Light Scattering from First and Second Sound near the λ Transition in Liquid He⁺

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We have observed the second-sound contribution to the Brillouin spectrum of liquid He⁴ near the λ transition at saturated vapor pressure and at 25.1 atm. Its spectral shape is compared with theoretical predictions. We have determined the intensity ratio of second-and first-sound scattering between 10^{-5} °K $< T_{\lambda} - T < 10^{-1}$ °K, and we present data on the dispersion and damping of high-frequency first and second sound.

We have used Brillouin scattering to measure the spectrum of the density fluctuations in liquid He^{4} ,^{1,2} near the λ line.³ The scattered intensity at a frequency shift $\omega/2\pi$ is proportional to the dynamic structure factor $S(K, \omega)$, which is the space and time Fourier transform of the density correlation function.⁴ The wave vector of the fluctuation being observed, K, is determined by the wave vector of the light in the medium, k_0 , and the scattering angle θ through the relation $K = 2k_0 \sin(\theta/2)$. $S(K, \omega)$ contains contributions from all normal modes of the fluid. Above the λ temperature, T_{λ} , the modes are those characteristic of normal liquids: adiabatic sound and entropy diffusion. Below T_{λ} the latter mode becomes second sound. At saturated vapor pressure (svp) the coupling of second sound to the density is very weak since the thermal expansion is small; previous Brillouin scattering experiments have not observed the second-sound contribution to the spectrum.⁵ The thermal expansion, however, increases with pressure near the λ line. Therefore, at high pressures the scattering from second sound is expected to be much stronger than at svp as noted first by Ferrell et $al.^{6,7}$ These authors also pointed out that close to T_{λ} one should observe the effect of the critical fluctuations on $S(K, \omega)$ using light scattering.

We have derived a model $S(K, \omega)$ by solving the initial-value problem⁸ for the linearized equations of two-fluid hydrodynamics.⁹ This model spectrum¹⁰ should represent the actual $S(K, \omega)$ except in the critical region very close to T_{λ} . Under the conditions of this experiment the firstsound contribution to $S(K, \omega)$ can be represented by a pair of Lorentzian lines centered at $\omega_1 = \pm u_1 K$ with full width at half-height $2u_1\alpha_1$. u_1 and α_1 are the velocity and attenuation coefficients of first sound, respectively, at the frequency $\omega_1/2\pi$. The second-sound contribution to $S(K, \omega)$ is found at much smaller frequency shifts and has the following form in the limit $u_2/u_1 \ll 1$, where u_2 is the second-sound velocity:

$$S_{2}(K,\omega) \propto \frac{\gamma - 1}{\gamma} \frac{D_{\xi} K^{2} (u_{2}K)^{2} + D_{\kappa} K^{2} \omega^{2}}{(\omega^{2} - u_{2}^{2}K^{2})^{2} + (D_{\xi} + D_{\kappa})^{2} K^{4} \omega^{2}}, \quad (1)$$

with the damping constants

$$D_{\zeta} = (\rho_s / \rho_n) [\rho^{-1}(\frac{4}{3}\eta + \zeta_2) + \rho \zeta_3 - 2\zeta_1],$$

$$D_{\kappa} = \kappa / C_{\rho}.$$

Here ρ_s , ρ_n , and ρ are the superfluid, normalfluid, and total densities; κ , η , and ξ_i are the thermal conductivity, shear viscosity, and second viscosity coefficients, respectively, as defined by Khalatnikov⁹; $\gamma = C_p/C_v$ is the ratio of the specific heats at constant pressure and volume. Well below T_{λ} , $u_2 K \gg (D_{\xi} + D_{\kappa})K^2$, and $S_2(K, \omega)$ approaches a pair of Lorentzian lines centered at $\omega_2 = \pm u_2 K$ whose width is given by $2u_2\alpha_2 = (D_{\xi} + D_{\kappa})K^2$ with α_2 being the attenuation coefficient of second sound at the frequency $\omega_2/2\pi$.

The measured spectra were compared with this theory by using a computer to convolve $S(K, \omega)$ with the measured instrumental profile and to take into account the small spread of K values accepted by the spectrometer. The parameters involved in $S(K, \omega)$ were then adjusted to give the best fit between the computer-generated spectra and the experimental traces.

The light source was a single-mode, frequencystabilized He-Ne laser, having a power of 20 mW. The spectrum of the light, scattered through an angle close to 90°, was analyzed with a spherical Fabry-Perot interferometer of free spectral range 148.98 MHz. The total instrumental width, including the jitter of the laser frequency, was found to be 4.0 MHz. The temperature of the scattering cell was stabilized within \pm 0.015 mK; T_{λ} was determined within the same accuracy. Experiments were performed at svp and at 25.1 atm ($T_{\lambda} = 1.846$ K), where the corresponding val-



FIG. 1. Brillouin spectra showing first and second sound in superfluid He^4 . In the central region of the upper trace (a) the gain is increased by a factor of 10. The counting rate in the second sound in (a) is about 3 Hz.

ues of K near T_{λ} were 1.449×10^5 and 1.457×10^5 cm⁻¹.¹¹

Representative experimental spectra are shown in Fig. 1. One free spectral range of the interferometer is displayed with zero frequency shift in the center. Note, there is no elastically scattered stray light present. The upper trace was taken at svp. The weak second-sound lines are just visible above the noise. The much stronger first-sound lines have a frequency shift of several free spectral ranges. At the higher pressure of 25.1 atm the scattering from second sound becomes comparable to that from first sound as can be seen from the spectra of Figs. 1(b) and 1(c).

The ratio of the intensity in the second-sound components to that in the first sound is given in the hydrodynamic theory to a very good approximation by $I_2/I_1 = \gamma - 1$. Figure 2 shows the intensity ratios measured at 25.1 atm in the temper-



FIG. 2. Ratio of the intensity in the second-sound components to that in the first sound at 25.1 atm. The vertical error bars indicate the uncertainty of the computed values for C_p/C_v-1 ; the horizontal ones, the uncertainty of $T_{\lambda}-T$.



FIG. 3. Velocity (solid symbols) and attenuation coefficient (open circles) of first sound of wave vector $K = 1.457 \times 10^5$ cm⁻¹ at 25.1 atm. The broken curves are guides to the eye.

ature range $0.01 < T_{\lambda} - T < 100$ mK, together with the ratio $\gamma - 1$ which diverges at T_{λ} . γ , which is equal to $\beta_T \rho u_1^2$, was computed using Grilly's data¹² on the isothermal compressibility β_T and our data on the first-sound velocity. The measured ratio is in agreement with $\gamma - 1$ far away from T_{λ} , but begins to fall below it at $\Delta T = T_{\lambda}$ $-T \sim 0.4$ mdeg and remains finite at $T = T_{\lambda}$. This behavior¹³ indicates that close to T_{λ} the correlation length $\xi(T)$, associated with the critical fluctuations, is becoming comparable to K^{-1} , and therefore two-fluid hydrodynamics is no longer applicable. A calculation of I_2/I_1 , which includes an Ornstein-Zernike correction similar to that used at the gas-liquid critical point, has been carried out by Stephen.¹⁴ Using his theory, together with the temperature dependence¹⁵ of $\xi(T)$ $=\xi_0 (\Delta T/T_\lambda)^{-2/3}$, we determined $\xi_0 = 2.0 \pm 0.4$ Å at 25.1 atm. This value is in satisfactory agreement with estimates of ξ_0 at svp.¹³

Figure 3 shows the data we obtained at 25.1 atm on the velocity and attenuation of ≈ 800 -MHz first sound. Included for comparison is the zerofrequency velocity $u_1(0)$ whose absolute value is matched to agree with our data at $\Delta T = 10$ mdeg.¹⁶ The minimum of $u_1(0)$ occurs directly at T_{λ}^{17} ; in contrast, our data for ≈ 800 MHz show the



FIG. 4. Frequency shift $u_2 K$ and linewidth $(D_{\zeta} + D_{\kappa}) K^2$ of second sound at 25.1 atm. Circles, our data; the solid and dashed curves show extrapolations of low-frequency data taken from Refs. 19 and 20.

minimum to be located at $\Delta T = 5.5 \pm 1$ mdeg. This remarkable displacement of the minimum is due to the dispersion of the velocity which at T_{λ} amounts to at least 4.1 ± 0.3 m/sec. Our data on ≈ 500 -MHz first sound at svp yield at T_{λ} a dispersion $u_1(500$ MHz) - $u_1(0) = 2.3 \pm 0.2$ m/sec which is much larger than the values observed previously at lower frequencies.¹⁸

Figure 4 shows our measured values for the frequency shift $u_2 K$ and the linewidth $(D_{\zeta} + D_{\kappa})K^2$ of second sound at 25.1 atm. Our analysis based on Eq. (1) gives, in addition, the ratio $D_{\chi}/(D_{\chi})$ $+D_{\kappa}$) which is found to be 0.6 ± 0.1 for $\Delta T < 4$ mdeg. Included in Fig. 4 for comparison are extrapolations based on values for u_2^{19} and D_{ζ} $+D_{b}^{20}$ measured at wavelengths larger by about 4 orders of magnitude. Our linewidth data agree well with the extrapolation for $\Delta T \gtrsim 10$ mdeg. However, closer to T_{λ} they do not increase with temperature in contrast to the low-frequency results which indicate a $\Delta T^{-1/3}$ divergence. ^{6, 15, 20} That deviation from the hydrodynamic behavior is unexpected since it is already evident at temperatures where $K\xi \sim 0.2$. It also is quite surprising since our data on the frequency shift (see Fig. 4) show no measurable velocity dispersion at all temperatures studied. It is as yet unexplained.

At present there is no theory available which predicts $S_2(K, \omega)$ for temperatures $|\Delta T| < 0.5$ mdeg where $K\xi \ge 1$. However, it has been suggested⁶ that the hydrodynamic form for $S_2(K, \omega)$ might also describe the spectrum in this temperature region. The special form considered in VOLUME 30, NUMBER 10

Ref. 6 corresponds in Eq. (1) to setting $D_{\zeta} = 0$. A computer fitting at $\Delta T = 1$ mdeg with $D_{\chi} = 0$ is illustrated in Fig. 1(c) as the dotted curve. It obviously does not describe the valley region. On the other hand, a fit with $D_{\zeta} \neq 0$, represented by the solid curve, matches the experimental trace remarkably well. We have continued our analysis based on Eq. (1) for $\Delta T \leq 0.5$ mdeg and have found no deviation from Eq. (1) within the noise of the counting statistics. However, an exact comparison is made difficult by the fact that the frequency shifts are smaller than the instrumental width at these temperatures. In the future, higher-resolution experiments will be necessary to decide upon the possible presence of an additional, but weak central component⁶ in $S(K, \omega)$ very close to T_{λ} .

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