Light Scattering in Superfluid Helium Under Pressure*

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We report here a calculation, based on Landau's hydrodynamical equations, of the intensities of the Brillouin spectral components due to density (first-sound) and entropy (second-sound) fluctuations in pure He II under pressure. In the hydrodynamic regime of temperature, the intensities of second-sound Brillouin components are found to be as strong as *one tenth* of those of the first-sound components under optimum pressure.

As early as 1943 Ginzburg[1] pointed out that Brillouin scattering of light from pure He II should exhibit two doublets corresponding to two independent thermodynamical fluctuations, density and entropy. Indeed, if one ignores the thermal-expansion coefficient of He II, the density fluctuation gets completely decoupled from the entropy fluctuation. A brief thermodynamical analysis shows that the above assumption leads to the condition that $\gamma-1$ is zero, γ being the ratio of specific heats. C_P is very nearly equal to C_V for He II away from the λ temperature under normal pressure. Since light is scattered mainly by the density fluctuation in pure He II, and the intensity is already very weak,[2] the experimental observation of the high-frequency (10^2-10^3 Mc/sec) entropy fluctuation (so-called "second sound") in He II by Brillouin light scattering is exceedingly difficult.

There has been some theoretical work[3] on the possibility of *indirectly* observing sound in ^{3}He :He II mixtures by way of scattered light from ^{3}He concentration fluctuations near critical opalescence. The two basic assumptions involved in this theory are (1) that the entropy fluctuations couple strongly to the concentration fluctuations and (2) that the dielectric constant at optical frequencies has an appreciable dependence on the concentration of ^{3}He impurities. Recently Pikes, Vaughan, and Vinen[4] have reported the detection, with a very low signal-to-noise ratio, of such Brillouin components in ^{3}He:He II mixtures.

A propagating sound wave is always associated with spatial density variation. This density fluctuation in the presence of a finite thermal-expansion coefficient should give rise to a temperature fluctuation in He II. This is, admittedly, a very weak effect in He II under normal conditions. But since there is a marked *increase* in the ratio of the specific heats, γ, with pressure,[5] the coupling between the density fluctuation and the entropy fluctuation may become appreciable at some pressure. This opens up the possibility of observing directly the second-sound Brillouin components in the spectrum of light scattered from first sound. The purpose of this Letter is to report the calculation of the intensities of Brillouin components for first and second sound in He II under pressure and to explore the feasibility of observing them experimentally.

The Kamarov-Fisher-Pecora theory[6] in conjunction with the Wiener-Khintchine theorem[7] states that the spectrum of light [that is, the differential cross section for the photon-scattering process $(\vec{k}_1, \omega_1) \rightarrow (\vec{k}_2, \omega_2)$] is given by the expression

$$k_B T \sum_{\alpha,\beta} (\partial\epsilon/\partial\alpha)_\beta (\partial\epsilon/\partial\beta)_\alpha \chi_{\alpha\beta}''(\vec{k}, \omega)/\omega. \qquad (1)$$

Here ϵ is the optical dielectric constant of the scatterer which is assumed to be a function of thermodynamical variables α and β, k_B is the Boltzmann constant, and $\chi_{\alpha\beta}''$ is the absorptive part of the response function. In order to calculate the absorptive response function we adopt a linear-response theory. According to this theory, if a generalized external perturbing force —say, $F_\beta(\vec{r}, t)$— couples to the thermodynamical variable $\beta(\vec{r}, t)$ of the system, then

$$\chi_{\alpha\beta}''(\vec{k}, \omega)/\omega = \mathrm{Re}[F_\beta(\vec{k}, Z)/\alpha(\vec{k})]_{\omega+i0^+}, \qquad (2)$$

where $F_\beta(\vec{k}, Z)$ is the Laplace transform in time and Fourier transform in space of the adiabatic-disturbance operator $F_\beta(\vec{r}, t)$, and $\alpha(\vec{k})$ is the Fourier transform of $\alpha(\vec{r}, t)$ at $t=0$. The final procedure in calculating $\chi_{\alpha\beta}''(\vec{k}, \omega)$ is to recall that the *linear* response of a disturbed system having slow variations in space and time of physical variables may alternatively be described by a set of linearized hydrodynamical equations.[8] The complicated structure of the absorptive response function can be inferred by requiring that these two descriptions should coincide in the long-wavelength, low-frequency ($\omega\tau < 1$, $kl < 1$) hydrodynamic regime.[9] The analytic structure of the absorptive part of the response function

should contain all the information about the Brillouin components. The *positions* and the *widths* of the spectral lines should be given, respectively, by the real and the imaginary parts of the *poles*, and the corresponding *intensities* should be given by the *residues* at these poles. In what follows we calculate the relevant response functions using existing hydrodynamical equations for He II, which ought to be a valid description in the hydrodynamic temperature regime.

The nondissipative linearized Landau hydrodynamical equations for He II result in two wave-like equations[10]

$$\ddot{\rho}(r,t)-\nabla^2 P(\vec{r},t)=0, \tag{3a}$$

$$\ddot{\rho}-(\rho_s/\rho_n)\sigma_0^{\,2}\nabla^2 T(\vec{r},t)=0. \tag{3b}$$

These two equations, under the assumption that the coefficient of thermal expansion can be neglected, give rise to two independent hydrodynamical modes, corresponding to density and entropy waves. With the help of the Laplace-Fourier transforms (in time and space, respectively) of Eqs. (3a) and (3b), we have derived the following expressions for the relevant absorptive response function from Eq. (2):

$$\chi_{\rho\rho}''(\vec{k},\omega)/\omega=-\gamma\omega^2 u_{10}^{\,-2}\{\omega^2-[u_{20}^{\,2}-\gamma^{-1}(\gamma-1)(\gamma-2)^{-1}u_{10}^{\,2}]k^2\}R(\vec{k},\omega), \tag{4a}$$

$$\chi_{\sigma\sigma}''(\vec{k},\omega)/\omega=-\gamma\omega^2\sigma_0^{\,2}(\rho_s/\rho_n)u_{20}^{\,-2}\{\omega^2-[\gamma^{-1}(\gamma-1)u_{20}^{\,2}-u_{10}^{\,2}]k^2\}R(\vec{k},\omega), \tag{4b}$$

$$\chi_{\rho\sigma}''(\vec{k},\omega)/\omega=-(\gamma-1)\omega^2(\partial\rho/\partial T)_\sigma[\omega^2-(u_{10}^{\,2}+u_{20}^{\,2})k^2]R(\vec{k},\omega). \tag{4c}$$

The resonance function $R(\vec{k},\omega)$ is equal to $\mathrm{Im}[(Z^2-u_1^{\,2}k^2)^{-1}(Z^2-u_2^{\,2}k^2)^{-1}]$ at $Z=\omega+i0^+$, with

$$u_1^{\,2}\simeq(\gamma\beta_T/\rho)[1+\gamma^{-1}(\gamma-1)u_{20}^{\,2}(u_{10}^{\,2}-u_{20}^{\,2})^{-1}], \tag{5a}$$

$$u_2^{\,2}\simeq u_{20}^{\,2}[1-\gamma^{-1}(\gamma-1)u_{10}^{\,2}(u_{10}^{\,2}-u_{20}^{\,2})^{-1}]; \tag{5b}$$

u_{10} and u_{20} are the respective velocities of first and second sound at normal pressure and β_T is the isothermal bulk modulus. In deriving the above expressions for response functions, we have *not* ignored the coefficient of thermal expansion in that we have retained the thermodynamical derivatives such as $(\partial\rho/\partial T)_\sigma$ and $(\partial\sigma/\partial P)_\rho$. This leads to a coupling between first- and second-sound velocities, as can be seen from Eqs. (5a) and (5b). The product involving these thermodynamical derivatives can be expressed in terms of $u_{10}^{\,2}$, $u_{20}^{\,2}$, and γ using the relation

$$(\partial P/\partial\sigma)_\rho(\partial\rho/\partial P)_\sigma(\partial T/\partial\rho)_\sigma(\partial\sigma/\partial T)_\rho=\gamma^{-1}(\gamma-1). \tag{6}$$

The poles of the spectral correlation functions indicate that the Brillouin spectral lines due to first and second sound should occur at $\pm u_1 k$ and $\pm u_2 k$, respectively. The positions of these lines shift with pressure because u_1 and u_2 depend on pressure through $\gamma(P)$. For $\gamma=1$, which amounts to ignoring the thermal-expansion coefficient, $\chi_{\rho\rho}''(\vec{k},\omega)/\omega$ has poles at the first-sound velocity, $\chi_{\sigma\sigma}''(\vec{k},\omega)/\omega$ has poles at the second-sound velocity, and $\chi_{\rho\sigma}''(\vec{k},\omega)/\omega$ vanishes, reflecting a complete decoupling between pressure and temperature fluctuations. Since the coupling between light and the entropy fluctuation is negligibly small [because $(\partial\epsilon/\partial\sigma)_\rho\simeq 0$], we will concentrate mainly on the light scattered from the density fluctuation.

The relative intensities I_1 and I_2 of the Brillouin components due to first and second sound, respectively, can easily be obtained from the density-density correlation function by finding the residues of $\chi_{\rho\rho}''(\vec{k},\omega)/\omega$ at the poles at $\pm u_1 k$ and $\pm u_2 k$. I_1 and I_2 are found to be

$$I_1=(k_B T)(\partial\epsilon/\partial\rho)_\sigma^{\,2}\pi\gamma u_{10}^{\,-2}(u_1^{\,2}-u_2^{\,2})^{-1}\{(\gamma^{-1}u_{10}^{\,2}-u_{20}^{\,2})+u_{10}^{\,2}\gamma^{-1}(\gamma-1)[u_{20}^{\,2}\gamma^{-1}(u_{10}^{\,2}-u_{20}^{\,2})^{-1}+(\gamma-2)^{-1}]\} \tag{7a}$$

and

$$I_2=(k_B T)(\partial\epsilon/\partial\rho)_\sigma^{\,2}\pi\gamma u_{10}^{\,-2}(u_1^{\,2}-u_2^{\,2})^{-1}\{\gamma^{-1}(\gamma-1)u_{10}^{\,2}[u_{20}^{\,2}(u_{10}^{\,2}-u_{20}^{\,2})^{-1}-(\gamma-2)^{-1}]\}. \tag{7b}$$

If C_P is equal to C_V, then the intensity of Brillouin components at the second-sound velocity, I_2, vanishes, which is an expected result. To see how I_1 and I_2 depend on pressure we have to determine the pressure dependence of γ. In order to do this we fall back on Eq. (5a). The use of a few simple thermodynamical relations leads to

$$\gamma(P)=[(\partial\rho/\partial P)_T u_1^{\,2}(P)+u_{20}^{\,2}(u_{10}^{\,2}-u_{20}^{\,2})^{-1}]/[1+u_{20}^{\,2}(u_{10}^{\,2}-u_{20}^{\,2})^{-1}]. \tag{8}$$

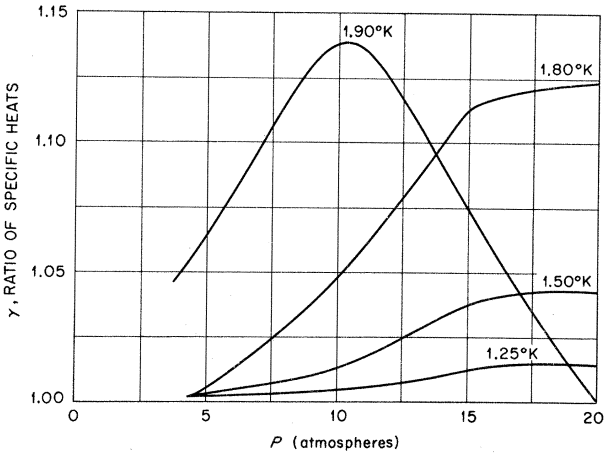

FIG. 1. The pressure dependence of the ratio of the specific heats, C_P/C_V, at different temperatures.

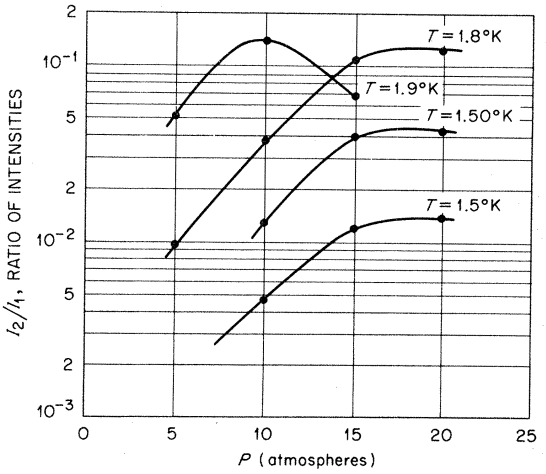

FIG. 2. The pressure dependence of the ratio of the intensities of Brillouin spectral components due to second sound and first sound at different temperatures.

The density of liquid ^{4}He as a function of pressure has been measured by Keesom and Keesom.[11] Using their data we have calculated the values of $(\partial\rho/\partial P)_T$ for different pressures and temperatures. The values of the velocity of first sound under different pressures and temperatures are taken from the data of Atkins and Stasior.[12] Taking the values of $u_{10} = 235$ m/sec and $u_{20} = 20.1$ m/sec at 1.5°K under saturated vapor pressure, we have calculated γ as a function of pressure at different temperatures. This is shown in Fig. 1. There is a sudden drop in $\gamma(P)$ at 1.9°K under about 10 atm pressure. This is probably due to the shift of λ temperature[13] in the vicinity of 1.9°K under such pressure. The intensity of Brillouin components due to first sound is found to remain more or less constant over the pressure range 0-20 atm and between temperatures of 1.25 and 1.90°K, at most 2% higher than the value at saturated vapor pressure. On the other hand, there is a marked increase in I_2 with pressure between 1.25 and 1.9°K. In Fig. 2 we have plotted the ratio I_2/I_1 against pressure on a semilogarithmic scale. As can be seen from this figure, there is an appreciable increase in this ratio with pressure. The value of I_2/I_1 at 1.9°K under 10 atm pressure is about 0.14 ($\simeq\frac{1}{7}$), and at 1.8°K under 15 atm pressure it is about 0.12 ($\simeq\frac{1}{8}$). This prompts us to predict the most favorable conditions for observing the second-sound Brillouin components to be $T \simeq 1.8$-1.9°K and $P \simeq 10$-15 atm. The results obtained at 1.9°K under 10-15 atm pressure should be viewed with caution because the λ temperature may lie very close to 1.9°K, in which case the hydrodynamical approach in calculating the spectral correlation functions is invalidated.

In the preceding calculations we have not included the dissipative terms in the hydrodynamical equations. This led to δ-function–type poles of the response functions. Inclusion of dissipative terms will displace the poles of the resonance function from the real axis and will give rise to finite widths of the spectral components[14] which may again vary with pressure and temperature. This can be demonstrated by calculating the attenuation coefficients of first and second sound in He II under pressure since the linewidth is directly related to the attenuation coefficients. The attenuation coefficient α_1 for first sound in He II under pressure is given by

$$\alpha_1 = 2\pi^2 f^2 \rho^{-1} u_1^{-3} [\tfrac{4}{3}\eta + \zeta_2 + (\gamma - 1)K/C_P], \qquad (9)$$

where f is the frequency of the sound wave, η and ζ_2 are the respective coefficients of the normal and second viscosities, and K is the thermal kinetic coefficient. Dransfeld, Newell, and Wilks[15] have measured α_1 as a function of pressure and temperature for 14.4-Mc/sec sound waves. In the hydrodynamic regime of temperature (1-2°K) the major contribution to α_1 comes from the second viscosity ζ_2, and above 1.4°K α_1 is small and practically independent of pressure. This indicates that the width of the first-sound Brillouin peaks should not diverge between 1.5 and 1.9°K under 10-20 atm pressure. To the author's knowledge no experimental data are available for the attentuation of second sound (α_2) at different pressures. A detailed theoretical analysis due to Khalatnikov[16] shows that the major contribution

to α_2 comes from the thermal conductivity and that α_2 is exceedingly small above 1.5°K. In the light of the above discussion we do not feel that there will be any anomalously large broadening of the Brillouin spectral components in the hydrodynamic regime. A word of caution must be added on this point, namely, that in the Brillouin-light-scattering experiments one is dealing with high-frequency sound waves and the widths of the Brillouin spectral components are already quite large because of the frequency effect. Finally, we would like to make a few comments regarding the resolution of the Brillouin spectral components in He II under pressure. The application of pressure should increase the resolution of the first-sound peaks for the following two reasons: Firstly, pressure increases the velocity of first sound considerably, which will push first-sound Brillouin components away from the incident laser frequency, making them easy to resolve. Secondly, since the attenuation of first sound decreases with pressure, the first-sound Brillouin components should become sharper at higher pressures. The situation is very different for the second-sound Brillouin spectrum. The application of pressure decreases the second-sound velocity[17] and this will make the resolution of Brillouin components comparatively difficult at higher pressures. One may think that the resolution can be increased considerably below 1°K where the velocity of the second sound increases sharply.[18] Unfortunately, the attenuation of second sound is so large below 1°K that it may not be at all possible to resolve the second-sound Brillouin components experimentally.

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