Neutron Scattering from Liquid ^{3}He

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The dynamical form factor $S(Q,\omega)$ of liquid ^{3}He, including both the coherent and the incoherent contributions, is calculated within the random-phase approximation. At low temperature a sharp zero-sound mode is obtained. At higher temperature this mode becomes a broad resonance in the particle-hole spectrum. This temperature effect may help account for the difference in $S(Q,\omega)$ observed in two recent neutron-scattering experiments.

Recent measurements[1-3] of neutron scattering from liquid ^{3}He yield a scattering intensity proportional to the dynamic form factor[4]

$$S(Q,\omega) = (\sigma_c/\sigma)S_c(Q,\omega) + (\sigma_i/\sigma)S_i(Q,\omega). \qquad (1)$$

Here $\hbar Q$ ($\hbar\omega$) is the momentum (energy) transferred from the neutron to the liquid. This form factor has a coherent part

$$S_c(Q,\omega) = N^{-1}\int_{-\infty}^{+\infty} \langle \rho(-Q,0)\rho(Q,t)\rangle e^{-i\omega t}\,dt, \qquad (2)$$

proportional to the correlation in the Qth Fourier component of the total particle density at time t, $\rho(Q,t) = \rho_\uparrow(Q,t) + \rho_\downarrow(Q,t)$, and an incoherent part,

$$S_i(Q,\omega) = 4N^{-1}\int_{-\infty}^{\infty} \langle G_Z(Q,0)G_Z(Q,t)\rangle e^{-i\omega t}\,dt \qquad (3)$$

which for an isotropic system is proportional to the correlations in the Qth Fourier component of the spin density, $G_Z(Q,t) = \frac{1}{2}[\rho_\uparrow(Q,t) - \rho_\downarrow(Q,t)]$. Here $\rho_\uparrow$ ($\rho_\downarrow$) is the density of particles with spin up (down), $\sigma = \sigma_c + \sigma_i$ is the sum of the coherent (σ_c) and the incoherent (σ_i) scattering cross section, and N is the total number of particles.

In the pioneering experiments on liquid ^{3}He at 1.3 and 0.63 K (Refs. 1 and 2, respectively), a broad scattering intensity was observed over a wide range of frequency for wave vector 1.0 A^{-1} $< Q <$ 2.8 A^{-1}. No sharp structure corresponding to a zero-sound mode—known to exist at small Q and low temperature[5]—was found. Recent experiments[3] at 15 mK do show structure characteristic of a well-defined zero-sound mode superimposed on a broad scattering intensity. In 1965 Pines[6] proposed the existence of zero sound at finite Q. Pines and Nozières[7] have investigated $S_c(Q,\omega)$ in detail. Recently several calculations of $S_c(Q,\omega)$ at $T = 0$°K, either in random-phase approximation[2,8] (RPA) or using the continued-fraction method,[9] have appeared.

The purpose of this Letter is (a) to compare these different experimental results and (b) to describe the broad features of the scattering within a simple model. This is done by calculating both $S_c(Q,\omega)$ and $S_i(Q,\omega)$ at finite temperatures. We domonstrate (a) that a zero-sound mode predicted by the model at $T = 15$ mK can appear as a broad resonance within the single particle-hole spectrum at $T = 0.63$ K and above, and (b) that, when the zero-sound mode does exist, the single-particle-hole spectrum is dominated by $S_i(Q,\omega)$.

The simple model consists of calculating the dynamic susceptibility, χ, within the RPA and replacing the bare particle-hole interaction appearing in it by the empirical Landau parameters.[10]

The coherent and the incoherent susceptibilities are

$$\chi_c^{\mathrm{RPA}}(Q,\omega) = \frac{\chi^0(Q,\omega)}{1 + v_s(Q)\chi^0(Q,\omega)},$$
$$\chi_i^{\mathrm{RPA}}(Q,\omega) = \frac{\chi^0(Q,\omega)}{1 + v_a(Q)\chi^0(Q,\omega)}, \qquad (4)$$

with

$$S^{\mathrm{RPA}}(Q,\omega) = 2[n(\omega) + 1]\,\mathrm{Im}\chi^{\mathrm{RPA}}(Q,\omega). \qquad (5)$$

Here $\chi^0(Q,\omega)$ is the noninteracting Fermi-liquid susceptibility for both spin states, $v_s = v_{\uparrow\uparrow} + v_{\uparrow\downarrow}$ and $v_a = v_{\uparrow\uparrow} - v_{\uparrow\downarrow}$ are the spin-symmetric and spin-antisymmetric components of the particle-hole interaction, respectively, and $n(\omega)$ is the Bose function.

To parametrize $v(Q)$ we make the following assumptions:

(1) We interpret $v(Q)$ as a scattering amplitude

and replace it by the corresponding Landau quasiparticle-hole amplitude, A, valid for the limit $Q\rightarrow 0$. With this choice the zero angular momentum component ($l=0$) of the Landau amplitude guarantees that as $Q\rightarrow 0$ coherent (density fluctuation) χ reproduces the experimental zero-sound velocity.[5] Explicitly, the interaction[11] is

$$\frac{dn}{d\omega}v_s\rightarrow A_s=F_0^s+\frac{F_1^s}{1+F_1^s/3}\lambda^2,$$
$$\frac{dn}{d\omega}v_a\rightarrow A_a=F_0^a+\frac{F_1^a}{1+F_1^a/3}\lambda^2, \qquad (6)$$

where F_0^s (F_0^a) and F_1^s (F_1^a) are the spin-symmetric (-antisymmetric) components of the $l=0$ and $l=1$ components of the quasiparticle interaction. F_1^s is related to the effective mass, m^*, by $m^*=m(1+F_1^s/3)$. Here $dn/d\omega=3\hbar/2\epsilon_F$ is the density of frequency states per unit volume at the Fermi surface, ϵ_F is the Fermi energy, and $\lambda=\omega/Qv_F$ is ratio of the phase to Fermi velocity. Equation (6) has been justified by Babu and Brown[11] for $Q\rightarrow 0$ but its validity at finite Q remains to be tested.

With this choice of parameters, the RPA $S_c(Q,\omega)$ satisfies the f-sum rule exactly:

$$\int_{-\infty}^{\infty}\frac{d\omega}{2\pi}\,\omega S_c^{\text{RPA}}(Q,\omega)=\frac{\hbar Q^2}{2m}. \qquad (7)$$

(2) In addition to the scattering from the single particle-hole states, the experiments[1-3] suggest scattering contributions from multi-particle-hole (MPH) states. We therefore write $S_c(Q,\omega)$ as

$$S_c(Q,\omega)=S_c^{\text{RPA}}(Q,\omega)+S_c^{\text{MPH}}(Q,\omega)$$

where S_c^{MPH} is the remainder of S_c not included in the RPA. It is believed[7] that at low Q, S_c^{MPH} makes a contribution to the f-sum rule proportional to Q^4. Preliminary calculations[12] suggest that at $Q=1.5$ Å^{-1}, approximately one-half of the observed scattering at $T=0.63$ K is contained in S_c^{MPH}. To account for this we require S_c^{RPA} to satisfy the modified sum rule,

$$\int_{-\infty}^{\infty}\frac{d\omega}{2\pi}\,\omega S_c^{\text{RPA}}(Q,\omega)=\frac{\hbar Q^2}{2m}(1-KQ^2), \qquad (8)$$

and choose K so that $KQ^2=\frac{1}{2}$ at $Q=1.5$ Å^{-1}. Equation (8) requires the modified scattering amplitude

$$A_s(Q)=A_s\beta(Q),$$

where

$$\beta(Q)=\left[1-\frac{3}{F_1}\frac{KQ^2}{1-KQ^2}\right].$$

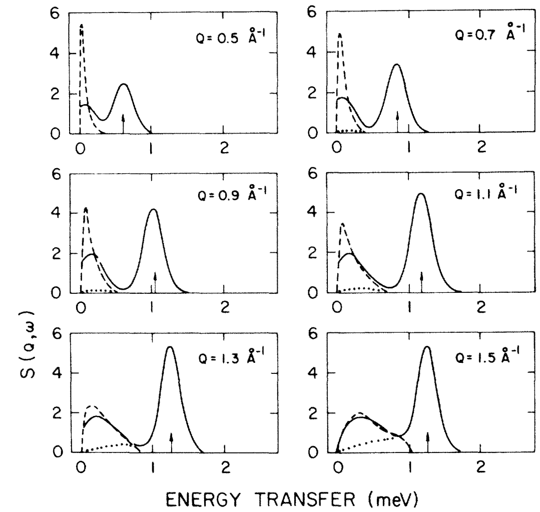

FIG. 1. The dynamic form factors at $T=15$ mK: solid curve, $S(Q,\omega)$ folded with a Gaussian to simulate an instrument energy resolution of 0.33 meV; dashed curve, $(\sigma_i/\sigma)S_i(Q,\omega)$ within the single particle-hole spectrum; dotted curve, $(\sigma_c/\sigma)S_c(Q,\omega)$, with the arrow denoting the zero-sound mode. The theoretical results for $S(Q,\omega)$ are in units of 10^{-12} s or 0.24 meV^{-1}.

The spin-antisymmetric scattering amplitude, A_a, could also be modified to account for multiparticle-hole contributions to $S_i(Q,\omega)$. However, since A_a is small, any reasonable choice of $\beta(Q)$ affects $S_i(Q,\omega)$ little so we have chosen to retain $A_a(Q)=A_a$. Then $S_i^{\text{RPA}}(Q,\omega)$ satisfies the sum rule

$$\int_{-\infty}^{+\infty}S_i^{\text{RPA}}(Q,\omega)\omega\frac{d\omega}{2\pi}=\frac{\hbar Q^2}{2m^*}(1+\tfrac{1}{3}F_1^a).$$

These two assumptions entirely determine $v(Q)$. To calculate χ^0 we have used the finite-temperature result derived by Khanna and Glyde.[13] The parameters[10] $F_0^s=10.8$, $F_0^a=-0.67$, $F_1^s=6.3$, and $F_1^a=-0.7$ are fixed by other experiments on liquid ^{3}He. We have used $\sigma_c=4.9$ b and $\sigma_i=1.2$ b.[14]

Figure 1 shows $S^{\text{RPA}}(Q,\omega)$ at $T=15$ mK. There we see a well-defined zero-sound mode which is outside the single particle-hole spectrum when $Q<1.6$ Å^{-1}, arising from the coherent part. The remaining single-particle spectrum is then dominated by the incoherent part. The latter leads to a second maximum in the scattering at low energy transfer. Figure 2 shows the temperature dependence of S^{RPA} for $Q=1.6$ Å^{-1}. At high temperature the zero-sound mode becomes a resonance within the single particle-hole spectrum. This resonance, when convoluted with a finite instrumental energy resolution, leads to a broad scat-

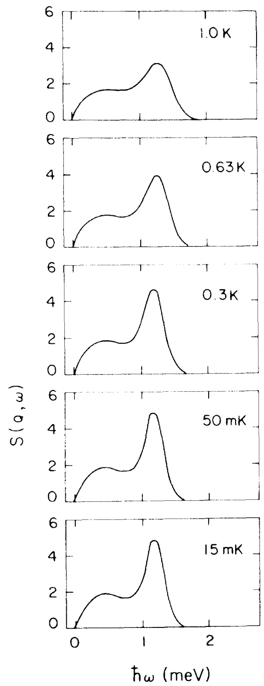

FIG. 2. The temperature dependence of the folded dynamic form factor, $S(Q,\omega)$, for $Q=1.6$ Å^{-1} in units of 10^{-12} s.

tering spectrum at high temperature. An example of the scattered intensity at $T=0.63$ K, converted to a constant scattering angle and including an instrumental resolution (full width at half-maximum $=0.33$ meV), is compared with the experimental results in Fig. 3. At $Q=1$ Å^{-1}, the lowest wave-vector transfer studied at 0.63 K, the zero-sound mode is close to but still outside the single-particle-hole spectrum. For higher values of Q the zero-sound mode lies inside the particle-hole spectrum. The calculated intensity distribution in Fig. 3 is strongly affected by the incoherent neutron scattering. Finally Fig. 4 shows the $T=15$ mK single-particle-hole and zero-sound mode spectra.

In conclusion, the present model gives a reasonable value for the zero-sound mode and the particle-hole spectrum, dominated by $S_i(Q,\omega)$, peaked at low ω as observed at $T=15$ mK. However, this simple model extension of Landau theory holds up to $Q\cong 1.8$ Å^{-1} only, beyond which $v(Q)$ is too small and m^* is too large. As temperature is increased, the separation between the zero-sound mode and the particle-hole band decreases substantially, making an isolated zero-sound mode more difficult to observe at $T=0.63$ K than at $T=15$ mK. There remains, however, more structure in $S_{\mathrm{RPA}}(Q,\omega)$ calculated here than

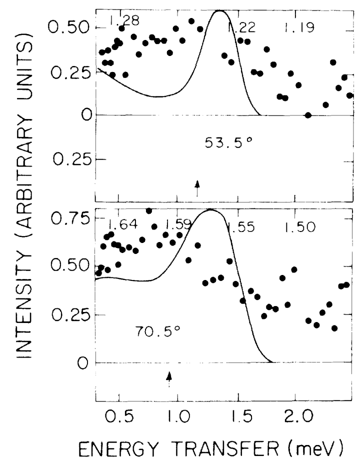

FIG. 3. The observed scattered neutron intensity (dots) at $T=0.63$ K for two scattering angles. Solid lines are the present RPA results. The normalization is arbitrary. The wave-vector transfer (Q) scale is given at the top of each figure.

observed at $T=0.63$ K. This structure would be reduced by (a) reduced effective mass m^* at high temperature[15] and (b) the multi-particle-hole excitations which also presumably increase in number and in importance with temperature.

It should be emphasized that, even in the long-wavelength limit, Landau theory is valid only at very low temperatures ($T\leqslant 0.2$°K). The particle-hole calculations at higher temperature ($T=0.63$°K) give only a qualitative description of

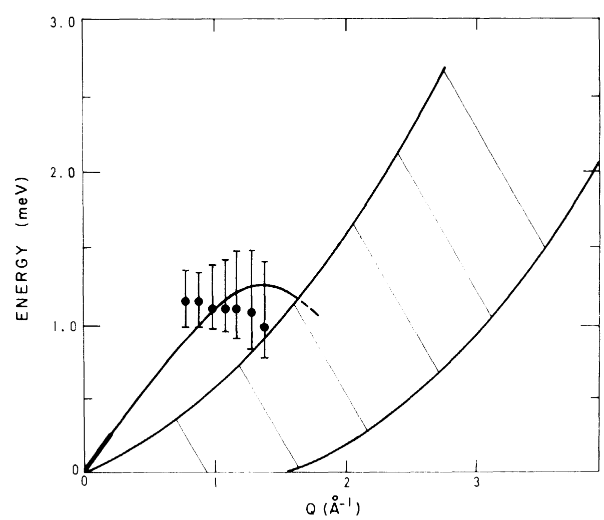

FIG. 4. The zero-sound dispersion curve (solid line) is compared with the experimental results at 15 mK. The heavy solid line shows the zero-sound velocity $S=194$ m/sec. The shaded area shows the particle-hole spectrum at $T=0$ K.

the excitation spectrum in liquid ^{3}He. Only a microscopic calculation of the particle-hole interaction at finite temperature can lead to a proper description of the dynamical structure factor at finite values of Q and ω over a wide temperature range.

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Neutron-Scattering Study of the Structure of Adsorbed Helium Monolayers and of the Excitation Spectrum of Few-Atomic-Layer Superfluid Films

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Elastic diffraction of neutrons from helium adsorbed on Grafoil shows that the surface monolayer forms as a highly compressed, ordered triangular lattice incommensurate with the structure of the underlying graphite basal planes. Inelastic scattering from thin superfluid films on top of the surface monolayers reveals the existence of bulklike rotons in films containing on average as few as three layers of atoms.

We have recently completed a neutron-scattering study of adsorbed helium and would like to report here some of the results relating to both the excitation spectrum of few-atomic-layer superfluid films and the structure of the surface monolayer. Our measurements were made with helium adsorbed on Grafoil,[1] an exfoliated graphite foil widely used for thermodynamic investigations. As has been noted,[2] Grafoil is an excellent substrate for neutron studies because it is relatively transparent to neutrons and at the same time offers large specific areas and surfaces which are remarkably uniform and free from impurities. In addition, these surfaces (primarily basal planes) exhibit a considerable degree of preferred orientation—a useful property for inelastic investigations because the adsorbed film planes can be partially oriented either parallel or perpendicular to the direction of momentum transfer, thereby giving different weightings to the in-plane and out-of-plane components of the scattering.

Studies of the heat capacity of the helium-Grafoil system[3,4] show that at monolayer completion the atoms form a solid with a density of 0.115 atoms/Å^2. The completed second layer is found to have a much lower density (0.081 atoms/Å^2) and does not solidify unless compressed by a third layer.[5] Superflow begins at about $2\frac{1}{2}$ layers,[6] suggesting that layers subsequent to the second are liquid.

All of our measurements were made with a triple-axis neutron spectrometer. For elastic scans