Emission of the second sound with an expanding ^{3}He-concentrated droplet and phase-separation kinetics in a superfluid ^{3}He-^{4}He mixture

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We study the growth kinetics of a droplet of the ^{3}He-concentrated phase in a superfluid ^{3}He-^{4}He supersaturated mixture. The growth equation, which generalizes the Rayleigh-Plesset equation for a radial expansion of bubbles in the normal fluids, is derived under the assumption of an arbitrary boundary condition for the normal velocity. The total intensity of the first- and second-sound emissions for a droplet expanding in the superfluid mixture is calculated. The emission of the second-sound mode is found to be predominant due to the smallness of the second-sound velocity compared with the velocity of the first sound. In contrast to demixing normal mixtures the diffusion and heat conduction processes play a minor role in the phase-separation kinetics of the supersaturated ^{3}He-^{4}He superfluid mixtures.
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The supersaturated ^{3}He-^{4}He superfluid mixtures, separating into the ^{3}He-concentrated and ^{3}He-dilute phases, provide us a unique possibility for investigating the phase separation kinetics in binary mixtures at very low temperatures down to absolute zero. In this connection a great portion of recent interest both experimental[1,2] and theoretical[3–6] has been focused on the problems associated with the separation of a mixture via macroscopic quantum tunneling. However, much less work has been done for the clarification and understanding of the physical processes accompanying separation of a superfluid mixture.

Unlike the case of a superfluid mixture the study of the phase separation in the classical mixtures has a very long history and it is a fact of common knowledge that the diffusion processes play a significant role in the phase separation kinetics. The separation of the superfluid ^{3}He-^{4}He mixtures should have a series of specific features compared with that of the classical mixtures. In fact, besides the usual diffusion the ^{3}He impurities in a superfluid liquid can also be transferred in a convective way together with the flow of the normal component. Such convective flow of the ^{3}He impurities can provide an expanding c-phase droplet with a necessary amount of ^{3}He atoms and replace the diffusion flows responsible for the separation of the classical mixtures. In addition, the flow of the normal component will take care of carrying away the latent heat released in the course of the phase transition.

The aim of the work is to derive a correct equation which the c-phase droplet expanding in the environment of the superfluid d-phase obeys. So far all the efforts[3–5] to derive such a growth equation were made under the assumption of absolute incompressibility of a superfluid ^{3}He-^{4}He mixture. Two factors are in favor of this approximation. The first one is trivial and connected with the negligible thermal expansion of a mixture at low temperatures. The second is completely associated with the assumption of a sufficiently low growth rate of a droplet. This means the slowness of the growth rate, at least, with the respect to the sound velocity, i.e., sufficiently small supersaturations of a mixture. However, in a superfluid one has two types of sound propagation in contrast to normal liquids. The velocity of the first sound, associated mainly with the pressure oscillations, is large enough and, in principle, the incompressible liquid approximation can be justified in the phase-separation experiments.

As concerns the second-sound mode, the situation is not so transparent. The point is that the second-sound velocity in a saturated ^{3}He-^{4}He mixture is relatively small compared with the case of pure ^{4}He, being about 10 m/s.[7] Thus the condition for the slowness of the growth rate of a c-phase droplet should be much stronger and more difficult to satisfy, especially, for the large supersaturations and small droplets[5] which play a key role in the nucleation experiments. In addition, we will take into account the heat effects ignored completely in the previous works and involve the coupling between the first- and second-sound modes resulting from the dependence of density ρ on the ^{3}He mass concentration c. The single approximation we use is that the growth rate of the c phase does not exceed the both sound velocities.

So, let us consider a c-phase droplet of radius $R(t)$, expanding spherically at the rate $\dot{R}(t)$ in the superfluid d phase. Our starting point is the linearized hydrodynamic equations for the superfluid ^{3}He-^{4}He mixtures.[8] We are employing these equations in order to describe the state and the motion of the d-phase surrounding the c-phase droplet. At the first stage, for the sake of simplicity, we omit the dissipative flows. Then the general solution of these equations represents two spherically divergent waves of the first- and second-sound modes propagating away from the c-phase droplet at the velocities u_1 and u_2, respectively. The relations between the normal and superfluid velocities and the variable amplitudes of pressure, temperature, and concentration for the plane waves can be found elsewhere.[9]

Exploiting the spherical symmetry of the problem, we can straightforwardly express the normal $\boldsymbol{v}_n(r,t)$ and superfluid $\boldsymbol{v}_s(r,t)$ velocities in terms of the two velocity potentials as

$$\boldsymbol{v}_n(r,t)=-\left(1-\beta\frac{\rho_s}{\rho_n}\right)\nabla_r\left(\frac{A_1(t-r/u_1)}{4\pi r}\right) \qquad (1)$$

$$-(1+\beta)\nabla_r\left(\frac{A_2(t-r/u_2)}{4\pi r}\right),$$

$$\boldsymbol{v}_s(r,t)=-(1+\beta)\nabla_r\left(\frac{A_1(t-r/u_1)}{4\pi r}\right)$$

$$+\left(1-\beta\frac{\rho_s}{\rho_n}\right)\frac{\rho_n}{\rho_s}\nabla_r\left(\frac{A_2(t-r/u_2)}{4\pi r}\right).$$

Here, as usual, ρ_n and ρ_s are the normal and superfluid densities and $\overline{\sigma}=\sigma-c\partial\sigma/\partial c$ where σ is the entropy of a mixture per unit mass. The coefficient $\beta=(c/\rho)\partial\rho/\partial c$, which determines the coupling between the first- and second-sound modes, should be involved since, in general, it is not small in the concentrated ^{3}He-^{4}He mixtures.

Correspondingly, we have for the deviations of the pressure $\delta P(r,t)$, temperature $\delta T(r,t)$, and concentration $\delta c(r,t)$ from their values P_0, T_0, and c_0 at infinity

$$\delta P(r,t)=\rho\frac{\dot{A}_1(t-r/u_1)}{4\pi r}+\beta\rho\frac{\dot{A}_2(t-r/u_2)}{4\pi r},$$

$$\delta T(r,t)=-\beta\frac{\rho_s}{\rho_n}\frac{\overline{\sigma}}{u_1^2}\frac{\partial T}{\partial\sigma}\frac{\dot{A}_1(t-r/u_1)}{4\pi r}+\frac{\overline{\sigma}}{u_2^2}\frac{\partial T}{\partial\sigma}\frac{\dot{A}_2(t-r/u_2)}{4\pi r}, \tag{2}$$

$$\delta c(r,t)=-c\beta\frac{\rho_s}{\rho_n}\frac{1}{u_1^2}\frac{\dot{A}_1(t-r/u_1)}{4\pi r}+c\frac{1}{u_2^2}\frac{\dot{A}_2(t-r/u_2)}{4\pi r}.$$

The unknown amplitudes A_1 and A_2 must be determined from the two boundary conditions at the interface of the c and d phases. The most convenient way is to match the mass flow $\jmath=\rho_n v_n+\rho_s v_s$ and the normal velocity v_n at the interface. The first condition is obvious and expresses the conservation law of the total mass

$$\jmath(R(t),t)=-\Delta\rho\dot{R}(t).$$

Here $\Delta\rho=\rho'-\rho$ is the difference in the densities of the c and d phases at the interface.

As it concerns the second boundary condition for the normal velocity, the well-defined point of view is absent. One approach, based on the dissipationless model of the demixing kinetics,[3,5] derives the boundary condition under assumption of the lack of any ^{3}He diffusion flow. But within the framework of this model there arises a problem associated with the impossibility to satisfy simultaneously one more necessary requirement due to nonzero latent heat, namely, the continuity of the entropy flow across the interface. The other approach[10,4] is based on the assumption that the normal component of a superfluid liquid, connected with the normal excitations, should stick to the surface of the normal c-phase droplet. Unfortunately, present experiments cannot support any point of view. That is why we impose the boundary condition for the normal velocity in a general form in order to embrace any case

$$v_n(R(t),t)=\gamma\dot{R}(t).$$

Here γ is the accommodation coefficient which describes the sticking of the normal component to the surface of the c-phase droplet, γ being $(1-\rho'c'/\rho c)$ for the dissipationless case and $\gamma=1$ for the second assumption.

To determine the unknown amplitudes A_1 and A_2 within our approximation, it is sufficient to restrict ourselves by first-order time derivative alone. Then we have

$$A_1(t)=-\frac{\gamma\beta+(1+\beta)\Delta\rho/\rho}{1+\beta^2\rho_s/\rho_n}\dot{V}(t),$$

$$A_2(t)=\frac{\gamma+(1-\beta\rho_s/\rho_n)\Delta\rho/\rho}{1+\beta^2\rho_s/\rho_n}\dot{V}(t), \tag{3}$$

where $V(t)=4\pi R^3(t)/3$ is the volume of an expanding droplet. Thus we arrived at the full description of the state of the superfluid d phase.

We are now in the position to derive the growth equation, using, e.g., the law of the energy conservation. The total energy of the system is a sum of the surface energy $E_s=4\pi\alpha R^2$ and bulk E' and E energies of the c and d phases. Obviously, one can write the time derivative for the total energy of the system

$$\partial(E'+E+E_s)/\partial t=Q'-Q$$

or, identically, as a boundary condition at $r=R$

$$(Q'-4\pi R^2\rho'\epsilon'\dot{R})-(Q-4\pi R^2\rho\epsilon\dot{R})-\frac{2\alpha}{R}4\pi R^2\dot{R}=0.$$

Here $\rho'\epsilon'$ and $\rho\epsilon$ are the energies of the c and d phases per unit volume, α is the surface tension. The fluxes $Q'\equiv 0$ and $Q=4\pi R^2 q$ are the energy fluxes across the droplet surface and the density of the energy flux q is given by[8]

$$\boldsymbol{q}=\jmath(\phi+v_s^2/2)+(\rho\sigma T+\rho cZ)\boldsymbol{v}_n+\rho_n\boldsymbol{v}_n\cdot(\boldsymbol{v}_n-\boldsymbol{v}_s)\boldsymbol{v}_n+\boldsymbol{q}_{\mathrm{dis}}.$$

Here $\boldsymbol{q}_{\mathrm{dis}}$ is an additional dissipative flux associated with the processes of viscosity, diffusion, and heat conduction in the d phase, $\phi=\mu_4/m_4$ is the ratio of the chemical potential of a ^{4}He atom to its mass, and $Z=\mu_3/m_3-\mu_4/m_4$ is the second thermodynamic potential of a mixture.

In the course of the calculation of the energy flux we will omit the terms proportional to $\dot{R}^3$ and higher orders. Thus, since the energy flux vanishes at $\dot{R}=0$, it is sufficient to take into account only the first correction for the deviations of the thermodynamic quantities from their values at infinity. For calculating the dissipative contribution into the total energy flux at the droplet surface, it is convenient to use the dissipative function of a mixture and estimate the total dissipative flux $Q_{\mathrm{dis}}=4\pi R^2 q_{\mathrm{dis}}$ as an integral over the whole bulk of the d phase from the dissipative function

$$Q_{\mathrm{dis}}=\int_{r>R(t)}W_{\mathrm{dis}}\,4\pi r^2\,dr, \tag{4}$$

where the dissipative function W_{dis} is equal to the energy dissipation per unit volume and unit time reads[8]

$$W_{\rm dis}=\frac{\eta}{2}\left(\frac{\partial v_{ni}}{\partial r_k}+\frac{\partial v_{nk}}{\partial r_i}-\frac{2}{3}\delta_{ik}\frac{\partial v_{nl}}{\partial r_l}\right)^2 +\rho D\frac{\partial Z}{\partial c}\left(\nabla c+\frac{k_T}{T}\nabla T+\frac{k_P}{P}\nabla P\right)^2+\kappa\frac{(\nabla T)^2}{T}. \quad (5)$$

Here η is the viscosity, D, $k_T D$, and $k_P D$ are the coefficients of the diffusion, thermo-, and barodiffusion, and κ is the heat conductivity. In the above expression we consciously omitted the three terms proportional to the second coefficients $\zeta_\imath$ of viscosity since these terms, arising from nonzero div $\boldsymbol{v}_n$ and div$\mathbf{J}$, will give no contribution within the accuracy of our approximation.

Let the index *''0''* refer to the equilibrium values of the all thermodynamic quantities taken away at infinity. Involving the thermodynamic relation $\rho\epsilon=P+\rho\sigma T+\rho c Z+\rho\phi$ and boundary conditions valid for first-order approximation in the slowness of $\dot{R}(t)$,

$$-\rho' c'\dot{R}=\rho_0 c_0(v_n-\dot{R}),$$

$$-\rho'\sigma'\dot{R}=\rho_0\sigma_0(v_n-\dot{R}),$$

we arrive at the following equation:

$$dU/dt=-Q^{(2)}-Q_{\rm dis}.$$

Here U is the energy of the formation of a droplet

$$U(R)=4\pi\alpha R^2+[\rho'(\phi'-\phi_0)-(P'-P_0)+\rho'\sigma'(T-T_0) +\rho'c'(Z'-Z_0)]4\pi R^3/3 \quad (6)$$

and the flux $Q^{(2)}$ across the droplet $r=R$ surface equals

$$Q^{(2)}=[J\delta\phi+v_n\delta(\rho\sigma T+\rho c Z)+\dot{R}\phi_0\delta\rho-\dot{R}\delta(\rho\epsilon)]4\pi R^2.$$

Omitting the algebraic calculations, we find using Eqs. (1), (2), and (3) that $Q^{(2)}$ can be represented as

$$Q^{(2)}=\frac{d}{dt}\left(\frac{1}{2}\rho_{\rm eff}\frac{\dot{V}^2(t)}{4\pi R(t)}\right)+\rho_0\frac{\ddot{V}^2(t)}{4\pi u_{\rm eff}}.$$

The effective density $\rho_{\rm eff}$ of a droplet is given by the expression in which, for brevity, we omit the index *''0''*

$$\frac{\rho_{\rm eff}}{\rho}=\left(\frac{\Delta\rho}{\rho}\right)^2+\frac{\rho_n}{\rho_s}\left(\gamma+\frac{\Delta\rho}{\rho}\right)^2 +(1+\beta)(\gamma-1)\frac{\gamma+(1-\beta\rho_s/\rho_n)\Delta\rho/\rho}{1+\beta^2\rho_s/\rho_n}\frac{\sigma T+cZ}{u_2^2}. \quad (7)$$

As expected, in the $u_2=\infty$ limit the effective density of a droplet goes over into the expression found in the model of an absolutely incompressible mixture.[3] Within the latter model this term, considered as a kinetic energy of a droplet, can be estimated as an integral of the kinetic energy density over the whole bulk of a mixture. Note only that, if the accommodation coefficient γ differs noticeably from unity, the correction from the second-sound mode becomes of the order of unity.

The second term describes the emission of the first- and second-sound waves from a spherical droplet expanding at the rate $\dot{R}(t)$ in the superfluid liquid. The total intensity of the emission is determined by the effective velocity $u_{\rm eff}$ depending on both sound velocities u_1 and u_2:

$$\begin{aligned}(1+\beta^2\rho_s/\rho_n)u_{\rm eff}^{-1}=&\frac{\rho_n}{\rho_s}[\gamma+(1-\beta\rho_s/\rho_n)\Delta\rho/\rho]^2\\ &\times\left(1+\frac{(1+\beta)^2}{1+\beta^2\rho_s/\rho_n}\frac{\rho_s}{\rho_n}\frac{\sigma T+cZ}{u_2^2}\right)u_2^{-1}\\ &+(\gamma\beta+(1+\beta)\Delta\rho/\rho)^2\\ &\times\left(1-\frac{(1+\beta)(1-\beta\rho_s/\rho_n)}{1+\beta^2\rho_s/\rho_n}\right.\\ &\left.\times\frac{\gamma+(1-\beta\rho_s/\rho_n)\Delta\rho/\rho}{\gamma\beta+(1+\beta)\Delta\rho/\rho}\frac{\sigma T+cZ}{u_2^2}\right)u_1^{-1}.\end{aligned} \quad (8)$$

This sound emission term becomes comparable with the kinetic energy term at the growth rates $\dot{R}_u\sim(\rho_{\rm eff}/\rho)u_{\rm eff}$. In a saturated ^{3}He-^{4}He mixture owing to the inequality $u_2\ll u_1$ the main contribution is connected with the second-sound mode and $\dot{R}_u=u_2$. Only if the excitation of the second-sound mode is suppressed, i.e., $\gamma^2<(\rho_s/\rho_n)(\Delta\rho/\rho)^2$, $\dot{R}_u$ exceeds u_2 and for $\gamma=0$ one has $\dot{R}_u\approx u_2\rho/\rho_n$. In any case the involvement of the sound emission becomes significant if one deals with the damping of the radial pulsations of a droplet.

Let us turn now to evaluating the dissipative flow $Q_{\rm dis}$ Eq. (4) which can be represented as a sum of three contributions due to viscosity, diffusion, and heat conduction

$$Q_{\rm dis}=Q_\eta+Q_D+Q_\kappa.$$

The simple calculation, using Eqs. (5), (1), and (2), yields

$$Q_\eta=16\pi\gamma^2\eta R\dot{R}^2.$$

This viscous flux corresponds completely to the usual Stokes force which hinders the radial expansion of a droplet in the superfluid liquid.

The last two contributions into $Q_{\rm dis}$ have the same structure and can be represented using the absorption coefficients $a_D^{(1,2)}$ and $a_\kappa^{(1,2)}$ in the sound damping $\gamma_{D,\kappa}^{(1,2)}=a_{D,\kappa}^{(1,2)}\omega^2/2$ due to diffusion and heat conduction in the first and second modes, respectively,

$$Q_D+Q_\kappa=\rho[(b_1\sqrt{a_D^{(1)}u_1}+b_2\sqrt{a_D^{(2)}u_2\rho_n/\rho_s})^2 +(b_1\sqrt{a_\kappa^{(1)}u_1}+b_2\sqrt{a_\kappa^{(2)}u_2\rho_n/\rho_s})^2]\frac{\ddot{V}^2(t)}{4\pi R}. \quad (9)$$

Here b_1 and b_2 denote

$$b_1=\frac{\gamma\beta+(1+\beta)\Delta\rho/\rho}{\sqrt{1+\beta^2\rho_s/\rho_n}};\quad b_2=\frac{\gamma+(1-\beta\rho_s/\rho_n)\Delta\rho/\rho}{\sqrt{1+\beta^2\rho_s/\rho_n}}.$$

The expressions for $\gamma_{D,\kappa}^{(1,2)}$ can be found, e.g., in Ref. 11. The second-sound mode plays a main role in the heat conduction flux Q_κ.

To conclude, the growth equation for the evolution of a spherical droplet in a superfluid ^{3}He-^{4}He mixture can be represented in the following general form:

$$\frac{d}{dt}\left(4\pi\rho_{\mathrm{eff}}R^3\frac{\dot{R}^2(t)}{2}+U(R)\right)$$
$$=-16\pi\gamma^2\eta R\dot{R}^2-\rho\frac{\ddot{V}^2}{4\pi u_{\mathrm{eff}}}-\rho\frac{\nu_{\mathrm{eff}}}{u_{\mathrm{eff}}^2}\frac{\ddot{V}^2}{4\pi R}, \qquad (10)$$

where the physical parameters are given by Eqs. (6), (7), (8), and the last term is given by a sum of the diffusion and heat conduction fluxes (9). The physical meaning of the growth equation is obvious. The dissipative function on the right-hand side determines at which rate the droplet energy, being a sum of the kinetic and potential energies, dissipates. The first term in the dissipative function corresponds to the viscous Stokes force. The second is an intensity of the sound emission provided a body, immersed into a superfluid mixture, changes its volume. Unlike the well-known case of a normal liquid, the intensity of the emission depends on the both second- and first-sound velocities (8).

The last term is responsible for the irreversible energy losses due to diffusion and heat-conduction processes in the bulk of a mixture. From the dimensional speculations one may expect $\nu_{\mathrm{eff}}\propto u_{\mathrm{eff}}\ell(T)$ where $\ell(T)$ is the mean free path of excitations. Compared with the sound emission intensity, in the limit of large droplets these energy losses are of minor importance due to the hydrodynamic inequality $R\gg\ell(T)$. The latter circumstance is in drastic contrast to normal liquid mixtures where the diffusion and heat-conduction energy losses dominate over the sound emission in the hydrodynamic $R\gg\ell(T)$ limit.

Essentially, the equation (10) for a radial expansion of a droplet represents a generalization of the well-known Rayleigh-Plesset equation[12] for the bubble growth in a normal fluid to the case of a superfluid mixture. On the appropriate choice of the potential energy $U(R)$ of a droplet the growth equation (10) can also be used for studying, e.g., dynamics of the formation of electron bubbles[13] or, if $\Delta\rho=-\rho$ and $U(R)=4\pi\alpha R^2+4\pi PR^3/3$ where P is the applied external pressure, for studying pulsation dynamics of the cavitation bubbles in superfluid ^{4}He.[14] The parameters of Eq. (10) can be found experimentally by studying the time dependence of the droplet radius under various conditions, in particular, by measuring frequency and damping of the droplet pulsations in the ultrasound field. This can give certain information on the value of the accommodation coefficient γ since the other physical quantities determining parameters in Eq. (10) are known. For sufficiently low temperatures, when the *c*-phase droplet undergoes the transition into the superfluid state, one may expect a variation of the accommodation coefficient.

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