see, for example, H. Risken, in *Progress in Optics*, edited by E. Wolf (North-Holland, Amsterdam, 1970), Vol. 8.

[5]We use the same notation as Graham, except that we have altered the scales of w and t as noted in the text. We also characterize the deviation from threshold by a, which differs from Graham's ϵ. We repeat Graham's notation for the reader's convenience: Rayleigh number $R=g\beta\Delta T l^3/\nu\kappa$; Prandtl number $P=\nu/\kappa$; $Q=\pi^2(Q_1+Q_2)/(1+P)$; $Q_1=g\beta kT/\rho C_p\nu^2\Delta T$; $Q_2=kT/\rho\nu^2 l$; l and V, thickness and volume of the convection cavity; ΔT, externally maintained temperature difference between the top and bottom of the convection cavity; T, average fluid temperature; (the following fluid properties have their value for acetone at 20°C given in parentheses) ρ, density (0.79 gm/cm^3); C_p, specific heat (2.18 J/g mC deg); β, volume expansion coefficient (0.0015/C deg); κ, thermometric conductivity (1.04×10^{-3} cm^2/sec); and ν, kinematic viscosity (4.11×10^{-3} cm^2/sec). The acceleration of gravity is g and Boltzmann's constant is k.

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[7]The evaluation of these initial derivatives was greatly facilitated by performing algebraic manipulations on a computer using the ALTRAN language. See A. D. Hall, Commun. Assoc. Comput. Mach. 14, 517 (1971).

[8]R. Graham, private communication. This is an expression for the spontaneous reversal time.

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Neutron-Scattering Study of the Momentum Distribution of ^{4}He†

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Neutron inelastic scattering at a momentum transfer of about 15 Å^{-1} has been used to measure the scattering law $S(Q,\omega)$ with high accuracy for ^{4}He at 1.2 and 4.2°K. At this momentum transfer the impulse approximation is nearly valid and momentum distributions for ^{4}He can be determined directly from the data. The momentum distributions for the two temperatures show features not previously observed and provide new information to compare with theoretical results.

When neutrons are scattered with a sufficiently high momentum transfer Q from a material, the scattering may be thought of as taking place from individual atoms and collective effects have little importance. For most materials the value of the momentum transfer Q needed to be in the single-particle regime is prohibitively high; however for liquid ^{4}He, a material of extreme interest, the value of Q necessary is about 15 Å^{-1}, making experiments difficult but possible with neutrons available from a reactor.[1-7] Two prior experiments have been performed on ^{4}He with momentum transfers of this magnitude, the first by Harling[8] with a time-of-flight technique and the second by Mook, Scherm, and Wilkinson[9] with a triple-axis spectrometer. The ^{4}He momentum distribution n_p depends on the derivative of the scattering distribution and the experimental data must have high accuracy and resolution to obtain a satisfactory result for n_p. Harling's data were of sufficient accuracy to obtain bulk quantities such as the average kinetic energy for the helium atoms but were unsuitable for extracting detailed information like n_p. The Mook, Scherm, and Wilkinson experiment concentrated around the peak of the scattering distribution in order to observe Bose-Einstein condensation and was of high accuracy in this region. However the data in the tails and part of the sides of the scattering distribution were not of sufficient accuracy to produce a reliable n_p. The data presented in this paper are of sufficient quality for the entire scattering distribution that detailed information about n_p is available for the first time.

n_p has been determined for two temperatures, 4.2 and 1.2°K. Both distributions show surprising structure and give information about ^{4}He that was not previously available. The 1.2° distribution exhibits a peak of 0.2 Å^{-1} that is clearly indicative of Bose-Einstein condensation. The remainder of the distribution shows structure that has not been observed in previous calculations for n_p and thus suggests that more refined theoretical work is necessary. The data at 4.2°K show a large hump at a momentum of 2.25 Å^{-1} which is in the roton region of the excitation spectrum. If a connection is made between the excitation spectrum and n_p, one is led to the surprising conclusion that rotons persist as distinct excitations at temperatures up to twice the tran-

sition temperature $T_\lambda = 2.19°K$.[10] It is hoped that the measurements will encourage theoretical calculations of n_p as a function of temperature.

For the case of liquid helium, where the neutron can be thought of as interacting only with a single atom, conservation of energy and momentum for the scattering event gives the following relation for the scattering cross section:

$$\frac{d^2\sigma}{d\Omega\, d\omega} = c\frac{k_f}{k_i}S(Q,\omega) = c\frac{k_f}{k_i}\sum_p n_p \delta\left(\omega - \frac{Q^2}{2m} - \vec{Q}\cdot\frac{\vec{p}}{m}\right), \quad (1)$$

where c is a constant, k_f and k_i are the final and incident neutron wave vectors, p is the momentum of the helium atom before the scattering event, m is the ^{4}He mass, and $\hbar$ has been set equal to 1. In reality the neutron will not interact solely with one helium atom and final-state effects must be considered. In this case Eq. (1), commonly known as the impulse approximation, must be modified and the scattering law $S(Q,\omega)$ is given by

$$S(Q,\omega) = \sum_p n_p R(Q, \omega - Q^2/2m - \vec{Q}\cdot\vec{p}/m), \quad (2)$$

where

$$2\pi R = \int_{-\infty}^{\infty} dt\, e^{-i(\omega - Q^2/2m - \vec{Q}\cdot\vec{p})t} R(Q,t), \quad (3)$$

and $R(Q,t)$ is a complicated expression involving the liquid's one-particle and two-particle off-diagonal density matrices.[3] The function R has been evaluated for several momentum transfers of interest including 15 Å^{-1}.[11] Evaluating n_p using the approximate expression (1) is almost equivalent to obtaining n_p from experimental data taken with finite resolution. Sharp structure like the condensate contribution will be modified significantly; however, the n_p which is obtained from the main body of $S(Q,\omega)$ will not be greatly affected. Since the experiment itself must be done with finite resolution and unfolding the resolution from an arbitrary scattering function probably only results in a deterioration of the data, it appears that the best thing one can do in a straightforward manner is to evaluate n_p directly from the scattering data using Eq. (1). The experimental resolution contributes about 0.3 Å^{-1} to the resolution width of n_p and the function $R(Q,t)$, representing the final-state effects, about 0.4 Å^{-1}. It has been suggested that the calculated $R(Q,t)$ might slightly overestimate the effective width contributed by the final-state effects.[11] Thus the n_p obtained using Eq. (1) would have an effective resolution somewhat under 0.5 Å^{-1} which is narrow enough to observe a great deal of interesting structure. Perhaps attempts may be made to improve this effective resolution in n_p with the aid of Eq. (2) at a later date.

Continuing then with Eq. (1) and converting to an integral, we obtain, after differentiation,

$$pn_p|_{p_{\min}} = (4\pi Q^2\rho/m^2)\partial S(Q,\omega)/\partial\omega, \quad (4)$$

where ρ is the density and $p_{\min} = [\omega - \omega(Q)](Q/m)^{-1}$, with $\omega(Q)$ the neutron energy transfer for the momentum Q. Unfortunately Eq. (4) must be modified in a complicated manner to obtain pn_p since the measurement was made at constant scattering angle rather than constant momentum transfer. In this case Eq. (4) becomes

$$pn_p|_{p_{\min}} = \frac{\pi^2\rho}{cm}\left(1 - \frac{\omega}{\epsilon_i}\right)^{-1/2}\left\{8\omega(Q)\frac{\partial}{\partial\omega} + \left[\frac{4\omega(Q)}{\epsilon_i - \omega} - 1 + \left(1 - \frac{\omega}{\epsilon_i}\right)^{-1/2}\cos\theta\right]\right\}\frac{d^2\sigma}{d\Omega\, d\omega} \times\left\{1 + \tfrac{1}{8}\left(1 + \frac{\omega}{\omega(Q)}\right)\left[1 - \left(1 - \frac{\omega}{\epsilon_i}\right)^{-1/2}\cos\theta\right]\right\}^{-1}, \quad (5)$$

where ϵ_i is the incident neutron energy and θ is the scattering angle.

A neutron scattering experiment with a momentum transfer of ~15 Å^{-1} is very difficult in that neutrons with energies of about 0.2 eV must be used, which is much higher than the peak in the Maxwell spectrum from the reactor, and good resolution must be used since the width of $S(Q,\omega)$ is only about 0.015 eV. The data were taken with the cross-correlation time-of-flight spectrometer at the high-flux isotope reactor that utilizes a magnetically pulsed beam.[12] This instrument can produce a 4-μsec pulse for a 1-in. by 1½-in. beam and can be programmed for any type of correlation pulse chain. Total data collection time was 2½ weeks at each temperature. Similar data taken only over the peak of $S(Q,\omega)$ with a triple-axis spectrometer took five months.[9] The incident energy was 0.1894 eV and the scattering angle was 151.75 deg giving $Q = 14.79$ Å^{-1}. The energy resolution was about 0.003 eV and the statistical error about 3% on all the data points.

The data are shown in Fig. 1 for the cross sec-

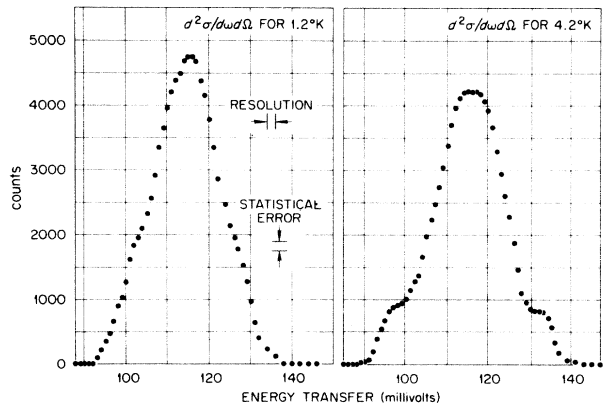

FIG. 1. Scattering cross section $d^2\sigma/d\Omega\, d\omega$ for a scattering angle of 151.75° plotted as a function of the energy transfer for ^{4}He at 1.2 and 4.2°K. The cross section scale is arbitrary since only the relative cross sections were determined for the two temperatures.

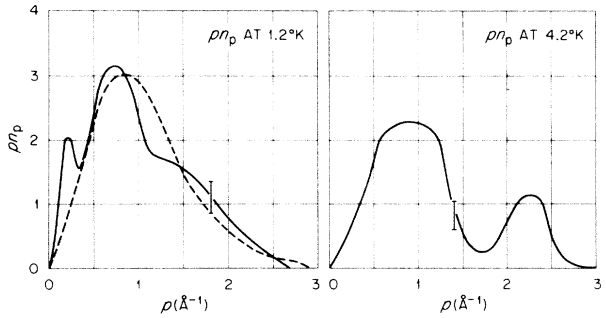

FIG. 2. pn_p distributions for ^{4}He at 1.2 and 4.3K. The dashed line is a result calculated by Kalos.

tions at 1.2 and 4.2°K. One can see that the curves show considerable structure so that pn_p will also have structure. pn_p obtained from the cross section by use of Eq. (5) is shown in Fig. 2. The pn_p curves were determined in the following manner. The cross section was differentiated point by point and pn_p obtained for each side of the cross-section curve. The two sets of points for pn_p were then combined and the solid curves obtained by a Gaussian smoothing procedure. pn_p is the most logical quantity to discuss rather than n_p since pn_p is derived directly from the data and the statistical uncertainty is roughly constant over the entire curve. The dashed line shown with the 1.2°K data is a calculation made by Kalos.[13] The calculation is for zero temperature; however, it is thought that the pn_p distribution would not be modified significantly between 0 and 1.2°K.[1] The Kalos calculation seems to fit the data better than the one made by Francis, Chester, and Realto[14] and considerably better than a result by McMillan.[15]

The peak at about 0.2 Å^{-1} appears in no calculation for pn_p and is certainly the result of Bose-Einstein condensation below T_λ. Instrumental resolution and final-state effects certainly greatly affect the sharpness of this peak; however, its area should not be changed, and if the Kalos calculation is used to extrapolate the main body of the pn_p distribution to zero, a separation can be made between the condensate and noncondensate part of the distribution. This results in a value of the condensate fraction of $(1.8 \pm 1)\%$ which is in agreement with the value of $(2.4 \pm 1)\%$ obtained from the triple-axis experiment.[9] The rest of the pn_p curve at 1.2°K contains structure that differs from the calculated results and hopefully serves to encourage better calculations.

The pn_p curve for 4.2°K shows no condensate peak but contains other interesting structure. Apparently as the ^{4}He is heated up from 1.2°, more and more energy goes into the roton excitations with a wave vector of around 2.25 Å^{-1} and these rotons persist as the ^{4}He is heated through the λ point to 4.2°K. It seems surprising that the roton hump is so distinct as one might expect the distribution to look more like the Maxwell-Boltzmann distribution as the temperature is raised from T_λ.

The single-particle density matrix can be obtained from the pn_p curves and, from this, other properties of ^{4}He that are of interest. The mean kinetic energy per atom can be obtained in a trivial manner from the pn_p curves by calculating the mean squared momentum. This is a bulk property of the ^{4}He and is not greatly affected by the fine structure in the scattering distribution. Values of 13.2 and 16.9°K are obtained from the 1.2 and 4.2°K data, respectively. The 1.2°K result is in good agreement with earlier work[4] but the 4.2°K mean kinetic energy is slightly higher, probably because the earlier work overlooked some of the roton excitations in the tails of the scattering distribution.

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Experimental Determinations of the Excluded-Volume Exponent in Different Environments

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The excluded-volume exponent ν is determined from a neutron scattering experiment by polystyrene chains, in the "intermediate" momentum-transfer range. The chains were successively dispersed in a good solvent, in a Θ solvent, and in the bulk material. The new result is the value of ν for the bulk material, which is discussed below.

The excluded-volume interaction introduces a characteristic exponent ν in the scaling law[1] of the end-to-end distribution function of chain segments separated by n steps,

$$P_n(r) = (1/n^{3\nu}) f(r/n^{\nu}). \quad (1)$$

The theoretical value of ν is very close[2] to $\frac{3}{5}$ in the presence of an excluded-volume interaction, and $\frac{1}{2}$ in the case of a free chain. Two observables can be derived from Eq. (1):

(a) The mean squared end-to-end distance $\langle R^2\rangle$, for a polymer chain of N segments,

$$\langle R^2\rangle = KN^{2\nu}, \quad (2)$$

where K is a constant independent of N. Relation (2) provides an experimental test of the value of ν in the momentum-transfer range $q < (\langle R^2\rangle)^{-1/2}$.

(b) The pair correlation function

$$g(r) = Be^{-r/Nl} r^{-(3\nu-1)/\nu} \quad (3)$$

in the range $l < r < (\langle R^2\rangle)^{1/2}$, where l is the step length and B is a constant independent of N. Equation (3) suggests a scattering experiment in the intermediate momentum-transfer range

$$(\langle R^2\rangle)^{-1/2} < q < l^{-1}. \quad (4)$$

The scattering law is

$$S(q) = \frac{12}{(ql)^{1/\nu} + O(1/N)}. \quad (5)$$

The value of ν is determined by the q dependence of the scattered intensity.

Relations (2) and (5) may be directly tested in a scattering experiment, provided that the single chain can be observed in its environment. This problem was discussed in earlier papers,[3,4] where it was shown that the neutron scattering technique is the only suitable one for the bulk material.

Real chains are nonintersecting and the measured value of ν is expected to be $\frac{3}{5}$, as in the case of a theoretical chain with excluded-volume interaction. This is observed in good solvents,[5] in the limit of zero polymer concentration. The environment of the chain may, however, produce situations in which the chain behaves as if it were free. A well-known case is the Θ solvent at the Flory[6] temperature $T = \Theta$. Another situation of interest is the bulk material, for which it is also thought[6] that the individual chains display no excluded-volume interaction. This was recently tested[7] by a neutron scattering experiment by deu-