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Lattice dynamics of hcp ^{4}He at high pressure

J. Eckert, W. Thomlinson, and G. Shirane
Brookhaven National Laboratory, Upton, New York 11973

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The neutron-inelastic-scattering technique was used to measure the phonon dispersion relations in two high-density crystals of hcp ^{4}He with molar volumes of 11.61 and 9.41 cm^3/mol. These densities are on the order of twice that of ^{4}He at 30 bars. The crystals were grown from the melt in the fcc phase, and cooled across the fcc-hcp phase transition. The observed phonon spectra show anharmonic effects much less prominent than those observed in earlier measurements at 21 cm^3/mol. In particular, multiphonon interference effects were not found to be very pronounced, while multiphonon scattering appeared to influence phonon line shapes strongly for large wave-vector and energy transfers. Elastic constants determined from the initial slopes of the dispersion curves were found to be in good agreement with those calculated by Goldman using the first-order self-consistent-phonon theory. The dependence on volume of the phonon energies is discussed with reference to the earlier studies at 21.1 and 16.0 cm^3/mol. No significant dispersion of mode Grüneisen parameters was found unlike the case of fcc He. Grüneisen parameters were found to depend on volume approximately in a manner given by Ahlers for the thermodynamic Grüneisen parameter.

I. INTRODUCTION

In two previous papers[1,2] we reported extensive neutron scattering studies of the lattice dynamics of fcc ^{4}He of high densities. Crystals with molar volumes between 9 and 10 cm^3/mol were studied over the range of temperatures at which the fcc phase is stable for those particular densities. Used in conjunction with the results of the only previous neutron scattering study[3] on fcc ^{4}He, at a density of 11.72 cm^3/mol, these data provide a description of both the quasiharmonic volume shifts (mode Grüneisen parameters) and the anharmonic temperature shifts of the phonon energies. The isochoric temperature shifts were found to be unexpectedly large, i.e., as much as 15% over a temperature range of about 16 K at a density of 9.03 cm^3/mol. Where comparisons were possible with studies[4-7] at the lowest densities of solid He, particularly those in the bcc phase,[6,7] the magnitudes of the anharmonic effects were found to be much reduced in the high-density crystals. However, the importance of multiphonon interference effects was clearly established in the high-density crystals, as well as that of multiphonon scattering for large energy and momentum transfers. Recent theoretical calculations prompted by these studies proved to give an excellent description of the phonon-dispersion relations[8,9] and good agreement with observed phonon line shapes.[9] However, the rather large isochoric temperature shifts of the phonon energies could only partially be accounted for by the calculations.[9] In view of these important results, it was considered to be of considerable interest to extend these studies to the hcp phase of solid He, where a much wider range of densities is experimentally accessible by the present methods.

The initial neutron scattering measurements[4,5] of phonon-dispersion relations in solid ^{4}He were performed on crystals in the hcp phase because of the fact that it is the stable phase at low pressures with the exception of the very small region of stability for the bcc phase. Most of the theoretical activity[10] concerned with the lattice dynamics of solid He, however, has been centered around experiments done in the bcc[6,7] and fcc[3] phases for reasons of the relative computational simplicity of the cubic structures. Thus, while refined calculations[10,11] using higher order self-consistent-phonon theories have been very successful in accounting for the observed lattice-dynamic response of the bcc and fcc crystals, the only calculation of hcp He phonon-dispersion relations, that of Gillis *et al.*[12] in the self-consistent-harmonic approximation, gives only fair agreement with experiment. In particular, this calculation shows a poor volume dependence of the phonon energies when applied to the two different experiments at 21.1 and 16 cm^3/mol. This difficulty is generally recognized to result mainly from the inflexibility of the methods used to treat short-range correlations.[10] In analogy with the case of the cubic crystals one would also expect the inclusion of cubic anharmonicities in the calculations to improve agreement with experiment significantly.

Comparison of the measured phonon energies in two previous experiments on hcp He at[4,7] 21.1 and[5] 16.0 cm^3/mol gave rise to some ambiguity concerning the magnitude of the volume shifts of the phonon energies, i.e., the mode Grüneisen parameters. Reese *et al.*[5] concluded that their phonon energies at 16.0 cm^3/mol were related to those of Minkiewicz *et al.*[4] by a factor of 1.91, approximately independent of branch and wave vector. More recent data on the LO[001] by Minkiewicz *et al.*[7] appeared to contradict this uniform scaling assumption. One would not *a priori* expect the mode Grüneisen parameters to be the same for all modes.

In this paper we discuss the lattice dynamics of two crystals of hcp He with densities of 9.43 and 11.62 cm^3/mol. Following presentation of the experimental data and phonon-dispersion relations the present results will be compared with the earlier measurements on hcp He at[4,7] 21.1 and[5] 16.0 cm^3/mol.

II. EXPERIMENTAL DETAILS

The He crystals used in the present experiment were grown isothermally directly from the melt at high pressures by a method described in earlier work.[1,13] Since at the densities of interest in the present experiment the fcc phase is stable[14] at temperatures just below the melting line, the crystals to be studied had to be cooled through the fcc-hcp phase transition. The first crystal was grown at 19.4 K and 1.68 kbar and was cooled directly into the hcp phase to 8.3 K at which point the search for a suitable crystallite was begun. The largest crystal found had a volume of about 0.3 cm^3 and was oriented in a (hhl) zone. It was later tilted into an $(h0l)$ zone for measurements of phonons along the [100] symmetry direction. The lattice parameters of this crystal were determined from the positions of several Bragg reflections to be $a = 3.011 \pm 0.002$ Å and $c = 4.908 \pm 0.002$ Å, corresponding to a molar volume of 11.61 cm^3/mol.

The second crystal used in the experiment was grown at 33.9 K and 4.1 kbar ($V = 9.43$ cm^3/mol) and was utilized for measurements of phonon-dispersion relations in the cubic phase.[2] Subsequently, it was cooled carefully into the hcp phase, such that the original cubic crystal with a $[01\bar{1}]$ orientation, transformed completely into the corresponding hcp crystal oriented in a $(h0l)$ zone. The lattice parameters of this crystal were found to be $a = 2.807 \pm 0.002$ Å and $c = 4.580 \pm 0.002$ Å, such that $c/a = 1.632$ and $V = 9.41$ cm^3/mol. The volume change across the hcp-fcc phase transition was therefore 0.02 ± 0.02 cm^3/mol. Both crystals had mosaics of about 30 min full width at half maximum (FWHM).

The measurements of the phonon dispersion relations were done on triple-axis spectrometers at the Brookhaven high-flux beam reactor in the constant-Q mode of operation using fixed incident energy of 13.7 or fixed final energies of 14.8 and 24 meV. Pyrolytic graphite filters were used to eliminate higher order contamination in the 13.7- and 14.8-meV beams. Monochromator and analyzer were pyrolytic graphite crystals, the former being cylindrically curved while the latter was flat. Horizontal beam collimations of 20- or 40-min FWHM were used depending on energy and resolution requirements.

III. RESULTS AND DISCUSSION

A. Phonon spectra

Representative phonon groups for the 9.41-cm^3/mol crystal are shown in Fig. 1. The figure at the top is typical of the data for acoustical modes at small reduced wave vectors used for the determination of the elastic constants. Shown in the middle of Fig. 1 is the zone-center optical mode, whose energy will be compared with Raman scattering measurements below. The figure at the bottom illustrates the difficulties in observing phonons at large $\vec{Q}$ and high energy transfers to be discussed later in this section. The solid lines represent a fit to the data by a Gaussian with a linearly energy-dependent background. The peak positions thus obtained were taken to be the experimental phonon energies and are given in Tables I and II for the 11.61-cm^3/mol crystal and Table III for the 9.41-cm^3/mol crystal. The data shown are not corrected for shifts due to the resolution of the neutron spectrometer since for their range of values of reduced wave vector and energy transfer the shifts can be expected to be small (less than 1% in most cases, and no more than 2%). The errors given in Tables I–III are statistical as given by the Gaussian fitting procedure. Inclusion of systematic errors such as the analyzer crystal setting and the uncertainty due to the omission of resolution corrections are estimated to approximately double the errors shown. For some phonons with closely overlapping peaks (principally the LA and LO [100] branches at high energy transfers) peak positions were determined graphically, and correspondingly large uncertainties assigned.

Phonon-dispersion relations representing the data of Tables I–III are shown in Figs. 2 and 3 for the 11.61-cm^3/mol crystals and in Fig. 4 for the 9.41-cm^3/mol crystal. The solid lines are simply guides to the eye. No attempt was made to observe the TO[100] mode which previous experiments[4,5] on hcp He were unable to detect. Transverse modes along [100] with polarization in the basal

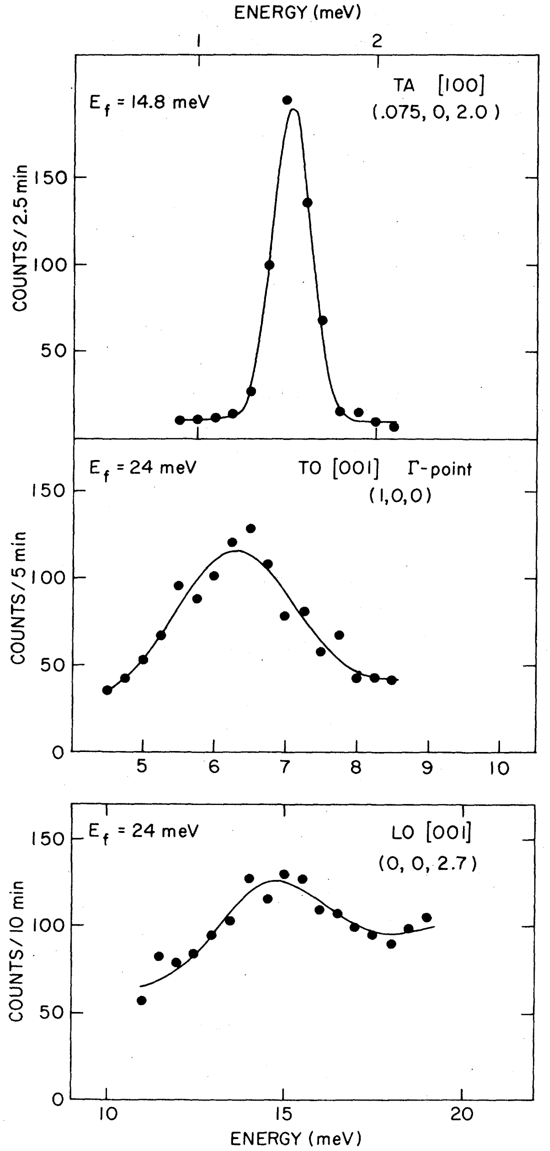

FIG. 1. Representative phonon scans for hcp ^{4}He at 9.41 cm^3/mol and 10 K. Solid lines are fitted Gaussians. Scans were taken with fixed final energies $E_f = 14.8$ and 24 meV.

plane could not be observed in the scattering zones used in the present experiment. No theoretical calculations at these densities are available at present to compare with the present data. Morley and Kliewer[15] calculated dispersion curves for hcp ^{3}He and ^{4}He for densities up to 10 cm^3/mol in the lowest-order self-consistent-harmonic theory without short-range correlations. When scaled to the densities used in the present experiment, their results overestimate the measured frequencies by

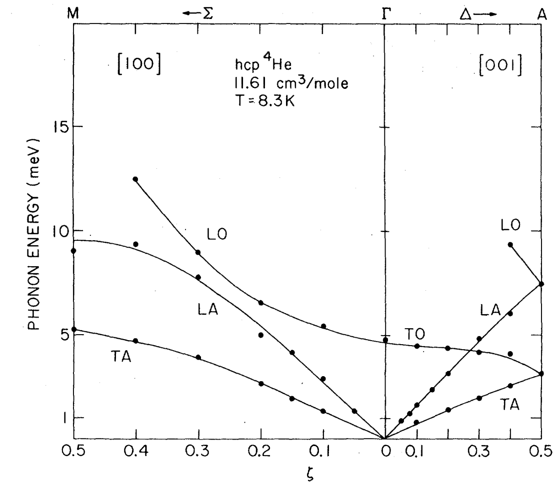

FIG. 2. Phonon-dispersion relations in the [100] and [001] directions for hcp ^{4}He at 11.61 cm^3/mol and 8.3 K. Solid lines are a guide to the eye only. Polarization of TA mode is perpendicular to the basal plane.

nearly 50%. Chell *et al.*[16] estimated the correction to Morley and Kliewer's calculation from the inclusion of cubic anharmonic terms to be of a similar magnitude. This has been shown to be a typical result for the self-consistent-harmonic approximation[10,17] (SCH). The addition of the first anharmonic cubic correction (SCH + C) is found to produce phonon energies of only slightly lower values than experimental ones,[9,17] while inclusion of all cubic anharmonic terms (SC1 in Ref. 17) results in frequencies slightly higher than experiment.[1,2,10,17] Both approaches, SCH + C and SC1,

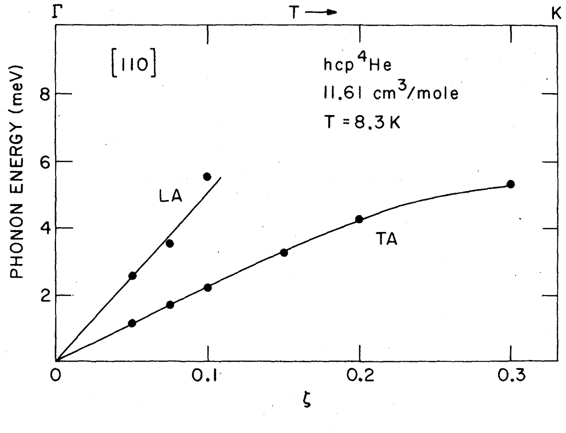

FIG. 3. Phonon-dispersion relations for the [110] direction of solid ^{4}He at 11.61 cm^3/mol and 8.3 K. Solid lines are a guide to the eye only.

TABLE I. Experimental phonon energies (meV) for hcp He at 11.61 cm^3/mol and 8.3 K. Errors given are statistical only. Overall errors are approximately twice as large.

		[001]		
ξ	TA	LA	TO	LO
0.00			4.80 ± 0.34	
0.05		0.926 ± 0.036		
0.075		1.26 ± 0.03		
0.10	0.821 ± 0.011	1.66 ± 0.02	4.48 ± 0.07	
0.15		2.39 ± 0.03		
0.20	1.44 ± 0.02	3.16 ± 0.03	4.41 ± 0.11	
0.30	2.01 ± 0.02	4.69 ± 0.03	4.22 ± 0.07	
0.40	2.58 ± 0.03	6.09 ± 0.04	4.10 ± 0.20	9.44 ± 0.48
0.50	3.15 ± 0.03	7.44 ± 0.11	3.15 ± 0.02	7.44 ± 0.11

TABLE II. Experimental phonon energies (meV) for hcp He at 11.61 cm^3/mol and 8.3 K. Errors given are statistical only. Overall errors are approximately twice as large. The polarization of the TA mode is perpendicular to the basal plane.

	[100]			[110]	
ξ	TA	LA	LO	TA	LA
0.05		1.38 ± 0.08		1.16 ± 0.01	2.57 ± 0.02
0.075				1.71 ± 0.02	3.54 ± 0.04
0.10	1.37 ± 0.03	2.94 ± 0.16	5.45 ± 0.05	2.24 ± 0.02	5.56 ± 0.36
0.15	1.97 ± 0.02			3.30 ± 0.03	
0.20	2.70 ± 0.05	5.01 ± 0.05	6.56 ± 0.06	4.29 ± 0.04	
0.30	3.97 ± 0.04	7.08 ± 0.2	8.9 ± 0.3	5.28 ± 0.08	
0.40	4.76 ± 0.05	9.3 ± 0.5	12.5 ± 0.5		
0.50	5.29 ± 0.06	9.09 ± 0.07			

TABLE III. Experimental phonon energies (meV) for hcp He at 9.41 cm^3/mol and 10 K. Errors given are statistical only. Overall errors are approximately twice as large.

		[100]			[001]		
ξ	TA	LA	LO	TA	LA	TO	LO
0.00			6.28 ± 0.07			6.28 ± 0.07	
0.05		2.24 ± 0.02			1.28 ± 0.02		
0.075	1.52 ± 0.02	3.29 ± 0.03			1.85 ± 0.03		
0.10	1.99 ± 0.02	4.27 ± 0.04	7.23 ± 0.07	1.09 ± 0.03	2.43 ± 0.03	6.47 ± 0.05	
0.125		5.44 ± 0.04					
0.15	2.95 ± 0.03	6.23 ± 0.06		1.59 ± 0.02	3.73 ± 0.03		
0.20	3.87 ± 0.03	7.42 ± 0.20	10.09 ± 0.10	2.07 ± 0.02	4.66 ± 0.08	6.25 ± 0.04	15.52 ± 0.32
0.30	5.54 ± 0.05	11.35 ± 0.05	12.73 ± 0.17	3.06 ± 0.04	7.44 ± 0.06	6.05 ± 0.05	14.59 ± 0.18
0.40	7.20 ± 0.06	14.25 ± 0.25	16.2 ± 0.5	4.06 ± 0.05	9.44 ± 0.04	5.16 ± 0.06	12.71 ± 0.19
0.50	8.32 ± 0.07			4.61 ± 0.03	11.48 ± 0.14		11.48 ± 0.14

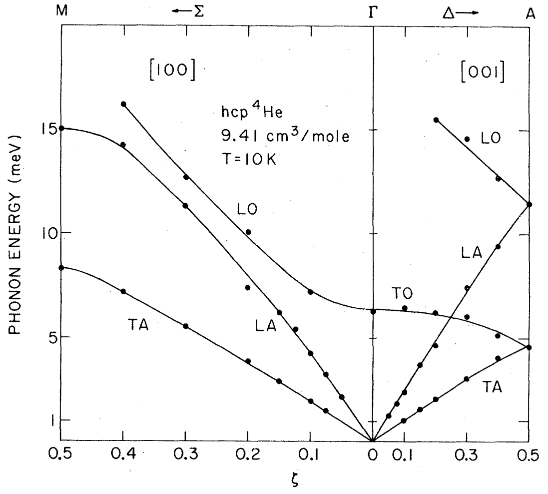

FIG. 4. Phonon-dispersion relations for hcp ^{4}He at 9.41 cm^3/mol and 10 K. Solid lines are a guide to the eye only.

without short-range correlations yield good agreement[1,2,9] with experiment for high-density fcc He and should work equally well when applied to the present hcp crystals. The treatment of short-range correlations explicitly in high-density He is under investigation[8] and their importance in these cases is yet to be determined conclusively.

As in previous experiments on the hcp[4,5] and fcc[1-3] phases of solid He some phonons of large energy and wave-vector transfers could not be identified in the present experiments (note in particular the missing portions of the LO branches in Figs. 2 and 4). Both multiphonon scattering and multiphonon interference effects have been shown to contribute to the reduction of visibility of phonons at large $\vec{Q}$.[9,18] Collins and Glyde[9] have calculated $S(\vec{Q},\omega)$ including one- and two-phonon terms as well as interference terms between one- and two-phonon scattering for high-density fcc He. They found the two-phonon scattering to increase rapidly at large $\vec{Q}$ and energies corresponding to about twice the energy range of the first maximum in the one-phonon density of states.[1] This results in extremely asymmetric line shapes with only a very small drop in intensity from the one-phonon peak and a long, high energy tail. Calculations[9] for a series of L[111] phonons that were studied in the previous work[2] on fcc He were able to demonstrate the rapid increase in the multiphonon background as $\vec{Q}$ approaches (1.5, 1.5, 1.5). With the inclusion of multiphonon terms beyond the two-phonon part it may be possible to more closely simulate the observed line shapes which show a complete obscuring of the one-phonon peak.

The multiphonon interference term may influence both the intensity and the background of the one-phonon peaks.[18] It consists of a symmetric contribution which decreases or increases the apparent intensity of the one-phonon peak depending on the sign of the reduced wave vector, and a second contribution which gives an asymmetric contribution to the background. If the intrinsic linewidth of the phonon is relatively large this will results in asymmetric line shapes, which may be severe enough to shift the apparent position of the one-phonon peak (see Fig. 6 in Ref. 6).

Such effects were studied extensively in the earlier work[6,7] on bcc He as well as in hcp He,[4,5] and recently in fcc He.[1,2] No extensive efforts were undertaken in the current experiment to study the intensity anomalies resulting from the symmetric part of the interference term, as initial observations revealed anomalies much smaller than those found in hcp He at low pressure (see Fig. 9 in Ref. 7). We did, however, observe many phonons with sloping or asymmetric background, as well as highly asymmetric line shapes for high energy phonons at large $\vec{Q}$. An example thereof is the LO[001] phonon shown in the bottom part of Fig. 1. In view of the recent calculations of Collins and Glyde[9] discussed above the large background on the high-energy side should be taken to result primarily from two-phonon scattering. LO[001] phonons at energies above 16 meV were completely obscured by such multiphonon background, similar to the L[111] branch in fcc He at 9.03 cm^3/mol (Fig. 7 in Ref. 2). It may be assumed that an analogous calculation for hcp He to that for fcc He by Collins and Glyde[9] would demonstrate the strong effects of multiphonon scattering at large $\vec{Q}$ equally well.

Shown in Table IV are the elastic constants for both of the hcp He crystals that could be determined from the present measurements. Measurements of transverse-acoustical modes with polarization in the basal plane are necessary to obtain the remaining hcp elastic constants. No particular effort was made to make extensive measurements at small $\vec{q}$ for an accurate determination of the elastic constants. Therefore no resolution correction was applied to the phonons included in the analysis, as past experience[1] has shown such a correction to be small ($<2\%$) for the phonons of reduced wave vector and energy transfer used in this analysis. The experimental pressures were estimated by extrapolation of the thermodynamic data of Ahlers.[19] Uncertainties quoted are statistical but include the above uncertainty due to resolution effects. The calculated values are the results of a self-consistent phonon calculation (SC1) of Goldman.[8] The agreement with the experimental values

TABLE IV. Elastic constants and pressure for hcp He (kbar). c_{12} and c_{13} cannot be obtained from the present data. The calculated values are due to Goldman (Ref. 8).

	$V=9.41$ cm^3/mol $T=10$ K	$V=9.41$ cm^3/mol $T=0$ K (calc)	$V=11.61$ cm^3/mol $T=8.3$ K	$V=11.6$ cm^3/mol $T=0$ K (calc)
c_{11}	28.2 ±0.8	26.8	11.3± 0.7	10.53
c_{12}	···	15.9	···	5.78
c_{33}	32.0 ±0.6	31.2	12.6 ±0.4	12.30
c_{44}	5.66±0.12	5.4	2.40±0.12	2.35
c_{13}	···	11.5	···	4.03
P	3.7 ±0.1	···	1.4 ±0.1	1.3

is good with the exception of c_{11}. The theoretical expression[20] for c_{11} contains an additional term due to internal strains, which does not affect the values of c_{33} and c_{44}. Thus this discrepancy may be due in part to an inadequate representation of this effect.

B. Grüneisen parameters

The volume dependence of the phonon frequency $\omega_j(\vec{q})$ is given by the mode Grüneisen parameters $\gamma_j(\vec{q})=d\ln\omega_j(\vec{q})/d\ln V$. In principle, the $\gamma_j(\vec{q})$ may be different for the various modes, and may depend on temperature as well as on volume. The present two sets of experimental phonon energies as well as those of previous studies[4,5] can be used to compute "experimental" mode Grüneisen parameters whenever a phonon of a particular reduced wave vector $\vec{q}$ has been measured at two different densities. In this way, information can be obtained on any possible significant variation of the mode γ's for different symmetry directions and wave vectors. With the large range of molar volumes for which phonon energies in hcp He have now been determined the dependence of the Grüneisen parameters on volume may also be studied.

As far as any dependence of the mode γ's on wave vector is concerned, a comparison of the first two experiments[4,5] on hcp He suggested that all the observed frequencies could be scaled uniformly within experimental error by a factor of 1.91 (i.e., $\gamma=2.34$). This result may be contrasted with the case of fcc He and Ne where a variation of the mode γ's with branch type and wave vector was evident.[2,11] In a subsequent work[7] on the hcp, phase data were added at 21.1 cm^3/mol for the LO[001] branch which scaled by a factor of 2.6 ($\gamma\sim3.0$) to the data[5] at 16 cm^3/mol. Difficulties in assigning the correct one-phonon frequency to the severely broadened profiles of this mode (see Fig. 9 in Ref. 7) were suggested[7] to possibly account for this difference in scale factor. Recent Raman scattering results on hcp He for densities up to about 17 cm^3/mol showed for the Raman-active zone-center TO mode a value for γ of 2.6 ± 0.1 when assumed to be independent of volume. Since there is no *a priori* reason individual mode γ's should be the same, such discrepancies are not unexpected.

The present data at 11.61 and 9.41 cm^3/mol were therefore used in conjuction with that of previous work[4,5,7] at 21.1 and 16.0 cm^3/mol to obtain several sets of mode γ's. Two such results are shown in Figs. 5 and 6 comparing the present data at 11.61 cm^3/mol with those at 16.0 and at 9.41 cm^3/mol. It is quite apparent that within experimental errors the phonon frequencies do scale by approximately a constant factor in each case, similar to the conclusion reached by Reese *et al.*[5] It should be pointed out, however, that small variations of the mode γ's with branch type and wave vector in hcp He may be masked by the relatively large uncertainties in the values of the mode γ's, each of which is derived from four separate experimental quantities.

Comparson of Figs. 5 and 6, however, does show a definite volume dependence, as the unweighted average value of γ for the 16.0–11.61 cm^3/mol range is 2.44, while it is 1.77 for the 11.61–9.41 cm^3/mol range (Table V). Reese *et al.*[5] analyzed their data at 16.0 cm^3/mol with respect to the 21.1-cm^3/mol data[4] for a possible volume dependence of the Grüneisen parameter. The assumption of a linear dependence of γ on volume led to the conclusion that $\gamma=0.1262V$. A much weaker volume dependence for the thermodynamic Grüneisen parameter is given by Ahlers[19] as the result of the analysis of his thermodynamic data, i.e., $\gamma_G=1.02+0.083V$ for molar volumes down to about 13.7 cm^3/mol. Shown in Table V are values for the Grüneisen parameter as obtained from extrapolating the analysis of Reese *et al.*[5] and Ahlers[19] to the molar volumes of the present study. Comparison with the experimental values would

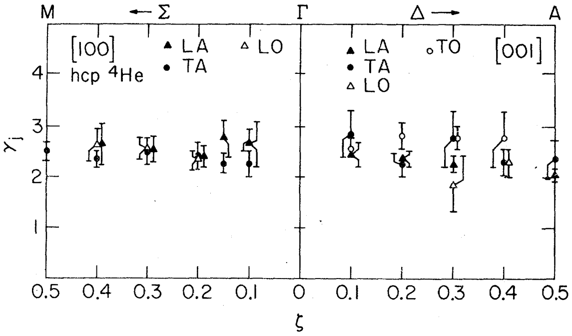

FIG. 5. Mode Grüneisen parameters for hcp ^{4}He over the range $V_1 = 11.61$ cm^3/mol (this work) to $V_2 = 16.0$ cm^3/mol (Ref. 5). Error bars shown represent a combination of 1.5 times the statistical errors given in each work.

suggest that the Grüneisen parameter depends on volume more nearly in the way suggested by Ahlers.[19]

The Raman scattering data[21] on the zone-center transverse-optical mode for densities up to ~17 cm^3/mol may also be used in conjunction with the present data for the elucidation of the volume dependence of γ. Shown in Fig. 7 are the experimental phonon energies for the TO mode at the zone center as a function of molar volume. All of the neutron scattering data for this mode as well as the point at the lowest molar volume of the Raman scattering work[21] are included. The lines indicate the calculated variation of the phonon energy of this mode for three cases: a constant value of the Grüneisen parameter of 2.6, and volume-dependent Grüneisen parameters as obtained by Ahlers and Reese *et al.* It may be seen that in this large range of molar volumes over which phonons in hcp He have now been measured the Grüneisen parameter appears to depend only weakly on volume. Even a constant value of γ is consistent with the

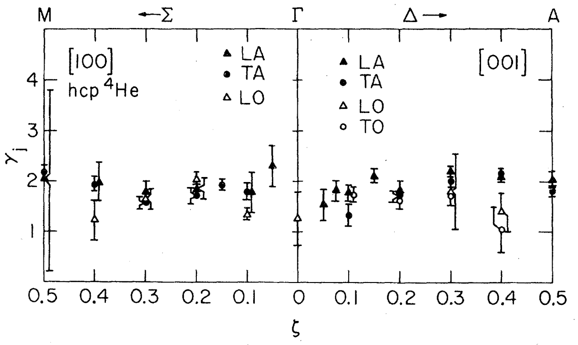

FIG. 6. Mode Grüneisen parameters for hcp ^{4}He over the range $V_1 = 9.41$ cm^3/mol to $V_2 = 11.61$ cm^3/mol. Error bars shown represent a combination of 1.5 times the statistical errors of each data set.

TABLE V. **Grüneisen** parameters in solid He. Experimental values are unweighted averages of mode Grüneisen parameters calculated from experimental phonon energies. (1) is an extrapolation of the analysis of Reese *et al* . (Ref. 5) with $\gamma = 0.1262V$. (2) is the volume dependence of γ_G obtained by Ahlers ($\gamma_G = 1.02 + 0.083$) from specific-heat data (Ref. 19).

Volume range (cm^3/mol) V_1	V_2	Experiment	Analysis (1)	(2)
21.1	16.0	2.34[a]	2.34[a]	2.56
16.0	11.61	2.44	1.74	2.17
11.61	9.41	1.77	1.33	1.89

[a] Reference 5. A value of 3.0 was obtained for phonons of the LO[001] branch (Ref. 7). As the phonons of this branch in both experiments are severely broadened this value carries a correspondingly large uncertainty and therefore does not represent a significant disagreement with the value of 2.34 quoted above.

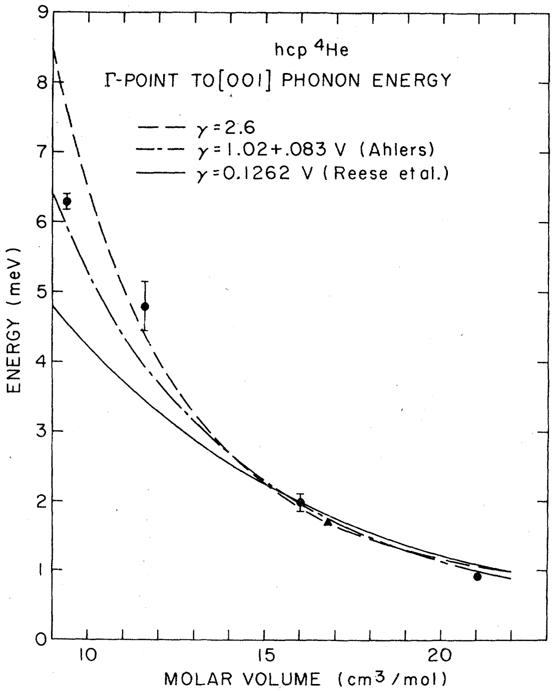

FIG. 7. Dependence on molar volume of the zone-center transverse-optical phonon. Solid points are neutron scattering results. Triangle represents the point at lowest molar volume of the Raman scattering work (Ref. 21).

available data. This agrees with the conclusion reached on the Raman scattering data,[21] which covers the much smaller range of densities from approximately 20.5 to 16.8 cm^3/mol. It could be analyzed in terms of a volume independent γ of 2.6 ± 0.1 or equally well to agree with the volume dependence obtained by Ahlers.[19] Neither the Raman scattering data nor the neutron scattering data are accurate enough to determine unambiguously the volume dependence of the Grüneisen parameter.

IV. CONCLUDING REMARKS

With the results of the present neutron scattering study on hcp He and the previous work[1,2] on fcc He at high pressure, we have presented a rather complete picture of the lattice dynamics of He quantum solids at high densities. As was the case for the fcc crystals, anharmonic effects were found to be still pronounced at these densities in the hcp phase, though reduced in magnitude. While it appears that multiphonon interference effects are not nearly as prominent at the temperatures and densities of the crystals in this study as at the lowest densities, phonon line shapes at large momentum and energy transfers are dominated by multiphonon scattering processes. Calculations of phonon line shapes performed for the fcc phase[9] give a fairly detailed account of this effect and would be valuable for the present work as well. Phonon energies in hcp He have now been measured over a range of densities differing by more than a factor of 2, which provides a unique opportunity for a test of the general validity of the lattice-dynamic-model calculations. Among the questions still to be answered conclusively is the degree of importance of short-range correlations in high-density He. As the theoretical methods for treating short-range correlations have been shown to have an inadequate density dependence,[8,10] calculations of the volume dependence of phonon energies should be of use in determining their form. Such calculations would also be an interesting test of the apparent lack of a significant dispersion of the experimental mode Grüneisen parameters. An extension of these measurements to still higher densities may well be of interest, in particular with the respect to the question of the stability of the fcc phase down to $T = 0$ K at higher pressures.

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