Momentum Density of hcp ^{4}He

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The momentum density of the quantum solid hcp ^{4}He has been measured at 1.70 K at the molar volumes 18.20 and 19.45 cm^3. The measurements were made with pulsed incident neutrons of energy 505 meV, and with momentum transfers in the range of 12 to 22.5 Å^{-1}. The momentum density is obtained from the inelastic-scattering spectrum at each of eight scattering angles. The measured distribution is Gaussian and the derived kinetic energies are larger than proposed by existing theories.

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The solid heliums (both ^{3}He and ^{4}He) provide extreme examples of quantum solids which have attracted much theoretical and experimental interest over the years. Theoretical descriptions of the properties of solid helium are complicated by the relatively large kinetic energy and strong correlations of the atomic motion. Thus, theoretical methods that accurately describe the dynamics of most solids are not adequate to describe the properties of the solid heliums.[1] Although considerable progress has been made in theoretical estimates of the properties of solid heliums, many uncertainties remain. For example, theoretical estimates of vacancy-tunneling frequencies differ by more than an order of magnitude.[2] These uncertainties also preclude *ab initio* estimates of particle-exchange integrals which are of interest, for example, in the magnetism of solid ^{3}He.

Information about the real-space wave function of solid helium is obtainable via the single-atom momentum distribution. In this Letter we report the first momentum density measurements on a solid helium which provide a detailed test for ground-state calculations. Our measurements of the spherically averaged momentum distribution made on hcp ^{4}He at two molar volumes, furthermore, provide the variation with sample density.

It has been known for years that neutron scattering at high energies offers a technique to obtain information about the wave function of solid helium through the momentum distribution, $n(\vec{p})$, which is the square of the Fourier transform of the real-space wave function. However such experiments are difficult with reactor sources because the relatively low epithermal neutron flux requires long counting times. Spallation neutron sources, which have recently begun to come on line around the world, give one the opportunity to do such experiments reliably and with short counting times, 40 h per sample in the case of the present experiment. If the dynamic structure factor, $S(\vec{Q},E)$, is measured as a function of energy transfer E at sufficiently high wave vectors $\vec{Q}$, the scattering can be treated as a collection of scattering events involving isolated atoms of mass M and the dynamic structure factor can, with good accuracy, be written in the form of the impulse approximation (IA)[3]

$$S(\vec{Q},E)=\int n(\vec{p})\delta[E-(\hbar^2Q^2/2M) -\hbar^2(\vec{Q}\cdot\vec{p}/M)]\,d^3p. \quad (1)$$

If the impulse approximation is valid, we expect the data collected at a given scattering angle to be centered about an energy transfer value, referred to as the recoil energy, of

$$E_r=(\hbar^2Q^2/2M). \quad (2)$$

This is the relation that is valid for scattering of neutrons from free helium atoms.

Many systems have a momentum density that is well represented by a Gaussian of the form

$$n(\vec{p})=C\exp(-p^2/\hbar^2A). \quad (3)$$

While there is no *a priori* reason that $n(\vec{p})$ for helium should be of this form, our data indicate that a Gaussian accurately represents this function, as discussed below. Inserting Eq. (3) into Eq. (1), one finds the dynamic structure factor to be

$$S(\vec{Q},E)=C\exp\left[\frac{-(E-E_r)^2}{\hbar^4Q^2A/M^2}\right]. \quad (4)$$

The dynamic structure factor is thus a Gaussian centered at the recoil energy E_r, of variance

$$\sigma_Q^2 = (\hbar^4 Q^2 A/2M^2). \quad (5)$$

Measurement of the inelastic scattering cross section at suitably large momentum and energy transfer would thus yield A, the width parameter in $n(\vec{p})$. Further, the form of Eq. (3) corresponds to a kinetic energy

$$\langle K \rangle = \tfrac{3}{4}\hbar^2(A/M) \quad (6)$$

so this also provides a test for ground-state kinetic energy calculations.

In the above discussion, information about $n(\vec{p})$ is deduced using the impulse approximation. The IA is expected to be valid when the recoil energy is very large relative to the dynamic excitation energies of the system which, in hcp ^{4}He, extend to about 7 meV.[4] Thus recoil energies of say 100 meV and above are required, implying wave vectors $Q \sim 14$ Å^{-1} and higher. Earlier neutron scattering studies of solid helium[5] indicated an approach to single-particle scattering at Q up to about 5.5 Å^{-1}; however, these were unable to yield critical information about the momentum density.

While Eq. (6) has been deduced using the IA, we point out that the relation between $\langle K \rangle$ and the second moment of $S(\vec{Q},E)$ is established by the existence of a rigorous sum rule, $\sigma_Q^2 = (\frac{4}{3})E_r\langle K \rangle$, valid for all Q, independent of the IA. However, numerical determination of moments is somewhat difficult. Because our experimentally determined $S(\vec{Q},E)$ can, at constant Q, be empirically fitted very well by a Gaussian, we use the fit to establish the variance of $S(\vec{Q},E)$. For this reason, our measurement of $\langle K \rangle$ is independent of the validity of the IA.

The data were taken on the low-resolution chopper spectrometer, a time-of-flight spectrometer at the Intense Pulsed Neutron Source (IPNS-1) at Argonne National Laboratory.[6] The neutron detectors were positioned at mean scattering angles from 47.7° to 115.5°; this gave data for momentum transfers in the range 12.2 to 22.5 Å^{-1} and energy energy transfers in the range 77 to 264 meV for the 505-meV incident neutrons. Samples of hcp ^{4}He were grown at molar volumes 19.45 and 18.20 cm^3; the molar volume was determined from the pressure and temperature at which each sample solidified. Data were taken with each sample held at 1.7 K which, when compared with the Debye temperatures (30 and 36 K for the two densities), implies that the samples were essentially in the ground state.

The samples were cylindrical, 2.7 cm diameter by 5.7 cm high, and were contained in an aluminum sample cell to minimize the scattering from the cell. The high thermal conductivity of aluminum also promoted the growth of polycrystalline samples; the extent to which the samples were polycrystalline was checked by observing the neutron diffraction pattern from the high-density solid sample. We chose to prepare polycrystalline samples because it was not clear from theory the extent to which $n(\vec{p})$ was expected to be isotropic,[7,8] and we wanted to measure an average $n(\vec{p})$ in this experiment.

The raw time-of-flight data were converted to the scattering function after the background data, obtained with the sample cell empty, were subtracted. Detector efficiencies were incorporated using the known filling pressure and energy variation of the ^{3}He absorption cross section. The sample transmission was 93%; multiple-scattering effects represent about 4% of the signal and will not significantly affect the shape of the recoil peak. The data were obtained with the scattering angle held constant; the resulting structure factor is denoted as $S(\phi,E)$, whereas the form of Eq. (4) is for a constant-$\vec{Q}$ scan. An analysis of the peak shape for a constant angle scan shows that if the constant-$\vec{Q}$ scan has a Gaussian peak of variance σ_Q^2 the constant angle scan will to the first approximation also have a Gaussian peak, but of variance σ_ϕ^2 given by

$$\sigma_\phi^2 = \sigma_Q^2(1 - C_\phi)^{-2} \quad (7)$$

where

$$C_\phi = \frac{m}{M}\left[\frac{k_0}{k_1}\cos\phi - 1\right]; \quad (8)$$

m is the neutron mass and k_0 and k_1 are its incident and scattered wave vectors. We confirmed the validity of Eq. (7) by interpolating the many observed $S(\phi,E)$ to get $S(\vec{Q},E)$, and fitting Gaussians to the two sets of data. Having calculated σ_Q, one must correct for the finite instrument resolution, the calculation of which is reported separately.[9] The resolution was also deduced from scattering from a vanadium sample and convoluted with $S(\vec{Q},E)$ calculated for a room-temperature vanadium sample. Good agreement was obtained with the measured spectra, thus providing a check on the resolution functions calculated for our hcp ^{4}He measurement.

The measured $S(\phi,E)$ was fitted to a Gaussian with a linear background, i.e.,

$$S(\phi,E) = C_1 \exp[-(E - E_{pk})^2/2\sigma_\phi^2] + C_2E + C_3, \quad (9)$$

and a typical fit to the data is shown in Fig. 1. In

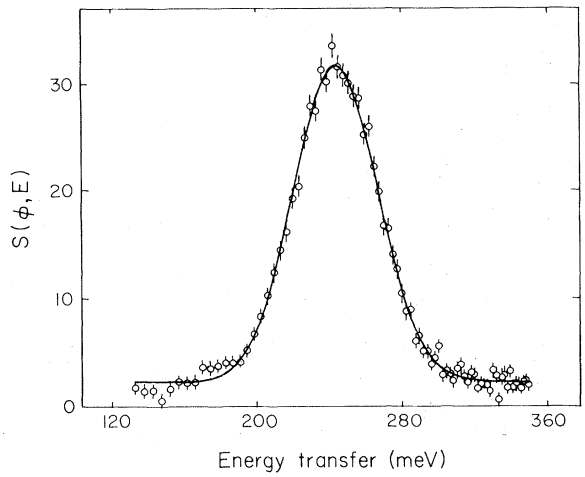

FIG. 1. The measured dynamic structure factor in arbitrary units is shown for the 19.45 cm^3/mole sample with the scattering angle at 105.9°. The statistical error bars are shown, together with a least-squares fit to the Gaussian form [Eq. (9)]. For this data peak $\sigma_Q = 28.9$ meV and the resolution width is 11.3 meV.

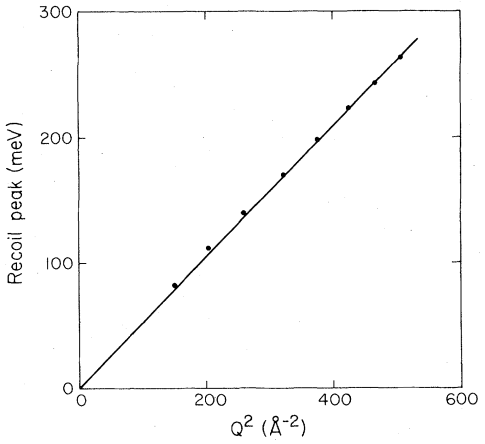

FIG. 2. The peak positions fit to $S(\phi,E)$ are shown together with the straight line calculated for single-particle scattering. The error in the fitted values is given by the size of the points, and the values for all the samples are within the errors.

Fig. 2 we have plotted the values of the fitted E_{pk} as a function of Q^2. The line is a plot of Eq. (2) with M being the ^{4}He atomic mass. That the measured recoil energies lie on the line indicates that the scattering events occur between a single neutron and a single ^{4}He atom. This is a necessary, though not sufficient, condition for IA applicability. From the fitted values of σ_ϕ we obtain σ_Q using Eq. (7). According to Eq. (5), σ_Q should be a linear function of $\vec{Q}$; in Fig. 3 we have plotted the results of our measurement for the two hcp ^{4}He samples together with a linear fit constrained to pass through the origin. We believe some of the scatter to be caused by anisotropy effects evident in diffraction data taken on the high-density hcp sample which indicated some degree of nonrandom orientation.

The diffraction data taken on the high-density solid indicates that the sample was made up of fairly large grained crystalline regions. The observed anisotropy in this sample was sufficient to explain why the points in Fig. 3 tend to lie further from the fit than their uncertainty. A simple model based on the picture of a helium atom constrained inside a Wigner-Seitz cell appropriate for 18.20 cm^3/mole hcp ^{4}He accounts for this anisotropy and, though such a cell model is not capable of accurately modeling the properties of solid helium,[10] we find it explains the apparent anisotropy in the data. This anisotropy is averaged over in our data analysis so that our results are appropriate for the spherically averaged $n(\vec{p})$. Further experiments are necessary on single-crystal ^{4}He samples before one can make quantitative statements about the anisotropy of $n(\vec{p})$ in the solid.

From the values of σ_Q extracted from the fits in Fig. 3, we find the following values of the single-atom kinetic energy, in temperature units, in the two samples:

$$\langle K \rangle = 34.3 \pm 0.9 \text{ K at } 18.20 \text{ cm}^3\text{/mole}$$

and

$$\langle K \rangle = 31.1 \pm 0.9 \text{ K at } 19.45 \text{ cm}^3\text{/mole}.$$

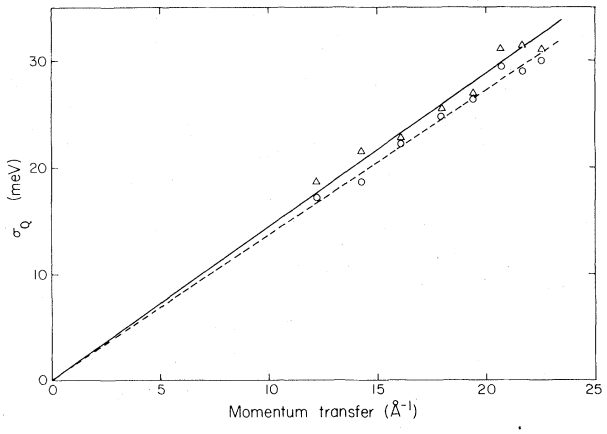

FIG. 3. The standard deviation of the fit to the data is shown as a function of the momentum transfers; the size of the points is the estimated error in the variance. The straight line fit was constrained to pass through the origin. The full line and the triangles correspond to the 18.20 cm^3/mole solid sample, while the dashed line and the circles correspond to the 19.45 cm^3/mole sample.

A Monte Carlo calculation has been reported[8] that yields a ground-state kinetic energy and momentum density for fcc solid helium, with atoms interacting via a Lennard-Jones potential, at a molar volume of 19.12 cm^3. These authors quote a ground-state kinetic energy of $\langle K \rangle = 27.2 \pm 0.1$ K. This value is 15% lower than our measured values and, although their calculation yields a somewhat larger calculated value for the hcp phase,[11] crystal structure apparently does not account for the difference in the measured and predicted values. However, the experimentally determined dependence of $\langle K \rangle$ on the molar volume agrees well with the values calculated by these authors.

In conclusion, the momentum density of hcp ^{4}He measured in this experiment is Gaussian, within statistics and resolution, and it is broader than that theoretically predicted. This experiment is representative of the techniques that are well suited to spallation neutron sources due to the high flux of epithermal neutrons available.

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[1]R. A. Guyer, in *Solid State Physics,* edited by F. Seitz, D. Turnbull, and H. Ehrenreich (Academic, New York, 1969), Vol. 23, p. 143; C. M. Varma and N. R. Werthamer, in *The Physics of Liquid and Solid Helium,* edited by K. H. Bennemann and J. B. Ketterson (Wiley, New York, 1976), Vol. I, Chap. 6.

[2]H. A. Goldberg and R. A. Guyer, J. Low Temp. Phys. 28, 449 (1977).

[3]P. C. Hohenberg and P. M. Platzman, Phys. Rev. 152, 198 (1966).

[4]R. A. Reese, S. K. Sinha, T. O. Brun, and C. R. Tilford, Phys. Rev. A 3, 1688 (1971); V. J. Minikiewicz, T. A. Kitchens, F. P. Lipschultz, R. Nathans, and G. Shirane, Phys. Rev. 174, 267 (1968).

[5]T. A. Kitchens, G. Shirane, V. J. Minikiewicz, and E. B. Osgood, Phys. Rev. Lett. 29, 559 (1972).

[6]D. L. Price, J. M. Carpenter, C. A. Pelizzari, S. K. Sinha, I. Breshof, and G. E. Ostrowski, in Proceedings of the Sixth Meeting of the International Collaboration on Advanced Neutron Sources, Argonne National Laboratory, June 1982, Argonne National Laboratory Report No. ANL-82-80 (unpublished), pp. 207–215.

[7]L. H. Nosanow, Phys. Rev. 146, 120 (1966).

[8]P. A. Whitlock, D. M. Ceperley, G. V. Chester, and M. H. Kalos, Phys. Rev. B 19, 5598 (1979).

[9]D. L. Price and S. K. Sinha (to be published).

[10]N. Bernardes, Phys. Rev. 112, 1534 (1958).

[11]P. A. Whitlock, M. H. Kalos, G. V. Chester, and D. M. Ceperley, Phys. Rev. B 21, 999 (1980).