tains an extra factor of 2. Finally, the integration can be restricted to positive frequencies if one includes in the integrand the detailed-balance factor $1-e^{-\hbar\omega/kT}$.

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$$\vec{Q}\cdot\vec{u}_1=(3N)^{-1/2}\sum_{s=1}^{3N}\xi_s$$

and assume that the quantities ξ_s are statistically independent and $\langle\xi_s\rangle=0$. With no further assumptions it then follows that the terms in (3) are of order 1, N^{-1}, N^{-2}, $\cdots$, respectively, so that only the first term survives in the limit $N\rightarrow\infty$.

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Single-Particle Scattering from Solid ^{4}He†

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An investigation of the high-energy, large-momentum-transfer neutron scattering from the low-density hcp and bcc phases of ^{4}He has revealed scattered neutron groups with the same characteristics as those associated with single-particle scattering in liquid ^{4}He. The observed groups of this type are independent of the crystallographic phase and of direction within the accuracy of this investigation. Such neutron groups have been observed at energies as low as 1.5 times the maximum phonon energy in the bcc phase.

During an extension of research on inelastic neutron scattering from solid ^{4}He,[1,2] some observations of scattered neutron groups from the low-density phases of ^{4}He in the high-energy, high-momentum-transfer (high-ω, high-Q) regime have been made. The observations reported here indicate that in yet another facet the solid phases of helium are liquidlike. In the low-density solid phases of helium, single-particle excitations appear to lie at energies as low as they do in the liquid. These observations are the first of single-particle excitations in any solid produced with thermal neutrons.

Because, as we shall demonstrate later, the high-ω, high-Q neutron scattering in the solid phases is quite like that from the liquid, we will briefly review the present understanding of the liquid. There has been a great deal of activity in this area since Hohenberg and Platzman explained that high-energy neutron scattering experiments would be able, in principle at least, to test the theoretical results of Bose liquid theory.[3] For Q large enough such that the scattering atoms may be considered to act independently, the dynamic structure factor for the momentum transfer $\hbar Q$ and an energy transfer $\hbar\omega$ can be written as

$$S(Q,\omega)=\frac{\hbar}{N}\sum_{\vec{p}}n(\vec{p})\delta\left(\hbar\omega-\frac{\hbar^2Q^2}{2M_4}-\frac{\hbar^2\vec{Q}\cdot\vec{p}}{M_4}\right), \qquad (1)$$

where $n(\vec{p})$ is the number of particles with an initial momentum $\hbar\vec{p}$, and the δ function is due to the conservation of momentum and energy.[4] The scattering from a Bose liquid will be the superposition of the scattering from the fraction of atoms in the zero momentum state (the Bose condensate) and a symmetric broad background, also centered at $\hbar^2Q^2/2M_4$, whose width is determined by the Doppler term in the δ function. This "Doppler approximation" was discussed earlier by Sjölander[4]; he found that the full width due to the normal atoms would be roughly 2.7 times the geometric mean of the mean kinetic energy of the atoms, $\overline{E}_k$, and the mean recoil energy transferred to the atoms, $R=\hbar^2Q^2/2M_4$. Cowley and Woods[5] have investigated this problem for Q between 3 and 9 Å^{-1}. Harling[6] has extended the region to $Q=20$ Å^{-1}, and various workers have analyzed these experiments.[7] Gersch and Smith[8] have recently completed a detailed analysis and conclude that the data are consistent with not more than 3% Bose condensate; the neutron group profile is due almost completely to the more classical aspects of the liquid. More pertinent to this study, Sears[9] and Gersch, Rodriguez, and Smith[10] have considered the effects of He-He interactions and other corrections and agree that the peak energies will be reduced from the independent-particle value R and that the profile will be asymmetric with the larger wing extending to lower energies.

As Q is reduced from large values, the scattering can no longer be characterized by single-particle scattering and is better considered as multiphonon scattering.[4] The difficulty in making the largely semantic distinction between multiphonon and single-particle scattering is that the average energy of the observed scattering should be exactly R for either case. This is due to the first-moment sum rule

$$\hbar^2 \int S(Q,\omega)\omega\, d\omega = R, \qquad (2)$$

calculated first by Placzek,[11] and demonstrated for liquid ^{4}He by Cowley and Woods.[5] Therefore any distinction between multiphonon scattering and single-particle scattering must rely on the line shape. From Eq. (1) it is expected that the line shape of the single-particle scattering would be symmetric about R for any reasonable $n(\vec{p})$. The n-phonon scattering, on the other hand, depends on the nth convolution of the phonon density of states and thermal weighting factors. Only for quite low n does any of the residual character of the density of states appear in the n-phonon scattering; generally the multiphonon scattering is quite diffuse, but it is not symmetric about R for low Q.[4,12] Cowley and Woods[5] have argued that for $Q>3$ Å^{-1} the neutron groups are of single-particle scattering nature. We shall show that the characteristics of scattering from the low-density solid phases of He^4 are the same as those of the scattering from the liquid phase.

To our knowledge, single-particle-like dispersion—that is, a dispersion relation of the form $\hbar\omega=\hbar^2Q^2/2M$—has not been observed for neutron scattering from a solid. This can be understood from the fact that most solids have much larger Debye-Waller factors $\exp(-2W) \equiv \exp[-2B(Q/4\pi)^2]$ (i.e., much smaller B) and binding energies than helium. Ambegaokar, Conway, and Baym[13] have shown that, even accounting for interference effects from multiphonon scattering, the first-moment sum rule applied to the dynamic structure factor for *single*-phonon scattering, $S_I(Q,\omega)$, will be a fraction of the total Placzek sum rule, the fraction being equal to the Debye-Waller factor e^{-2W}. Thus e^{-2W} must be small before much scattered intensity is available for the multiphonon or single-particle scattering; but, conceptually, it might be expected that $\hbar^2Q^2/2M$ must be larger than the binding energy less a correction for thermal fluctuations before single-particle scattering becomes dominant. The possibility of channeling or mass-fluctuation-wave-type[14] excitations may considerably reduce this necessary energy. However, hindrance to single-particle motion due to the other atoms in the system may make the energy necessary for single-particle scattering much larger. For both liquid and solid helium, the binding energies are nearly equal and are less than 1 meV, suggesting that the Debye-Waller condition is more restrictive. (For comparison, in neon the binding energy is about 20 meV and the energy condition may be more restrictive.)

This investigation was carried out on the Brookhaven high-flux beam reactor using neutrons with an initial energy of 44 meV on a three-crystal spectrometer operated in the constant-Q mode. The neutron beam resolution was 2 meV or better in these measurements and no corrections for resolution have been applied in the data shown here; the widths of the neutron groups are nearly an order of magnitude larger. Such corrections, if applied to the data, typical examples of which are displayed in Fig. 1, would tend to make the line shapes even more asymmetrical but would leave the peak positions unaltered.

The bcc helium crystal was grown as described before in the cryostat used in our earlier investigations.[1,2] The hcp helium sample was grown

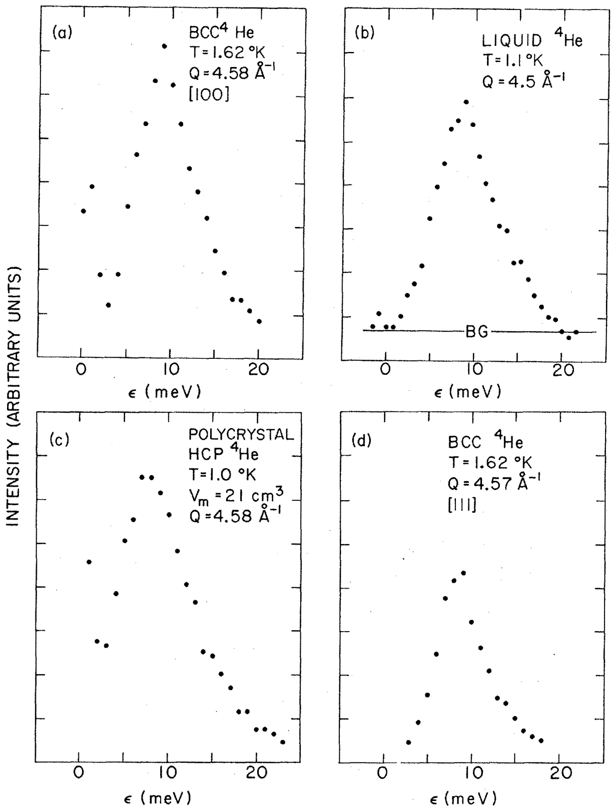

FIG. 1. Comparison of scattered neutron groups from the liquid and the low-density solid phases of ^{4}He. The background in these cases is negligible. The counting time in (d) was $\frac{1}{2}$ that used for (a) and (c). The data for the liquid phase were taken from the work of Cowley and Woods (Ref. 5). The liquid was at low pressure and at low temperature (1.1°K), and the data were taken on a three-crystal spectrometer with a final neutron energy of 62 meV.

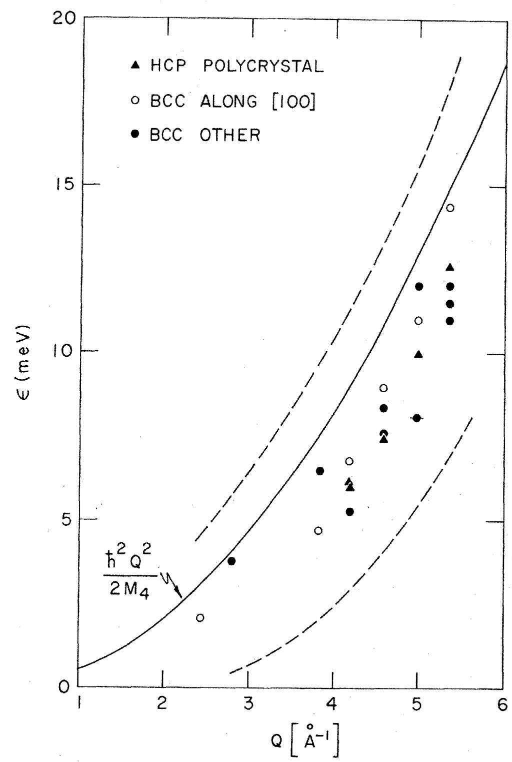

FIG. 2. Energy versus transferred momentum for neutron groups scattered from the low-density solid phases of ^{4}He. Solid line, expected relationship for single-particle scattering; dashed lines, approximate positions of the intensities equal to $\frac{1}{2}$ the peak of the observed neutron groups.

"through" the bcc phase by holding the cell pressure at 27.7 atm, establishing some liquid at the top of the cell and cooling the bottom of the cell to ~1.56°K, well below the hcp-bcc transition. When the cell top cooled from the bcc-liquid transition temperature the mass input indicated a molar volume of 20.8 cm^3. This molar volume is outside the bcc phase, assuring us of an hcp sample.[15] Bragg scattering verified the phase to be hcp and that the sample was of several crystallites, but not many crystallites.

Some typical neutron groups scattered from these samples are shown in Fig. 1. We have chosen to present data near $Q = 4.5$ Å^{-1} to facilitate comparison with the liquid scattering data collected in a similar way by Cowley and Woods.[5] These data are shown in part (b) of this figure. Quite clearly, the characteristics of the scattering observed in the liquid and solid phases are nearly identical. The figure also clearly demonstrates that the scattering appears to be the same for the bcc phase in the [100] and [111] directions. Measurements for other directions confirm that the scattering is isotropic. Data for the hcp sample composed of several crystals were also isotropic and, as seen from Fig. 1, of the same character as the bcc and the liquid data for the same momentum transfer Q.

The dispersion relation for the data collected for both phases is shown in Fig. 2 along with the expected single-particle relation, $R = \hbar^2 Q^2/2M_4$. The dashed lines are the approximate energies where the observed neutron groups are $\frac{1}{2}$ as intense as they are at peak intensity. The effective mass for the least-squares fit to the peak intensities is $M^* = 1.27M_4$ while that for the mean of the half-height energies is $M^* = 1.25M_4$. However, the observed deviations from the single-particle relation are quite consistent with the liquid data and with the calculations by Sears[9] and by Gersch, Rodriguez, and Smith.[10] The asymmetry of the profiles is, however, of the opposite character to those calculated by Gersch, Rodriguez, and Smith, the larger wing extending to higher energies. The observed asymmetry appears to have the characteristics of multiphonon scattering.

The data taken for $Q<3$ Å^{-1} show the very weak single-phonon peaks since the Debye-Waller factor $e^{-2W}<0.10$ for $Q>2.4$. These groups also have more structure than those for $Q>3$ Å^{-1} as might be expected from low-order multiphonon scattering. For $Q\gtrsim 3$ Å^{-1} the neutron groups have the same character as the single-particle-like groups shown in Fig. 1 and lie as low as 5–7 meV, less than 50% more energy than the most energetic phonon seen in the bcc phase.[1]

Interestingly, the widths of these neutron groups are not inconsistent with the assumption that the single-particle lifetime is limited by a collision with a near neighbor. The uncertainty principle suggests that the linewidth expected for a single-particle excitation of energy E_s due to a collison after traveling a distance d is $2E_s/Qd$. If we assume as the shortest "free distance" the nearest-neighbor distance less the hard-core diameter, the linewidth at $E=13$ meV ($Q\sim 5$ Å^{-1}) would be 5.2 meV, slightly less than that observed after resolution corrections. Furthermore, the liquid linewidths are also consistent with this assumption. The Doppler-approximation width would be over twice this value, or somewhat greater than that observed. Although these preliminary data were not extensive enough to check the sum rule of Eq. (2), the integrations do suggest that the sum rule is complete; that is, there are no substantial contributions to the groups outside the distribution characterized in Fig. 1.

Finally, these results also suggest that previous experimental results on the phonon spectra for hcp ^{4}He for medium[16] and low[17] densities should be re-examined for effects of single-particle scattering.

In summary, this paper has presented even more direct evidence of the similarity of the low-density solid phase of ^{4}He with the liquid phase than other recent results.[18,19] The scattering from the hcp and bcc phases in the high-energy, high-momentum-transfer regime is isotropic and independent of crystallographic phase and is nearly identical to that in the liquid. For $Q>3$ Å^{-1}, the scattering appears to be consistent with that from single particles. This work constitutes the first observation of such scattering from a solid phase.

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