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LIQUID STRUCTURE FACTOR OF He^4 BY X-RAY SCATTERING AT SMALL MOMENTUM TRANSFER*

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The liquid structure factor of helium-4 has been determined in the momentum-transfer range 0.2 to 0.8 $Å^{-1}$. At 0.32 K the structure factor exhibits a gentle but decided change of slope near 0.4 $Å^{-1}$ in qualitative agreement with certain theoretical predictions. Below 0.4 $Å^{-1}$ the liquid structure factor agrees with that deduced from the Bijl-Feynman dispersion relation. At 4.99 K and 680 mm pressure we have shown that helium gas is distinctly nonideal.

We have measured the intensity of copper $K\alpha$ x rays scattered from helium-4 as a function of scattering angle both in the gas phase near 5 K and in the liquid phase at several lower temperatures. By normalization against neon at 77 K, structure was observed in the helium gas and a determination of the structure factor of the liquid was made. At. 0.32 K the liquid structure factor shows a gentle but decided change of slope near the momentum transfer 0.4 $Å^{-1}$. This gentle shoulder is in qualitative agreement with the shoulder suggested by Miller, Pines, and Nozières,[1] but is much weaker than the shoulder given by Massey.[2] The liquid structure factor in the range 0.2 to 0.8 $Å^{-1}$ is in good agreement with a recent calculation by Campbell and Feenberg.[3] Our measurements disagree somewhat with those of Gordon, Shaw, and Daunt[4] and also with those of Achter and Meyer.[5]

The Bijl-Feynman dispersion relation[6]

$$E(k)=\hbar^2k^2/2MS(k), \tag{1}$$

where $S(k)$ is the liquid structure factor, k the momentum transfer, and M the mass of helium atom, relates the energy of an elementary excitation in a condensed Bose system to the liquid structure factor at absolute zero. If the excitations are phonons, we expect

$$S(k)=\hbar k/2Mc_1, \tag{2}$$

where c_1 is the velocity of first sound in liquid helium.

It has been shown[7,8] that for finite temperatures the structure factor in the limit of vanishing momentum transfer can be written as

$$\lim_{k\to 0} S(k)\equiv S(0)=nk_BTX_T, \tag{3}$$

where n is the number density, k_B the Boltzmann constant, and X_T the isothermal compressibility. Experiments to date in the momentum transfer range below 0.8 $Å^{-1}$ indicate in some cases[4,9] that $S(k)$ approaches the expected value of nk_BTX_T as $k\to 0$. In other experiments[10] abpve 0.8 $Å^{-1}$ the extrapolated values of $S(0)$ are in close agreement with (3) although the extrapolations themselves are open to errors. In all of these investigations the structure factor was seen to be insensitive to changes in temperature for values of the momentum transfer greater than about 0.8 $Å^{-1}$. This led Jackson[11] and independently Miller, Pines, and Nozières[1] to notice that the experiments did not extrapolate correctly to (2). Miller, Pines, and Nozières suggested that if the data down to 0.8 $Å^{-1}$ were valid at temperatures near absolute zero, then a shoulder should appear on the structure factor curve in the momentum transfer range where the expected linear dependence "joined up" with the measured structure factor. We have investigated the range 0.2 $Å^{-1} < k < 0.8$ $Å^{-1}$ in some detail at temperatures as low as 0.32 K to study this problem.

The intensity of x rays scattered from a sample of an ideal gas is given by the expression

$$I = ATN(\xi_e + \xi_I), \tag{4}$$

where A is the Thompson cross section, T the transmission factor, N the number of atoms, and ξ_e and ξ_I the coherent and incoherent scattering factors. If the sample consists of a liquid or a nonideal gas, the expression for the scattered intensity becomes

$$I = ATN(\sigma_e S + \sigma_I), \tag{5}$$

where S is the structure factor. These two expressions can be divided to obtain a relation for S in terms of the observed scattering intensities. Since the scattering volume for a particular angle of scatter is a fixed quantity, we can replace the number ratio by the ratio of number densities to obtain

$$S(k) = \frac{I_L T_G \rho_G}{I_G T_L \rho_L} \frac{\xi_e + \xi_I}{\sigma_e} - \frac{\sigma_I}{\sigma_e}. \tag{6}$$

This expression can be used to determine the structure factors for both liquid and gaseous helium. We have used neon at 77 K as the ideal gas which provides our normalization.[12]

The density of helium gas was computed from virial coefficients as given by Kilpatrick, Keller, and Hammel[13] and also from the Leiden[14] virial coefficients. The Berlin virial equation was used to obtain the density of neon.[15] Using (3) it is possible to obtain $S(0)$ for these gases from the virial equations. Table I shows the result of such a calculation. We note that neon is more nearly ideal than helium. As we can see from (6), geometrical correction factors are eliminated for the most part by this type of analysis.

The spectrometer, target cell, and cryostat were built in our laboratory. The cryostat was designed by Dr. R. S. Safrata and has been described elsewhere.[16] The spectrometer and target cell will be discussed in detail in a forthcoming paper. A General Electric CA-7H x-ray tube is used as the source of primary x rays. The main beam is derived from the line source and is collimated by a series of knife edges and Soller slits. The target chamber is a demountable Mylar cylinder whose axis is the rotation axis of the scattered-beam detector. The target is attached to the base of a closed-cycle helium-3 refrigerator which is capable of maintaining temperatures as low as 0.30 K for several weeks at a time. The temperature was measured by two calibrated carbon resistors.[17] The detector is a sealed xenon-filled proportional counter with methane as a quench gas. The detector was moved to provide automatic angular increments of 0.5 deg. The angular resolution for this work is about 0.6 deg.

Table I. Values of $S(0)$ as computed from the virial equations.

Sample	Temperature (K)	Pressure (mm Hg)	$S(0)$
Neon	77.35	382	1.002
Neon	77.35	620	1.004
Helium	4.99	680	1.46
Helium	1.24	Liquid	0.045
Helium	0.32	Liquid	0.01

The data were acquired using a single-channel analyzer to select the copper K line and discriminate against noise. A nickel foil was used to reduce the $K\beta$ contribution to about 1% of the $K\alpha$ contribution. A 1% angle-independent multiple-scattering correlation has been applied to the data. The first and last liquid runs were both at 0.32 K and the data were reproducible to about 1 %. The gain of the detector was checked periodically by observing the single-channel analyzer window on a multichannel analyzer. This gain remained essentially constant over the entire run as did the room background as observed at the detector.

Some of our results are shown in Figs. 1 and 2. In Fig. 1 we present the structure factor of helium gas at 4.99 K and 680 mm pressure. We

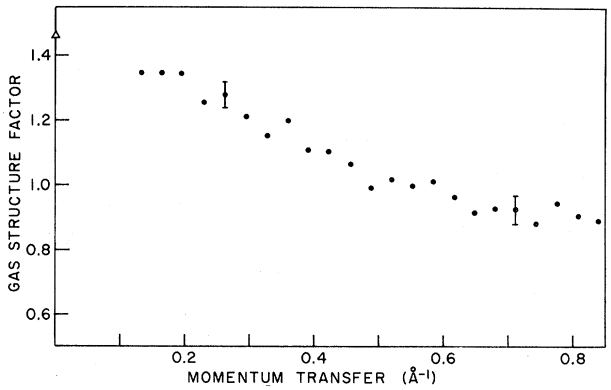

FIG. 1. The structure factor as computed from Eq. (6) for helium gas at 4.99 K and a pressure of 680 mm. The closed circles represent unsmoothed experimental points while the open triangle represents $S(0)$ as computed from Eq. (3). These data demonstrate conclusively that helium gas is distinctly nonideal for these values of temperature and pressure. The error bars are indicative of statistical error alone.

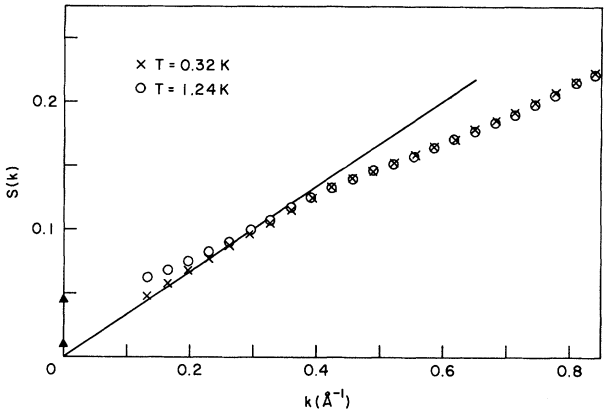

FIG. 2. The smoothed liquid structure factor at 0.32 and 1.2 K. The straight line is obtained from Eq. (2) using $c_1 = 238$ m/sec. The closed triangle represents $S(0)$ as computed from (3) in each case. We note that the data extrapolate well to the expected value (closed triangle) in each case.

also show the extrapolated zero-momentum-transfer structure factor obtained from (3). Note that these data were taken with a beryllium target cell while the results presented in Fig. 2 were obtained from data taken with the Mylar cell. The data of Fig. 2 have been smoothed in the following manner: The empty-target scattering was subtracted from the scattering produced by the target with the gas or liquid sample present. These reduced data points were then plotted and smooth curves drawn through them. Points read from these curves were then used to obtain the $S(k)$ values shown in Fig. 2. Curves without this smoothing also show the change of slope apparent in this figure although the statistical error is of course greater.

We see that for values of $k \lesssim 0.4$ Å^{-1} the liquid structure factor is essentially linear in the momentum transfer with a slope near that deduced from the Bijl-Feynman dispersion relation. Above 0.4 Å^{-1} our data fall below this line. We believe this to be a real effect. Hence, we have shown that the Bijl-Feynman dispersion relation is valid for momentum transfers up to about 0.4 Å^{-1}. We are at present actively extending our measurements to both lower temperatures and wider angles. We also expect to study expression (3) at temperatures above the lambda point.[18]

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