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Spatial structure changes in ^{4}He at fixed density as a function of temperature

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X-ray scattering techniques have been used to determine changes in spatial order when cooling ^{4}He below T_λ at several fixed values of the density. The results show surprisingly little density dependence and are relevant to the discussion of condensate fraction determinations in ^{4}He.

For many years both x-ray and neutron scattering techniques have been of great value in determinations of the spatial structure of matter. In the case of liquid helium the superfluid transition has been a particularly interesting area of study. Scattering techniques provided a crucial test of the early idea[1] that superfluid helium could be characterized by a diamond lattice which disappeared[2] upon warming through the λ transition. That idea was put to rest, but the neutron work of Henshaw[3] showed clearly that the temperature dependence of the spatial order in liquid helium undergoes a change in the vicinity of T_λ. Recent work[4,5] has displayed the details of this temperature dependence.

Measurements show that as liquid helium is cooled toward T_λ the spatial order shows a gradual increase as might be expected due to the decrease in thermal disorder. Further cooling from the vicinity of T_λ reveals a decrease in spatial order. This decrease has been well documented at saturated vapor pressure[5] and fixed low density.[4,5] Some limited[6] and preliminary[7] results at elevated density have also been presented. The present paper presents the results of a new systematic set of measurements of the temperature dependence of the spatial structure in ^{4}He at several fixed values of the density and yields the surprising conclusion that *relative changes in spatial order are remarkably insensitive to the density.*

There are two popular reasons advanced for the generally observed decrease in spatial order with a decrease in temperature. One idea concerns the possible existence of a Bose condensate in ^{4}He. The physical argument centers around the idea that if, upon cooling, a finite fraction of the ^{4}He atoms condense into a macroscopic state of precise momentum, those atoms become spatially delocalized and thus the overall spatial order must decrease. From this point of view, as the temperature is lowered and more atoms enter the condensed state, the observed spatial order should decrease. If this decrease in the observed spatial order is due to the growth of a finite condensate fraction n_0, it might be possible to deduce the magnitude of the temperature-dependent condensate from the experimental data. Following this line of reasoning, Cummings *et al.*[8] have suggested that the prescription

$$F=1-\left[\frac{g(r,T^-)-1}{g(r,T^+)-1}\right]^{1/2} \tag{1}$$

may be used to compute the condensate fraction from the pair correlation function at the temperature of interest, $g(r,T^-)$, and the pair correlation function above[9] T_λ, $g(r,T^+)$. The work of Ref. 8 suggests $n_0=F$ and is not without its critics. In particular, Fetter[10] has shown a counterexample in which a system with an explicit Bose condensation does not fit Eq. (1). Griffin[11] has argued that the theoretical basis upon which Eq. (1) has been derived is weak and at best incomplete. Nonetheless, it is argued[12] that Eq. (1) may in fact be a proper prescription for the experimental determination of n_0.

An alternate point of view has been advanced by Reatto and his co-workers[13–15] who argue that the increase in spatial structure seen on warming ^{4}He below T_λ is due to the increasing population of thermal rotons and not directly related to questions of a condensate fraction in the liquid. The physical picture here is that the roton is an entity with substantial short- and intermediate-range correlations among the atoms which make it up. Thus an enhancement in the number of rotons (e.g., with an increase in temperature) results in an increase in the observed spatial order in the helium. This effect should persist for temperatures well above the λ point but the effects of finite roton lifetime wash it

out.

These two points of view result in very different predictions for the temperature evolution of the spatial order as a function of density. The condensate fraction is expected[16] to be a rather strong function of density *falling* from $n_0 \simeq 10\%$ at low temperature and at saturated vapor pressure to a few percent at low temperature near the melting curve; the increased interactions among the atoms at elevated density are generally expected to deplete the condensate. The argument based on rotons predicts an *enhancement* of the changes in spatial order with an increase in density due in part to the increase in the number density of rotons.

Given these rather different predictions of what one might expect for ^{4}He, we have made detailed measurements of the spatial structure of ^{4}He at three fixed values of the density, $\rho = 171.0$, 162.5, and 150.3 kg/m^3, over the temperature range $1.16 \leq T \leq 2.80$ K. X-ray scattering techniques which have been described in some detail previously[17,18] were used to determine the liquid structure factor $S(k)$ as a function of the momentum transfer k over the range $0.2 \leq k \leq 5.1$ Å^{-1}. A rotating-anode x-ray source provided Cu $K\alpha$ x rays of wavelength $\lambda = 1.54$ Å. The pair correlation function is determined from the liquid structure factor by a Fourier transform. To facilitate the transforms the $S(k)$ data is smoothed and interpolated to provide a dense data grid over the momentum-transfer range $0 \leq k \leq 5$ Å^{-1}. The intercepts at $k = 0$ are in excellent agreement with expectations[17] based on thermodynamics.

The results of the present work show clearly that the temperature-dependent changes in spatial order at fixed density do not decrease with an increase in density. This can be illustrated in two ways which we now describe.

Direct application of Eq. (1) to the data results in a comparison of the temperature-dependent spatial structure in ^{4}He at a given temperature to that just above the λ point. We have computed this comparison from the data for each of the three fixed values of the density studied with the results shown in Fig. 1. Contrary to what might be expected for the condensate fraction, we observe the relative changes in spatial order to remain essentially independent of changes in the density. There is a shift with T_λ, but no change as a function of T/T_λ. In fact, the entire data set is well represented by the single function

$$F = A[1-(T/T_\lambda)^B]$$

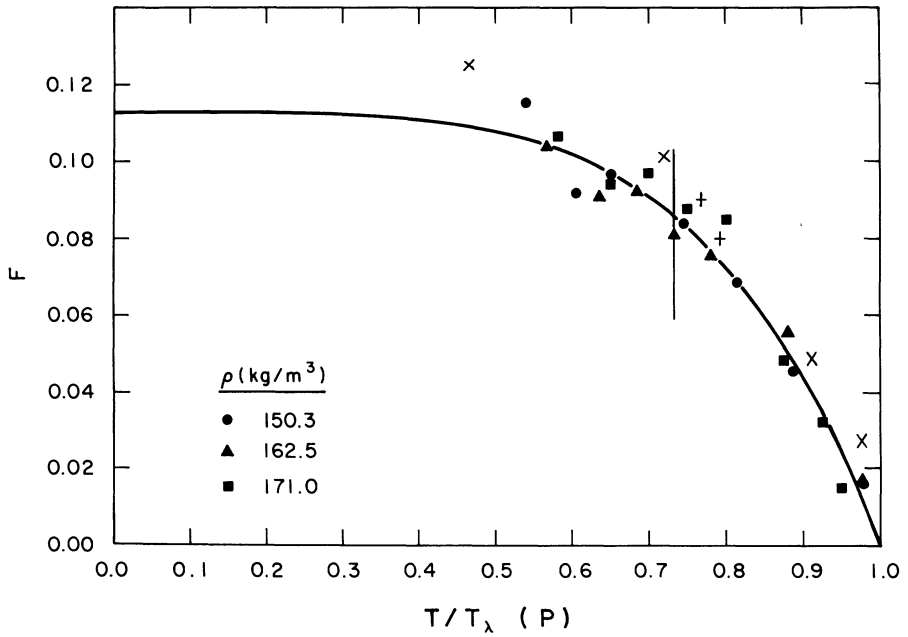

FIG. 1. Values of F which result (solid symbols) from application of the prescription, Eq. (1), to the fixed-density structure-factor measurements vs T/T_λ. Solid curve represents $F = A[1-(T/T_\lambda)^B]$ with fitting parameters $A = 0.112 \pm 0.006$ and $B = 4.64 \pm 0.48$ with T_λ appropriate for each density studied. Values of (A,B) for each separate density are found to be 150.3 ($0.115 \pm 0.009, 4.27 \pm 0.83$), 162.5 ($0.103 \pm 0.006, 5.54 \pm 0.70$), and 171.0 ($0.119 \pm 0.012, 4.31 \pm 0.92$). Also shown is representative standard vapor pressure neutron work (Refs. 5 and 20) ($\times$) and earlier (Ref. 4) x-ray work at saturated vapor pressure and $\rho = 150.3$ kg/m^3 ($+$). A typical error bar generally representative of the data is shown.

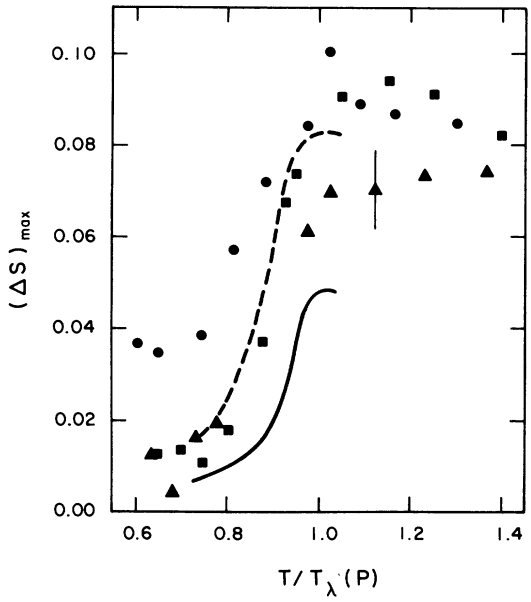

FIG. 2. Temperature dependence of $(\Delta S)_{max}$ for the data at each density. Lines are predictions from Ref. 12: —— for 145 kg/m^3, — — — for 174 kg/m^3.

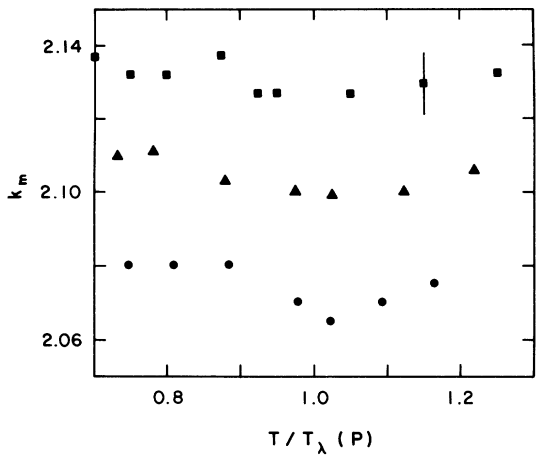

FIG. 3. Pressure and temperature dependence of k_m, the location of the principal maximum of $S(k)$.

with fit parameters $A=0.112\pm0.006$ and $B=4.64\pm0.48$. This suggests that either current theoretical understanding of the condensate fraction as a function of density is in need of serious reexamination *or* that Eq. (1) is in fact inappropriate for helium.

The present data can be compared directly with the work of Gaglione *et al.*[14] who predict that the spatial structure will strengthen rather than diminish as a function of density. At each density the maximum value of

$$\Delta S=S(k,T)-S(k,T_0)\ ,$$

which we denote as $(\Delta S)_{max}$, has a temperature dependence (see Fig. 2) which is in general agreement with predictions[14,19] based on the Green's-function Monte Carlo calculations at elevated density due to Whitlock *et al.*[16] Here T_0 is the lowest available temperature at each density. However, the expected substantial increase in amplitude with an increase in density is not observed. In addition it is predicted[14] that the location of the principal maximum of $S(k)$, k_m, will shift (by 0.1 Å^{-1}) to smaller values of the momentum transfer as the temperature is increased toward T_λ. Our results do not confirm this latter prediction in quantitative detail. Rather there is a small decrease $|\Delta k| \lesssim 0.02$ Å^{-1} in the vicinity of T_λ with a general trend for k_m to be independent of temperature (see Fig. 3).

On the basis of these new data we conclude that liquid helium behaves in a manner which is more complex than previously considered. If, as suggested by recent inelastic neutron determinations[20] of n_0 at saturated vapor pressure, Eq. (1) is appropriate for ^{4}He, then new theoretical work is needed to explain why the condensate is not suppressed by an increase in density. On the other hand, it may be that the changes in the spatial order as a function of density expected due to (1) Bose condensation and (2) the thermal population of rotons nearly cancel out. In this later case one would conclude that the prescription, Eq. (1), may in general be inappropriate for the determination of n_0 for ^{4}He. Inelastic neutron results of elevated density would be of particular interest.

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