Lattice dynamics of fcc ^{4}He†

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The lattice dynamics of solid fcc ^{4}He, at a molar volume of 11.7 cm^3, has been investigated using inelastic-neutron-scattering techniques. Well-defined phonon peaks, whose widths could be accounted for by the instrumental resolution, were observed in the vicinity of the zone center; however, these phonon peaks, when taken on opposite sides of a reciprocal-lattice point, exhibited intensity differences which could not be accounted for by conventional harmonic theory. These observations indicate the presence of interference terms between one-phonon and multiphonon scattering. From approximately halfway to the zone boundary the phonon peaks of the $L[100]$, $T_2[111]$, and $L[111]$ branches exhibited appreciable broadening which was particularly pronounced in the vicinity of the point X of the Brillouin zone. Close to the zone boundary these phonons were found to be asymmetric with a rather long tail on the high-energy side. These observations are in qualitative agreement with the theoretical calculations of the scattering function. The measured dispersion curves, along the [100], [110], and [111] symmetry directions, were found to be in good agreement with Horner's theory. The latter theory is found to be in more satisfactory agreement with the experimental observations than the cutoff theories used in the heavier rare-gas solids.

I. INTRODUCTION

In recent years there has been a considerable amount of theoretical work on the lattice dynamics of quantum crystals.[1] The starting point of all theories of the quantum crystals is some form of the self-consistent-phonon (SCP) theory of lattice dynamics, which has been successfully applied to moderately anharmonic solids like the heavier-rare-gas crystals.

In the SCP theory one assumes that the ground state of the system can be represented by a collection of simple harmonic oscillators and variational or perturbation methods are employed for obtaining their frequencies in a self-consistent manner.[1a,1c,1g] However, for a quantum crystal, this simple ground-state wave function is a very poor approximation, because the large vibrational amplitudes in these crystals cause the atoms to experience large anharmonic forces due to the repulsive hard core of the interatomic potential. The ground-state wave function of the system must, therefore, be modified to include the short-range correlations induced between particles by the hard core of the interatomic potential. This can be most simply achieved by multiplying the ground state by a Nosanow-Jastrow function[2] which goes rapidly to zero as the interatomic distance becomes comparable to the hard-core diameter of the interatomic potential.

The hard-core problem has also been approached by the T-matrix methods, familiar from nuclear physics.[3] Since these methods were devised to cope with the hard-core problem in nuclear matter, they are equally appropriate for the solid-state problem. These methods were first applied[4] to the evaluation of the T matrix for the single-particle wave functions introduced by Nosanow. Since correlated wave functions are essential to treat the dynamics of quantum crystals, the method has been generalized by Glyde and Khanna[5] to consider the important case of correlated Gaussian functions. A third theoretical approach to the lattice dynamics of the quantum crystals has been developed by Horner and others.[1b,1c,1f,1h] The advantage of Horner's theory over the other methods is that correlations between the particles are introduced from the beginning so that the hard-core interactions are treated on the same footing as any other aspect of the problem. Clearly, the phonon spectra, measured in inelastic-neutron-scattering experiments, provide a sensitive test of the theory.

In a neutron-scattering experiment information regarding the dynamics of a solid is obtained by measuring the scattering function $S(\vec{Q},\omega)$ in terms of the momentum $\hbar\vec{Q}$ and energy $\hbar\omega$ transferred to the crystal. If a single phonon is excited by the incoming neutron beam the scattering function has a strong maximum at a frequency ω equal to the energy of the excited phonon. The neutrons may also excite multiples of phonons, particularly at large momentum transfers. This multiphonon contribution to the scattering function usually occurs at higher energy transfers, varies slowly with frequency, and in general can be separated from the scattering by single phonons. Thus, in most cases of interest the experimental results can be compared directly with the calculated one-phonon scattering function.

The situation is more complicated in the case of crystals with strong anharmonic interactions like solid ^{4}He. In this case a single phonon excited by the incoming neutron may decay, through anharmonic interactions, into several phonons. Also several phonons excited by the incident neutron may recombine into a single phonon in an intermediate state of the multiphonon scattering process. As a consequence, there is interference between single phonon and multiphonon scattering[1f,6] and the scattering function reads

$$S(\vec{Q},\omega)=S_1+S_{\rm int}+S_{\rm mult}=S_p+S_{\rm mult}\ ,$$

where S_1 and $S_{\rm mult}$ are the one-phonon and multiphonon contribution, respectively, $S_{\rm int}$ is the interference term between one-phonon and multiphonon scattering, and $S_p(\vec{Q},\omega)=S_1+S_{\rm int}$. Detailed calculations of $S(\vec{Q},\omega)$ have been performed and can be compared with the experiments.

Early inelastic-neutron-scattering experiments[7,8] on hcp ^{4}He demonstrated that the low-lying excitations were phononlike with frequencies in agreement (to within 30%) with the predictions of the self-consistent harmonic approximation (SHA). The phonon peaks observed[7,8] in hcp ^{4}He showed considerable line broadening, particularly at high frequencies, 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. However, detailed theoretical calculations based on the SCP theory, modified to take into account the hard-core part of the interatomic potential, have been performed only for the cubic quantum solids. The need for a direct comparison between these theories and experiment motivated inelastic scattering studies of the bcc[9,10] and fcc[11] phases of solid ^{4}He.

In this work inelastic-neutron-scattering techniques have been used to investigate the dynamical properties of the fcc phase of ^{4}He. The excitation spectrum at a molar volume of 11.7 cm^3, was determined along the [100], [110], and [111] symmetry directions. Preliminary results, obtained using a constant-incident-energy (20-meV) triple-axis spectrometer were reported in an earlier communication.[11] More recently, we performed a complete set of measurements using a variable-incident-energy instrument. In the present paper we present a complete report of these experiments.

II. EXPERIMENTAL

The apparatus used in the present experiment consisted of a He cryostat modified so that it could be connected to a mechanical pressurizer. The sample chamber was a 0.05-in. wall stainless-steel bomb, 2 in. long and 0.375 in. in diameter. The bomb was in thermal contact with the helium bath and was connected to the pressurizer by a stainless-steel capillary, enclosed in an exchange gas chamber immersed in the He bath. Heater coils and appropriate thermocouples were attached to the top and bottom of the bomb and capillary. During the pressurizing process the exchange gas chamber was evacuated and the bomb and capillary heaters were kept on. The bomb was thus pressurized with high-purity He gas to a pressure of 1.36 kbar. The heaters on the bomb were then switched off and the He pressure was kept constant as the temperature of the sample chamber decreased below 17 °K, the solidification point of He at this pressure. After solidification the bomb was sealed by switching off the capillary heaters and introducing He gas in the exchange gas chamber. The high-pressure valve, connecting the capillary to the pressurizer, was then shut off and the cryostat was disconnected from the pressurizer.

The measurements were performed using the triple-axis spectrometers at the 5-MW Ames Laboratory Research Reactor. The cryostat containing the solid He was mounted on a two-circle goniometer which was used to search for and orient the crystals. Large single crystals (1–2 cm^3) were grown by melting the solid in the bomb and then slowly cooling down below the melting point. Since under a pressure of 1.36 kbar the fcc phase of solid He exists in the range of 15–17 °K the temperature of the crystal was controlled to within 0.25 °K at 15.5 °K. 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.

Crystals oriented so that the scattering plane coincides with the (100) or (1$\bar{1}$0) planes, were used to measure the phonon spectra in the [100], [100], and [111] symmetry directions. Two sets of measurements were performed using the constant-Q method. The first set of measurements[11] was taken using a constant incident neutron energy of 20 meV, and the second with variable incident energy and fixed scattered neutron energy of 13.6 meV. In the latter set of measurements pyrolitic graphite was used as a monochromator and analyzer and a pyrolitic-graphite filter was placed in the scattered beam to eliminate higher-order contamination. For intensity reasons all measurments were performed in the vicinity of the (1,1,1) and (0,0,2) reciprocal-lattice points where the maximum of the factor Q^2e^{-2W} appearing in the one-phonon neutron-scattering cross section, occurs. For larger scattering vectors this factor de-

creases rapidly as a result of the large vibrational amplitude of the He nuclei.

III. EXPERIMENTAL RESULTS AND DISCUSSION

Since in quantum crystals multiphonon and interference effects are of importance, the proper way of analyzing the experimental results is not by assigning peak positions and widths to the observed neutron groups but by convolution of the resolution function $R(Q,\omega)$ of the instrument with theoretical results for the scattering functions. However, such an analysis cannot be performed reliably in view of the rather large experimental uncertainties which are due mainly to the presently limited neutron intensities. In particular, it is very difficult to extract the multiphonon contribution to the scattering function from the background. In the following discussion we will adopt the conventional concepts of dispersion relation and one-phonon frequency in comparing the experimental results with numerical calculations.

In the present experiment the excitation spectrum was determined along the [100], [110], and [111] symmetry directions. The general features of the scattering function have been determined by measurements on a large number of crystals.

Well-defined phonon peaks, whose widths could be accounted for by the instrumental resolution, were observed in the vicinity of the zone center. However, these phonon peaks, when taken on opposite sides of a reciprocal-lattice point, exhibited intensity differences which could not be accounted for by conventional harmonic theory (see Fig. 1). These observations indicate the presence of interference terms between one-phonon and multiphonon scattering. Since the widths of these phonons are relatively small the interference terms cannot redistribute the intensity under the one-phonon scattering function and affect only the peak intensity. The leading interference term

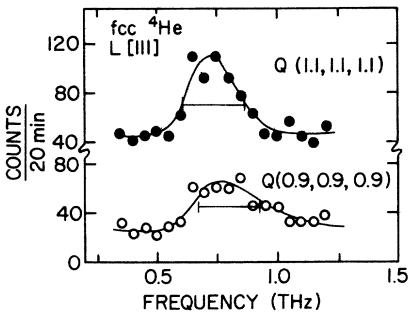

FIG. 1. Example of the difference in intensity observed for neutron groups taken at two equivalent positions in reciprocal space. The horizontal line is the instrumental resolution.

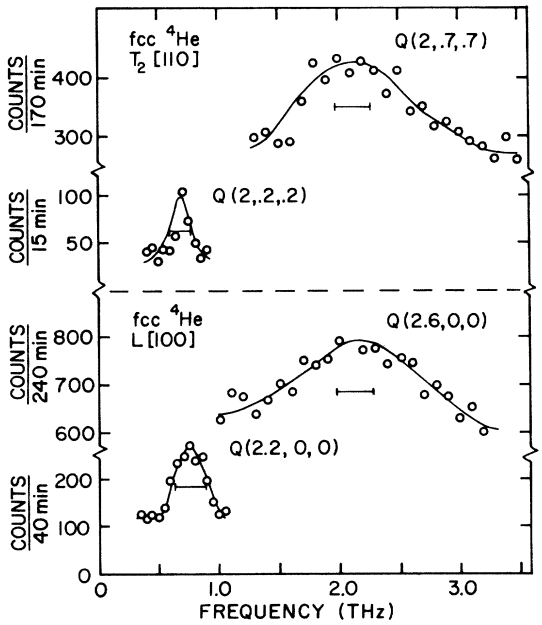

FIG. 2. Figure illustrates the broadening of the observed neutron groups in the vicinity of the point X of the Brillouin zone. Notice the rather long tail on the high-energy side of the phonon with $Q(2, 0.7, 0.7)$. The solid line is the instrumental resolution.

(one-phonon–two-phonon interference) is an odd function of the phonon wave vector and, therefore, introduces intensity differences between phonons with opposite wave vectors. In addition, multiphonon scattering masks the high-frequency part of $S_p(Q,\omega)$ whose leading term (one-phonon–two-phonon interference term) is negative[1c] for one of the phonons; as a result, the observed peak height of this phonon is enhanced since the negative high-frequency part of $S_p(Q,\omega)$ reduces the background. The interference effects, as expected are not as important as in the more anharmonic case of bcc He.[10]

From approximately halfway to the zone boundary, the phonon peaks in certain branches exhibited considerable broadening (Fig. 2). Close to the zone boundary these phonons were found to be asymmetric with a rather long tail on the high-energy side. The broadening of the phonon peaks was appreciable for the higher-lying $L[100]$, $T_2[100]$, and $L[111]$ branches and particularly pronounced in the vicinity of the point X of the Brillouin zone. The phonons of the other branches on the other hand, did not exhibit any appreciable broadening. These observations are in qualitative agreement with calculations of *one-phonon* widths using Horner's theory.[12] The intrinsic phonon widths were obtained by correcting the measured widths of the neutron groups for the instrumental

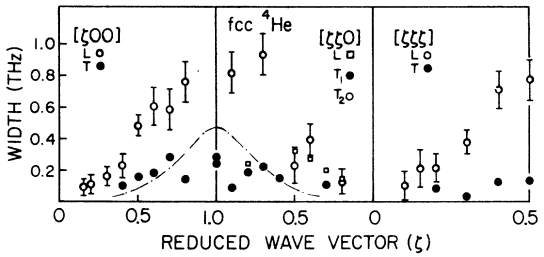

FIG. 3. Intrinsic phonon widths obtained by correcting the observed widths of the neutron groups for the instrumental resolution. The dashed curves are the calculated (Ref. 12) one-phonon widths for the L [100] and T_2[110] branches (see text).

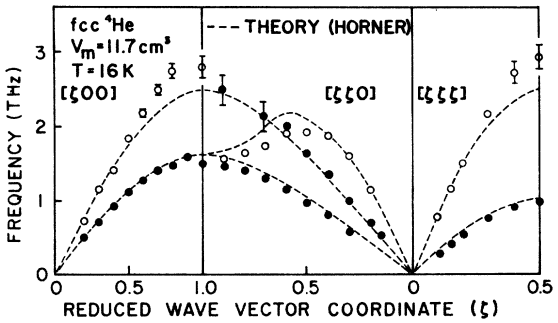

FIG. 4. Comparison of the experimental dispersion curves with the theoretical calculations of Horner (Ref. 12).

resolution. As a check on the accuracy of the resolution calculations, intrinsic widths of several phonons extracted from both constant-incident-energy and constant-scattered-energy scans were compared and were found to agree to within experimental error. The observed intrinsic phonon widths, plotted in Fig. 3, are approximately 50% higher than the *one-phonon* widths calculated by Horner.[12] This discrepancy, however, may be due to interference and multiphonon effects. As the zone boundary is approached in a direction in which there is significant energy broadening, $S_p(Q,\omega)$ merges with the increased multiphonon scattering on the high-energy side and the experimentally determined width is expected to be considerably larger than the one one-phonon width. No secondary peaks were observed on the high-energy side of the phonons close to the zone boundary as predicted by Horner's theory.[12,13] However, even if these structures were present[1c] they certainly could not be observed in the experiment because of the finite instrumental resolution.

For intensity reasons the measurements were performed at relatively large scattering vectors [$Q \gtrsim 1.5(2\pi/a)$]. For such scattering vectors the multiphonon contribution has been estimated to be more than 50%. Thus, the measured intensities cannot be reliable for a qualitative comparison with the apparent oscillatory behavior of the Debye-Waller factor predicted by the theory. However, an intensity analysis, using the Ambegaokar-Conway-Bayne sum rule for a selection of rather well-defined phonons, indicated larger intensities than expected by conventional theory at $Q \simeq 2.2(2\pi/a)$. Certainly, high-resolution measurements like those performed by the Brookhaven group[10] on bcc He are needed for a detailed comparison with theory on this point.

The measured phonon frequencies for the [100], [110], and [111] phonon branches are given in Table I and the experimental dispersion curves are compared with the theoretical calculations of Horner[12] in Fig. 4. It can be seen that the theoretical calculations agree to within 10% with the experimental data and the agreement is particularly good at low frequencies. The theoretical calculations of Horner[12] are in better agreement with the experimental results then previous theoretical calculations by the same author using the T-matrix method (see Ref. 11). Note that the discrepancies between theory and experiment for the L[100]

TABLE I. Measured phonon frequencies (in THz) in fcc ^{4}He at a molar volume of 11.72 cm^3.

				ν (THz)			
$\zeta = q/q_{max}$	L [ζ00]	T[ζ00]	L [ζζ0]	T_1[ζζ0]	T_2[ζζ0]	L [ζζζ]	T[ζζζ]
0.10	···	···	···	···	···	0.77±0.01	0.30±0.02
0.15	0.53±0.02	···	···	···	0.54±0.02	···	0.43±0.03
0.20	0.72±0.01	0.50±0.05	1.14±0.02	···	0.71±0.03	1.46±0.02	0.56±0.03
0.30	1.07±0.03	0.71±0.01	1.60±0.02	0.58±0.01	1.02±0.02	2.14±0.03	0.78±0.04
0.40	1.41±0.03	0.92±0.02	1.87±0.07	0.81±0.02	1.39±0.05	2.70±0.10	0.92±0.03
0.50	1.84±0.05	1.11±0.02	1.90±0.04	0.97±0.02	1.65±0.05	2.94±0.16	0.97±0.04
0.60	2.18±0.05	1.27±0.03	1.80±0.04	1.15±0.04	2.01±0.05		
0.70	2.50±0.07	1.40±0.03	1.73±0.03	1.30±0.03	2.14±0.17		
0.80	2.74±0.10	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	2.48±0.17		
1.00	2.80±0.15	1.55±0.03	···	1.50±0.03			

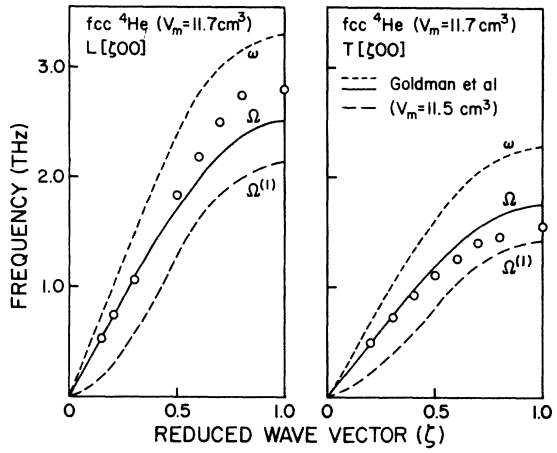

FIG. 5. Experimental dispersion curves along the [100] symmetry direction compared with the theoretical calculation of Goldman *et al.* (Ref. 14).

and $L[111]$ branches occur close to the zone boundary. In this region the experimentally determined frequencies may have been overestimated because the observed phonons are broadened and the high-energy side of their lines is certainly distorted by multiphonon effects. The rather remarkable agreement between the experiment and the theoretical calculations demonstrates that Horner's theory is a satisfactory approach to the problem of short-range particle correlations in the quantum solids.

It should be pointed out that Horner's calculations include only the usual cubic anharmonic term (single-bubble diagram). It is not presently clear whether the agreement between theory and experiment can be further improved by including higher-order anharmonic terms, in particular, higher-order bubble diagrams. These latter diagrams are of importance in theories[1b] which approximate the effects of short-range correlations by cutting off the hard core of the Lennard-Jones potential. Since theories using a cutoff have been used in interpreting the experimental results in heavier-rare-gas solids, it is instructive to compare these calculations with our experimental data on fcc ^{4}He. The calculations of Goldman *et al.*[14] performed at a molar volume of 11.5 cm^3 are compared with the experimental data for the $L[100]$ and $T[100]$ branches in Fig. 5. The ω curve in Fig. 5 represents their SHA calculations. The $\Omega^{(2)}$ curve includes the leading anharmonic contribution (single bubble diagram) and it may be seen that it exhibits anomalous upward curvature at long wavelengths. The curve labeled Ω has been calculated by summing the contributions of all linked bubble diagrams. It can be seen that the higher-order corrections have removed the anomalous behavior in the long wavelength region. However, the calculated dispersion curves do not agree as well with the experimental data as those obtained using Horner's theory; in particular note the discrepancy between the calculations and the experiment for the $T[100]$ branch for which the observed phonon groups were well-defined throughout the zone.

In summary, we find that the theories of quantum crystals as developed in the last decade, and in particular Horner's theory, account for the experimental observations of fcc ^{4}He. Interference and multiphonon effects provide a qualitative understanding, at this stage, of the observed general characterisitic of the scattering function and the calculated dispersion curves are, in view of the experimental uncertainties, in excellent agreement with the measurements.

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