Conf-720833--6

July 1972

Single Particle Excitations in Solid Helium*

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High-energy high-momentum transfer neutron scattering experiments on the low density phases of solid helium-4 indicate the existence of single particle excitations. The characteristics of the neutron groups are found to be isotropic and independent of the crystallographic phase of the solid sample within the accuracy of this exploratory investigation. The scattered neutron groups appear to have the same properties as those from liquid helium-4, thus illustrating that in yet another property this quantum solid is liquid-like. The neutron groups obeying the single particle dispersion relation are well formed for energies as low as 1-1/2 times the most energetic phonon in the bcc phase. This work appears to be the first observation of single particle dispersion relation from a solid sample. Reasons for this and the effect of multiphonon scattering will be discussed.

* Work performed under the auspices of the U.S. Atomic Energy Commission.

† Supported in part by the Advanced Research Project Agency.

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RECEIVED BY TJC AUG 25 1972

During an extension of research on the inelastic neutron scattering from solid ^4He ,^{1,2} some observations of scattered neutron groups from the low density phases of ^4He 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 liquid-like; in the low density solid phases of helium single particle excitations appear to lie at energies as low as they do in the liquid. To our knowledge these observations are the first of single particle excitations in any solid produced with thermal neutrons.

Most low temperature physicists are familiar with the basis of the measurements since the technique is nearly the same as that used in measuring the fraction of ^4He atoms in the zero momentum state in superfluid helium.³ For large enough momentum transfer $\hbar Q$ such that scattering atoms may be considered to act independently, the scattering law for the energy transfer $\hbar\omega$ can be written as

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

where $n(\mathbf{p})$ are the number of particles with an initial momentum $\hbar\mathbf{p}$, and the delta function is due to the conservation of momentum and energy. This "Doppler approximation" was discussed earlier by Sjölander⁴; he found that the full width is roughly 2.7 times the geometric mean of the mean kinetic energy of the atoms $\bar{E}_k$ and the mean recoil energy transferred to the atoms, $R = \hbar^2 Q^2 / 2M_4$.

As Q is reduced from large values, the scattering can no longer be characterized as 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,

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

calculated first by Placzek,⁵ and demonstrated for liquid ^4He by Cowley and Woods.⁶ 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 $n(p)$. The n -phonon scattering, on the other hand, depends on the n th 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 Cowley and Woods⁶ have argued that for $Q > 3 \text{ \AA}^{-1}$ the neutron groups are of single particle scattering nature. Our measurements, taken in the same momentum transfer range used by Cowley and Woods, show that the characteristics of scattering from the low density solid phases of ^4He 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 $\hbar\omega = \hbar^2 Q^2 / 2M$, 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 Baym⁷ 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 $\hbar^2 Q^2 / 2M$ 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 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 larger. An uncertainty principle argument shows the width of the observed neutron groups is consistent with the single particle state existing only until collision with a near neighbor.

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 than the resolution width. Such corrections, if applied to the data, 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 "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 $\sim 1.56^{\circ}\text{K}$, 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.⁸ Bragg scattering verified the phase to be hcp and that the sample was comprised 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 $Q = 4.5 \text{ \AA}^{-1}$ to facilitate comparison with the liquid scattering data collected in a similar way by Cowley and Woods.⁶ These data are shown in part B of this figure. Quite clearly, the characteristics of the scattering observed in the liquid and in the 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 1/2 as intense as they are at peak intensity. The effective mass for the least-squares fit to the peak intensities is $M^* = 1.27 M_4$.

However, such deviations from the single-particle relation are quite consistent with the liquid data.

The data taken for $Q < 3 \text{ \AA}^{-1}$ show 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 $Q > 3 \text{ \AA}^{-1}$ as might be expected from low order multiphonon scattering. For $Q \gtrsim 3 \text{ \AA}^{-1}$ 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, about 1.5 times the energy of the most energetic phonon seen in the bcc phase.¹ The Doppler approximation width is 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. These results also suggest that previous experimental results on the phonon spectra for hcp ^4He for medium and low densities⁹ should be reexamined for effects of single particle scattering.

In summary, we have presented yet more evidence of the similarity of the low density solid phases of ^4He with the liquid phase.¹⁰ 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 $Q > 3 \text{ \AA}^{-1}$, the scattering appears to be consistent with that from single particle excitations. This work constitutes the first observation of such scattering from a solid phase.

We are grateful to M. Rosso and T. Oversluizen for invaluable technical assistance and for many beneficial discussions with our colleagues, in particular J. D. Axe, M. Blume, R. A. Guyer, N. R. Werthamer, V. Korenman, and R. Prange.

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FIGURE CAPTIONS

- Fig. 1 Comparison of scattered neutron groups from the liquid and the low density solid phases of ^4He . The data for the solid phases were obtained using a three-crystal spectrometer with 44 MeV incident neutrons from the Brookhaven High Flux Beam Reactor. The background is negligible. The counting time in D was one-half that used for A and C. The data for the liquid phase were taken from the work of Cowley and Woods.⁶ The liquid was at low pressure and at low temperature, 1.1⁰K, and the data were taken on a three-crystal spectrometer with a final neutron energy of 62 MeV.
- Fig. 2 The energy vs. transferred momentum relationship for the neutron groups scattered from the low density solid phases of ^4He . The solid line corresponds to the expected relationship for single particle scattering. The dashed lines show the approximate position of the intensities equal to one-half the peak of the observed neutron groups.

FIGURE 1

FIGURE 2