PHONON SPECTRA OF fcc ^{4}He*

J.G. TRAYLOR, C. STASSIS, R.A. REESE**, S.K. SINHA
Institute for Atomic Research and Department of Physics,
Iowa State University, Ames, Iowa,
United States of America

Abstract

PHONON SPECTRA OF fcc ^{4}He.

Inelastic neutron scattering techniques have been used to investigate the dynamical properties of the fcc phase of ^{4}He. The phonon spectrum, at a molar volume of 11.7 cm^3, was determined along the [100] and [110] symmetry directions. The experimental data are in good agreement with the results of the anharmonic perturbation theory based on the self-consistent harmonic approximation.

1. INTRODUCTION

In recent years there has been a considerable amount of theoretical [1,2] and experimental [3,4] work on the lattice dynamics of quantum crystals. The classic approach of Born and von Kármán [5] is not applicable to a quantum crystal since the amplitude of the vibrational motion of the nuclei can be a substantial part of the interatomic distance. As a consequence of the large vibrational amplitudes, the atoms experience significant anharmonic forces, and the motions of immediately neighboring atoms become correlated to a high degree.

The self-consistent harmonic approximation [1] (SHA) and the anharmonic perturbation theory based on the SHA are now well established in the treatment of lattice dynamics of rare-gas solids [6]. The starting point of the SHA is to assume that the low-lying excitations of the system are phonons and hence that the ground state wave function may be written as a product of harmonic oscillator wave functions. The phonon frequencies are then calculated by averaging the second derivatives of the potential energy over the ground state of the system. Since the ground state wave function itself depends on the phonon frequencies, the procedure is repeated until self-consistency is achieved. This procedure is not directly applicable for the quantum solids where the potential produces a very strong repulsion for small atomic separations as the electronic cores of the atoms start to overlap. The problem is particularly critical in solid He with its very large zero-point vibrational amplitudes. The ground state crystal wave function must therefore be modified so that the probability is sharply diminished for the configurations of high repulsive potential energy. This is usually achieved by multiplying the ground state wave function by a function of the Jastrow form. The frequencies of the phonons are then obtained by the procedure outlined above.

---

* Work was performed in the Ames Laboratory of the US Atomic Energy Commission. Contribution No. 3188.

** Now with Union Carbide Corporation, Tarrytown, New York, USA.

Inelastic neutron scattering experiments [3,4] on hcp He^4 showed that the low-lying excitations were, indeed, phonons with frequencies in agreement with the predictions of SHA theory to within 30%. However, the volume dependence of the phonon frequencies predicted by the SHA was found to be incorrect. In addition, the neutron groups observed in hcp He^4 showed considerable line broadening, particularly for the higher frequency modes, and their intensities were not consistent with the one phonon neutron scattering cross-section formula for harmonic crystals. These observations indicated the need to go beyond the SHA theory to account for the dynamical properties of the quantum crystals.

Recent theoretical calculations have taken into account higher order effects using anharmonic perturbation theory based on the SHA. The effect of these higher order terms is to shift the phonon frequencies and to introduce finite lifetime for the decay of a phonon into other phonons. Because of the simpler structure of cubic crystals, these calculations have been performed only for the cubic quantum solids. The need for a direct comparison between these theories and experiment motivated the present inelastic neutron scattering experiment on fcc He^4.

2. EXPERIMENTAL

The single crystals used in the present experiment were grown in a .05 in. wall stainless steel bomb, 2 in. long and .375 in. in diameter. The bomb was in thermal contact with a helium bath and heater coils attached to the top and bottom of the bomb were used to control its temperature to within .25°K in the vicinity of 16°K. The bomb was connected to the pressurizer by a stainless steel capillary enclosed in an exchange gas chamber which was immersed in the He bath. The exchange gas chamber was evacuated and the capillary kept open by means of heater coils enabling the bomb to be pressurized with He^4 gas to a pressure of 1.36 kb. The heaters on the bomb were then switched off so that the temperature of the bomb would decrease to below 17°K, the solidification point of He^4 at this pressure. After solidification the bomb was sealed by switching off the capillary heaters and introducing He gas in the exchange gas chamber. Large crystals were grown by subsequently melting the solid in the bomb and then slowly cooling down below the melting point. Sometimes the size of the crystal could be increased by annealing. Since under a pressure of 1.36 kb the fcc phase of solid He exists in the range of 15°-17°K, the temperature of the crystal was controlled to within .25°K at 15.5°K.

The measurements were performed using a triple-axis spectrometer at the Ames Laboratory Research Reactor. The cryostat containing the crystal was mounted on a two circle goniometer which was used to search for and orient the crystals by standard neutron diffraction techniques. The data presented in the present communication were collected with the crystal oriented so that the scattering vector $\vec{Q}$ is in the (001) plane of the reciprocal lattice. The lattice constant of the crystals used in the present experiment was determined to be 4.28 Å corresponding to a molar volume of 11.72 cm^3. For intensity reasons, only a restricted region of reciprocal space could be used in the measurements. This is due to the fact that the large vibrational amplitude of the He^4 nuclei causes the Debye-Waller factor e^{-2W} to decrease rapidly with increasing scattering vector. Evidently the most favorable region to perform the measurements is in the vicinity of the maximum of the factor Q^2e^{-2W} (Fig. 1) appearing in the one-phonon neutron scattering cross-section. The measurements of the dispersion curves were made using the constant-Q method with a fixed incident neutron energy of 20 MeV.

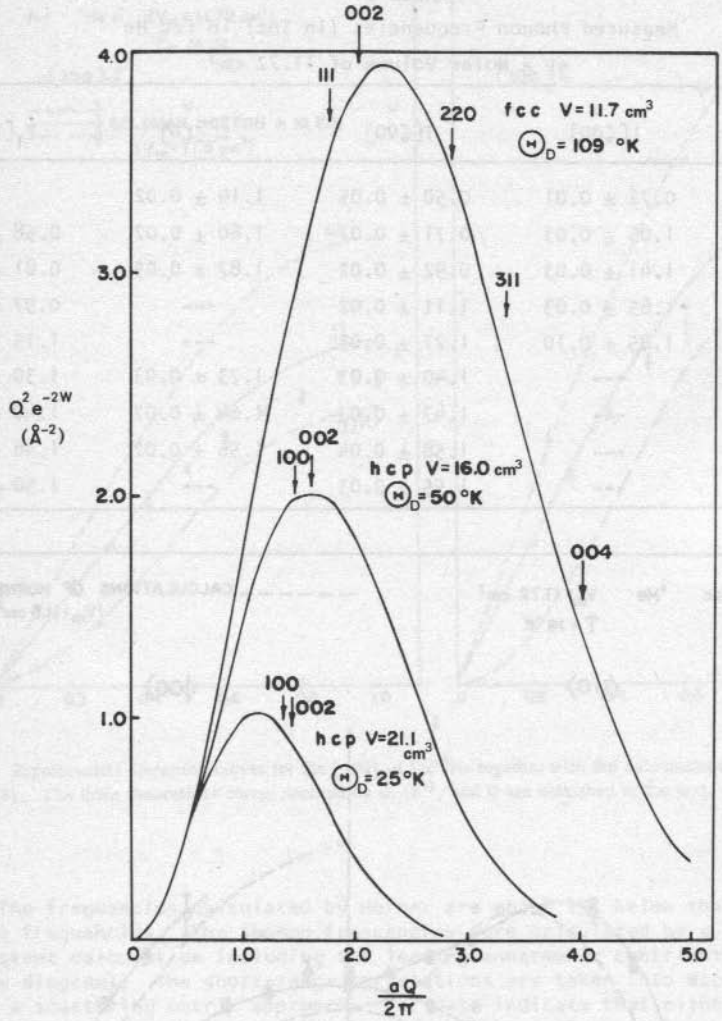

FIG.1. Plot of the function Q^2e^{-2W} vs Q in units of $2\pi/a$. The position of several reciprocal lattice points are indicated by arrows.

3. EXPERIMENTAL RESULTS AND DISCUSSION

Well-defined neutron groups were observed for the T[100], the T_1[110] and the lower frequency portion of the L[100] and L[110] branches. At higher frequencies, however, well-defined phonon peaks could not be observed. The measured phonon frequencies, defined as the mean positions of the observed neutron groups, are given in Table I and the experimental dispersion curves are plotted in Fig. 2. The experimental results are compared with the theoretical calculations of Horner [7] in Fig. 2 and with those of Goldman et al. [8] in Fig. 3. It is seen that the agreement between both these theories and the experimental data is reasonable.

TABLE I.

Measured Phonon Frequencies (in Thz) in fcc He^4 at a Molar Volume of 11.72 cm^3

$\zeta = q/q_{max}$	ν L[ζ00]	ν T[ζ00]	ν L[ζζ0]	ν T_1[ζζ0]
0.20	0.72 ± 0.01	0.50 ± 0.05	1.14 ± 0.02	---
0.30	1.06 ± 0.03	0.71 ± 0.01	1.60 ± 0.02	0.58 ± 0.01
0.40	1.41 ± 0.03	0.92 ± 0.02	1.87 ± 0.05	0.81 ± 0.02
0.50	1.65 ± 0.03	1.11 ± 0.02	---	0.97 ± 0.02
0.60	1.85 ± 0.10	1.27 ± 0.03	---	1.15 ± 0.04
0.70	---	1.40 ± 0.03	1.73 ± 0.03	1.30 ± 0.03
0.80	---	1.47 ± 0.03	1.64 ± 0.02	1.40 ± 0.03
0.90	---	1.58 ± 0.04	1.56 ± 0.02	1.46 ± 0.03
1.00	---	1.55 ± 0.03	---	1.50 ± 0.03

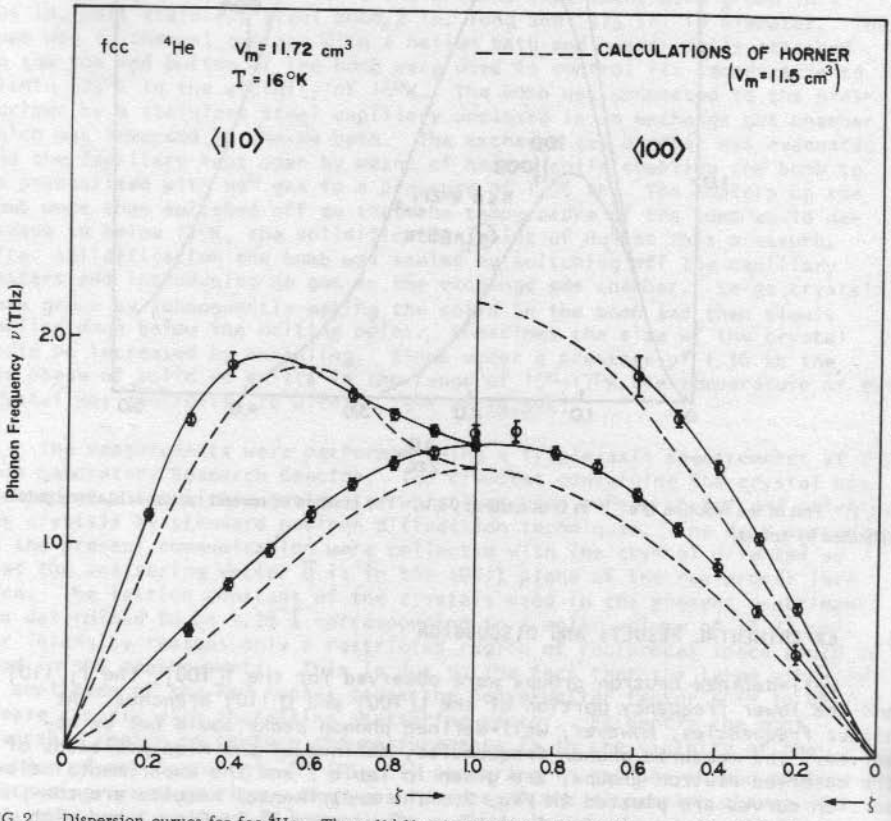

FIG. 2. Dispersion curves for fcc ^{4}He. The solid line is simply a guide for the eye. The dashed curve represents the calculations of Horner [7].

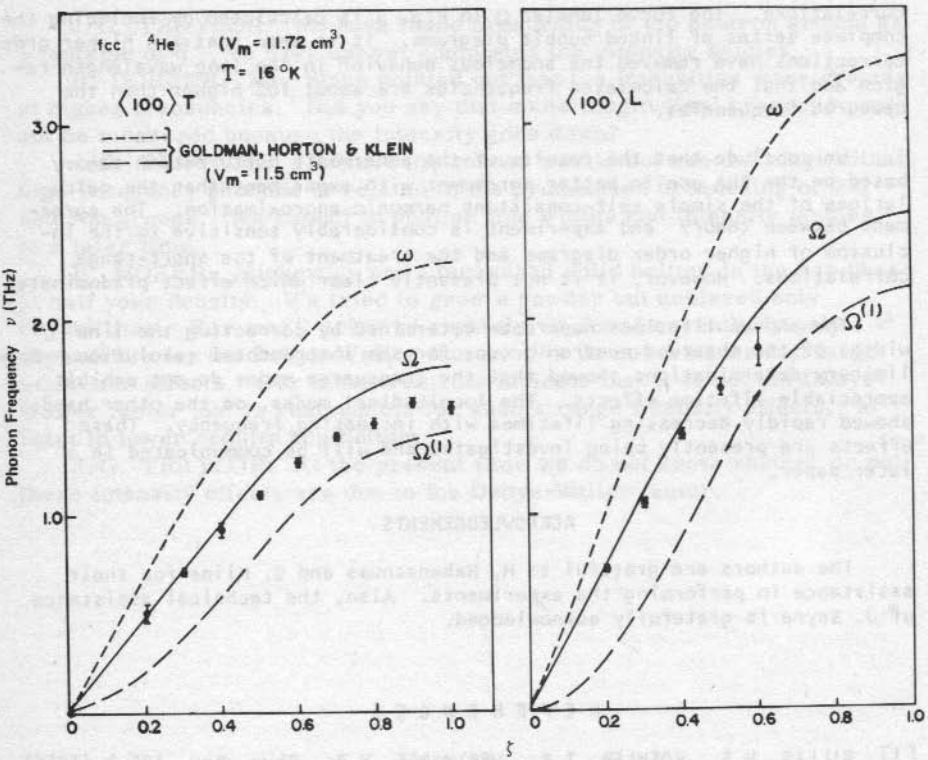

FIG.3. Experimental dispersion curves for the [100] of fcc ^{4}He together with the calculations of Goldman et al. [8]. The three theoretical curves denoted by ω, $\Omega^{(1)}$, and Ω are described in the text.

The frequencies calculated by Horner are about 15% below the experimental frequencies. The phonon frequencies were calculated by a self-consistent calculation including the leading anharmonic contribution (single bubble diagram). The short-range correlations are taken into account by using a scattering matrix approach. Our data indicate that either Horner's treatment overestimates the softening of the phonon frequencies by the short-range correlations or that the contribution of higher order diagrams is of the order of 15%.

The calculations of Goldman et al. [8] were performed at a molar volume of 11.5 cm^3 for the [100] direction only. In these calculations the short-range correlations were approximated by cutting off the hard core of the Lennard-Jones potential at a radius determined by a variational calculation. The ω curve in Fig. 3 represents their SHA calculations. The $\Omega^{(1)}$ curve includes the leading anharmonic contribution (single bubble diagram), and it may be seen that it exhibits anomalous upward curvature at long wavelengths. Although the present data do not extend to q values smaller than 0.2 ($2\pi/a$), the data extrapolate smoothly to a straight line through the origin; thus, there seems to be no experimental evidence for the above anomalous behavior. It should be noted that the $\Omega^{(1)}$ curve corresponds to Horner's calculations apart from the different treatment of the short-range

correlations. The curve labeled Ω in Fig. 3 is calculated by including the complete series of linked bubble diagrams. It is seen that the higher order corrections have removed the anomalous behavior in the long wavelength region and that the calculated frequencies are about 10% higher than the measured frequencies.

We conclude that the results of the anharmonic perturbation theory based on the SHA are in better agreement with experiment than the calculations of the simple self-consistent harmonic approximation. The agreement between theory and experiment is considerably sensitive to the inclusion of higher order diagrams and the treatment of the short-range correlations. However, it is not presently clear which effect predominates.

The phonon lifetimes have been determined by correcting the line widths of the observed neutron groups for the instrumental resolution. Preliminary determinations showed that the transverse modes do not exhibit appreciable lifetime effects. The longitudinal modes, on the other hand, showed rapidly decreasing lifetimes with increasing frequency. These effects are presently being investigated and will be communicated in a later paper.

ACKNOWLEDGEMENTS

The authors are grateful to M. Habenschuss and G. Kline for their assistance in performing the experiments. Also, the technical assistance of J. Sayre is gratefully acknowledged.

REFERENCES

[1] GILLIS, N.S., KOEHLER, T.R., WERTHAMER, N.R., Phys. Rev. 175 3 (1968) 1110. See also KOEHLER, T.R., Phys. Rev. Lett. 165 (1968) 942.

[2] WERTHAMER, N.R., Am. Jnl. Phys. 37 (1969) 763, and references therein.

[3] MINKIEWICZ, V.J., KITCHENS, T.A., LIPSCHULTZ, F.P., NATHANS, R., SHIRANE, G., Phys. Rev. 174 (1968) 267.

[4] REESE, R.A., SINHA, S.K., BRUN, T.O., TILFORD, C.R., Phys. Rev. A3 5 (1971) 1688.

[5] BORN, M., HUANG, K., Dynamical Theory of Crystal Lattices, Oxford Univ. Press, New York (1954).

[6] KOEHLER, T.R., Phys. Rev. Lett. 22 (1969) 777.

[7] HORNER, H., Solid State Comm. 9 (1971) 79.

[8] GOLDMAN, V.V., HORTON, G.K., KLEIN, M.L., Phys. Rev. Lett. 24 25 (1970) 1424.

DISCUSSION

B. DORNER: I think I remember hearing from Mr. S. Sinha that you find an unusual dependence of intensities on the phonon wave vector $\vec{q}$. Could you comment on this?