The role of dissipative processes in the supercritical growth kinetics of a stable phase in quantum liquids and their mixtures

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(Submitted 2 December 1993)
Zh. Eksp. Teor. Fiz. **105**, 881–885 (April 1994)

We discuss experiments on the growth kinetics of seeds of a stable phase in a quantum liquid at low temperatures. We emphasize the role of dissipative processes in the supercritical stage of seed growth, and show that the supercritical growth kinetics of a one-component system differ qualitatively from the kinetics of a two-component system. This is because viscosity-driven processes play a fundamental role in the first type of system, in contrast to the diffusion-driven processes that play a dominant role in the second type.

In recent years, serious attempts have been made to observe regimes of quantum tunneling during first-order phase transitions in various metastable systems. The discussion centers primarily on ^{3}He and ^{4}He and their mixtures, where phase transitions can be observed at temperatures down to absolute zero. Experimental work includes investigation of the solidification of superfluid ^{4}He under pressure (Ref. 1), stratification of superfluid solutions of ^{3}He–^{4}He (Refs. 2, 3), and preliminary studies of cavitation in superfluid ^{4}He (Ref. 4). These experiments were stimulated by theoretical work of Lifshits and Kagan[5] and Lifshits, Khokhlov, and Polesski[6] dating from the beginning of the 1970's. In our subsequent papers[7,8] we clarified the fundamental role of dissipative processes in the quantum-mechanical below-barrier formation of seeds of a stable phase. These processes determine the temperature dependence of the probability for forming a critical seed even for the case of weak dissipation; for strong dissipation they completely specify the growth dynamics of the critical seeds as well.

It is important to note that when we investigate the process of seed formation, the seeds we are forced to observe experimentally are not those of critical size, but are rather seeds with dimensions far above critical. The problem is that the characteristic dimensions of a critical seed are no more than a few tens of angstroms as a rule, i.e., extremely small compared to the usual macroscopic dimensions. Although seeds with larger radii could in principle exist at smaller values of the supersaturation, their critical dimensions cannot be measured in practice due to the large characteristic fluctuation times required to create them. Seeds of the new phase with such small dimensions, i.e., on the order of tens of angstroms, are very difficult to observe experimentally; in fact, no one has even addressed the problem of finding a way to detect seeds of such small size, at least in experiments performed up to now.[1–4] In the experiments that are actually performed, seeds with relatively large dimensions are observed, either visually[1] or by measuring their effect on the macroscopic characteristics of the system, e.g., discontinuities in pressure,[1,2,4] changes in the average velocity of sound and capacitance of the system,[3] or NMR response.[2,3] The information derived from these observations thus involves seeds of the new phase whose dimensions considerably exceed the size of a critical seed.

In light of this fact, it is necessary to study the supercritical growth time of a seed and compare it with characteristic times for subcritical fluctuations. In principle, the growth of a supercritical seed is described by the same equations as a seed in the subcritical region:[5,7]

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

here the Lagrange function L and the coefficient of friction $\mu(R)$ have the form

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

$$U(R)=4\pi\sigma R^2-(4\pi R^3/3)\delta\phi,$$

$$\mu(R)=16\pi\eta(\Delta\rho/\rho)^2Rf(R/l);$$

$$f(x)=1/(1+(\alpha x)^{-1}),\quad \alpha\sim 1,$$

where $\delta\phi=\{\phi(p)-\phi'(p')\}/v'(p')$ is the difference in the free energies of the metastable and stable phases divided by the specific volume of a seed; $\rho_{\text{eff}}=(\Delta\rho)^2/\rho$, where ρ is the density of the metastable phase and $\Delta\rho$ is the difference in the densities of the stable and metastable phases; and $l=l(T)$ is the mean-free path. For $R\gg 1$ the value of the friction coefficient $\mu(R)$ corresponds to hydrodynamic growth of the seed, while $R\ll 1$ corresponds to the Knudsen collisionless regime.

In what follows we will be interested in the case where the size of the seed $R\gg R_c$. In this case the potential energy takes the form

$$U(R)=-(4\pi R^3/3)\delta\phi,$$

where $\delta\phi>0$ and is small ($\delta\phi\Rightarrow 0$). The supercritical growth of the seed is determined by the kinetic coefficient $K(R)$, which characterizes the growth rate of the seed radius $\dot{R}$ for a given degree of metastability:

$$\dot{R}=K(R)\delta\phi\quad(\delta\phi\Rightarrow 0)\tag{1}$$

and which, generally speaking, depends on the size of the seed R.

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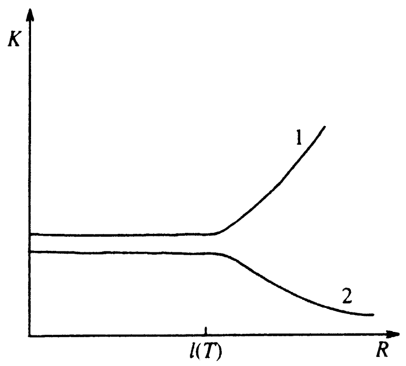

FIG. 1. Dependence of the growth coefficient $K(R)$ for a supercritical seed on the size of the seed R in a one-component (*1*) and two-component (*2*) system; $l(T)$ is the mean-free path of an excitation in the system.

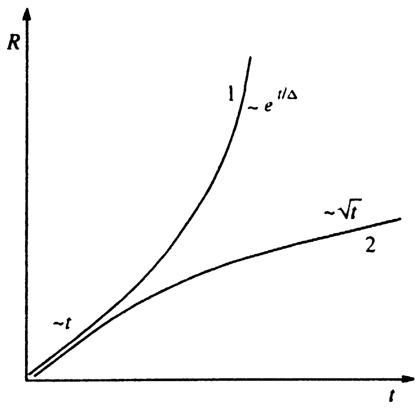

FIG. 2. Dependence of the size of a seed R on time t in the supercritical region for a one-component (*1*) and two-component (*2*) system for fixed temperature T and a constant degree of nonequilibrium $\delta\phi$.

The equation of motion for the supercritical growth of a seed can be written in the form

$$\rho_{\rm eff}R\ddot{R}+3\rho_{\rm eff}\dot{R}^2-\mu(R)(4\pi R^2)^{-1}\dot{R}=-\delta\phi. \qquad (2)$$

Generally speaking, $\dot{R}=\dot{R}(R,\delta\phi)$ is a complicated function of $\delta\phi$. We are interested in the limit $\delta\phi\Rightarrow 0$. It is not difficult to see that in this limit we need save only the frictional force, and neglect the term with the mass, i.e.,

$$\dot{R}=K(R)\delta\phi, \quad K(R)=4\pi R^2/\mu(R). \qquad (3)$$

Actually, the correction from the kinetic term will be

$$\dot{R}=K(R)\delta\phi\{1+O(\rho_{\rm eff}K^2(R)\delta\phi)\},$$

and, in order that (3) be valid, it is necessary to have

$$\delta\phi\ll 1/\{\rho_{\rm eff}K^2(R)\}.$$

The fact that the growth of the seed in the supercritical region is determined exclusively by the dissipative term as $\delta\phi\Rightarrow 0$ distinguishes this region from the subcritical region, i.e., the quantum regime, in which we can have both dissipative and nondissipative behavior as $\delta\phi\Rightarrow 0$ (Ref. 7).

From (3) it is easy to obtain the dependence of the kinetic growth coefficient $K(R)$ as a function of the size of the seed R by substituting in the expression for $\mu(R)$. For a seed radius R large enough that the mean-free path $l(T)$ is much smaller than R, so that the hydrodynamic description of the dynamics of seed growth can be used, we have

$$K(R)=(1/4)(\rho/\Delta\rho)^2R/\eta(T) \quad (l(T)\ll R). \qquad (4)$$

In the opposite limiting case, i.e., the Knudsen collisionless regime,

$$K(R)=(1/4)(\rho/\Delta\rho)^2 l(T)/\eta(T) \quad (l(T)\gg R). \qquad (5)$$

The function $K(R)$ is shown in Fig. 1 (curve *1*). The increase in seed size takes place according to essentially different laws, depending on the ratio of R to $l(T)$. At early times, when R is small, the seed grows linearly with time t. However, when R exceeds the mean-free path $l(T)$, the size of the seed increases exponentially (Fig. 2, curve *1*):

$$R(t)-R_0=K\delta\phi(t-t_0), \quad R(t)<l(T),$$

$$R(t)=R_0\exp\{(t-t_0)\delta\phi/\Delta\},$$

$$\Delta=(1/4)(\rho/\Delta\rho)^2R/\eta(T) \quad (l(T)>R).$$

We emphasize that the dependence of $K(R)$ on temperature can differ considerably from one system to the next. Since the viscosity $\eta\approx\rho vl$, we have $K(R)\sim\rho/(\Delta\rho)^2R/(v_F l(T))\sim T^2$ in ^{3}He, as in a normal Fermi liquid, when for $l(T)\ll R$, while $K(R)\sim[\rho/(\Delta\rho)^2]/v_F$, which does not depend on temperature, when $l(T)\gg R$. This dependence is dictated by the characteristic behavior of the temperature dependence of the mean-free path in a Fermi liquid, i.e., $l(T)\sim T^{-2}$. In superfluid ^{4}He the behavior of $K(R)$ is determined entirely by the normal fluid component and the mean-free time of an excitation. Since in this case[8] $\rho/\Delta\rho\sim 1$, we have

$$K(R)=R/(4\eta), \quad \eta=(1/5)c^2\rho_{ph}\tau_{ph}$$

in the hydrodynamic regime $l(T)\ll R$, where $\rho_{\rm ph}$ is the density of the phonon gas (for the normal component of ^{4}He) and $\tau_{\rm ph}$ is the phonon mean-free time; in the Knudsen regime $l(T)\gg R$ we have $K(R)\ \sim\ (\rho_{\rm ph}c)^{-1}\ \sim\ T^{-4}$.

Now let us turn to the supercritical growth kinetics for stratification of ^{3}He–^{4}He mixtures. In this case we find that all the dependence of the growth coefficient $K(R)$ on the size of the seed R and on temperature is radically changed. Specifically, in the two-component ^{3}He–^{4}He system we have a new mechanism for dissipation in addition to viscosity, which controls the seed growth in the one-component system: diffusion. When this process is taken into account, the friction coefficient $\mu(R)$ has the form:[8]

$$\mu(R)=16\pi\eta\left(\frac{\Delta\rho}{\rho}\right)^2R+4\pi(c'-c)^2\rho\frac{\partial Z}{\partial c}\frac{R^3}{D}. \qquad (6)$$

Accordingly, the leading term for large R is the second term, which is determined by diffusion. This is true for all $R\gg l(T)$. Therefore,

$$K(R)=D(c'-c)^{-2}(\rho\partial Z/\partial c)^{-1}R^{-1}. \qquad (7)$$

This result differs considerably from (4), where $K(R)$ depended linearly on R versus the R^{-1} behavior in (7). Thus, in these solutions the larger the seed, the slower it grows, in contrast to one-component systems in which the size of the seed depends exponentially on time in this region of parameters. According to (7), the radius of the seed in solution increases only according to a power law, with a small exponent equal to one-half (see Fig. 2, curve *2*):

$$R^2=R_0^2+\varkappa\delta\phi(t-t_0),\quad 2\varkappa=D(c'-c)^{-2}(\rho\partial Z/\partial c)^{-1}. \tag{8}$$

The temperature behavior (8), determined by D, is also in contradiction with the one-component system.

Note that the temperature of the surrounding medium may change in the growth process, e.g., when energy is released in the formation of the stable phase. This makes the seed growth time strongly sensitive to T. Furthermore, when the seed grows rapidly the value of $\delta\phi$ can change; in this case, the rate of growth of the seed can change abruptly, and at some time may stop entirely.

The fact that the two-component and one-component systems differ so much in their behavior makes the process of seed formation sensitive to even a very insignificant amount of impurities,[9,10] since, according to (6) and (7), the slowest process that establishes equilibrium in the impure system is the diffusion process. Therefore, in a system with impurities it is possible to establish partial equilibrium over all the internal parameters of the system except the impurity distribution. Subsequently, a uniform distribution is established with respect to impurities in the system by the diffusion process. The slowness of this process is the origin of the extreme sensitivity of the process of seed formation to even small amounts of impurities—see Ref. 10.

The authors are deeply grateful to Yu. Kagan for his numerous discussions of the problem. This work was supported financially by the Russian Fund for Fundamental Research (project 93-02-2540).

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Translated by Frank J. Crowne