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Second Sounds in Phase-Separating ^{3}He–^{4}He Mixtures

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Second-sound propagation is investigated in superfluid phases of ^{3}He–^{4}He mixtures in the presence of finely divided nonsuperfluid droplets. The attenuation is enhanced by diffusion of the entropy and the ^{3}He component through the interface. At relatively high frequencies, it is proportional to the surface area density. At very low frequencies, we predict anomalous damping due to first-order phase transition periodically taking place at the interface. The second-sound speed is considerably decreased at low frequencies.

Bodensohn and Leiderer[1)] have recently observed that second-sound damping in a superfluid phase is strongly enhanced in ^{3}He–^{4}He mixtures near the tricritical point in the presence of finely divided nonsuperfluid droplets. That is, the attenuation α per unit length increases with onset of nucleation and varies in proportion to the surface area density A of the droplets. The observed maximum of α is of the order of 1 cm^{-1}. In terms of the attenuation per wavelength, $\alpha_\lambda = 2\pi c_{\rm II}\alpha/\omega$, the corresponding maximum $(\alpha_\lambda)_{\rm max}$ is of the order of 10^{-2}. The second-sound velocity $c_{\rm II}$ and the acoustic frequency ω are around 8×10^2 cm/sec and 5×10^5 sec^{-1}, respectively. The wavelength used is of the order of 10^{-2} cm and is much longer than the characteristic domain size R and the interdomain distance. The aim here is to examine second-sound propagation in such two-phase states of ^{3}He–^{4}He mixtures at relatively low frequencies. In this letter, we neglect scattering of second sounds by droplets.[2)] It is known that the resultant attenuation $\alpha_{\rm scat}$ is proportional to ω^4 when the sound wavelength $\lambda = 2\pi c_{\rm II}/\omega$ is much longer than the domain size R. We expect that the scattering mechanism will be negligible at low frequencies as compared to a diffusion mechanism which we will propose in the following.

In the presence of nonvanishing superfluid mass density ρ_s, the superfluid velocity v_s and the chemical potential μ_4 of ^{4}He molecules are coupled to form the second-sound mode near equilibrium. In ^{3}He–^{4}He mixtures, we have an additional diffusive mode, "a concentration mode", which is decoupled from the second-sound mode to linear orders in deviations in homogeneous superfluid states. This mode constitutes a bottleneck which slows down phase separation processes in ^{3}He–^{4}He mixtures.[3)] When droplets of ^{3}He-rich normal fluids are dispersed, two types of diffusive relaxations are induced near the interface of the droplets, giving rise to extra dissipation of second sounds. Firstly, μ_4 must relax diffusively inside the droplets because of the nonexistence of ρ_s inside them. Its diffusion constant D_f is much larger than that of the concentration mode D_0 near the tricritical point.[4,5)]* Secondly, second sounds induce the concentration mode on both the superfluid and normal fluid sides separated by the interface. In the experiment, the latter slower diffusion process seems to have dominantly dissipated second sounds at least for $R/\xi \lesssim 100$, ξ being the correlation length. We furthermore expect $l_D < R$ there, $l_D = (D_0/\omega)^{1/2}$ being the diffusion length. Then, the volume fraction of the inhomogeneous region is of the orger of $l_D A$ and the resultant attenuation should be proportional to the surface area density A, as in the experiment.

Following ref. 4, we introduce two scalar dimensionless deviations,

$$\delta c_1 = -(k_B T)^{-1}\delta\mu_4 = (k_B T)^{-1}(s\delta T + X\delta\Delta), \tag{1}$$

* In ref. 5 D_f is written as D_2 and D_0 as D_1.

$$\delta c_2 = k_B^{-1}(-X\delta s + s\delta X), \tag{2}$$

where s is the entropy per particle, X is the molar concentration, and $\Delta = \mu_3 - \mu_4$ is the chemical potential difference per particle. Hereafter, we neglect the pressure deviation, so $\delta p = 0$. It is easy to see why δc_2 is decoupled from the second-sound mode.* The s and X are convected by the normal fluid velocity v_n as

$$\frac{\partial}{\partial t}(ns) = -\nabla\cdot(nsv_n), \tag{3}$$

$$\frac{\partial}{\partial t}(nX) = -\nabla\cdot(nXv_n), \tag{4}$$

where the dissipation has been neglected and n is the number density. For second sounds we have $\delta p \cong 0$ and $\rho_s v_s + \rho_n v_n \cong 0$, so that $v_n \cong -(\rho_s/\rho_n)v_s$. From eqs. (3) and (4), we find that δc_2 evolves only diffusively both in superfluid and normal fluid phases as

$$\frac{\partial}{\partial t}\delta c_2 = D_0 \nabla^2 \delta c_2. \tag{5}$$

Here we neglect the cross diffusion ($\propto \nabla^2 \delta c_1$) on the right-hand side,[5)] which does not change the following conclusions qualitatively. We note that s and X in definition (2) should be the average values on the superfluid side, to be precise. The δc_1 is coupled to v_s in superfluids as

$$m_4 \frac{\partial}{\partial t} v_s = -\nabla(\delta\mu_4), \tag{6}$$

where m_4 is the ^{4}He mass and the dissipation has been neglected. In normal fluids $\delta\mu_4$ obeys

$$\frac{\partial}{\partial t}\delta\mu_4 = D_f \nabla^2 \delta\mu_4. \tag{7}$$

However, note that D_0 and D_f take different values in the two coexisting phases. We will therefore write $D_0 = D_{0s}$ or D_{0n} and $D_f = D_{fs}$ or D_{fn} in the superfluid or normal fluid phase where confusion may occur.** In equilibrium, the long-wavelength Fourier components of δc_1 and δc_2 have Gaussian distributions with variances χ_1 and χ_2,

$$\langle \delta c_{ik} \delta c_{jk}^* \rangle = \chi_i \delta_{ij}. \tag{8}$$

The orthogonality follows from eqs. (1) and (2) in the superfluid phase (and in the two phases approximately for $T \cong T_t$). Near the tricritical point, we have $\chi_1 \sim 1/n$ and $\chi_2 \sim 1/nt_t$, where $t_t = 1 - T/T_t$. The kinetic coefficient $\lambda_{11} = D_f \chi_1$ grows strongly as $t_t^{-(1+p)/2}$ with $p = (1/3) + O(4-d)$, d being the dimensionality in the $\varepsilon = 4-d$ expansion.[4,5)] However, the kinetic coefficient $\lambda_{22} = D_0 \chi_2$ is nearly constant, so $D_0 \propto t_t$. We will also write $\chi_i = \chi_{is}$ or χ_{in} for the two phases to avoid confusion, if necessary.

Next we calculate the heat dissipation rate $\dot{Q}$ of a second sound per unit volume in the presence of spherical domains with a small volume fraction ϕ. It is related to the attenuation α by $\dot{Q} = 2c_{\rm II} E\alpha$, where $E \cong \rho_s \overline{v_s^2} \cong \overline{\delta c_1^2}/\chi_{1s}$ is the acoustic energy density averaged over the period $2\pi/\omega$ (see ref. 2). Due to inhomogeneities of δc_1 we find the following contribution:

$$\dot{Q}_1 = \int d\boldsymbol{r} D_f \frac{1}{\chi_1} |\nabla \delta c_1|^2$$
$$\cong [D_{fs} k^2 + 3\phi\omega \mathscr{F}_n(\omega R^2/D_{fn})]E, \tag{9}$$

where the integral is over a unit volume, $k = \omega/c_{\rm II}$, and

$$\mathscr{F}_n(x) = -\text{Im}\,[\coth\sqrt{ix}/\sqrt{ix}] - \frac{1}{x}. \tag{10}$$

In Fig. 1 we plot $\mathscr{F}_n(x)$ on a semilogarithmic scale. The $\mathscr{F}_n(x)$ tends to $(2x)^{-1/2}$ for $x \gg 1$ and to $(1/15)x$ for $x \ll 1$, and it attains a maximum of 0.12 at $x \cong 12$. The attenuation $\alpha_\lambda^{(1)}$ per wavelength due to $\nabla \delta c_1$ then becomes

$$\alpha_\lambda^{(1)} = \pi D_{fs}\omega/c_{\rm II}^2 + 3\pi\phi \mathscr{F}_n(\omega R^2/D_{fn}). \tag{11}$$

The first term is the usual term for homogeneous superfluids.*** The second term arises from integrals within the domains. We have calculated δc_1 inside the domains from eq. (6) by setting $\partial/\partial t = i\omega$ and imposing $\delta c_1 = \text{const.}$ at the interface. The δc_1 outside the domains is supposed to be homogeneous

* Instead of s and X, we may use the entropy σ per unit mass and the mass concentration c. This alternative choice does not essentially change the definition of δc_2 because $\delta\sigma/\sigma - \delta c/c = \delta s/s - \delta X/X$.

** In the superfluid phase D_{fs} appears in the second-sound damping (see eq. (11)).

*** We are neglecting the attenuation arising from the relaxation of the complex order parameter.

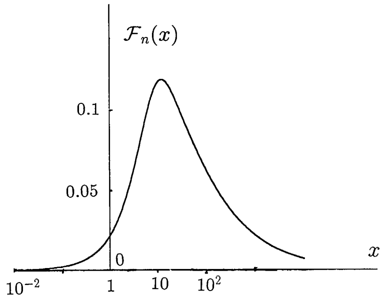

Fig. 1. $\mathscr{F}_n(x)$ versus x on a semilogarithmic scale.

on the scale of R because their small scale deviations are convected away from the droplets by counterflows. The second term attains a maximum of order ϕ at $\omega \sim D_f R^{-2}$. For $\omega \ll D_f R^{-2}$, eqs. (9) and (10) lead to

$$\alpha_\lambda^{(1)} \cong \pi \left[D_{fs}/c_{II}^2 + \frac{1}{5}\phi R^2/D_{fn} \right] \omega. \tag{12}$$

The domain contribution exceeds the usual damping for $\phi(R/\xi)^2 > 5D_{fs}D_{fn}/(c_{II}\xi)^2$, where the right-hand side is of the order of t_t^{-p} with $p \sim 1/3$ near the tricritical point. For $\omega \gg D_f R^{-2}$ we have

$$\alpha_\lambda^{(1)} \cong \pi [D_{fs}/c_{II}^2 + 3\phi(D_{fn}/2\omega R^2)^{1/2}]. \tag{13}$$

The dissipation rate $\dot{Q}_2$ from $\nabla \delta c_2$ is the integral of $D_0 |\nabla \delta c_2|^2/\chi_2$. To calculate the attenuation $\alpha_\lambda^{(2)}$ from $\nabla \delta c_2$ we must specify the boundary condition of δc_2 at the interface. We impose the continuity of δT, $\delta \Delta$, and $\delta \mu_4 = -s\delta T - X\delta \Delta$. Since s and X are discontinuous, we find at the interface,

$$\delta \Delta = -\left(\frac{\Delta s}{\Delta X}\right) \delta T = \left(\frac{\partial \Delta}{\partial T}\right)_{cxs} \delta T, \tag{14}$$

where $(\partial \cdots / \partial \cdots)_{cxs}$ is the derivative along the coexistence curve at fixed p. This relation means that the deviations on the interface are confined on the coexistence surface. After some calculations, the ratio $\delta c_2/\delta c_1$ at the interface is calculated near the tricritical point as

$$\delta c_2/\delta c_1 = A_0 \equiv T\left(\frac{\partial X}{\partial T}\right)_{cxs} \left[s - \left(\frac{\partial s}{\partial X}\right)_{cxs} X \right] \Bigg/ \left[s + \left(\frac{\partial \Delta}{\partial T}\right)_{cxs} X \right], \tag{15}$$

which is equal to $-\alpha_1/\alpha_2$ in the notation of ref. 5, and we find $A_0 \cong -1/0.696 \cong -1.44$ at the tricritical point at saturated vapor pressure, using data in ref. 6. Using eqs. (5) and (14), we find

$$\alpha_\lambda^{(2)} = 3\pi\phi A_0^2 \chi_{1s} \left[\frac{1}{\chi_{2n}} \mathscr{F}_n(\omega R^2/D_{0n}) + \frac{1}{\chi_{2s}} \mathscr{F}_s(\omega R^2/D_{0s}) \right], \tag{16}$$

where

$$\mathscr{F}_s(x) = \frac{1}{x} + \frac{1}{\sqrt{2x}}. \tag{17}$$

The first term in the brackets arises from the dissipation within the droplets, and the second term from that outside them.

Near the tricritical point[5)*] we have $\chi_1/\chi_2 \sim t_t \ll 1$ and $D_0/D_f \sim t_t^{(3+p)/2} \ll 1$. As a result, the domain contribution $\alpha_\lambda^{(1)}$ exceeds $\alpha_\lambda^{(2)}$ for $\omega > \omega_{cr}$ and vice versa for $\omega < \omega_{cr}$. The crossover frequency ω_{cr} is given by

$$\omega_{cr} \sim (\chi_1/\chi_2)^{2/3} D_f^{2/3} D_0^{1/3} R^{-2}, \tag{18}$$

which is between $D_0 R^{-2}$ and $D_f R^{-2}$ near the tricritical point. The experimental values are $t_t \sim 4 \times 10^{-2}$, $\omega R^2/D_{0s} \sim 5 \times 10^{-3}(R/\xi)^2 \gg 1$, and $D_{fn}/D_{0s} \sim 10^3$, where $\xi \sim t_t^{-1}$ Å is the correlation length. We may conclude that $D_{0s} \ll \omega R^2 \lesssim D_{fn}$ when α_λ was enhanced in the experiment, under which eqs. (10) and (14) become

$$\alpha_\lambda^{(1)} \sim \phi(D_{0s}/D_{fn})(\omega R^2/D_{0s}), \tag{19}$$

$$\alpha_\lambda^{(2)} \sim 10\phi t_t (\omega R^2/D_{0s})^{-1/2}. \tag{20}$$

We estimate $\alpha_\lambda^{(1)} \sim 10^{-5}\phi(R/\xi)^2$ and $\alpha_\lambda^{(2)} \sim 10\phi(\xi/R)$ for the experiment. Then $\alpha_\lambda^{(2)} \gtrsim \alpha_\lambda^{(1)}$ for $R/\xi \lesssim 10^2$ in accord with the observation of $\alpha_\lambda \propto A \sim \phi/R$. The maximum value $\alpha_\lambda \sim 10^{-2}$ in the experiment also follows from $\phi \sim 0.1$ and $R/\xi \sim 10^2$. However, $\alpha_\lambda^{(1)}$ should have exceeded $\alpha_\lambda^{(2)}$ roughly for $R/\xi \gtrsim 10^2$ or $R \gtrsim 5000$ Å (~the laser light wavelength).

Although not detected in the experiment, most surprising is the low-frequency behavior of $\alpha_\lambda^{(2)}$ for $\omega < D_{0s} R^{-2}$,

$$\alpha_\lambda^{(2)} \cong 3\pi\phi A_0^2 (\chi_{1s}/\chi_{2s})(D_{0s} R^{-2}) \frac{1}{\omega}. \tag{21}$$

* Because $\chi_1/\chi_2 \to 0$ as $T \to T_t$, $\alpha_\lambda^{(2)}$ vanishes as $T \to T_t$. The experiment of ref. 1 was performed not too close to the tricritical point, where $\alpha_\lambda^{(2)}$ is still important.

This behavior ($\propto 1/\omega$) is strikingly different from the usual behavior ($\propto \omega$). The origin is as follows. For $\omega \ll D_{0s}R^{-2}$, δc_2 is nearly homogeneous inside the droplets, but it can extend far from the droplet as

$$\delta c_2 \cong (A_0 \delta c_1) \frac{R}{r} \exp\left[-\sqrt{\frac{i\omega}{D_{0s}}}(r-R)\right], \quad (22)$$

where r is the radius from a droplet center. This is the solution of $i\omega\delta c_2 = D_{0s}\nabla^2\delta c_2$ under eq. (15) at $r=R$. We should note that the diffusion current $-D_{0s}\nabla\delta c_2$ is discontinuous at the interface $r=R$. This means that conversion between the superfluid and the normal fluid takes place at the interface periodically in time. The excess amount of δc_2 arising from the discontinuities of s and X is released at the interface, leading to the long-range variation of δc_2 given by eq. (22). Recall that the current nucleation theory supposes long-ranged variations of the form $\delta c_2 \propto 1/r$ analogous to eq. (22) around growing droplets.

Recently, I have found the same low-frequency, anomalous attenuation $\alpha_\lambda \propto \phi/\omega R^2$ against ordinary sounds in classical two-phase fluids due to latent heat generation at the interface.[7] There, we have shown that interference among droplets suppresses the growth of α_λ at very low frequencies. In our problem, eq. (21) is valid only for $\omega > \phi D_{0s}R^{-2}$. For $\omega < \phi D_{0s}R^{-2}$, $\alpha_\lambda^{(2)}$ becomes small as

$$\alpha_\lambda^{(2)} \cong \frac{\pi}{3\phi} A_0^2(\chi_{1s}/\chi_{2s})(\omega R^2/D_{0s}). \quad (23)$$

A more systematic calculation[7] yields an expression incorporating eqs. (21) and (23) for $\omega < D_{0s}R^{-2}$,

$$\alpha_\lambda^{(2)} = \pi A_0^2(\chi_{1s}/\chi_{2s}) \,\mathrm{Im}\,[1+3\phi D_{0s}/i\omega R^2]^{-1}. \quad (24)$$

So far we have been interested in the attenuation. Moreover, the second-sound velocity is considerably decreased in the presence of droplets in accord with the experiment. The Kramers-Kronig relation[8] implies that the complex sound velocity $c_D(\omega)$ is analytic for $\mathrm{Im}\,\omega > 0$. Here we suppose that the spatial average of δc_1 over volume elements containing many droplets behaves as $\exp[i\omega(t - x/c_D)]$. We find for $\omega > \phi D_{0s}R^{-2}$,

$$c_{\mathrm{II}}/c_D(\omega) - 1 \cong \frac{3}{2}\phi\left(\frac{\coth z_{1n}}{z_{1n}} - \frac{1}{z_{1n}^2}\right) + \frac{3}{2}\phi A_0^2 \chi_{1s}\left[\frac{1}{\chi_{2n}}\left(\frac{\coth z_{2n}}{z_{2n}} - \frac{1}{z_{2n}^2}\right) + \frac{1}{\chi_{2s}}\left(\frac{1}{z_{2s}^2} + \frac{1}{z_{2s}}\right)\right], \quad (25)$$

where $z_{1n} = (i\omega R^2/D_{fn})^{1/2}$, $z_{2n} = (i\omega R^2/D_{0n})^{1/2}$, $z_{2s} = (i\omega R^2/D_{0s})^{1/2}$, and c_{II} is the second-sound velocity without domains. For $\omega < \phi D_{0s}R^{-2}$, we use eq. (24) to obtain

$$c_{\mathrm{II}}/c_D(\omega) - 1 \cong \frac{1}{2}\phi + \frac{1}{2}A_0^2(\chi_{1s}/\chi_{2s}) \times \left[1 - \frac{1}{1+3\phi/z_{2s}^2}\right]. \quad (26)$$

In summary, we have investigated the attenuation and dispersion of second sounds in two-phase ^{3}He–^{4}He mixtures with emphasis on the case near the tricritical point. However, the general features of our predictions are also applicable even to mixtures far from the tricritical point. Systematic experiments with varying acoustic frequency ω and distance from the tricritical point are very informative. Particularly, the anomalous behavior shown in eq. (21) is of great interest and might be used to sensitively detect the onset of nucleation, although the characteristic frequency $D_{0s}R^{-2}$ seems to be too slow for experiments near the tricritical point. More extensive analysis will be performed in the future. We should also examine propagation of the first- and second-sound modes in superfluids in the presence of domains in a normal fluid or crystal phase.

Finally we propose different but closely related experiments on phase-separating ^{3}He–^{4}He mixtures. (i) By application of a stationary shear flow in concentric cylinders, for example, we may realize intriguing two-phase states of ^{3}He–^{4}He mixtures with fine domains and vortex lines. This is because shear breaks up large droplets and competes with the domain growth due to the thermodynamic instability.[9,10] Such two-phase states were investigated by light scattering in usual binary mixtures[11] and in polymer mixtures.[12] (ii) We may periodically drive fluids through an instability point by periodic pressure quenching.[13] Such periodic spinodal decomposition was realized in usual binary mixtures.[14] There, in "a disordered state", the structure factor is dramatically enhanced as $k^{-2.5}$ at small wave number k, while in "an unstable state", phase separation proceeds extremely slowly. In ^{3}He–^{4}He mixtures we furthermore conjecture parametric excitation of second sounds because of forced oscillation of ρ_s or c_{II}.

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