Temperature-Dependent Spatial Order in Superfluid and Normal ^{4}He

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Measurements of the structure factor $S(k)$ for ^{4}He at constant density above and below T_λ are reported. The results show a clear loss of spatial order on cooling through T_λ. Application of the theory of Cummings, Hyland, and Rowlands to the data allows a measurement of the condensate fraction for He II.

We report new results[1] for the static structure factor $S(k)$ for ^{4}He taken at several temperatures at constant density in the vicinity of the λ transition. The measurements indicate an increase in spatial order in the liquid on cooling to T_λ. Two features of the data show evidence for a *decrease* in spatial order on further cooling: (1) The amplitude of the principal structure-factor peak decreases with an associated increase in its width, and (2) the amplitude of the oscillations in the pair correlation function decreases. A direct application of the theoretical expression due to Cummings, Hyland, and Rowlands[2] results in a value for the condensate fraction, $n_0 = (9 \pm 3)\%$ at $T = 1.67$ K at saturated vapor pressure (SVP) in disagreement with earlier work with use of neutron scattering by Mook and co-workers[3] but in general agreement with recent neutron studies by Svensson *et al.*[4] At $T = 1.7$ K and density = 150.3 kg/m^3 we find $n_0 = (8 \pm 3)\%$.

The structure factor has been determined by use of an x-ray diffractometer[5] over the momentum-transfer range $0.2 \lesssim k \lesssim 5.2$ Å^{-1}. Geometric factors in the scattering geometry have been removed through the process of scattering[6] from nearly ideal neon gas at 77 K and a pressure of 760 Torr. The scattering intensity has been corrected both for long- and short-term variations in the main x-ray beam intensity, absorption in the scattering chamber by the helium or neon and multiple-scattering effects. A typical determination of $S(k)$ is shown in Fig. 1(a), where no smoothing to reduce statistical error has been applied to the data. The principal source of the scatter in the data at larger momentum-transfer values is the relatively weak scattering from the

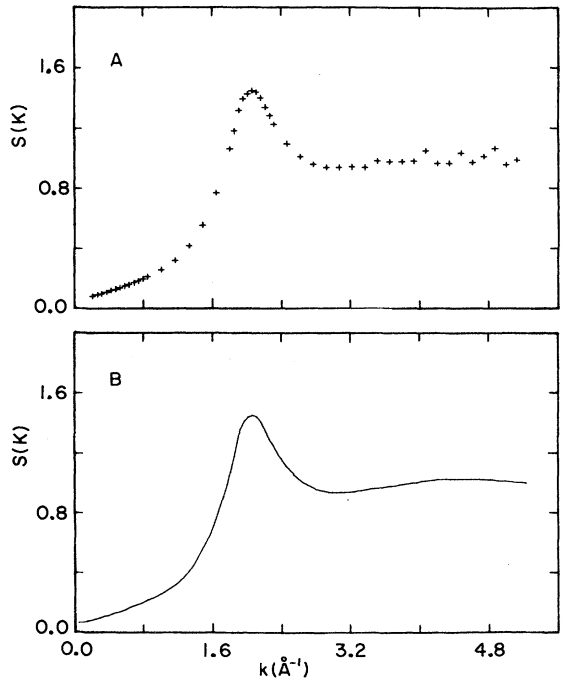

FIG. 1. (a) Structure factor for $T = 1.67$ K at saturated vapor pressure. No smoothing has been applied to the data. Much of the scatter at large momentum transfer is common to the various $S(k)$ determinations and is due to the neon normalization data. (b) Smoothed structure factor which results from the data of (a).

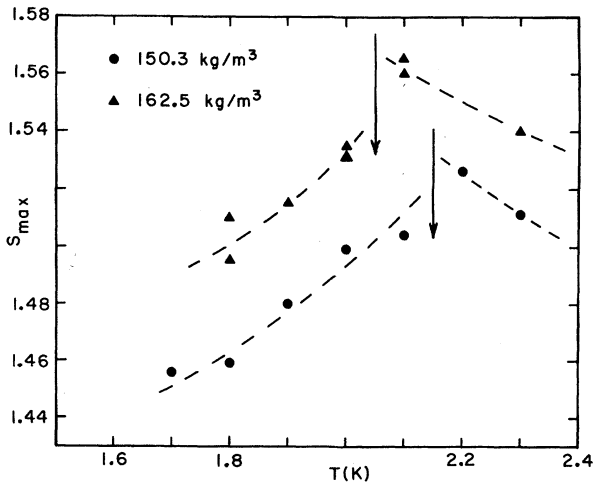

FIG. 2. Maximum height of the principal structure-factor peak at $\rho = 150.3$ and 162.5 kg/m^3 as a function of temperature. The arrow indicates T_λ for each density. The dashed lines are guides to the eye.

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neon used for normalization. In Fig. 1(b) we show the same $S(k)$ after a smoothing procedure[7] has been applied. The principal result of such smoothing is to remove the scatter in the data at large momentum transfer and to extrapolate the small-momentum-transfer results to the intercept $S(k=0)=\rho k_B T\chi_T$. Here ρ is the density, k_B the Boltzmann constant, T the temperature, and χ_T the isothermal compressibility. Because of the detailed data down to $k\simeq 0.2$ Å^{-1} and the limited extrapolation necessary, $g(r)$ at large r can be found with increased reliability.

The spatial order present in the fluid is visible directly in $S(k)$. Figure 2 is a plot of the maximum height of the main structure-factor peak as a function of the temperature at fixed density for the two densities $\rho=150.3$ and 162.5 kg/m^3. This maximum height, S_{max}, is seen to reach a maximum in the vicinity of T_λ and decrease on each side. In cooling a fluid one generally expects an increase in spatial order because of the decreased thermal motion of the atoms. Thus, the structure-factor peak should sharpen as the temperature is lowered. The data are consistent with both an increase in the peak height and a sharpening of the peak as the temperature is reduced.

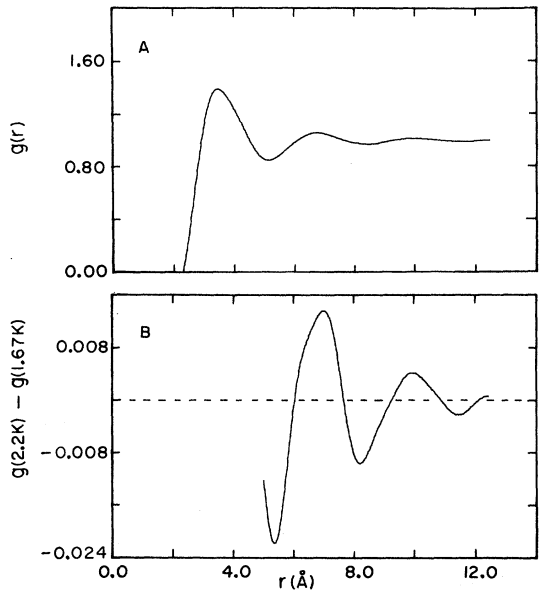

FIG. 3. (a) The pair correlation function $g(r)$ at $T=1.67$ K and saturated vapor pressure. Spurious structure in $g(r)$ for $r<2.0$ Å introduced by the Fourier transform have been suppressed. (b) The difference between two pair-correlation functions for $r>5$ Å. Plotted is $g(r, T=2.2\text{ K})-g(r, T=1.67\text{ K})$ as a function of r at saturated vapor pressure.

However, as the temperature is lowered below T_λ at constant density we observe a reduction in height and an increase in width of the peak, i.e., a loss of spatial order.

Fourier transformation of the data results in the pair correlation function $g(r)$. This quantity is the real-space measure of the spatial order in the fluid. In Fig. 3(a) we show the pair correlation function at SVP at $T=1.67$ K. There is less structure than in the pair correlation function at SVP at $T=2.20$ K. Since this difference would be difficult to see in a figure of the sort shown in Fig. 3(a), we show the difference $g(r, T=2.2\text{ K})-g(r, T=1.67\text{ K})$ in Fig. 3(b). The decrease in spatial order on cooling through T_λ is again evident.

Cummings, Hyland, and Rowlands[2] have suggested that the condensate fraction n_0 is related to the change in the large-r behavior of $g(r)$ that occurs on crossing the λ line. Explicitly,

$$n_0=1-\{[g(r, T^-)-1]/[g(r, T^+)-1]\}^{1/2}, \qquad (1)$$

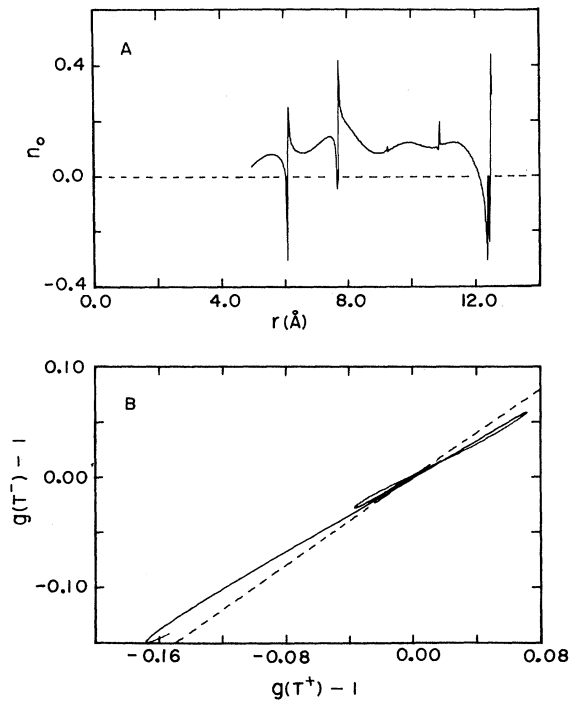

FIG. 4. (a) The quantity n_0 as a function of r for $r>5.0$ Å. The singularities result from near-zero values of $g(r, T=2.2\text{ K})-1$ and are of no significance. For these data at $T=1.67$ K and saturated vapor pressure we find $n_0=(9\pm 3)\%$. (b) An alternate way to illustrate $n_0\neq 0$. Plotted here is $g(r, T=1.67\text{ K})-1$ vs $g(r, T=2.2\text{ K})-1$. The dashed line has slope consistent with $n_0=0$ and clearly does not represent the data.

where $g(r, T^{\mp})$ is the pair correlation function below (−) and just above (+) the λ transition.[8] This expression is expected to be valid for the domain $r \gtrsim 4.5$ Å where the reduced one-particle density matrix approaches the condensate density n_0. It is instructive to apply Eq. (1) to the data with the understanding *firmly* in mind that the depth of the theoretical basis for Eq. (1) has yet to be established for the real ^{4}He system.

The application of Eq. (1) can be accomplished in more than one way. A direct application is shown in Fig. 4(a) for data taken at saturated vapor pressure and with $T^- = 1.67$ K and $T^+ = 2.20$ K. Since Eq. (1) becomes very sensitive to slight irregularities in the Fourier transform of $S(k)$ in the vicinity of $g(r) = 1$, the singularities which result should be ignored in favor of the more regular behavior encountered between the zeros of $g(r, T^+) - 1$. For the case shown the condensate fraction has the value $\simeq(9 \pm 3)\%$. In Fig. 4(b) we have illustrated the expression $g(r, T^-) - 1$ vs $g(r, T^+) - 1$ which, according to Eq. (1), is expected to have a slope equal to $(1 - n_0)^2$. Also shown on the figure is a dashed line drawn with slope unity (i.e., consistent with $n_0 = 0$). The merit of this type of plot is that the singularities

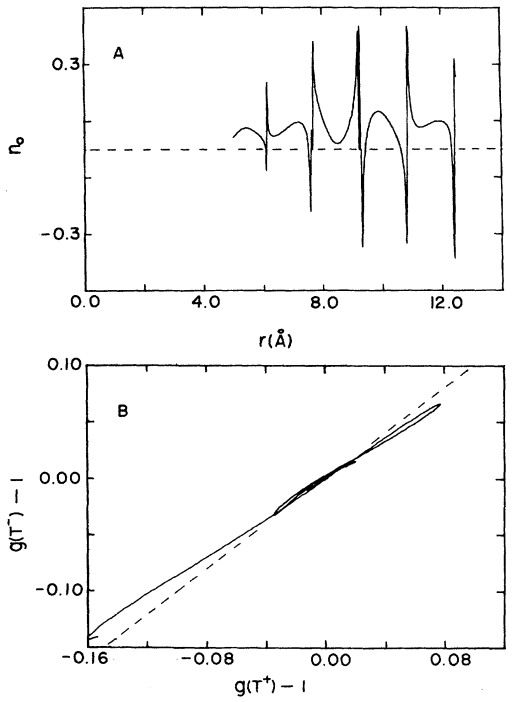

FIG. 5. Illustration of $n_0 \neq 0$ similar to that in Fig. 4 except here for the constant-density data 150.3 kg/m^3 at $T^+ = 2.2$ K and $T^- = 1.7$ K.

are confined to the region $g(r, T) - 1 \simeq 0$. A similar analysis applied to the constant-density data at 150.3 kg/m^3 and $T = 1.7$ K yields $n_0 = (8 \pm 3)\%$ and is displayed in Figs. 5.

A loss in spatial order in ^{4}He on cooling through T_λ has been observed by others.[9] We present more detailed evidence here that this behavior does in fact occur. There is no doubt that the spatial order in the liquid reverses its increase with decreasing temperature and decreases on cooling below T_λ. This loss in spatial order *may*[10] take place due to the formation of the condensate. To the extent that Eq. (1) can be applied to ^{4}He,[11] we observe a value for the condensate fraction well above that suggested by previous inelastic neutron results[3,12] and in reasonable agreement with theoretical expectations[13] and recent neutron work by Svensson *et al.*[4] A more careful theoretical investigation of the appropriateness of Eq. (1) would be most helpful.

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Pair Correlations and the Condensate Fraction in Superfluid ^{4}He

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By applying the method of Hyland, Rowlands, and Cummings to pair-correlation functions obtained from new high-accuracy neutron-diffraction measurements, the condensate fraction, n_0, in superfluid ^{4}He has been determined for seven temperatures in the range $1.00 \leq T \leq 2.15$ K. The results are very well described by the relation $n_0(T) = n_0(0)[1-(T/T_\lambda)^\alpha]$, with $n_0(0) = 0.133 \pm 0.012$ and $\alpha = 6.2 \pm 1.6$.

It is generally believed that the unique properties of superfluid ^{4}He result from the macroscopic occupation of the zero-momentum state, and numerous theoretical calculations[1-7] suggest that the fraction of atoms in this state, n_0, is in the range 0.08 to 0.13 at $T=0$.

In principle, the most direct method for the experimental determination of n_0 is, as proposed by Hohenberg and Platzman,[8] by means of neutron inelastic scattering at large wave-vector transfer, Q, where the dynamic structure factor $S(Q,\omega)$ reflects the momentum distribution of the atoms. In practice, however, the scattering at Q values accessible at present is distorted by a number of effects[9] and attempts[10-14] to determine n_0 by this method have yielded contradictory results with values ranging from 0.02 to 0.17 at $T \simeq 1.2$ K.

An alternative method in which the condensate fraction is determined through its effect on the spatial correlations of the atoms as reflected in the static pair-correlation function $g(r)$ was proposed some years ago by Hyland, Rowlands, and Cummings.[15] In this Letter we report the first critical experimental test of this method and obtain values of n_0 for seven temperatures which give a reasonably complete picture of its temperature dependence. Our values are consistent with the most recent theoretical estimates[5,7] but not with most of the values inferred from the high-Q experiments.[10-14]

The basic hypothesis of Hyland, Rowlands, and Cummings[15] is that the temperature variations of $g(r)$ at large r for superfluid ^{4}He are caused primarily by atoms condensing into the zero-momentum state where they no longer contribute to the spatial correlations of the atoms which give rise to the oscillations in $g(r)$. On the basis of *ad hoc*, although physically resonable, assumptions they then obtain

$$g(r) - 1 = (1-n_0)^2[g^*(r) - 1], \tag{1}$$

where $g(r)$ and n_0 refer to some temperature below T_λ, and $g^*(r)$ refers to a temperature T^* just above T_λ. The quantity $g(r)-1$, which is a measure of the spatial correlations, is thus proportional to $(1-n_0)^2$, the normalized probability that the two atoms are not in the condensate. A necessary condition for the validity of (1) is that $r > r_0$, where r_0 is the value above which the one-particle density matrix effectively attains its asymptotic limit, n_0.

There have been two previous attempts to determine n_0 from Eq. (1). Cummings, Hyland, and Rowlands[15] found from existing x-ray results that $n_0 \simeq 0.1$ at $T = 1.4$ K with an uncertainty of a factor of 2 or 3. Later, Raveché and Mountain[16] used neutron-diffraction results for pressurized liquid ^{4}He to obtain values of 0.1 at $T = 1.86$ K

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