Temperature Dependence of $S(Q, \omega)$ in Superfluid ^{4}He

A. D. B. Woods and E. C. Svensson
Atomic Energy of Canada Limited, Chalk River, Ontario K0J 1J0, Canada
(Received 20 June 1978)

The dynamic structure factor, $S(Q,\omega)$, for superfluid ^{4}He at finite temperatures is described as the sum of two components: a "superfluid" component with a shape characteristic of $S(Q,\omega)$ at $T=0$ and a weight ρ_s/ρ, and a "normal-fluid" component with a shape characteristic of $S(Q,\omega)$ for nonsuperfluid ^{4}He and a weight ρ_n/ρ.

Recent neutron-scattering measurements[1] on liquid ^{4}He for wave vectors near the first maximum of the dispersion relation have demonstrated, for the first time at short wavelengths, a distinct difference in the shape of $S(Q, \omega)$ for temperatures just above and just below the superfluid transition temperature, $T_\lambda = 2.17$ K. In this Letter we present an analysis of these results, and of new measurements at the roton-minimum position, which shows that, for a wide range of temperatures at each Q value, there is a direct relationship between the "microscopic" $S(Q, \omega)$ and the "macroscopic" quantities ρ_s (the superfluid density) and $\rho_n = \rho - \rho_s$. In particular, for each Q, the integrated intensities of the sharp "one-phonon" peaks are proportional to ρ_s. Previous demonstrations of the role of ρ_s in determining the properties of liquid helium have been restricted to thermodynamic or hydrodynamic properties which are related to $S(Q, \omega)$ only in the region $Q \to 0$, $\omega \to 0$, e.g., measurements of the velocity of second sound[2] or of the Brillouin scattering of light.[3]

The experiments described in Ref. 1 were carried out with a triple-axis crystal spectrometer operated in the constant-$|\vec{Q}|$ mode[4] with a fixed scattered-neutron frequency, E'/h, of 1.134 THz. The (111) planes of silicon and the (002) planes of pyrolitic graphite were used as monochromator and analyzer, respectively. A beryllium filter prevented higher-order neutrons from reaching the detector. The width of the elastically scattered peak from vanadium was 0.04 THz. Four values of Q, 0.80, 1.13, 1.30, and 1.40 Å^{-1}, were studied at nine temperatures, 1.00, 1.38, 1.77, 1.97, 2.07, 2.12, 2.15, 2.27, and 4.21 K. Subsequent measurements have also been carried out at $Q = 1.926$ Å^{-1} at most of these same temperatures under conditions of higher resolution (0.03 THz at the elastic position). Corrections have been made to the observed signal counting rate for fast-neutron background, specimen-holder and cryostat scattering, multiple scattering in the helium,[5] and the effect of higher-order contamination on the counting rate in the incident-beam monitor. The resultant spectra are directly proportional to $S(Q, \omega)$ broadened by experimental resolution.

Figure 1 shows the corrected spectra at $Q = 1.13$ Å^{-1}. The most striking feature of these spectra is the presence, at all temperatures below T_λ, of a reasonably sharp peak which gives rise to an abrupt change in slope at $\nu \approx 0.35$ THz ($\nu = \omega/2\pi$), and the absence of any evidence for such a peak above T_λ; no such signature on the superfluid has been previously observed in any experiment which probes the liquid on a scale of interatomic distances.

The presence of a reasonably well-defined peak whose intensity decreases with increasing temperature and appears to be zero for $T > T_\lambda$ suggests a two-component character for $S(Q, \omega)$ for $T < T_\lambda$ and we have found that our results are well described by

$$S(Q, \omega) = \frac{\rho_s(T)}{\rho} S_s(Q, \omega) + \frac{\rho_n(T)}{\rho} S_n(Q, \omega) \frac{1 - \exp(-\hbar\omega/kT_1)}{1 - \exp(-\hbar\omega/kT)}, \quad (1)$$

where $S_s(Q, \omega)$ is characteristic of the superfluid and $S_n(Q, \omega)$ is $S(Q, \omega)$ measured at $T = T_1 > T_\lambda$. The thermal population factor[6] is close to unity for the results reported in this Letter.

Since Eq. (1) assumes no Rayleigh scattering and, hence, is inappropriate for $T_1 = 4.21$ K, we use $T_1 = 2.27$ K in our analysis. At $T = 0$, $S_s(Q, \omega)$ is usually assumed[7] to consist of a δ-function "one-phonon" component of strength $Z(Q)$ plus a broad "multiphonon" component, $S_{\rm II}(Q, \omega)$, at higher frequencies. According to the theory of Landau and Khalatnikov[8] the "one-phonon" component will broaden as the temperature is increased but we would expect the "multiphonon" component to be relatively insensitive to temperature changes. The spectral form of the "one-phonon" peak at finite temperatures is not known and our resolution-broadened curves are not able to distinguish between the Lorentzian line shape suggested by Cohen[9] and some other shape such

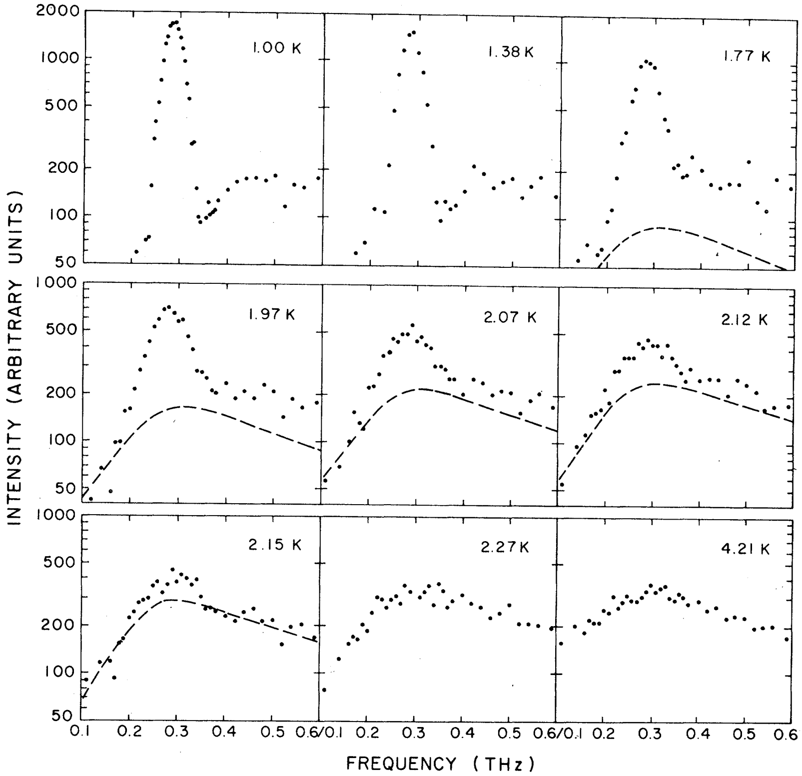

FIG. 1. The corrected scattering by liquid ^{4}He for $Q = 1.13$ Å^{-1} and $0.1 \le \nu \le 0.6$ THz. These results are directly proportional to the resolution-broadened dynamic structure factor, with the same constant of proportionality for each temperature. Note the logarithmic intensity scale. The dashed curves (which are off the low end of the scale for 1.00 and 1.38 K) represent the "normal-fluid" component, i.e., the second term in Eq. (1), based on the measurements at 2.27 K (see text).

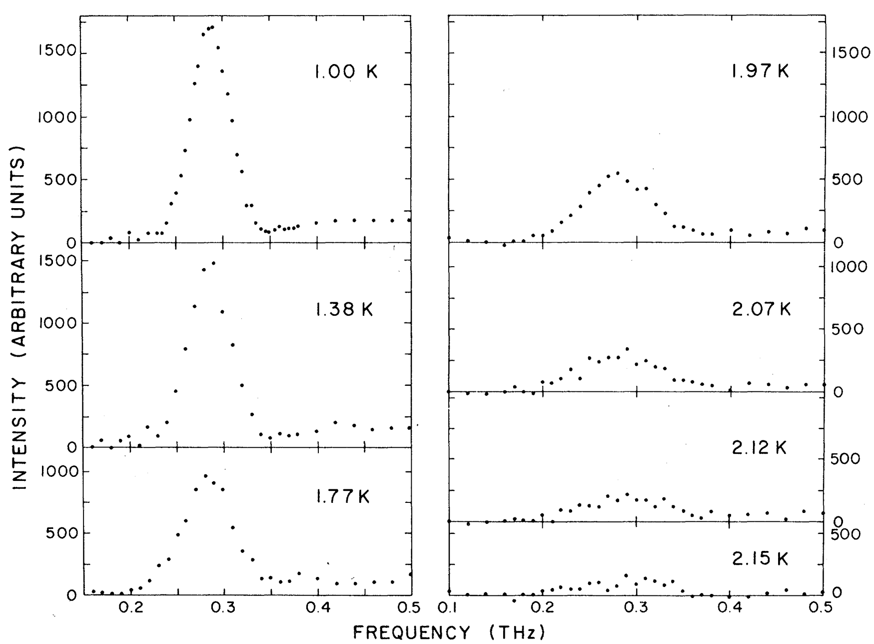

FIG. 2. The "superfluid" component for $Q = 1.13$ Å^{-1} obtained by subtracting the "normal-fluid" component from the distributions shown in Fig. 1.

as a Gaussian. If we assume a Lorentzian line shape and also that $\hbar\omega(Q) \gg kT$ we can write

$$S_s(Q, \omega) = \frac{Z(Q)}{\pi} \frac{\Gamma(T)}{\Gamma^2(T) + [\omega - \omega(Q)]^2} + S_{\text{II}}(Q, \omega) \qquad (2)$$

so that the "one-phonon" peak has a half-width $\Gamma(T)$ and a strength $Z(Q, T) = \rho_s Z(Q)/\rho$.

The second term in Eq. (1), i.e., the "normal-fluid component" based on the measurements at $T_1 = 2.27$ K, is shown as the dashed curves in Fig. 1. Subtraction of these "normal-fluid components" gives the results shown in Fig. 2. Each spectrum in Fig. 2 consists of a well-defined peak, which falls to zero intensity on its low-frequency side, and a weak component extending to high frequencies. Apart from a temperature-dependent broadening of the peaks the shapes of these spectra are similar at all temperatures, whereas the shapes of the original spectra in Fig. 1 are very sensitive to temperature. Similar behavior is observed when this subtraction procedure is applied to the spectra obtained at the other values of Q.

The variations with temperature of the reduced integrated intensities, $Z(Q, T)/Z(Q)$, of the peaks in Fig. 2 (assumed to be symmetrical) and of the similar peaks for the other values of Q are compared in Fig. 3(a) with ρ_s/ρ. The correlation between $Z(Q, T)/Z(Q)$ and ρ_s/ρ is very striking and strongly supports the form of Eq. (1). The peak widths, 2Γ, corrected for the Gaussian instrumental resolution on the assumption that the intrinsic line shapes are Lorentzian [see Eq. (2)] are shown in Fig. 3(b). Within errors these widths are independent of Q, a result which in turn implies that the lifetime-limiting mechanism is independent of Q at least in the range $0.80 \leq Q \leq 1.93$ Å^{-1}. As can be seen from Fig. 3 these widths agree remarkably well with those for rotons calculated from the theory of Landau and Khalatnikov.[8]

The increase of Γ with temperature shown in Fig. 3 is, especially near T_λ, less than previously reported[10] increases, all of which were determined from measurements carried out with fixed incident-neutron energy. Under such conditions the high-frequency part of the spectrum is suppressed[4] in a way which tends to obscure the separation of the sharp peak from the broad component and thus the previously reported widths near T_λ probably include contributions from the "normal-component" scattering. A reassessment of the neutron-scattering results on the basis of Eq. (1) removes at least some of the discrepancies noted by Brooks and Donnelly[11] in their calculations of various thermodynamic properties. For example, the value of Δ_t at $T = 2.12$ K from Table 22 of Ref. 11 is 0.159 THz, about 25% higher than previously reported neutron-scattering values[10] but in good agreement with the value of 0.163 THz determined directly from the center of the "one-phonon" peak (as in Fig. 2) at $Q = 1.926$ Å^{-1}.

The possibility that $S(Q, \omega)$ for superfluid ^{4}He might contain more than one component has been discussed by a number of authors. Pines,[12] in particular, drew attention to this possibility and put forward intuitive arguments connecting ρ_s and quantities related to $S(Q, \omega)$. He expressed surprise that phonons in helium, which he identified with zero sound, were apparently unaffected

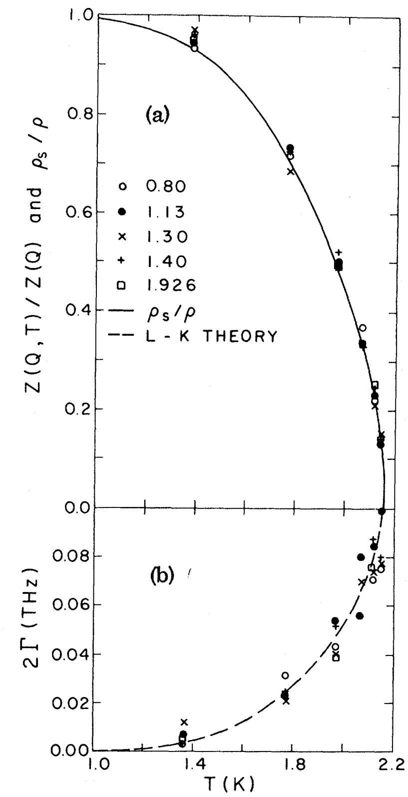

FIG. 3. (a) The reduced (i.e., normalized to ρ_s/ρ at 100 K) integrated intensities $Z(Q,T)/Z(Q)$ of the peaks in the "superfluid" component for the five values of Q and ρ_s/ρ (solid curve). (b) The intrinsic widths of the peaks in the "superfluid" component for the five values of Q and the widths (dashed curve) for rotons calculated from the theory of Landau and Khalatnikov (Ref. 8).

by the transition from the superfluid to the nonsuperfluid and pointed out that little difference in (the density-density part of) $S(Q, \omega)$ is expected for nonsuperfluid ^{3}He and nonsuperfluid ^{4}He. Comparison of the present results for $T > T_\lambda$ and those reported previously[13] for $T = 2.3$ K with those[14] for nonsuprefluid ^{3}He supports this point of view; except at very low frequencies where spin-spin correlations are important for ^{3}He all of these distributions are similar in shape.

In summary, the description presented in this paper of the temperature dependence of $S(Q, \omega)$ for liquid helium suggests a two-component structure with the two components directly related to the macroscopic quantities ρ_s and ρ_n. At the present time there is, however, no known theoretical relationship between these macroscopic quantities and the microscopic $S(Q, \omega)$.

The authors have benefitted from discussions with A. Jacobs, F. C. Khanna, and, in particular, D. J. Thouless whose remark was instrumental in stimulating the analysis presented here. We wish to thank H. F. Nieman and D. C. Tennant for valuable technical assistance.

[1]E. C. Svensson, R. Scherm, and A. D. B. Woods, in Proceedings of the Fifteenth International Conference on Low Temperatures Physics, Grenoble, France, 23–29 August 1978 (to be published).

[2]V. P. Peshkov, J. Phys. (U.S.S.R.) 10, 389 (1946).

[3]W. F. Vinen, C. J. Palin, and J. M. Vaughan, in *Low Temperature Physics, LT-13*, edited by K. D. Timmerhaus, W. J. O'Sullivan, and E. F. Hammel (Plenum, New York, 1974), Vol. 1, p. 524; G. Winterling, F. S. Holmes, and T. J. Greytak, *ibid.*, p. 537.

[4]B. N. Brockhouse, in *Symposium on Inelastic Scattering of Neutrons of Solids and Liquids* (International Atomic Energy Agency, Vienna, 1961), p. 113.

[5]V. F. Sears, Nucl. Instrum. Methods 123, 521 (1975).

[6]This factor ensures that the second term in Eq. (1), the only term giving a significant contribution for $\hbar\omega \lesssim kT$ satisfies detailed balance; it also preserves the first moment of $S(Q,\omega)$.

[7]A. Miller, D. Pines, and P. Nozières, Phys. Rev. 127, 1452 (1962).

[8]L. D. Landau and I. M. Khalatnikov, Zh. Eksp. Teor. Fiz. 19, 637 (1949). See also J. Wilks, *The Properties of Liquid and Solid Helium* (Clarendon, Oxford, 1967), Chaps. 6, 7.

[9]M. Cohen, Phys. Rev. 118, 27 (1960).

[10]K.-E. Larsson and K. Otnes, Ark. Fys. 15, 49 (1959); D. G. Henshaw and A. D. B. Woods, Phys. Rev. 121, 1266 (1961); R. A. Cowley and A. D. B. Woods, Can. J. Phys. 49, 177 (1971); O. W. Dietrich, E. H. Graf, C. H. Huang, and L. Passell, Phys. Rev. A 5, 1377 (1972).

[11]J. S. Brooks and R. J. Donnelly, J. Phys. Chem. Ref. Data 6, 51 (1977).

[12]D. Pines, in *Quantum Fluids*, edited by D. F. Brewer (North-Holland, Amsterdam, 1966), pp. 257, 338.

[13]A. D. B. Woods, E. C. Svensson, and P. Martel, Phys. Lett. 57A, 439 (1976).

[14]W. G. Stirling, R. Scherm, P. A. Hilton, and R. A. Cowley, J. Phys. C 9, 1643 (1976); K. Sköld, C. A. Pelizzari, R. Kleb, and G. E. Ostrowski, Phys. Rev. Lett. 37, 842 (1976).

Identification of the Isolated Ga Vacancy in Electron-Irradiated GaP through EPR

T. A. Kennedy and N. D. Wilsey
Naval Research Laboratory, Washington, D. C. 20375
(Received 27 July 1978)

An $S = \frac{1}{2}$ EPR spectrum is observed in electron-irradiated GaP:Zn with an isotropic $g = 2.0130 \pm 0.0015$ and resolved ligand hyperfine splitting from four equivalent $I = \frac{1}{2}$ neighbors. This spectrum is assigned to the isolated Ga vacancy. A molecular-orbital model for V_{Ga}^{--} is consistent with the experimental data. Analysis of the hyperfine parameters reveals some lattice relaxation although the full T_d symmetry is maintained.

Although electron paramagnetic resonance (EPR) has been successfully used to investigate vacancies in Si[1,2] and in the more ionic II-VI compounds[3-5], the current knowledge of the structure of native defects in III-V semiconductors is limited because of the lack of EPR results. The lack of EPR data is mainly due to large linewidths arising from hyperfine (hf) interactions with the group III and V nuclei, most of which have high nuclear spins. The only reported EPR identification of a native defect in a III-V semiconductor is that of P on a Ga site in GaP—an antisite defect.[6]

In this Letter, we report the observation of an