

## Lattice dynamics of solid neon at 6.5 and 23.7 K\*

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The low-frequency phonons of fcc neon have been studied along the high-symmetry directions at both 6.5 and 23.7 K (melting point, 24.6 K) using inelastic neutron scattering on a stress-free single crystal grown at a pressure near one bar. At 23.7 K the zero-sound-wave velocities are approximately 2.5% larger than the first-sound-wave velocities measured by Brillouin scattering. The sign of the result here is as expected and compares well with the expected larger difference of approximately 5% which was found in krypton at 114 K (melting point, 115.8 K). At 6.5 K with a lattice spacing of  $4.466 \pm 0.002$  Å, the phonon measurements are in excellent agreement with previous measurements on a clamped crystal with the stated lattice spacing  $4.454 \pm 0.002$  Å; the data show a frequency shift of  $(1.2 \pm 0.5)\%$ , whereas the calculated volume difference assuming a Grüneisen constant of 3 would suggest a shift of  $(2.4 \pm 0.5)\%$  (the present measurements are the first phonon measurements done on a neon sample with the equilibrium lattice parameter). An analysis of the data giving the long-wavelength elastic properties is presented. The integrated intensities of the peaked responses were also studied at 23.7 K and are shown to behave anomalously; this behavior has been found in studies of other quantum crystals indicating the existence of substantial multiphonon scattering and interference effects.

### I. INTRODUCTION

Inelastic neutron scattering has been used extensively in the study of the lattice dynamics of rare-gas solids. The main information desired is the  $\vec{q}$  dependence of the single-phonon energy response. This type of data is then most conveniently compared to theoretical calculations of the phonon dispersion in crystals. This comparison, in addition to calculations of other known physical properties, serves as a test of knowledge of the interatomic forces present and of the calculation methods. The rare-gas solids offer a simple group of substances for which the number of simplifying assumptions of a calculation can be minimized.

Neon has already been investigated by both Leake *et al.*<sup>1</sup> and Skalyo *et al.*<sup>2</sup> These studies were done on clamped crystals grown in a high-pressure cell after the manner of the study on krypton by Daniels *et al.*<sup>3</sup> These near isochoric studies however do not allow direct comparison with other experimental investigations performed on stress-free crystals; although the aforementioned two measurements were made on samples at 5 K which were nearly stress free. Recently measurements at Brookhaven have been done on stress-free crystals of krypton<sup>4,5</sup> and xenon<sup>6-8</sup>; comparisons have been made of the resulting data with bulk-modulus measurements near 5 K and with Brillouin-scattering measurements of the sound speeds near the melting point. Since many physical quantities are strongly dependent on the lattice constant, we have extended these studies on stress-free rare-gas crystals to neon.

Particularly useful quantities which serve as a

test for theoretical calculations are the elastic constants; the long-wavelength characteristics which are the second derivatives of the free-energy density with respect to strain. There have already been measurements of the elastic constants on a single crystal of neon near its melting point by Gewurtz *et al.*<sup>9</sup> using Brillouin scattering. Therefore we can make a direct comparison with the neutron-scattering results made at the same temperature to obtain a measure of the zero-, first-sound difference.<sup>10</sup>

This latter comparison is interesting in that one must keep in mind the frequency difference between the two probes which gives rise to the concept of first and zero sound. In neutron scattering the frequencies encountered are greater than 0.1 THz where the elastic waves propagate in a collision-free zero-sound regime ( $\omega\tau \gg 1$ , where  $\tau$  is the phonon lifetime). This contrasts with the collision-dominated or hydrodynamic regime ( $\omega\tau \ll 1$ ) of first sound which is probed by ultrasonic ( $\omega \approx 10^{-5}$  THz) and probably by Brillouin light-scattering ( $\omega \approx 10^{-3}$ -THz) experiments.

Theoretical estimates of the differences between zero and first sound near  $T_M$  have been made for argon,<sup>11</sup> and for krypton and xenon<sup>12</sup> with zero sound being larger by 2, 4, and 7%, respectively. The experimental results for krypton show a difference of about 5%<sup>4,13</sup> while the results for xenon<sup>6,14</sup> show first-sound speeds larger than the zero-sound speed (the latter result is incongruous with theoretical expectation).

As previously stated, measurements on neon at 5 K were done on near-stress-free crystals grown isochorically. The present experiment on a stress-free sample eliminates both a possible

minute systematic error due to instrumental calibration and an assumption as to the magnitude of the Grüneisen constant  $\gamma$  which could affect the derived stress-free properties. In particular an uncertainty in the lattice parameter  $\Delta a/a$  relates to an elastic-constant uncertainty according to  $\Delta c_{ij}/c_{ij} \approx -6\gamma\Delta a/a$ . Accurate elastic-constant measurements are important near 0 K as other methods have proven heretofore to be uncertain. An added feature of the low-temperature measurement is that the zero-, first-sound differences vanish. A comparison with theoretical values is important as a check on the validity of the interatomic potential utilized for neon. In the case of neon, it is noted that evidence of the magnitude of multibody forces cannot be obtained merely by noting the difference between  $c_{12}$  and  $c_{44}$  since large zero-point motion exists even at 0 K.

We have additionally measured the presence of single-phonon multiphonon interference effects<sup>15-18</sup> in the neutron response of neon at 23.7 K. While the single-phonon response is usually sought in order to determine the crystal forces, the neutron-scattering technique probes both single and multiphonon excitations. The multiphonon peak in neon has already been observed for large- $\vec{q}$  measurements even at 5 K.<sup>2</sup> In this case the single-phonon peak appeared to be discernible from the multiphonon peak even though the two peaks partially overlap one another.

The apparent separability of the two peaks is not, however, without question. The more general case occurs when one observes single phonon, multiphonon, and their interference. This latter case is already well documented in the case of helium<sup>18</sup> and it is clear from the work of Horner<sup>17</sup> that a direct determination of the phonon dispersion from the data for some  $\vec{q}$  values is not possible. An additional complicating feature of helium is its large zero-point vibrational amplitude and anharmonicity which combine to broaden the single-phonon peak and enhance the interference effect.

Horner shows that the same interference effect should also be observed in neon,<sup>19</sup> but to a much lesser extent than for helium due to the smaller zero-point vibrational amplitude (an atomic mass of 20 and Debye temperature of 75 K for neon compares with an atomic mass of 3 or 4 and a Debye temperature near 25 K for helium).

In particular, the single-phonon response obeys a sum rule given by Ambegaokar *et al.*<sup>16</sup> as

$$(1/2\pi) \int_{-\infty}^{+\infty} S_1(\vec{Q}, \omega) \omega d\omega = (Q^2/2m) e^{-2w(\vec{Q})}, \quad (1)$$

i.e., the fraction of the total observed first-frequency moment ( $Q^2/2m$  as given by the Placzek sum rule<sup>20</sup>) due to the single-phonon response is

given by the Debye-Waller factor. Equation (1) is difficult to fit experimentally due to the interference that takes place between the broad multiphonon response and the relatively narrow single-phonon response. The former generally appears as a contribution to the background while a relatively sharp peak usually attributed to the single-phonon response can be diminished or enhanced by interference effects. An intensity anomaly observed in solid hydrogen<sup>21</sup> may also be due to interference effects although to a smaller extent than in helium since solid hydrogen has a higher Debye temperature,  $\Theta_D \approx 120$  K.

## II. SAMPLE AND EXPERIMENTAL METHOD

The sample in the present experiment was grown at atmospheric pressure using a technique that has been fully described by Skalyo *et al.*<sup>5</sup> in their phonon measurements on a krypton single crystal. A natural isotopic mixture of research grade neon gas (99.999% pure) was first liquified in a cylindrical sample cell whose walls were made from 0.05-mm-thick kapton (polypyromelitimide); the cell itself was contained in a variable-temperature liquid-helium-cooled cryostat.

During the condensation the base of the cell was kept at 25 K and the top at 30 K. Temperature was controlled to within  $\pm 0.01$  K of the set point using germanium resistance thermometers and two ac electronic bridges. Then the temperature was continuously lowered at the rate of 0.6 K per day controlling both temperatures such that the temperature gradient across the cell was kept at 5 K. Once the crystal had begun to grow, a cooling period of 12 h was alternated with an annealing period of 12 h. Finally, when the whole cell was solidified, the temperature gradient was reduced, and the crystal equilibrated at 23.7 K.

The method produced a large single crystal with its  $[01\bar{1}]$  axis about  $12^\circ$  from the Dewar axis; this permitted measurements to be made in both the  $[01\bar{1}]$  and  $[001]$  zones of scattering by appropriate tilting. The sample cell was not completely filled by only one single crystal; however, the sample crystallite was cylindrical in shape with other crystallites forming layers above and below it. This was confirmed by neutron polaroid photographs of several Bragg peak reflections and the stray crystallites were easily masked from the neutron beam by cadmium. A comparison of phonon intensities with those from a standard sample indicated the sample volume was about  $5 \text{ cm}^3$ . At 23.7 K the crystal mosaic was less than 4-min full width at half-maximum (FWHM).

After the completion of the measurements at 23.7 K, the temperature was reduced at a rate

of 1 K/h until 18.5 K. The crystal mosaic at this temperature was measured and found to be 24-min FWHM. The crystal which was initially transparent became cloudy. Cooling then proceeded to 6.5 K within the next few hours resulting in a final mosaic spread of 36-min FWHM. The lattice spacing was measured to be  $4.466 \pm 0.002 \text{ \AA}$ , which is equal within the experimental error to the zero pressure 6.5-K value obtained by Batchelder *et al.*<sup>22</sup> by means of x-ray measurements.

The inelastic-neutron-scattering measurements were done on a triple-axis spectrometer at the Brookhaven high flux beam reactor. A cylindrically curved pyrolytic graphite monochromator<sup>23</sup> was used to produce incident neutron beams of 14.5, 13.5, and 5.5 meV. A pyrolytic graphite filter<sup>24, 25</sup> was used to eliminate higher-order contamination, the analyzer was a flat pyrolytic graphite crystal, and all spectrometer collimators were 20-min FWHM.

### III. RESULTS

#### A. Dispersion at 23.7 K

The analysis of the experimental data has been treated extensively in a series of rare-gas-solid experiments.<sup>1-8, 26</sup> Typical neutron groups taken at 23.7 K are shown in Figs. 1 and 2 taken with incident neutron energies  $E_i$  of 13.7 and 5.5 meV, respectively; measurements were limited to  $\vec{q} < 0.35 \text{ \AA}^{-1}$ , the region of interest for a zero-sound determination. The associated curves are Gaussians fit to the data. It is noted that the transverse phonons have energy widths that could be attributed to instrumental resolution alone while some of the longitudinal phonons had widths 30% larger than the instrumental resolution.

As in the previous studies the measured peak positions were corrected for slight shifts due to

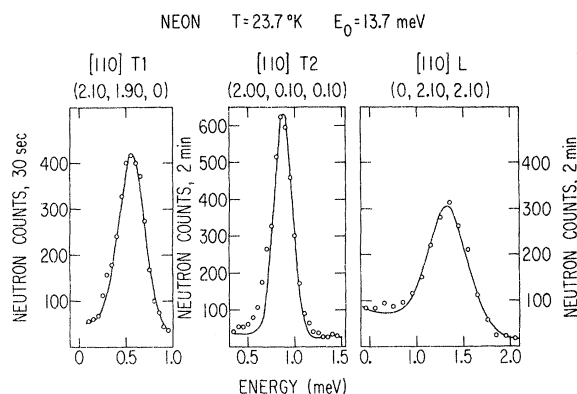


FIG. 1. Typical phonon scans at 23.7 K with  $E_i = 13.7$  meV. The solid lines are fitted Gaussians.

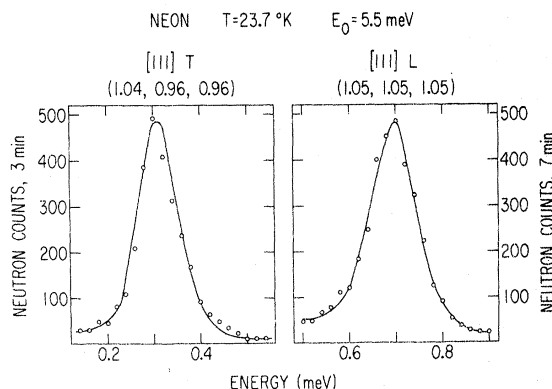


FIG. 2. Typical phonon scans at 23.7 K with  $E_i = 5.5$  meV. The solid lines are fitted Gaussians.

instrumental resolution. This procedure involves an iterative process of determining first a set of Born-von Karman force constants and then generating a resolution-influenced line profile using a modified version of a program by Pynn and Werner<sup>27</sup>; the shift in the peak of the generated profile from its expected position is the correction applied to the observed data.

The corrected phonon energies for the main crystallographic symmetry directions are given in Table I. The errors given in this table are due to both the precision of the resolution correction and to estimates of the error in the peak position obtained from the Gaussian fits. The corrected data are also shown in Figs. 3-5.

A Born-von Karman (BVK) force-constant analysis including the first two nearest neighbors was fit to the data in order to determine the elastic constants and the results are shown in Table II. An analysis in terms of more distant neighbors is not justified because of the limited range of the data in  $\vec{q}$ . The solid lines in Figs. 3-5 are a result of this fit. It is noted that for  $\vec{q} > 0$ , in the limited range of  $\vec{q}$  investigated, that the  $[110] T_1$  mode has a positive second derivative. This also occurs in neon at 5 K,<sup>2</sup> krypton at 114 K,<sup>4</sup> but not in krypton at 10 K.<sup>5</sup>

The individual branches have also been fitted by the function

$$E = \epsilon + \alpha q + \beta q^3, \quad (2)$$

where  $\epsilon$  and  $\alpha$  represent a possible systematic spectrometer misset and the sound velocity, respectively, and  $\beta$  is deviation of the dispersion from linearity ( $E$  is an odd function of  $q$ ). For this fit, the data have been limited to  $q < 0.25 \text{ \AA}^{-1}$ . The value for  $\epsilon$  was of the order of its estimated error and could be attributed to the least count of the spectrometer positioning and was therefore fixed to be zero in all scans taken in the  $[01\bar{1}]$

TABLE I. Phonon energies in neon at 23.7 K.

[100]			[110]			[111]			
$\xi$	$T$	$L$	$\xi$	$T_1$	$T_2$	$L$	$\xi$	$T$	$L$
0.04	0.242 ± 0.003	0.336 ± 0.009	0.02		0.167 ± 0.006		0.015		0.26 ± 0.01
0.05	0.305 ± 0.003	0.424 ± 0.007	0.03		0.258 ± 0.003		0.02	0.160 ± 0.005	0.337 ± 0.006
0.06	0.366 ± 0.003	0.517 ± 0.009	0.04		0.345 ± 0.003	0.52 ± 0.01	0.025		0.428 ± 0.005
0.07	0.424 ± 0.003	0.589 ± 0.007	0.05		0.429 ± 0.003	0.66 ± 0.01	0.03	0.244 ± 0.004	0.515 ± 0.005
0.08	0.488 ± 0.003	0.682 ± 0.008	0.06	0.30 ± 0.01	0.514 ± 0.003	0.80 ± 0.02	0.04	0.320 ± 0.007	0.686 ± 0.006
0.09		0.763 ± 0.007	0.07		0.598 ± 0.003		0.05	0.401 ± 0.004	0.86 ± 0.02
0.10	0.605 ± 0.003	0.85 ± 0.02	0.08	0.42 ± 0.01	0.682 ± 0.003	1.06 ± 0.02	0.06	0.485 ± 0.005	1.01 ± 0.02
0.12	0.73 ± 0.01		0.10	0.51 ± 0.01	0.84 ± 0.01	1.36 ± 0.02	0.07	0.565 ± 0.005	
0.15	0.91 ± 0.01	1.26 ± 0.02	0.12	0.619 ± 0.006	1.01 ± 0.01	1.64 ± 0.02	0.08	0.62 ± 0.01	1.38 ± 0.02
0.17	1.03 ± 0.01	1.42 ± 0.02	0.14	0.731 ± 0.006			0.10	0.77 ± 0.01	1.70 ± 0.01
0.20	1.22 ± 0.01	1.71 ± 0.05	0.15		1.26 ± 0.01	2.07 ± 0.02	0.12	0.94 ± 0.01	2.09 ± 0.02
0.25	1.54 ± 0.01	2.12 ± 0.03	0.16	0.84 ± 0.01			0.14	1.10 ± 0.01	
			0.18	0.94 ± 0.01					
			0.20	1.04 ± 0.01					

zone. Phonons taken in the [001] zone which included the [110]  $T_1$  and [100]  $L$  branches originally gave values of  $\epsilon = -0.010 \pm 0.002$  meV. This systematic error might have been caused by the large tilt of  $33^\circ$  used to obtain the [001] zone.

This correction was also applied to these branches for the Born-von Karman fit discussed previously and has already been effected in Table I. Elastic constants determined by a least-squares process on the values obtained for the individual branches is also shown in Table II. It is noted that Skalyo *et al.*<sup>2</sup> have determined that  $c_{11} = 167.7 \pm 2.3$  and

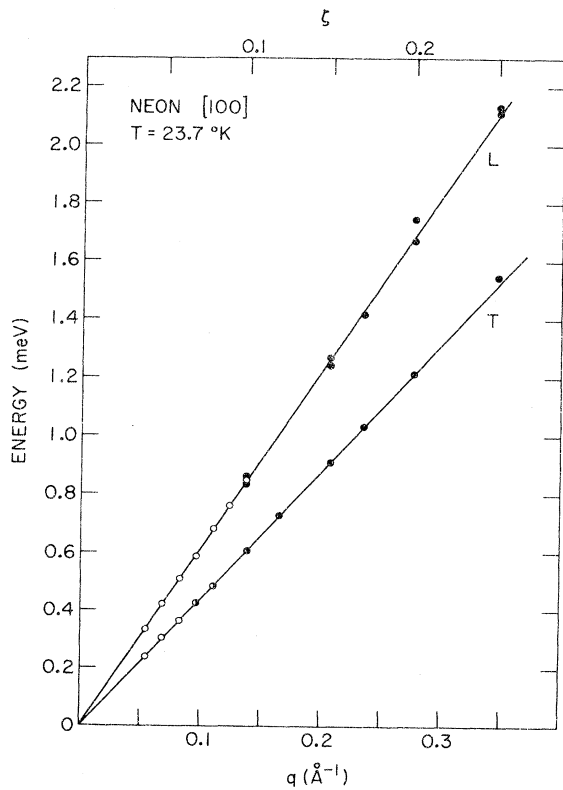


FIG. 3. Phonon dispersion at 23.7 K in the [100] direction. Open and filled circles are the data with  $E_i = 5.5$  meV and 13.7 meV, respectively. The solid lines are two neighbor BVK fit to the data.

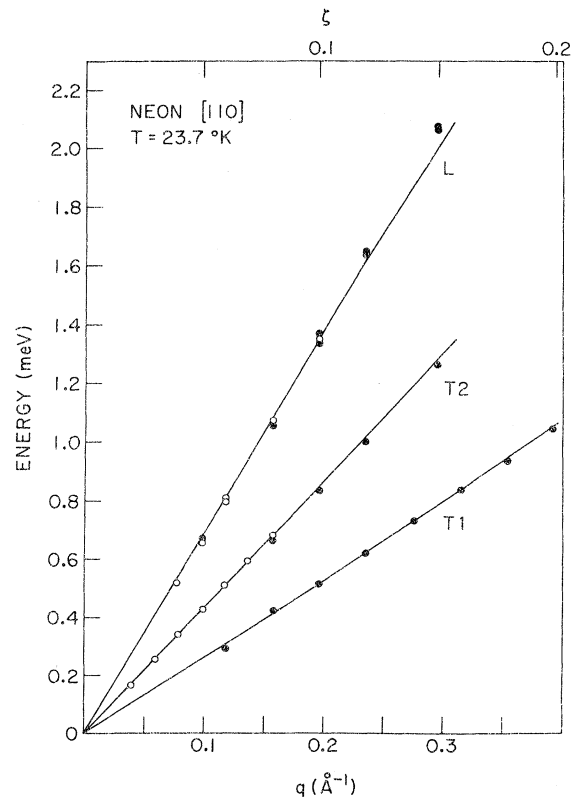


FIG. 4. Phonon dispersion at 23.7 K in the [110] direction.

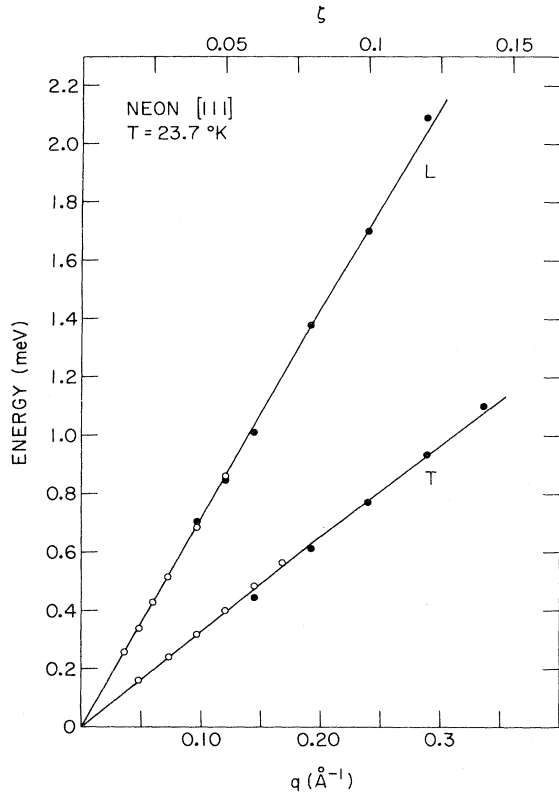


FIG. 5. Phonon dispersion at 23.7 K in the [111] direction.

$c_{44} = (88.5 \pm 0.6) \times 10^8 \text{ dyne cm}^{-2}$  for neon at 22 K with a lattice parameter of 4.45 Å, a 38 and 40% change, respectively, when compared to the equilibrium crystal measured with  $a = 4.529 \text{ Å}$  (therefore  $\gamma \approx 3.3$ ).

We additionally show in Table II values obtained by Gewurtz *et al.*<sup>9</sup> using Brillouin scattering. The results are generally within two standard deviations of one another, but the neutron-scattering values are systematically higher by about 5%, hence 2.5% higher in sound speed. Near the melting temperature, the elastic constants determined by neutron scattering for krypton<sup>4</sup> are about 10% higher than the Brillouin-scattering results<sup>13</sup> while for xenon<sup>6, 14</sup> the elastic constants are lower. The results for krypton and neon are qualitatively as predicted theoretically. As their Debye temperatures are comparable, the larger effect in krypton is no doubt due to its higher melting temperature and resultant effect on phonon population factors which enter the calculation.

#### B. Dispersion at 6.5 K

Extensive measurements on the phonon dispersion were carried out at 6.5 K primarily for two reasons. First, the work of Skalyo *et al.*<sup>2</sup> did not

TABLE II. Elastic constants of neon at 23.7 K (in  $10^8 \text{ dyne cm}^{-2}$ ):  $B_0 = \frac{1}{3}(c_{11} + 2c_{12})$  in  $10^8 \text{ dyne cm}^{-2}$ ,  $A = 2c_{44}/(c_{11} - c_{12})$ , and  $\delta = (c_{44} - c_{12})/c_{12}$ . The elastic-constant combination appropriate to the various branches is also given. Note that the Brillouin-scattering results were computed by the authors from the results by Gewurtz *et al.* (Ref. 9).

	BVK Model	Analysis with Eq. (2)	Brillouin scattering (24.3 K)
$c_{11}$	121.9 ± 0.9	120.6 ± 1.5	117.5 ± 2.0
$c_{12}$	76.2 ± 1.0	73.2 ± 1.5	74.0 ± 2.0
$c_{44}$	63.0 ± 0.3	63.3 ± 0.9	59.5 ± 1.5
$c[100]L$	121.9 ± 0.9	123.7 ± 1.2	117.5
$c[100]T$	63.0 ± 0.3	63.7 ± 0.9	59.5
$c[110]L$	162.0 ± 0.9	157.7 ± 1.4	155
$c[110]T_1$	22.8 ± 0.4	23.0 ± 0.9	21.7
$c[110]T_2$	63.0 ± 0.3	62.7 ± 0.8	59.5
$c[111]L$	175.4 ± 0.9	170.6 ± 0.9	167.8
$c[111]T$	36.2 ± 0.3	37.0 ± 0.9	34.3
$B_0$	91.4 ± 0.9	89.0 ± 1.5	88.5 ± 2.0
$A$	2.76 ± 0.04	2.63 ± 0.16	2.74 ± 0.25
$\delta$	-0.17 ± 0.01	-0.15 ± 0.02	-0.20 ± 0.03

contain the important [110]  $T_1$  branch due to the inability to tilt to a proper zone of scattering. It was also felt that the present measurements done on a stress-free crystal would eliminate a slight systematic error which could have existed in the lattice parameter of the clamped measurements and also obviate the assumption of a value for the Grüneisen constant (a value of three was taken by Skalyo *et al.*<sup>2</sup>).

The resolution-corrected data are presented in Table III. A direct comparison was made with the data of Skalyo *et al.*<sup>2</sup> for all phonons which were measured at the same  $\vec{q}$ . The percentage difference between the two experiments has been plotted in Fig. 6 as a function in reciprocal space. Some

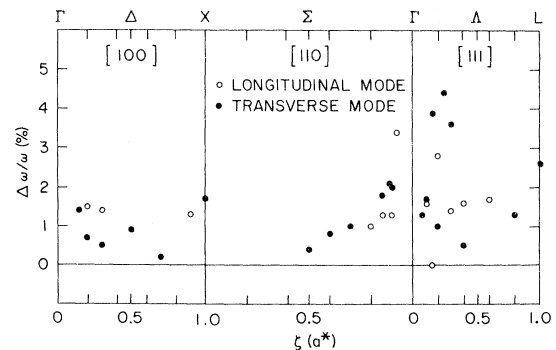


FIG. 6. Relative differences of phonon measurements between the present work at 6.5 K (lattice parameter  $a = 4.466 \pm 0.002 \text{ Å}$ ) and the work of Skalyo *et al.* (Ref. 2) at 5 K ( $a = 4.454 \pm 0.002 \text{ Å}$ ).

TABLE III. Phonon energies of an equilibrium crystal ( $a = 4.466 \pm 0.002 \text{ \AA}$ ) of solid neon at 6.5 K.

$\xi$	[100]T	[100]L		
0.06	0.437 ± 0.004			
0.15	1.08 ± 0.01	1.54 ± 0.02		
0.2	1.44 ± 0.01	1.97 ± 0.02		
0.3	2.11 ± 0.01	2.87 ± 0.07		
0.5	3.30 ± 0.02			
0.7	4.19 ± 0.02			
0.9	4.61 ± 0.02			
1.0	4.63 ± 0.02			
$\xi$	[110]T <sub>1</sub>	[110]T <sub>2</sub>	[110]L	
0.04		0.42 ± 0.03		
0.06		0.63 ± 0.01	0.88 ± 0.02	
0.08		0.82 ± 0.01	1.19 ± 0.01	
0.10	0.64 ± 0.03	1.00 ± 0.01	1.53 ± 0.02	
0.12	0.77 ± 0.01	1.21 ± 0.01		
0.15	0.97 ± 0.01	1.51 ± 0.02	2.34 ± 0.02	
0.2	1.30 ± 0.01	2.02 ± 0.01		
0.3		2.96 ± 0.01		
0.4	2.53 ± 0.01	3.86 ± 0.02		
0.5		4.65 ± 0.02		
0.6	3.64 ± 0.03			
$\xi$	[111]T	[111]L		
0.03		0.612 ± 0.006		
0.04	0.395 ± 0.005			
0.06	0.584 ± 0.006			
0.08	0.765 ± 0.008	1.61 ± 0.02		
0.10	0.990 ± 0.008	1.96 ± 0.01		
0.12	1.13 ± 0.01			
0.15	1.41 ± 0.02	2.93 ± 0.01		
0.2	1.85 ± 0.02	3.82 ± 0.02		
0.3		5.35 ± 0.05		
0.4	2.98 ± 0.02			
0.5	3.11 ± 0.02			

TABLE IV. Elastic constants ( $10^8 \text{ dyne cm}^{-2}$ ) and associated parameters for neon at  $T = 6 \text{ K}$ . Column *A* gives the results of the present measurement, column *B* are the corrected results of Skalyo *et al.* (Ref. 2), and column *C* is obtained by combining the present measurements with those of Skalyo *et al.* (Ref. 2).

	<i>A</i>	<i>B</i>	<i>C</i>
$c_{11}$	164.9 ± 3.0	162.1 ± 1.7	161.5 ± 1.2
$c_{12}$	90.3 ± 3.0	83.4 ± 2.1	84.6 ± 1.3
$c_{44}$	92.8 ± 0.8	92.9 ± 0.5	92.6 ± 0.3
<i>A</i>	2.49 ± 0.04	2.36 ± 0.08	2.41 ± 0.04
<i>B</i>	115.2 ± 3.0	109.7 ± 1.7	110.2 ± 1.2
$\delta$	0.03 ± 0.04	0.11 ± 0.03	0.09 ± 0.02

points at the smaller  $\vec{q}$  values exhibit large differences; this is no doubt due to the smaller energies of these points. Assuming a shift independent of  $\vec{q}$ , we have found an unweighted average shift of  $(1.2 \pm 0.5)\%$ . Assuming a Grüneisen value of three, Skalyo *et al.*<sup>2</sup> had used a shift of  $(2.4 \pm 0.5)\%$  to calculate the bulk modulus of an equilibrium crystal. The data of Leake *et al.*<sup>1</sup> and Skalyo *et al.*<sup>2</sup> have been shifted for the effects of volume and are shown plotted in Fig. 7 with the present data. The consistent agreement between all three experiments is excellent.

The present data have additionally been analyzed using a Born-von Karman general force-constant analysis inclusive of the first three nearest neighbors. The resultant long-wavelength properties are shown in column *A* of Table IV. Column *B* shows the shifted results of the data of Skalyo *et al.*<sup>2</sup> and in column *C* we show an analysis which

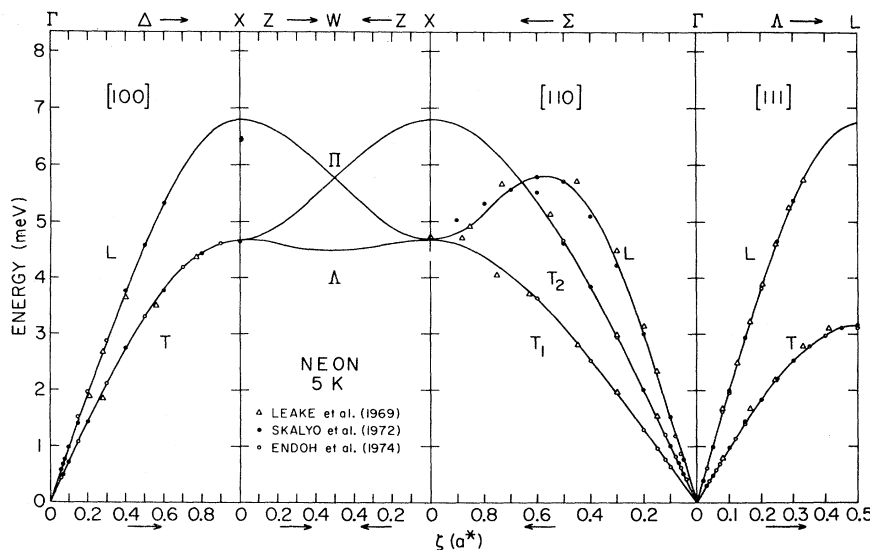


FIG. 7. Phonon dispersion in neon at 5 K inclusive of previous work; the previous work has been shifted to the equilibrium lattice parameter.

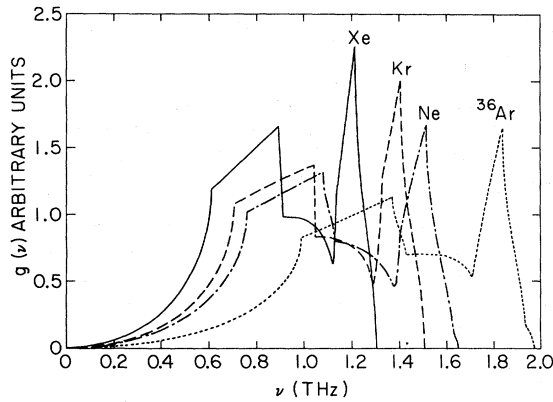


FIG. 8. Frequency density of states curves for the rare-gas solids.

includes data of both experiments in a general force-constant fit out to the fourth nearest neighbor. In Fig. 8 we show a  $g(\nu)$  frequency distribution resulting from the combined analysis along with previously determined  $g(\nu)$  for the other rare gases. For the case of argon we have made use of the combined data of Egger *et al.*,<sup>28</sup> Batchelder *et al.*,<sup>29</sup> and Batchelder *et al.*<sup>30</sup>

#### C. Cross-section interference

The search for interference effects in the peaked neutron response is carried out in the same manner as discussed by Minkiewicz *et al.*<sup>18</sup> for helium. Various phonons were measured at  $\vec{Q} + \vec{q}$  and  $\vec{Q} - \vec{q}$  along symmetry directions with  $\vec{Q}$  perpendicular to  $\vec{q}$  for transverse-type modes and  $\vec{Q}$  parallel to  $\vec{q}$  for longitudinal-type modes. Detailed balance can be used to rewrite Eq. (1) in the form

$$(1/2\pi) \int_0^\infty S_1(\vec{Q}, \omega) \omega (1 - e^{-\beta\omega}) d\omega = (Q^2/4m) e^{-2W(\vec{Q})}, \quad (3)$$

restricting the scan to the phonon-creation axis. The measured phonons were all well-defined neutron groups of Gaussian shape on a slightly energy-dependent background. The background-subtracted response was substituted for  $S_1(\vec{Q}, \omega)$  in Eq. (3) and the integral, which we define as  $M_1(\vec{Q})$ , determined.

The quantity  $M_1(\vec{Q})/Q^2$  is plotted on a log scale versus  $Q^2$  in Fig. 9. In the harmonic case the data should follow the illustrated dashed straight line; i.e.,  $W(\vec{Q})$  is proportional to  $Q^2$ . As Sears and

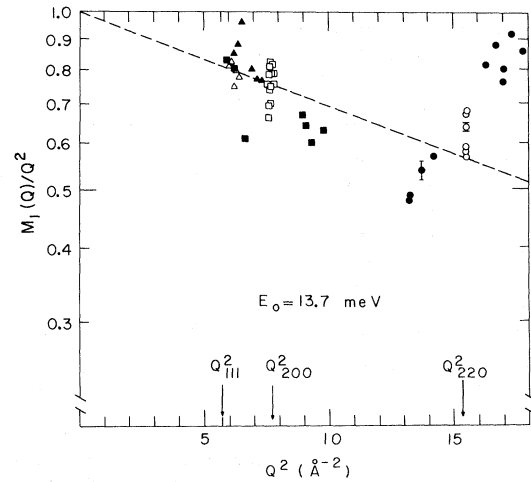


FIG. 9. Results of fitting the sum rule given by Eq. (3) to the observed data to indicate the presence of interference effects in neon at 23.7 K. The expected result is the dashed line. Longitudinal modes are closed marks, while transverse ones are open marks.

Khanna<sup>31</sup> have shown for helium, so too in the case of anharmonic neon one would not expect substantial deviation of the data from the straight line—provided we had integrated  $S_1(\vec{Q}, \omega)$ . The data deviate quite definitely from the line showing the effects of one-phonon multiphonon interference.

The findings here on neon at 23.7 K are very similar to the observations in helium<sup>18</sup> and support the predictions of Horner.<sup>19</sup> For the longitudinal modes where  $\vec{q}$  is parallel to  $\vec{Q}$  the illustration shows a varied dependence that appears to be an enhancement and diminution of  $M_1(\vec{Q})$  for measurements made on opposite sides of a Bragg peak. For transverse modes where  $\vec{q}$  is perpendicular to  $\vec{Q}$  and  $Q^2$  is only a slowly varying function of  $\vec{q}$ , the figure is only helpful in showing the existence of a variation in the interference. (The interference here must necessarily be an even function of  $\vec{q}$  due to the existence of symmetry.) Finally, a similar, but diminished search at 6.5 K for the effects of interference was null to within our precision.

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