. The processes involved in establishing equilibrium, whose characteristic relaxation times are longer than the characteristic growth time of the nucleus, make the motion of the liquid irreversible and therefore result in dissipation of the total energy of the nucleus.

For what follows we need to find the equation of motion that describes the growth of a nucleus. The flow of the liquid surrounding the nucleus leads to the fact that the nucleus has not only the potential energy (1) but also kinetic energy. These energies are dissipated as the nucleus grows. In order to find the kinetic energy of the nucleus and the amount of the total energy of the nucleus that is dissipated it is necessary to know the distribution of the velocity field  $\mathbf{v}(\mathbf{r})$  and the concentration distribution  $c(\mathbf{r})$  in both phases, expressed in terms of the radius of the nucleus  $R(t)$  and the velocity of its boundary  $\dot{R}(t)$ . At the boundary of the nucleus, which is a surface of discontinuity, boundary conditions expressing the continuity of the mass flux of the liquid and the mass flux of one component of the solution must be satisfied, i.e.,

$$\rho'(R) [v'(R) - \dot{R}] = \rho(R) [v(R) - \dot{R}], \quad (3)$$

$$\rho'(R) c'(R) [v'(R) - \dot{R}] + i'(R) = \rho(R) c(R) [v(R) - \dot{R}] + i(R). \quad (4)$$

Here the vector  $\mathbf{i}$  is the diffusion flux density.

In what follows we neglect the compressibility of the liquid, since for large nuclei, for which the departure from phase equilibrium is small, the velocity of motion is small compared with the velocity of sound  $s$ . Taking into account the compressibility of the liquid corresponds to taking into

account in the kinetic energy terms of order  $\dot{R}/s \ll 1$  and leads to small corrections to the transition probability.<sup>13</sup> Thus the instantaneous velocity field and the concentration distribution are determined from the equations of continuity for the metastable phase

$$\operatorname{div} \rho \mathbf{v} = 0, \quad (5)$$

$$\operatorname{div} (\rho c \mathbf{v} + \mathbf{i}) = 0 \quad (6)$$

and analogous equations for the stable phase.

The first equation in Eq. (5), together with Eq. (3), gives the well-known velocity profile of the liquid<sup>1</sup>

$$v(r) = \begin{cases} -\frac{\rho' - \rho}{\rho} R \frac{R^2}{r^2}, & r > R(t) \\ 0, & r < R(t) \end{cases} \quad (7)$$

The kinetic energy of the nucleus, defined as the integral of the kinetic energy density over the volume of the entire system, assumes the standard form<sup>1</sup>

$$K = \frac{1}{2} M(R) \dot{R}^2, \quad M(R) = 4\pi \frac{(\rho' - \rho)^2}{\rho} R^3. \quad (8)$$

In order to find from Eqs. (4) and (6) the concentration distribution  $c(\mathbf{r})$ , which is needed for calculating the dissipation in the system, it is necessary to have an expression for the diffusion flux. For small concentration gradients in the system and when the mean-free path of excitations  $l(T)$  in the metastable phase is small compared with the dimensions of the nucleus, the hydrodynamic expression for the diffusion flux can be employed:<sup>14</sup>

$$\mathbf{i} = -\rho D \nabla c, \quad (9)$$

where  $D$  is the diffusion coefficient, which, generally speaking, depends on the concentration. From Eqs. (4) and (6), substituting Eq. (7), we obtain the following concentration distribution in the system:

$$c(r) = \begin{cases} c - (c' - c) \frac{\rho'}{\rho} \frac{R \dot{R}}{D(c)} \frac{R}{r}, & r > R(t) \\ c', & r < R(t) \end{cases} \quad (10)$$

It is obvious that as the nucleus grows one component of the solution will be depleted near the surface of the nucleus, if the new phase corresponds to a higher concentration of the component  $c' > c$  and, vice versa, enrichment with the component will occur if we have  $c' < c$ . The distribution (10) in such a simple form occurs only if the nucleus grows so slowly that terms proportional to  $\dot{R}^2$  can be neglected and the diffusion coefficient  $D$  can be assumed, to the same accuracy, to be constant and independent of the concentration  $c$ . This restriction presumes that the following condition is satisfied:

$$\frac{R^2}{D} \ll \frac{R}{\dot{R}(\rho' - \rho)/\rho}, \quad (11)$$

which means that the characteristic time for a particle to diffuse over the distance  $R$  is short compared with the time required for the particle to traverse the same distance  $R$  purely mechanically together with the flow of the liquid. The condition (11) is also equivalent to studying motion corresponding to small Reynolds numbers:

$$\operatorname{Re} = v(R) R / D \ll 1. \quad (12)$$

It is obvious from Eqs. (7) and (10) that the growth of the nucleus in the solution is accompanied both by flow of the metastable liquid with the nonuniform velocity  $\mathbf{v}(\mathbf{r})$  of the macroscopic motion and by a nonuniform concentration distribution in the volume. This means that dissipative processes associated with relaxation of the solution to complete thermodynamic equilibrium arise in the solution. One dissipative process, governed by irreversible viscous transfer of momentum from sections of the medium moving with higher velocity to sections moving with lower velocity, also occurs in a one-component liquid.<sup>12</sup> The second dissipative process is associated with irreversible diffusion transfer of the solution components from sections of the medium with high concentration to sections with low concentration.

The hydrodynamic equations for an incompressible liquid solution can be used to find the energy dissipation in the hydrodynamic limit, when the size  $R$  of a nucleus is much larger than the mean-free paths of the excitations in the solution. Then the intensity of energy dissipation is determined by the relation<sup>14</sup>

$$\frac{dE}{dt} = -\frac{\eta}{2} \int d^3r \left( \frac{\partial v_i}{\partial x_k} + \frac{\partial v_k}{\partial x_i} \right)^2 - \rho D \frac{\partial Z}{\partial c} \int d^3r (\nabla c)^2, \quad (13)$$

where  $\eta(T)$  is the coefficient of viscosity of the metastable liquid surrounding the nucleus and  $Z(\rho, T, c)$  is the suitably defined chemical potential of the solution

$$Z = \mu_3/m_3 - \mu_4/m_4. \quad (14)$$

Here  $\mu_3$  and  $\mu_4$  are the chemical potentials of the components of the solution and  $m_3$  and  $m_4$  are the masses of the constituent particles of the solution. Substituting the velocity distribution of the liquid (7) and the concentration distribution (10), we obtain<sup>15</sup>

$$\begin{aligned} \frac{dE}{dt} = & -16\pi \left( \frac{\rho' - \rho}{\rho} \right)^2 \eta R \dot{R}^2 - 4\pi (c' - c)^2 \rho'^2 \\ & \times \frac{\partial Z / \partial c}{\rho D} R^3 \dot{R}^2, \quad R \gg l. \end{aligned} \quad (15)$$

The dependence of the rate of energy dissipation on the size of a nucleus in the solution is more complicated than in a one-component liquid. For large nuclei, to which there corresponds quite weak supersaturation of the solution, when

$$R \gg R_D(T) = 2 \frac{|\rho' - \rho|}{\rho'} \frac{1}{|c' - c|} \left( \frac{\eta D}{\rho \partial Z / \partial c} \right)^{1/2}, \quad (16)$$

the energy dissipation of a nucleus is determined entirely by diffusion processes. They will also determine the probability of formation of a critical nucleus.

As the temperature decreases the mean-free paths  $l(T)$  of the excitations, as a rule, increase rapidly and when they are of the order of the characteristic radius  $R_c$  of a nucleus the hydrodynamic relations are no longer applicable. In this case it is necessary to study the kinetic equations with definite boundary conditions, which are characterized by the interaction of excitations with the surface of the nucleus. In the Knudsen limit, when collisions are rare ( $l \gg R_c$ ), the problem simplifies significantly, since the excitations collid-

ing with the surface relax in the medium itself far from and independently of the nucleus.

In this case the rate of dissipation of energy of the nucleus becomes proportional to the area of the surface of the nucleus and can be found from the general equation of heat transfer,<sup>14</sup> which gives for the rate of irreversible dissipation of energy in the solution

$$-\frac{dE}{dt} = - \int d^3r \sigma_{ik} \frac{\partial v_i}{\partial x_k} + \int d^3r i \nabla Z, \quad (17)$$

where  $\sigma_{ik}$  is the tensor of the dissipative momentum flux density, which determines the internal friction forces acting in the solution. Using the equations of motion for a liquid solution, which are simplified owing to the slow growth  $\dot{R}$  of the nucleus, we obtain

$$dE/dt = 4\pi R^2 \sigma_{rr}(R) v(R) + 4\pi R^2 i(R) [Z(c) - Z(c(R))]. \quad (18)$$

Here  $\sigma_{rr}(R)$  is the additional force acting along the normal to the surface of the nucleus together with the usual pressure of the liquid. This force arises in connection with the motion of excitations in the metastable phase relative to the nucleus. The strength of this force is determined by the momentum flux transferred from excitations to the nucleus and is proportional to the velocity of the liquid

$$\sigma_{rr}(R) = -\vartheta v(R). \quad (19)$$

The coefficient  $\vartheta$  depends only on the properties of the excitations in the metastable phase and on the character of their interactions with the surface of the nucleus. In order of magnitude the coefficient  $\vartheta$  is the product of the density of the excitations by their characteristic velocity.

The second term in Eq. (18) is related to the presence of the diffusion flux in the metastable solution.

$$i(R) = \rho'(c' - c) \dot{R} \quad (20)$$

is the diffusion flux at the surface of the nucleus and can be found with the help of Eqs. (5) and (6). For further simplification we take into account the fact that the appearance of a difference  $c - c(R)$  between the concentration on the surface and far away from it is caused exclusively by the growth of the nucleus and the fact that the concentration difference is nonzero to the extent of the rate of growth  $\dot{R}$ . Expanding the difference of the potentials in Eq. (18) in powers of the concentration difference and using the relation between the diffusion flux density at the surface and the depletion (enrichment) of one of the solution components at the surface of the nucleus

$$i(R) = -\delta [c - c(R)], \quad (21)$$

we obtain for the diffusion term in Eq. (18) the expression

$$-4\pi R^2 [\partial Z(c)/\partial c] i^2(R) / \delta.$$

The coefficient  $\delta$  is also determined only by the properties of the metastable phase and the properties of the surface of the nucleus. In order of magnitude, it is also the product of the density of the excitations by their characteristic velocity.

Using Eqs. (7) and (20), we obtain finally

$$\begin{aligned} \frac{dE}{dt} = & -4\pi \left( \frac{\rho' - \rho}{\rho} \right)^2 \vartheta R^2 \dot{R}^2 \\ & -4\pi (c' - c)^2 \rho'^2 \frac{\partial Z / \partial c}{\delta} R^2 \dot{R}^2, \quad R \ll l. \end{aligned} \quad (22)$$

for the rate of dissipation of energy of the nucleus in the Knudsen limit. Dissipation in this case does not depend on the mean free path. The viscous contribution depends on the size of the nucleus in the same manner as does the diffusion contribution; this is different from the hydrodynamic limit (15).

When dissipative processes are present in the metastable medium surrounding the nucleus the dynamics of the development of the nucleus can be described by introducing a friction force  $F$ , which is a linear function of the rate of growth and opposes the growth of the nucleus. The growth of the nucleus is determined by the following equation of motion:<sup>12</sup>

$$\frac{d}{dt} \left( \frac{\partial L}{\partial \dot{R}} \right) - \frac{\partial L}{\partial R} = -\mu(R) \dot{R}, \quad (23)$$

where  $L$  is the Lagrangian of the system of interest

$$L(\dot{R}, R) = M(R) \dot{R}^2 - U(R).$$

The kinetic and potential energies are determined by the relations (8) and (1). As one can see from Eq. (23), the energy of the nucleus is dissipated at the rate

$$\frac{dE}{dt} = \frac{d}{dt} \left( \dot{R} \frac{\partial L}{\partial \dot{R}} - L \right) = -\mu(R) \dot{R}^2, \quad (24)$$

which makes it possible to establish a relation between the coefficient of friction  $\mu(R)$  and the dissipation of energy per unit time.

In order to calculate the probability  $W$  of the decay of the metastable state of the solution we employ the approach proposed in Refs. 10 and 11 and employed in Ref. 12 to calculate the probability of decay of a metastable quantum liquid. The probability of the formation of the nucleus of a new phase is given, to exponential accuracy, by the expression

$$W = \exp(-A), \quad (25)$$

where  $A$  is the extremal value of the effective action  $S_{\text{eff}}$ , defined in terms of the imaginary time as

$$\begin{aligned} S_{\text{eff}} = & \int_{-\beta/2}^{\beta/2} d\tau \left\{ \frac{M(R) \dot{R}^2}{2} + U(R) \right. \\ & \left. + \frac{1}{4\pi} \int_{-\beta/2}^{\beta/2} d\tau' [\gamma(R_\tau) - \gamma(R_{\tau'})]^2 \frac{(\pi T)^2}{\sin^2 \pi T (\tau - \tau')} \right\} \end{aligned} \quad (26)$$

with periodic boundary conditions  $R(-\beta/2) = R(\beta/2)$ . Here  $\beta = T^{-1}$  is the inverse temperature. The first two terms in Eq. (26) are the kinetic and potential energies. The vertex function  $\gamma(R)$  is determined by the classical mobility (24) according to the following relation

$$\mu(R) = (\partial\gamma/\partial R)^2, \quad (27)$$

and the last term in Eq. (26) corresponds, in real time, to the

$$\gamma(R) = \begin{cases} \frac{1}{2} \left[ 4\pi \left( \frac{\rho' - \rho}{\rho} \right)^2 \vartheta + 4\pi (c' - c)^2 \rho'^2 \frac{\partial Z / \partial c}{\delta} \right]^{1/2} R^2, & R \ll l \\ \frac{2}{3} \left[ 16\pi \left( \frac{\rho' - \rho}{\rho} \right)^2 \eta \right]^{1/2} R^{3/2}, & R \gg l, R \ll R_D \\ \frac{2}{5} \left[ 4\pi (c' - c)^2 \rho'^2 \frac{\partial Z / \partial c}{\rho D} \right]^{1/2} R^{5/2}, & R \gg l, R \gg R_D \end{cases} \quad (28)$$

As one can see from the relation (26), the dissipative processes in solution, which are associated with viscosity (internal friction) and diffusion, reduce the probability of subbarrier formation of a critical nucleus as compared with nondissipative kinetics<sup>1,9</sup> by the amount in the exponent of the exponential  $\sim \mu(R_c)R_c^2$ . Since the nucleation kinetics in the Knudsen regime  $R_c \ll l$  and in the hydrodynamic viscous regime  $R_c \gg l$  and  $R_c \ll R_D$  is entirely analogous to the nucleation kinetics in a one-component liquid ( $R_D = \infty$ ) in the corresponding regimes and with correspondingly redefined kinetic coefficients and is examined in detail in Ref. 12, we shall analyze below only the case of the hydrodynamic regime  $R_c \gg l, R_D$ , which has no analog in the one-component metastable phase. The diffusion regime is realized not only when the density of the metastable solution is almost equal to the density of the new stable phase, but also in the important case of the limit of a weakly supersaturated solution ( $R_c \rightarrow \infty$ ), where diffusion processes completely determine the quantum kinetics of nucleation processes. This is what makes a solution fundamentally different from a one-component system, where the kinetics of subbarrier nucleation in a weakly nonequilibrium metastable liquid is dictated by the kinetic energy  $K$  of the medium, which in turn is determined by the fluxes of the liquid accompanied by growth of fluctuations of the new phase.

So, in the diffusion hydrodynamic limit  $R_c \gg l, R_D$  the effective action is determined by the expression (26) with the vertex function  $\gamma(R)$ , which is given by the bottom line in Eq. (28). As usual, there exists a stationary extremal trajectory  $R(\tau) = R_0 = \frac{2}{3}R_c$ , which passes through the maximum of the potential energy  $U(R)$  (1) and gives the classical exponential for the probability of nucleation, equal to the height of the potential barrier

$$A(T) = U_0/T, \quad U_0 = (4/27)4\pi\sigma R_c^2. \quad (29)$$

The second extremal trajectory is explicitly time dependent, and in order to analyze it it is convenient to distinguish two cases: weak and strong dissipation. In the first case, when supersaturation of the solution is still quite large and the critical radius  $R_c$  satisfies the inequality

$$R_c^3 \ll \frac{(\Delta\rho)^2}{(\rho'\Delta c)^4} \frac{\sigma\rho D^2}{(\partial Z/\partial c)^2}, \quad (30)$$

the nonlocal term in Eq. (26) can be regarded as a perturbation of the nondissipative regime. As a result we have

friction force  $F$  in Eq. (23). Correspondingly, in the Knudsen and hydrodynamic limits the vertex function  $\gamma(R)$  depends nonlinearly on the radius  $R$  and has the form

$$A(T) = A_0 + A_1(T), \quad (31)$$

where  $A_0$  is given by<sup>1</sup>

$$A_0 = \frac{5 \cdot 2^{1/2} \pi^2}{16} (\sigma\rho_{eff})^{1/2} R_c^{1/2}, \quad \rho_{eff} = \frac{(\rho' - \rho)^2}{\rho}. \quad (32)$$

The transition from the activational regime (29) to the quantum regime (31) occurs at the temperature  $T_0$ , for which

$$A_0 + A_1(T_0) = U_0/T_0,$$

and occurs as a first-order phase transition. In the limit of low temperatures  $T \ll T_0$  or  $\pi T\tau_c \ll 1$ , where  $\tau_c = (|\Delta\rho|/\rho)(\rho R_c^3/2\sigma)^{1/2}$  is the transit time along the null-temperature extremal of the local part of the action (26), we obtain the following expression for the correction  $A_1(T)$ , calculated by making an expansion in the temperature:

$$A_1(T) = \frac{16\pi}{25} (c' - c)^2 \rho'^2 \frac{\partial Z / \partial c}{\rho D} R_c^5 \left[ \frac{J_{3/2}}{\pi} + \beta_{3/2} (\pi T\tau_c)^2 \right], \quad (33)$$

where

$$J_{3/2} = 1,36 \dots, \quad \beta_{3/2} = \frac{1}{3\pi} \left[ \frac{231\pi^2}{1024} - \frac{2048}{1225} \right] = 0,059 \dots$$

We shall now transfer to the more interesting case of strong dissipation, when the inequality opposite to (30) holds. In this limit the evolution of the nucleus under the barrier is entirely determined by the diffusion term, and the kinetic energy plays the role of a correction. The extremal trajectory is determined from the effective action, which in the corresponding variables has the universal form

$$A(T) = \frac{16\pi}{25} (c' - c)^2 \rho'^2 \frac{\partial Z / \partial c}{\rho D} R_c^3 s_{3/2}(t). \quad (34)$$

The functional  $s_{3/2}[x_\tau]$  is given by the expression

$$s_{3/2} = \int_{-1/2t}^{1/2t} d\tau \left[ x_\tau^2 (1 - x_\tau) + \frac{\pi t^2}{4} \int_{-1/2t}^{1/2t} d\tau' \frac{(x_\tau^{1/2} - x_{\tau'}^{1/2})^2}{\sin^2 \pi t (\tau - \tau')} \right], \quad (35)$$

and the reduced temperature  $t$  is the ratio of the tunneling exponential to the activational exponential

$$t = \frac{4}{25} \frac{T}{\sigma\rho D} (c' - c)^2 \rho'^2 \frac{\partial Z}{\partial c} R_c^3.$$

For zero temperature the value of  $s_{3/2}(0)$  is of the order of unity  $s_{3/2}(0) = 2.8 \dots$ . In the regime of strong dissipation

the temperature  $T_0$  of the transition into the activation regime is defined as the temperature at which the action on the stationary trajectory  $x_\tau = 2/3$  is equal to the action for the nonstationary trajectory, i.e., from

$$t_0 s_{5/2}(t_0) = \frac{4}{27}, \quad t_0 = \frac{4}{25} \frac{T_0}{\sigma \rho D} (c' - c)^2 \rho'^2 \frac{\partial Z}{\partial c} R_c^3. \quad (36)$$

The transition from the classical activation trajectory to the quantum trajectory in this case also occurs abruptly and the curve  $s_{5/2}(t)$  has a break at the point  $T_0$ .

Thus in metastable solutions with sufficiently small degrees of supersaturation the main process limiting the rate of quantum nucleation is diffusion of the solution components accompanying growth of fluctuations. As one can see from Eq. (34), a larger diffusion coefficient corresponds to a shorter lifetime of the supersaturated solution. A distinguishing feature of the probability of quantum nucleation is that the exponential depends much more strongly on the critical size of the nucleus than in the case of the activation mechanism.

### 3. QUANTUM NUCLEATION IN A SUPERFLUID SOLUTION

We now study nucleation processes in a superfluid supersaturated liquid solution consisting of a  $^3\text{He}$ - $^4\text{He}$  solution in the region of the lower phase. As in the preceding section, the problem reduces in practice to finding the classical equation of motion (23), describing the growth of fluctuations of radius  $R$ . The potential energy  $U(R)$  has the previous form (1). The effective mass  $M(R)$  and the coefficient of friction  $\mu(R)$  in turn differ from their values in the normal solution. This is connected with the presence of two types of motions in the superfluid solution and with the fact that the dissipative processes are different.

To describe the superfluid solution we employ the equations of two-velocity hydrodynamics for mixtures and we also assume that the impurities ( $^3\text{He}$ ) participate only in the normal motion.<sup>16</sup> In what follows we do not study the case of extremely low temperatures, when there arises the possibility for a phase transformation with a superfluid transition of the impurity component ( $^3\text{He}$ ) and the motion of such a liquid with two superfluid components is now described by the equations of three-velocity hydrodynamics.<sup>17</sup>

We also emphasize that we do not study the region of temperatures close to the temperature of the transition into the superfluid state. The point is that as the transition point is approached the relaxation times necessary for establishing the equilibrium value of the superfluid order parameter as well as the correlation radius of its fluctuations increase and become the longest relaxation times. For this reason the density of the superfluid component of the liquid, associated with the order parameter, can be expressed as a function of other quantities (density, temperature) as long as the critical radius of the nucleus and the characteristic time of the motion of the nucleus are large compared with the correlation radius of the fluctuations and the relaxation time of the order parameter. Close to the  $\lambda$  point, aside from singling out the "significant" variables related with the nucleus, it is also necessary to include among the "important" variables the order parameter also. For this reason, the complete equations of motion of the liquid must also include the equa-

tions determining the evolution of the order parameter.<sup>18</sup>

As before, we assumed that the rate of growth  $\dot{R}(t)$  of a nucleus is small and the liquid is incompressible. In this approximation the equations of motion have the form

$$\text{div } \mathbf{j} = 0, \quad (37)$$

$$\text{div}(\rho c \mathbf{v}_n + \mathbf{i}) = 0. \quad (38)$$

The mass flux density in the superfluid liquid is equal to

$$\mathbf{j} = \rho_n \mathbf{v}_n + \rho_s \mathbf{v}_s, \quad \rho = \rho_n + \rho_s, \quad (39)$$

where  $\rho_n$  and  $\rho_s$  are the densities of the normal and superfluid components of the liquid. The boundary conditions under which the mass flux of the entire liquid and of one of the components are continuous, taking into account zero velocities and absence of fluxes inside the nucleus, have the form

$$-\rho' \dot{R} = j(R) - \rho \dot{R}, \quad (40)$$

$$-\rho' c' \dot{R} = \rho c(R) [v_n(R) - \dot{R}] + i(R). \quad (41)$$

One other boundary condition is needed in order to find the distribution of the normal and superfluid velocities as well as the concentration. The normal velocity  $v_n$  is related with the motion of normal excitations, which have all the properties of a conventional viscous liquid. For this reason, we set the normal velocity  $v_n$  at the surface of the nucleus equal to the velocity of this surface, i.e.,

$$v_n(R) = \dot{R}. \quad (42)$$

This boundary condition, corresponding to attachment of normal excitations to the surface, is equivalent to the assumption that the excitations are in thermal equilibrium with the surface. This means that heat is not transferred between the nucleus and the liquid and there is no temperature jump at the boundary. As a result, we obtain for the velocity distribution

$$v_n(r) = \begin{cases} \dot{R} R^2 / r^2, & r > R(t) \\ 0, & r < R(t) \end{cases}, \quad (43)$$

$$v_s(r) = \begin{cases} -[(\rho' - \rho_s) / \rho_s] \dot{R} R^2 / r^2, & r > R(t) \\ 0, & r < R(t) \end{cases}.$$

For velocities of the normal and superfluid components that are low compared with the velocity of sound the dependence of the thermodynamic quantities on the relative velocity  $|v_n - v_s|$  can be neglected. Then the kinetic energy associated with the growth of the nucleus assumes the form

$$K = 1/2 \int \rho_n v_n^2(r) d^3 r + 1/2 \int \rho_s v_s^2(r) d^3 r, \quad (44)$$

$$K = M(R) \dot{R}^2 / 2, \quad M(R) = 4\pi \rho_{\text{eff}} R^3.$$

Here  $\rho_{\text{eff}}$  is the effective density, equal to

$$\rho_{\text{eff}} = \rho_n + (\rho' - \rho_s)^2 / \rho_s \quad (45)$$

and exceeds the analogous density (8) in a normal solution.

One of the interesting and unusual consequences of (45) is that the lifetime of a metastable superfluid liquid at low temperatures, when the contribution of rotons and phonons to the normal density  $\rho_n$  can be neglected, should increase as the impurity  $^3\text{He}$  is added. The point is that in

this case the normal velocity will be determined entirely by the concentration of the impurities  $\rho_n = \rho^*c$  and increasing their concentration will result in an increase of the effective density  $\rho_{\text{eff}}$ , and therefore according to Eq. (32), it will lead to a decrease of the nucleation probability. If the characteristic observation times of nucleation range from one second to one hour, then, as will be evident from what follows, the exponent of the exponential in Eq. (25) must be equal to  $A_0 \sim 80$ . But the change of the exponent as a result of a change  $\delta c$  of the concentration will be of the order of

$$\delta A \sim \frac{A_0}{2} \frac{\rho \rho^*}{(\rho' - \rho)^2} \delta c \quad (46)$$

so that a change of the impurity concentration by an infinitesimal amount  $\delta c \sim 10^{-4} - 10^{-3}$  will change the lifetime of the metastable liquid by several factors. Such a picture should be observed, for example, for overcompressed metastable He II near the liquid-crystal phase transition.

It is obvious that growth of a nucleus is accompanied not only by nonuniform flows of liquid, but also nonuniform distribution of the concentration  $c(r)$  in solution. In order to find  $c(r)$  it is necessary to know the relation between the flux  $i(r)$  and the concentration. For this reason, we shall first study the region of weak supersaturations of the solution, when the critical radius  $R_c$  of a nucleus is large compared with the mean free path length  $l(T)$  of the excitations and the hydrodynamic approximation<sup>16</sup>

$$i = -\rho D \left( \nabla c + \frac{k_T}{T} \nabla T \right), \quad R \gg l, \quad (47)$$

where  $D$  is the diffusion coefficient and  $k_T D$  is the thermo-diffusion coefficient, can be employed. The fact that the thermodiffusion term must be included in the diffusion flux is dictated by the fact that in a superfluid solution under stationary conditions a concentration gradient can occur together with a temperature gradient. Indeed, the equation of motion for the superfluid velocity, neglecting terms  $\sim \dot{R}^2$  and the coefficients of second viscosity  $\zeta_i$ , since  $\text{div } \mathbf{j} = 0$  and  $\text{div } \mathbf{v}_n = 0$ , has the form<sup>16</sup>

$$\nabla (\mu - Zc) = 0, \quad (48)$$

where  $\mu(T, c)$  is the thermodynamic potential of a unit mass of the solution and is equal to

$$\mu = c \frac{\mu_3}{m_3} + (1-c) \frac{\mu_4}{m_4}. \quad (49)$$

Thus the motion of the superfluid component satisfies the condition that the chemical potential of He II is constant in the volume, i.e.,  $\mu_4(T, c) = \text{const}$ . From Eq. (48) with the help of the thermodynamic identity  $d\mu = -s dT + dP/\rho + Zdc$  we find a relation between the concentration and temperature gradients

$$\nabla T = \frac{\partial Z / \partial c}{c \partial (S/c) / \partial c} \nabla c, \quad (50)$$

where  $S$  is the entropy per unit mass of the solution. Now, substituting Eq. (50) into Eq. (47) we find that the diffusion flux is related to the concentration gradient through the effective diffusion coefficient

$$\mathbf{i} = -\rho D_{\text{eff}} \nabla c,$$

$$D_{\text{eff}} = D \left( 1 + \frac{k_T}{T} \frac{\partial Z / \partial c}{c \partial (S/c) / \partial c} \right). \quad (51)$$

The relations found make it possible to determine the diffusion flux and concentration distributions in the solution:

$$i(r) = \begin{cases} \rho' c' \dot{R} R^2 / r^2, & r > R \\ 0, & r < R \end{cases}, \quad (52)$$

$$c(r) = \begin{cases} c - (c' \rho' / \rho D_{\text{eff}}) \dot{R} R^2 / r, & r > R \\ c', & r < R \end{cases}.$$

Near the surface of a nucleus the dissolved impurity is depleted and, according to Eq. (50), the temperature increases. The depletion of impurity and the increase of the temperature are proportional to the rate of the process. The distributions (43), (50), and (52) obtained above make it possible to calculate the rate of dissipation of energy of the nucleus from the relation<sup>16</sup>

$$\frac{dE}{dt} = -\frac{\eta}{2} \int d^3 r \left( \frac{\partial v_{ni}}{\partial x_i} + \frac{\partial v_{nh}}{\partial x_i} \right)^2 - \int d^3 r \frac{\partial Z / \partial c}{\rho D} i^2 - \int d^3 r \frac{\kappa (\nabla T)^2}{T} \quad (53)$$

and at the same time the coefficient of friction  $\mu(R)$  (24) in terms of the kinetic coefficients of the solution: the viscosity  $\eta$ , the diffusion coefficient  $D$ , and the thermal conductivity  $\kappa$

$$\mu(R) = 16\pi\eta R + 4\pi c'^2 \rho'^2 \frac{\partial Z / \partial c}{\rho D} R^3 + 4\pi c'^2 \rho'^2 \left( \frac{\partial Z / \partial c}{c \partial (S/c) / \partial c} \right)^2 \frac{\kappa}{T \rho^2 D_{\text{eff}}^2} R^3, \quad R \gg l. \quad (54)$$

In the hydrodynamic limit the dissipation of energy for a superfluid solution as a function of the radius  $R$  of the nucleus is analogous to the dependence for the normal solution (15). For this reason all results of the preceding section can be transferred to this case. A distinguishing element, apart from the different temperature and concentration dependences of the coefficients, in the superfluid solution is the presence of an additional mechanism of dissipation—heat conduction. In the limit of weak supersaturations the kinetics of nucleation will be determined by the mechanisms of diffusion and heat conduction.

In the Knudsen regime  $R_c < l$  each dissipation mechanism makes a contribution proportional to the area of the nucleus, and analogously to Eq. (18) for the dissipation of energy of the nucleus per unit time we have

$$dE/dt = 4\pi R^2 \{ \sigma_{rr}(R) v_n(R) + i(R) [Z(c) - Z(c_R)] + (q - Zi)_R (T - T_R) / T \}. \quad (55)$$

When the nucleus is not growing,  $\dot{R} = 0$ , all fluxes, together with the corresponding deviations of the thermodynamic quantities from equilibrium, vanish. For this reason, for slow rates of growth the fluxes can be expanded in powers of the departure from equilibrium and only the linear terms need be retained. Then, for the rate of dissipation of energy we obtain

$$\frac{dE}{dt} = -4\pi R^2 \left[ \frac{\sigma_{rr}^2(R)}{\vartheta} + \frac{\partial Z}{\partial c} \frac{i^2(R)}{\varepsilon} + \chi \frac{(T - T(R))^2}{T} \right], \quad (56)$$

where the coefficients  $\vartheta$  and  $\delta$  can be determined in terms of the relations (19) and (21), and  $\chi$  is the thermal conductivity of the nucleus-solution boundary and relates the heat flux through the boundary with the temperature difference. In order of magnitude  $\chi$  is the product of the heat capacity of the gas of excitations by their characteristic velocity and density. Using the relations (41) and (50) and the dependence of  $\sigma_{rr}$  on  $v_n(R)$  and  $T - T(R)$  and the dependence of  $i$  on  $c - c(R)$  and  $T - T(R)$  we represent the dissipation in a form analogous to Eq. (22) and from the same contributions as in Eq. (54)

$$\frac{dE}{dt} = -4\pi R^2 \left[ \vartheta + c'^2 \rho'^2 \frac{\partial Z / \partial c}{\delta} + c'^2 \rho'^2 \left( \frac{\partial Z / \partial c}{c \partial (S/c) / \partial c} \right)^2 \frac{\chi_{\text{eff}}}{T \delta_{\text{eff}}^2} \right] \dot{R}^2, \quad R \ll l. \quad (57)$$

The characteristics of nucleation in a superfluid solution in the region of the Knudsen regime are the same as in a normal metastable solution in the corresponding regime, since the functional dependence on the size of the nucleus is the same. Differences appear only as a result of the different behavior of the kinetic coefficients as functions of the temperature and concentration. In the superfluid solution they are also determined entirely only by the properties of the interaction of the normal excitations with the surface of the nucleus. The contribution of the dissipative term to the exponential for the nucleation probability (25) is of order  $\sim \mu(R_c) R_c^2$ .

#### 4. EXPERIMENTAL OBSERVATION OF QUANTUM NUCLEATION

The dynamics of the development of a nucleus of a stable phase at a certain temperature is determined by three energies. The first energy is the potential energy of formation of a nucleus of the stable phase as a function of the size of the nucleus. The second energy is the kinetic energy associated with the rate of growth of the nucleus. The third energy characterizes the interaction of the evolving nucleus of the stable phase with the surrounding metastable liquid and determines the dissipation of the energy of the nucleus as the nucleus grows. We are interested in relatively slow growth of a nucleus with rates much slower than the velocity of sound. It is precisely in this case that the kinetic energy can be systematically separated from the dissipative part of the interaction. Such slow growth of a nucleus occurs with relatively small degree of metastability of the unstable phase, when the nucleus has a macroscopic size. It is of no interest to analyze the formation of the critical nuclei of a smaller size, namely, microscopic size, since the formation time of such a nucleus is short and is determined by the times of separate microscopic acts. Indeed, the probability of formation of a critical nucleus includes, aside from the exponentials, a large preexponential factor  $W_0 \sim \omega_0 N$ , consisting of two factors, where  $\omega_0$  is the characteristic frequency of small oscillations near the bottom of the potential (1) and  $N = V(4\pi R_c^3/3)^{-1}$  is

the number of possible centers of nucleation. For the volume of the system  $V \sim 1 \text{ cm}^3$ ,  $R_c \sim a$ , and  $\omega_0 \sim 10^{12} \text{ s}^{-1}$  we have  $W_0 \sim e^{80} \text{ s}^{-1}$ . If the characteristic observation time ranges from one second to one hour, then the exponent  $A$  for the nucleation probability (25) must fall in the range  $A \sim 80$ . Such a large exponential factor is possible only for a critical nucleus of macroscopic size, while microscopic critical nuclei lead to instantaneous formation of a stable phase, analysis of which falls outside the scope of the approach studied in this paper.

At high temperatures, when thermoactivational formation of a nucleus occurs, the probability of formation of a nucleus is determined primarily by the potential energy only, more precisely, only by the height of the potential barrier which fluctuations of the new phase must overcome in order for a phase transition to occur in the metastable system. The barrier height is dictated by the effective surface tension at the interface between the phases. If impurities are introduced into the metastable system, then their role reduces primarily to changing the interphase surface tension. When a small quantity of impurities is introduced, generally speaking the change in the surface tension and together with it the change in the activational exponential must be small and proportional to the impurity concentration, if, of course, the impurities are not centers of nucleation or are not sorbed for any reasons on the interphase boundary, when a high degree of metastability of the unstable phase becomes much less likely or even impossible.

In the quantum region  $T < T_0$ , where  $T_0$  is the temperature separating two regimes and is proportional to the interphase surface tension, the effect of impurities on the lifetime of the metastable system is more complicated. Aside from changes in the thermodynamic quantities, such as, for example, the surface tension, introducing impurities into a metastable system will change the kinetic coefficients describing the dissipative processes of viscosity, diffusion, and heat conduction, which accompany growth of fluctuations of the new phase. Since this occurs at low temperatures, these changes can be significant and they can even result in other temperature dependences. This is clearly seen when  $^3\text{He}$ -impurities are introduced into superfluid He II, where at sufficiently low temperatures dissipative processes will be determined entirely by the impurity component. The introduction of the impurity component increases dissipation in the formation of the critical nucleus and therefore results in an increase of the lifetime of the metastable system. Moreover, in the limit of strong dissipation the probability of formation of a critical nucleus is determined entirely by dissipative processes and at temperatures  $T < T_0$  it does not depend on the surface tension between them.

A new qualitative element appears in the superfluid liquid. Here the presence of impurities ( $^3\text{He}$ ) affects not only the dissipative part of the effective action (26) but also its dynamic part. The reason is that in a superfluid liquid there exist two types of motion of the liquid—superfluid and normal—and also that the impurities participate only in one type of motion—in the normal motion. The difference in the motion of impurities and the liquid as a whole results in an increase of the effective density (45) in the mass for the kinetic energy of the nucleus. The increase in the kinetic energy of a nucleus accompanying introduction of impurities

into the system also increases the lifetime of the metastable phase. Since at sufficiently low temperatures the density of the normal component of the liquid is almost equal to the density of the impurity, the introduction of even an infinitesimal quantity ( $\delta c \sim 10^{-4}$ ) of impurities into  $^3\text{He}$  will result in an appreciable increase (46), by several factors, of the lifetime of the metastable superfluid He II near the liquid-solid phase transition.

A new qualitative element is also present in a two-component system (solution) as compared with the case of a single-component metastable liquid. In this case, in the limit of large critical nuclei the main dissipation process becomes diffusion, which is absent in a single-component system.

The observation of the effects studied above, depending on the degree of metastability of the system, can present definite difficulties, caused by the sharp dependence of the probability of decomposition on the critical radius. As pointed out above, because of the large value of the preexponential factor, the exponent  $A$  is also a large quantity and is equal to  $A \sim 80$ . Since  $A$  is also a nonlinear function of  $R_c$ ,  $A \propto R_c^n$  ( $n = 2-5$ ), the experimentally admissible range of variation of  $R_c$  is only several percent. A change of  $R_c$  by several tens of percent, however, would result in an enormous change of the characteristic required observation time, on the order of four orders of magnitude.

One way to overcome this difficulty is to formulate a dynamic experiment. As the degree of metastability of the system, for example, the difference of the chemical potentials of the phases  $\delta\mu(t)$ , gradually increases,  $R_c$  decreases and the probability of formation of the critical nucleus  $W(\delta\mu)$  increases, so that at some moment in time there occurs a phase transition, which is recorded. When the experiment is repeated, because of the probabilistic character of the process, the phase transition, generally speaking, will occur at a different value of  $\delta\mu$ . The distribution of the number of creations of a nucleus as a function of the degree of metastability  $P(\delta\mu)$  is related with the probability of nucleation  $W(\delta\mu)$  by the simple expression

$$P(\delta\mu) d(\delta\mu) = W(\delta\mu) dt \left[ 1 - \int_0^{\delta\mu} P(\delta\mu) d(\delta\mu) \right],$$

which makes it possible, in principle, to determine  $W(\delta\mu)$ . An analogous relation was used to analyze the probability of macroscopic quantum tunneling in superconducting Josephson contacts.<sup>19</sup> The width of the distribution  $P(\delta\mu)$  is related to the value of the exponential in the expression for the nucleation probability. Measuring the width of the distribution  $P(\delta\mu)$  as a function of the temperature and concentration will make it possible to determine the character of the nucleation processes occurring in phase transitions.

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