Fluid-structure-factor measurements in ^{4}He for $5.5 \leq T \leq 7.0$ K

D. A. Ewen, H. N. Robkoff, and R. B. Hallock
Laboratory for Low Temperature Physics, Department of Physics and Astronomy, University of Massachusetts, Amherst, Massachusetts 01003
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We report precise measurements of the structure factor $S(k)$ as a function of the momentum transfer k for ^{4}He at $T=5.5$ and 6.5 K in the density range $10 \lesssim \rho \lesssim 155$ kg/m^3. No theoretical predictions that can be tested against the data are available. The recent theoretical work by Bruch *et al.* at $T=7.0$ K, $\rho=69.6$ kg/m^3 is in only limited agreement with our experimental data at $T=7.0$ K, $\rho=69.0$ kg/m^3.

The techniques of x-ray scattering allow a unique opportunity to investigate matter at the atomic level.[1] Measurements of the scattering intensity as a function of the angle of scatter allow a direct determination of the fluid structure factor[2] $S(k)$. In general for a fluid

$$S(k)=1+\rho\int e^{i\vec{k}\cdot\vec{r}}[g(r)-1]\,d^3r \quad , \tag{1}$$

where k is the momentum transfer, ρ the density, and $g(r)$ is the pair correlation function. Here $k=(4\pi/\lambda)\sin(\frac{1}{2}\theta)$ where λ is the wavelength of the x ray and θ is the scattering angle. For a monatomic fluid such as helium $g(r)$ is isotropic.

In the past, precise systematic studies of $S(k)$ for helium as a function of density and temperature have been limited. This has been due in part to the fact that helium both scatters x rays weakly and is inherently of low density. The lack of systematic measurements has helped curtail the development of an adequate theoretical description of helium as a function of density[3,4] and temperature.[5] The present measurements were carried out primarily at 5.5 and 6.5 K to allow a systematic study of $S(k)$ over as wide a range of densities as possible so as to provide a stringent test of theoretical work as it becomes available.

The data we present here were obtained through the use of a cryogenic transmission x-ray diffractometer which incorporates many improvements[6] over that used previously for studies of ^{4}He at saturated vapor pressure.[7] The x-ray beam is derived from a Rigaku RU-200 PL rotating anode source, and the scattered x rays are detected by means of a PGT intrinsic germanium detector. A pulse-height spectrum is taken at each scattering angle and the 280-eV resolution of the detector allows computer analysis to separate the scattered Cu $K\alpha$ x rays from the $K\beta$ and others. Geometrical factors inherent in the diffractometer are removed through the use of normalization scattering[8] from neon gas at 77.3 K and a pressure of 1.0×10^5 Pa. Temperature measurement is by use of a calibrated germanium resistance thermometer in intimate contact with the scattering chamber. Temperature measurement is accurate to ±0.005 K, and pressure measurements are made to $\pm1.4\times10^3$ Pa. The statistical accuracy of the scattering data is reflected in the scatter of the data we shall present shortly. The accuracy decreases with an increase in momentum transfer due primarily to the weak scattering from the dilute neon. To correct for small drifts in the x-ray-source intensity, the diffractometer incorporates two Xe-Co_2 proportional counters. One monitors the main beam directly and the other monitors the x rays scattered from the helium at the fixed momentum transfer $k=2.0$ Å^{-1}. The scattering chamber which

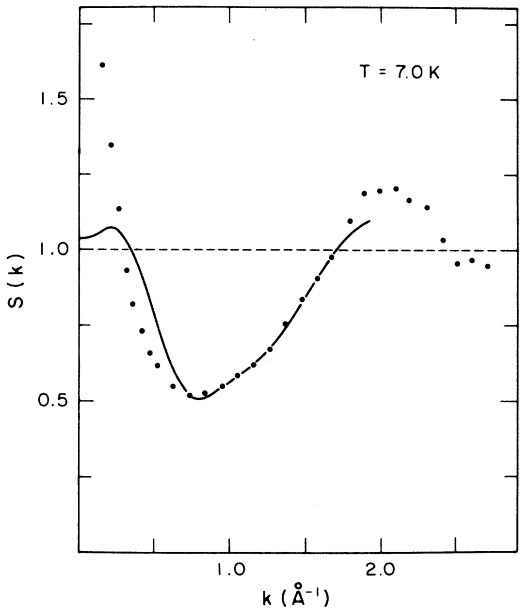

FIG. 1. Structure factor measurements for ^{4}He at $T=7.0$ K and $\rho=69.0$ kg/m^3. The scatter in the data reflects the errors due to counting statistics alone. The solid line is the result of the theoretical calculation of Bruch *et al.* (Ref. 5) at $T=7.0$ K and $\rho=69.6$ kg/m^3.

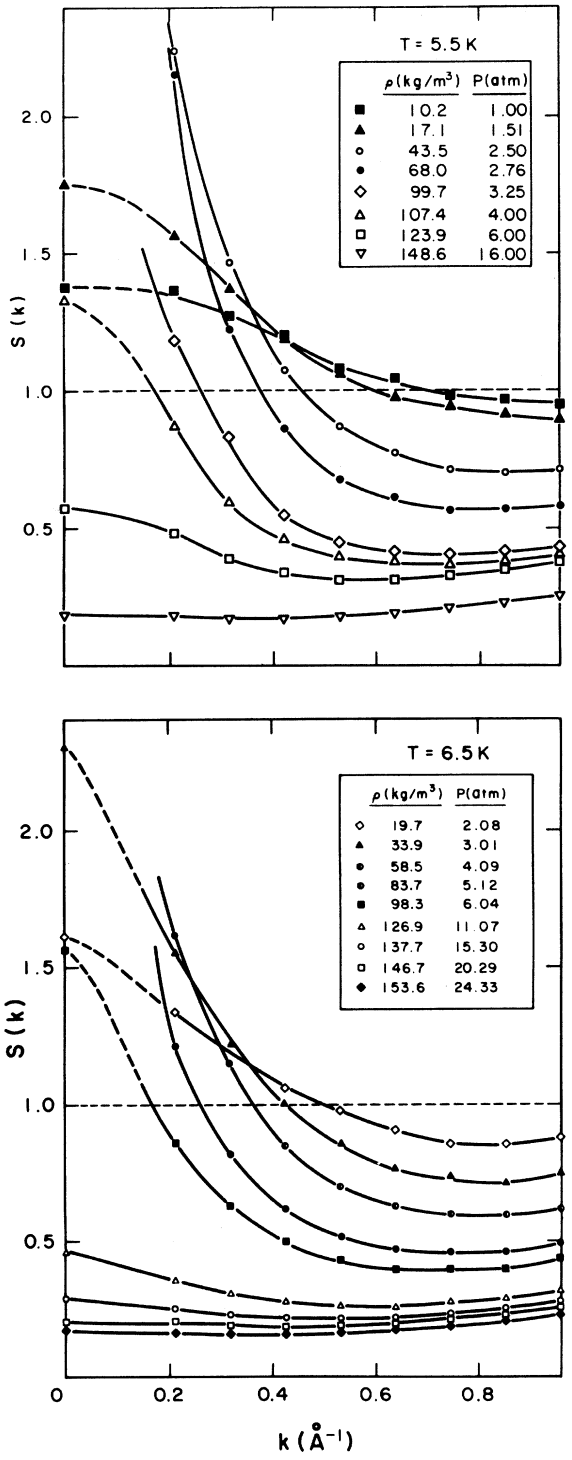

FIG. 2. Experimental structure factor values at small momentum transfer as a function of density at $T = 5.5$ and 6.5 K. The smooth curves which connect the data points are drawn as a guide to the eye and do not represent a theory. The intercepts, $S(0)$, were computed from the thermodynamic tables of McCarty (Ref. 12) as described in the text.

contains the helium target was machined from beryllium and is a hollow cylinder of diameter 0.95 cm and wall thickness 0.25 mm. The present configuration of the apparatus allows measurements for $1.4 < T < 10$ K, $P \lesssim 30$ atm over the momentum transfer interval $k \lesssim 5$ Å^{-1}.

The liquid structure factor is obtained directly from the observed scattering intensities by use of[7]

$$S(k) = \frac{\rho_{Ne} T_{Ne} (I_{He} - I_{HeC})}{\rho_{He} T_{He} (I_{Ne} - I_{NeC})} \frac{(\xi_e + \xi_i)}{\sigma_e} - \frac{\sigma_i}{\sigma_e} , \quad (2)$$

where the subscripts Ne and He refer to neon and

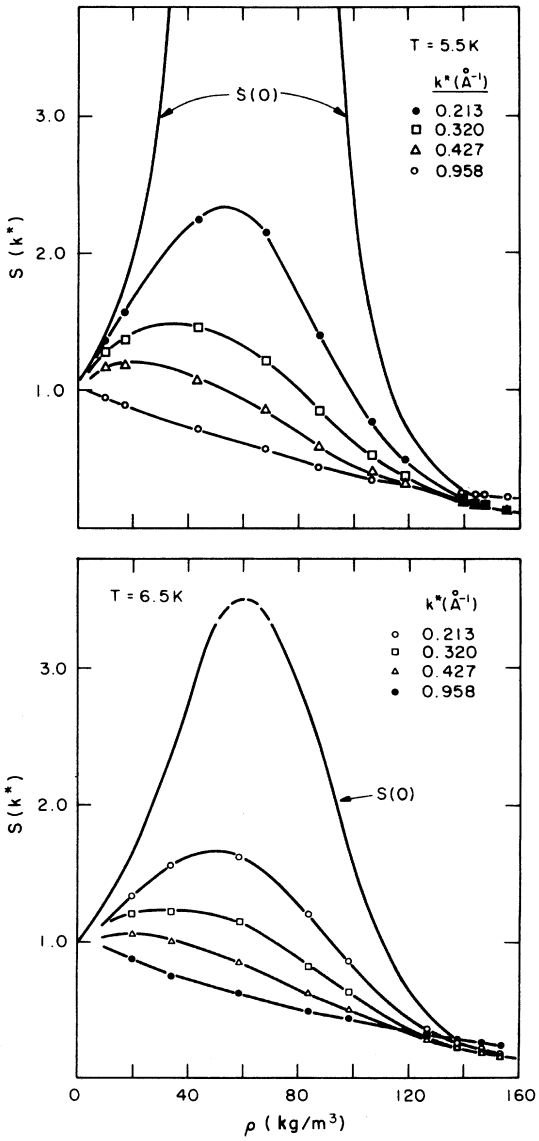

FIG. 3. The experimental structure factor at several fixed values of the momentum transfer k^* as a function of the density at $T = 5.5$ and 6.5 K. $S(0)$ computed from thermodynamics is shown as the solid-dashed curve. The smooth curves through the data are guides to the eye.

helium. Here ρ is the number density, T the transmission factor, I the scattered intensity, ξ_e and ξ_i the coherent and incoherent scattering factors[9] for neon, σ_e and σ_i the respective factors for helium, and the subscript C refers to the empty scattering chamber. Results typical of those obtained in this work are shown in Fig. 1. The data points shown have been computed directly from the scattering intensities through use[7] of Eq. (2), and no effort has been made to smooth the data. Such smoothing of, for example, the neon normalization data or the empty-cell background contribution is not necessary for our present purposes. For the data in the range $k < 1.0$ Å^{-1} the statistical error is substantially less than 1%. Overall systematic errors contribute at most an additional 2%. The assumption that neon is ideal and hence $S_{\rm Ne} \equiv 1$ introduces less than a 1% error at $k = 0$ Å^{-1}. This effect decreases with increasing momentum transfer and becomes invisible at the momentum transfer values used for this work.

The solid line in Fig. 1 has been taken from the recent theoretical work of Bruch *et al.*[5] in which an effective potential approximation was used to allow ^{4}He to be treated as a fictitious classical fluid. The results of this finite temperature calculation are seen to be in encouraging but limited agreement with the experimental data. Two major shortcomings of the theory are that it yields less structure than the experiment in the vicinity of the first maximum in $S(k)$ and that it displays an improper intercept at $S(k=0)$. The first of these shortcomings may be partially due to the choice of the Lennard-Jones potential.[10] The second is due to the difficulties with $g(r)$ at large[11] values of r.

The detailed evolution of the structure factor in the small momentum transfer domain as a function of density is illustrated in Fig. 2 for two temperatures. The intercepts at zero momentum transfer have been obtained through the use of $S(0) = \gamma k_B T / mc^2$ where k_B is the Boltzmann constant, T the temperature, γ the ratio of specific heats, m the ^{4}He atomic mass, and c the speed of first sound. The specific heats and sound velocity were determined from the extensive tables of McCarty[12] by graphical interpolation. Although the data do not extend to particularly small values of the momentum transfer, they are clearly consistent with the expected intercepts. No theoretical predictions are yet available which illustrate this detailed evolution as a function of momentum transfer.

A more dramatic representation of the detail a successful theory must reproduce is shown in Fig. 3. To produce this figure we have plotted the structure factor $S(k^*)$ at selected, fixed values of the momentum transfer, k^*, as a function of density. The zero momentum transfer limit as computed from thermodynamics is shown as the solid line. Difficulties with interpolation from McCarty's work result in larger errors in the vicinity of the $S(0)$ peak and hence it is drawn as a dashed line in the 6.5-K case. This figure is not meant to suggest that x-ray techniques should be used to obtain thermodynamic information; rather, a successful finite temperature theory of the structure of helium carried out as a function of density must illustrate this substantial dependence on momentum transfer.

We have obtained the first systematic data on the finite temperature structure factor for ^{4}He over a *wide* range of density. These data will provide a stringent test for finite temperature theory as a function of density and it is hoped they will act as a stimulus for further theoretical development.

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[9]C. Tavard, D. Nicholas, and M. Rovault, J. Chem. Phys. 64, 540 (1967); Y. Kim and M. Inokuti, Phys. Rev. 165, 39 (1967).

[10]A characteristic of this potential is that it produces a peak in $S(k)$ near $k = 2.0$ Å^{-1} which is both too low and too broad. (See Ref. 3.)

[11]L. W. Bruch (private communication).

[12]R. D. McCarty, J. Phys. Chem. Ref. Data 2, 923 (1973).