Temperature dependence of the zero-sound elastic constants of crystalline xenon*

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Neutron inelastic scattering has been used to measure the low-frequency phonon dispersion relations for a single crystal of xenon at temperatures of 159.6, 111.0, and 10.0 K. From these data, the sound speeds and zero-sound elastic constants have been deduced, as well as the bulk modulus, anisotropy, and deviation from the Cauchy relation. The data are compared against Brillouin-scattering measurements at 156 K with the result that there are differences in c_{11} and c_{12} which cannot be explained as zero-sound–first-sound differences. The experimentally observed temperature dependence of the elastic constants is not well described by theory, although good agreement is found for a Monte Carlo calculation near the melting temperature. Our result for the departure from the Cauchy relation at low temperature indicates that calculations of many-body effects probably overestimate their importance for xenon.

I. INTRODUCTION

It is now well established that the rare-gas single crystals provide an important class of materials for helping to understand the fundamental dynamics of crystal lattices. One of the reasons for this is that these materials constitute a relatively simple system of solids allowing detailed calculations to be performed, and thus fruitful comparisons can be made between experiment and theory. Among the goals of such studies is a better understanding of the interatomic potentials, including possible effects of three-body forces. Also of interest in the rare-gas solids are anharmonic effects. To these ends much effort has been devoted to the theory of these crystals which, with improved many-body techniques and better high-speed computers, has progressed rapidly. Complementary experiments have also progressed, due partly to improved techniques and partly to the higher quality crystals which can now be grown and maintained.

Particularly useful quantities which serve as a testing ground for the theories are the elastic constants and their temperature dependence. In this paper we present such measurements for crystalline xenon using the technique of inelastic neutron scattering. With this method the single-phonon responses are directly observed and the phonon dispersion relations can be mapped out in any desired direction.

Many measurements of the elastic constants of neon, argon, and krypton have been reported, but there has been only one measurement on a single crystal of xenon to date, that being a Brillouin-light-scattering study close to the melting temperature by Gornall and Stoicheff.[1] Only a few measurements of the elastic constants of rare-gas crystals have been made using ultrasonic techniques; these are difficult experiments and of uncertain reliability. For xenon, one measurement has been reported of the sound velocity in a polycrystal using ultrasonic techniques,[2] but this alone is not sufficient to give elastic constants. Several theoretical predictions of the temperature dependence of the elastic constants have been made,[3–5] but until now no experimental data existed over the large temperature region below the melting point.

When comparing neutron scattering determinations of the elastic constants with results of other methods, one must take care to distinguish between first and zero sound.[6,7] For neutron scattering where the frequencies are generally $> 10^{11}$ Hz, elastic waves propagate in the collision-free or "zero-sound" regime, $\omega\tau \gg 1$, where τ is the thermal-phonon lifetime. In ultrasonic experiments, $\omega < 10^{9}$ Hz, one is in a collision-dominated or hydrodynamic regime ($\omega\tau \ll 1$). This region of sound-wave propagation is known as "first sound." Brillouin light scattering is thought to be in the first-sound region, but may possibly be in the fuzzy transition between the two.

Theoretical estimates of the differences expected between zero- and first-sound velocities and elastic constants have been made for argon[8] and for krypton and xenon.[9] Differences in the sound velocities of the order of $\frac{1}{2}$ and 2% were estimated for argon at 20 K and near melting, respectively, with the zero sound being the larger. For krypton and xenon near melting the zero-sound and first-sound velocities were estimated to differ by less than 4%. The calculated differences in elastic constants for xenon were between 5 and 7%. A recent comparison of the light-scattering results for krypton[10]

with an earlier neutron scattering determination[11] showed about a 10% difference in the elastic constants near the melting point. It is of interest to further investigate this point in the other rare-gas solids.

We report here measurements of the sound speeds and elastic constants in a single crystal of xenon at three temperatures: 159.6, 111.0, and 10.0 K, using inelastic neutron scattering. As will be seen later, the neutron technique for determining elastic constants at a given temperature is very laborious, and therefore our measurements were limited to three temperatures. We present results for the sound speeds in the principal symmetry directions, the elastic constants, the bulk modulus $B=\frac{1}{3}(c_{11}+2c_{12})$, the anisotropy $A=2c_{44}/(c_{11}-c_{12})$ and the deviation from the Cauchy relation, $\delta=(c_{44}-c_{12})/c_{12}$. The data were analyzed with detailed consideration given to the effects of instrumental resolution in an effort to obtain the highest possible accuracy.

The remainder of the paper is divided into four sections. Section II describes briefly the methods used to grow and cool the crystal. The experimental details and procedures used to correct the data are discussed in Sec. III. This is followed by a description of the method of analyzing these data to get elastic constants and the results. Finally, in Sec. V, the results are compared with theoretical calculations and with the light-scattering measurements.

II. CRYSTAL GROWTH

A large single crystal of xenon was grown from the melt at its equilibrium vapor pressure using a procedure similar to that used previously in this laboratory for krypton.[11-13] A large crystal was necessary because of the high neutron absorption in natural xenon (24 b) and the small coherent scattering cross section (2.9 b). The sample cell consisted of a 3-cm-diameter cylinder 4.5-cm high fabricated from Kapton with wall thickness 0.05 mm. A stainless-steel top contained a heater and a calibrated platinum resistance thermometer. The aluminum base of the cell was mounted in contact with the copper block of a variable-temperature cryostat. A piece of copper wire connected the top and bottom of the cell to insure thermal equilibrium. A viewing port in the cryostat permitted observation of the crystal.

Xenon gas of 99.99% purity (purchased from Air Products and Chemicals, Inc.) was condensed in the cell until it was filled with liquid. Starting with the base of the cell at 162 K and the top at 165 K, the temperature was continuously lowered (melting point 161.4 K) at the rate of 0.75 K/day, maintaining the 3-K gradient. After the crystal had begun to grow, the cooling was continued at the same rate only during the day (~8 h) and was held constant overnight both to permit growth to keep up with the cooling rate and to anneal. Full growth was completed in about 5 days, following which the gradient was eliminated and the crystal equilibrated at 159.6 K.

This procedure yielded a large single crystal filling the bulk of the cell oriented with the [111] axis within 1° of the cylinder axis. The volume of the crystal was estimated to be about 12 cm^3 using the integrated intensity of several phonons along with an absolute calibration of the spectrometer. Neutron Polaroid photographs of several Bragg reflections confirmed this estimate. The lattice parameter measured with neutrons at 159.6 K was $a=6.350\pm0.002$ Å, in good agreement with molar-volume measurements.[14] The crystal mosaic was approximately 1' full width at half-maximum (FWHM). The [111] zone is not particularly useful for phonon measurements; however, the cryostat containing the crystal was capable of being tilted sufficiently to reach a [211] zone of scattering where most of the measurements were made.

Cooling of the crystal was accomplished using a procedure similar to that of Batchelder *et al.*[15] The crystal was sublimated by pumping in an effort to pull it away from the cell walls. Once a free-standing crystal was obtained, it was cooled first to 111 K (over a period of 24 h) and then to 10 K (over a 72-h period). The initial pumping procedure broadened the crystal mosaic from 1' to about 6'. Cooling broadened it further to about 11' at 10 K, still quite satisfactory for neutron measurements.

Upon completion of the measurements in the [211] zone, a new crystal was inadvertently obtained after attempting to warm up to the original temperature of 159.6 K. At this temperature recrystallization occurred yielding a new crystal with a [001] axis about 20° from the cylinder axis, and a mosaic of about 8' FWHM. The volume of the second crystal was also approximately 12 cm^3. The second crystal was cooled without first attempting to free it from the cell walls, and no change in mosaic was observed during cooling to 10 K.

The temperature of the crystals was maintained using an ac electronic bridge along with calibrated platinum and germanium resistance thermometers. The temperature was normally held constant to within ±0.010 K during the measurements.

III. EXPERIMENTAL METHOD

The neutron-inelastic-scattering measurements of the phonons were performed on the triple-axis crystal spectrometer at the Brookhaven High Flux Beam Reactor. A bent pyrolytic-graphite monochromator was used to produce incident neutron beams of 13.5, 5.5, and 5.0 meV for the present

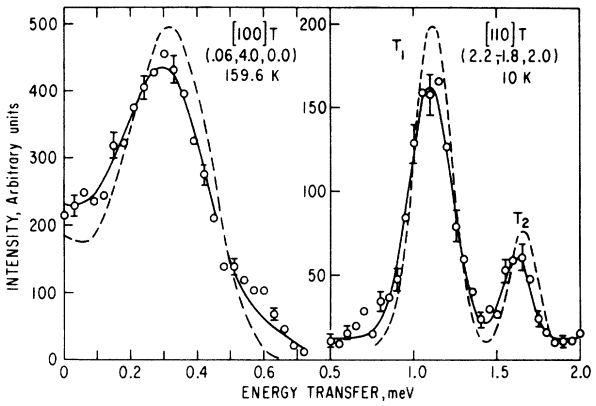

FIG. 1. Examples of the experimental data (circles). The error bars shown represent counting statistics only. The solid lines are Gaussian fits to the data. The dashed lines are resolution-broadened line-shape calculations as discussed in the text. Both measurements shown were taken with an incident energy of 13.5 meV and 20′ collimation.

measurements. A large-mosaic pyrolytic-graphite filter effectively removed the higher-order contamination from the beam at 13.5 and 5.5 meV. At 5.0 meV a refrigerated beryllium filter served the same function. The analyzer was a flat graphite crystal. Various combinations of 20′ and 40′ collimations were used. The constant-Q mode of operation was used for all the measurements. The particular scattering zones available made determination of the [110]T_2 branch extremely difficult at 159.6 K, and our efforts to observe it were not successful. At this temperature, the [100]L branch was also difficult to measure and only a few poorly determined phonons were observed. Because the quality of these latter points was significantly poorer than the other data, they were not used in the analysis. At 111 K, the [110]T_2 branch was also not observed but the [100]L mode was measured. Measurements of the [110]T_2 branch were successful at 10 K, but only for wave vectors larger than 0.14 of the zone boundary. For many of the branches, phonons with wave vectors as low as 0.05 of the zone-boundary wave vector were able to be seen. Many of the phonons were measured at several places in the scattering zone and with more than one incident energy.

All of the experimental data were analyzed using the following procedure: The experimental constant-Q scan was fitted to a Gaussian plus background to determine the phonon energy. The set of such energies and their corresponding wave vectors were used to fit harmonic (Born-von Kármán) atomic force constants allowing first-nearest-neighbor interactions only.[16] These force constants were used to generate resolution-broadened line shapes using a modified version of a program written by Pynn and Werner.[17] Such line shapes can be compared directly with the measurements. Some examples of the measured data, the corresponding Gaussian fits, and the calculated line shapes are shown in Fig. 1. It should be emphasized that the line-shape calculations shown in figures do not make use of any adjustable parameters. All of the instrumental parameters were fixed independently. The calculations were normalized to give the same area as the data.

By comparing the center of the computer-generated peak with the frequency used to generate it, the shift in frequency due to the resolution function of the instrument can be determined. Except at the smallest wave vectors, the shifts observed in this fashion are of the order of the angular uncertainties of the instrument (0.01°). All the experimental data were corrected for the resolution shifts, and the corrected data for the three temperatures measured are given in Table I. One standard deviation of the least-squares-fitted center of the Gaussian tends to underestimate the true uncertainty. Certain instrumental uncertainties are not taken into account by the Gaussian fit; this is borne out by previous experience in fitting models to this type of data.[12] We have therefore taken two standard deviations to represent what we believe is a more realistic indication of the true uncertainty. These are the errors quoted in Table I. The corrected data for the three temperatures are shown in Fig. 2 along with the near-neighbor force-constant fits. The wave vectors quoted in the table and figure are in reduced units, $\zeta = q(a/2\pi)$, where a is the lattice parameter.

IV. RESULTS AND ANALYSIS

Several approaches may be used to get the zero-sound elastic constants from the corrected phonon dispersions at low q. Since the slopes of these spectra represent the sound speeds in the corresponding symmetry directions, the sound speeds may be obtained from the data. Using the well-known relationships between the sound speeds and the elastic constants, the latter may be determined. We have used this approach and fit our corrected data to the form[11]

$$\hbar\omega = \epsilon + v_s q + Bq^3, \tag{1}$$

where ϵ, v_s, and B were determined by a least-squares routine. The intercept ϵ should be zero if there is no systematic error present in the measurements. The parameter B allows for possible nonlinearity in the dispersion relation in the low-q region measured. The sound speeds v_s may then be used to determine the elastic constants using the relations

TABLE I. Phonon energies for xenon (in meV).

ζ[a]	T=159.6 K	T=111 K	T=10 K	ζ	T=159.6 K	T=111 K	T=10 K
	[100]T				[110]T_1		
0.06	0.27±0.01	0.31±0.01	•••	0.05	•••	•••	0.27±0.01
0.07	0.31±0.01	0.36±0.01	•••	0.07	•••	•••	0.37±0.02
0.08	0.34±0.01	0.41±0.01	•••	0.08	•••	0.38±0.01	•••
0.09	•••	0.46±0.01	0.54±0.01	0.09	•••	•••	0.49±0.02
0.10	0.43±0.01	0.51±0.01	•••	0.10	0.37±0.01	0.47±0.01	0.57±0.03
0.12	0.52±0.01	0.62±0.01	0.74±0.01	0.12	•••	•••	0.63±0.02
0.14	0.60±0.01	0.71±0.01	•••	0.14	0.53±0.01	•••	0.76±0.01
0.15	0.65±0.01	0.76±0.01	0.90±0.01	0.15	•••	0.69±0.01	0.82±0.01
0.18	0.79±0.01	0.90±0.01	1.08±0.01	0.17	0.64±0.01	0.77±0.01	0.93±0.01
0.20	0.86±0.01	1.00±0.01	1.19±0.01	0.20	0.76±0.02	•••	1.08±0.03
0.22	0.96±0.02	1.11±0.01	•••	0.22	0.82±0.01	0.98±0.01	1.19±0.01
0.25	1.08±0.02	1.24±0.01	1.44±0.01	0.25	•••	•••	1.35±0.02
	[100]L				[110]T_2		
0.08	•••	0.58±0.02	•••	0.14	•••	•••	1.16±0.04
0.10	•••	0.73±0.01	0.78±0.02	0.15	•••	•••	1.24±0.03
0.12	•••	0.85±0.01	•••	0.17	•••	•••	1.41±0.02
0.15	•••	1.06±0.02	1.19±0.03	0.20	•••	•••	1.62±0.02
0.20	•••	1.41±0.02	1.51±0.04	0.22	•••	•••	1.82±0.02
0.22	•••	1.53±0.02	•••	0.25	•••	•••	2.03±0.04
0.25	•••	1.76±0.03	•••				
	[111]T				[110]L		
0.02	0.12±0.01	•••	•••	0.04	•••	0.45±0.01	0.49±0.02
0.03	0.17±0.01	0.20±0.01	0.24±0.01	0.05	0.48±0.01	0.58±0.01	0.64±0.03
0.04	0.23±0.01	0.27±0.01	0.33±0.01	0.06	0.58±0.01	0.69±0.01	•••
0.05	0.28±0.01	0.34±0.01	•••	0.07	0.71±0.02	0.81±0.01	•••
0.06	•••	0.39±0.01	0.49±0.01	0.08	•••	0.91±0.02	0.96±0.03
0.07	•••	0.47±0.01	•••	0.09	0.90±0.01	1.03±0.01	•••
0.08	0.45±0.01	0.55±0.01	0.58±0.02	0.10	0.95±0.03	1.11±0.01	1.29±0.03
0.10	0.56±0.01	0.66±0.01	0.76±0.02	0.15	1.42±0.04	1.66±0.02	1.88±0.02
0.12	0.67±0.01	0.79±0.01	0.92±0.01	0.20	1.93±0.04	2.17±0.03	2.44±0.02
0.15	0.84±0.01	0.96±0.01	1.11±0.03	0.25	2.35±0.05	2.67±0.03	•••
0.20	•••	1.27±0.01	1.47±0.01				
0.25	•••	•••	1.76±0.02				
	[111]L						
0.03	•••	0.42±0.01	•••				
0.04	0.49±0.01	0.57±0.01	•••				
0.05	0.61±0.01	0.70±0.02	0.83±0.02				
0.06	0.74±0.01	0.87±0.01	0.96±0.02				
0.08	0.96±0.02	1.16±0.02	1.28±0.02				
0.10	1.24±0.03	1.45±0.02	1.64±0.01				
0.12	1.47±0.03	1.74±0.02	1.96±0.02				
0.13	•••	•••	2.11±0.02				
0.14	1.75±0.05	2.00±0.03	•••				
0.15	•••	•••	2.44±0.02				
0.16	1.99±0.03	2.30±0.02	•••				
0.20	•••	•••	3.11±0.03				

[a]The measured lattice parameters of the crystal are: a=6.350±0.002 Å at 159.6 K; a=6.251 ±0.002 Å at 111.0 K; a=6.129±0.002 Å at 10.0 K.

$$
\begin{aligned}
&v^2_{100T} = v^2_{110T_2} = c_{44}/\rho,\\
&v^2_{100L} = c_{11}/\rho,\\
&v^2_{110T_1} = (1/2\rho)[c_{11} - c_{12}],\\
&v^2_{110L} = (1/2\rho)[c_{11} + c_{12} + 2c_{44}],\\
&v^2_{111L} = (1/3\rho)[c_{11} + 2c_{12} + 4c_{44}],\\
&v^2_{111T} = (1/3\rho)[c_{11} - c_{12} + c_{44}],
\end{aligned}
\tag{2}
$$

where ρ is the density.

In most cases, the intercepts ϵ were near zero within the accuracy of the instrumental resolution. The cubic coefficients B were usually small and poorly determined. For those few branches where

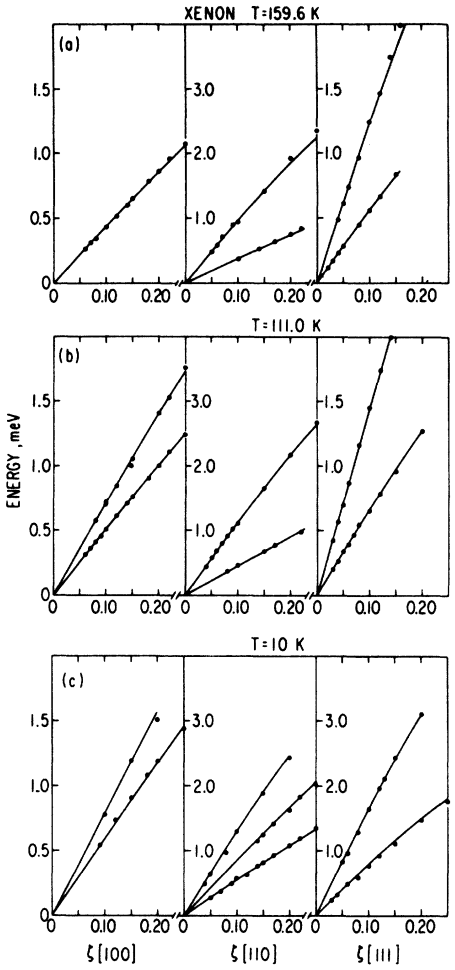

FIG. 2. Corrected low-q dispersion relation for xenon in the three high-symmetry directions. The lines represent near-neighbor harmonic-force-constant fits to the data: (a) 159.6 K, (b) 111.0 K, (c) 10.0 K.

a nonzero value of ϵ was found with reasonable accuracy, the data were corrected; such corrections were always less than 0.01 meV. This procedure yielded the sound speeds which are tabulated in Table II. Also given in the table are the sound speeds at 156 K calculated from the light-scattering measurements of Gornall and Stoicheff[1] using the relations (2). No errors are given for these values since the authors did not report the correlations between elastic constants. The errors quoted for our sound speeds are two standard deviations of the errors given by the least-squares fits.

Since there are generally more sound speeds measured (up to seven for all the high-symmetry directions) than elastic constants (three), the latter are overdetermined. One then calculates the best set of elastic constants in a least-squares fit to Eq. (2).

An alternative approach to get the elastic constants is to fit harmonic force constants to the dispersion relations. We have also used this procedure. Since only the small-wave-vector region was investigated here, a nearest-neighbor model was used. As seen in Fig. 2, a few points at the highest-q values depart slightly from the first-nearest-neighbor fits. The reason for this is that the first-neighbor model is not adequate to describe the data at large wave vectors. Including second and third neighbors would alleviate this difficulty while making little or no change in the elastic constants. At 111 and 10 K, second- and third-nearest-neighbor models were also calculated with the result that there is a slight difference in the quality of the fits, but little change in the elastic constants. The force constants determined from the data have little meaning in themselves, but they may be used to calculate the elastic constants.

The results from the two different approaches to analyzing the data were very close. They have been averaged and the results are given in Table III. The differences in the results from the two procedures are within the errors quoted in the table. Also included are the bulk modulus, the anisotropy, and the departure from the Cauchy relation. The errors quoted for these parameters take into account correlations between the fitted elastic constants.

V. DISCUSSION

It is of interest to compare our results at 159.6 K with the only other experimental measurement of the elastic constants of xenon, those of Gornall and Stoicheff[1] measured at 156 K by Brillouin scattering. The results for c_{44} are very close and within the experimental errors. Our measurement of c_{11} is about 5% lower than the light-scattering result, but this is still within two standard deviations of both experiments. Our value of c_{12} is about 10% lower than the light-scattering value and

TABLE II. Sound speeds in the high-symmetry directions for xenon. The light-scattering results at 156 K have been calculated from the measurements of Gornall and Stoicheff (Ref. 1). The units are m/sec (1 m/sec $= 0.658 \times 10^{-2}$ meV Å).

	159.6 K	156 K (light)	111 K	10 K
$v_{100T} = v_{110T_2}$	666 ± 8	660	779 ± 6	873 ± 20
v_{100L}	...	936	1068 ± 12	1150 ± 36
v_{110T_1}	407 ± 4	367	511 ± 4	568 ± 4
v_{110L}	1060 ± 26	1074	1228 ± 10	1331 ± 40
v_{111T}	494 ± 8	501	587 ± 6	673 ± 14
v_{111L}	1089 ± 6	1115	1259 ± 12	1393 ± 8

TABLE III. Elastic constants (10^8 dyn cm^{-2}) and associated parameters for xenon. The light-scattering results are those of Gornall and Stoicheff (Ref. 1).

Temperature (K)		c_{11}	c_{12}	c_{44}	A	B	δ
159.6	neutron	283 ± 5	173 ± 5	150 ± 2	2.73 ± 0.05	210 ± 5	−0.13 ± 0.03
156	light	298 ± 5	190 ± 4	148 ± 4	2.74 ± 0.30	226 ± 4	−0.22
111	neutron	414 ± 6	240 ± 6	211 ± 4	2.43 ± 0.06	298 ± 6	−0.12 ± 0.03
10	neutron	527 ± 9	282 ± 8	295 ± 4	2.41 ± 0.04	364 ± 8	+0.05 ± 0.04

this is well outside the errors. The interesting point here is that where the neutron scattering and light-scattering measurements disagree, the neutron measured values are lower than the light-scattering values in contrast to what would be expected for a difference between first and zero sound.[6–9] The latter has been attributed to the approximately 10% difference observed between light scattering and neutron scattering in krypton,[10] and in this case the discrepancies are in the expected direction. Allowance must be made for the fact that we are comparing results at slightly different temperatures. However, considering the predicted temperature variation of the elastic constants near melting, this is not expected to explain the differences observed.

The results at lower temperatures can only be compared against theoretical calculations of the elastic constants, of which there are many.[3–5,18] All of these calculations are based on a Lennard-Jones 6-12 potential. It is known that this potential is not satisfactory for describing certain thermodynamic properties, although it is very good for others.[19] Figure 3 shows the data and a number of these calculations. Gornall and Stoicheff's[1] conclusion that their data favor the all-neighbor calculations over the near-neighbor calculations seems to be still valid. At very low temperatures, where the anharmonic effects are small and the quasiharmonic approximation is reasonably good, our data are in fairly good agreement with the all-neighbor calculations.[5,18] Only c_{11} is outside the estimated uncertainty (see Table VI). None of the calculations of the temperature dependence has the shape given by experiment. Admittedly, the shape is not well defined experimentally with so few points; nonetheless, the theories are well outside the estimated errors of the measured points, and not in a systematic way. A Monte Carlo calculation by Klein and Murphy[20] with an all-neighbor Lennard-Jones (6-12) potential but with no account of three-body forces (shown as crosses in Fig. 3) gives rather good agreement at high temperatures, but no results were given for lower temperatures.

The observed disagreement between experiment and theory for the temperature dependence probably has to do with the lack of a good two-body potential. This condition, from which all of the calculations on xenon suffer, makes it difficult to use the experiments as a basis to evaluate the methods for treating anharmonic effects (e.g., quasiharmonic with perturbation theory, self-consistent, improved self-consistent).[21] Potentials for xenon along the lines of those already obtained for argon[22] and krypton[23] would be highly desirable.

Our determination of the bulk modulus, $B_0 = \frac{1}{3}(c_{11} + 2c_{12})$, is summarized in Table IV. If there were no difference between zero and first sound, as at $T = 0$ K, B_0 would be identical to the adiabatic bulk modulus B_s. For comparison we have also shown

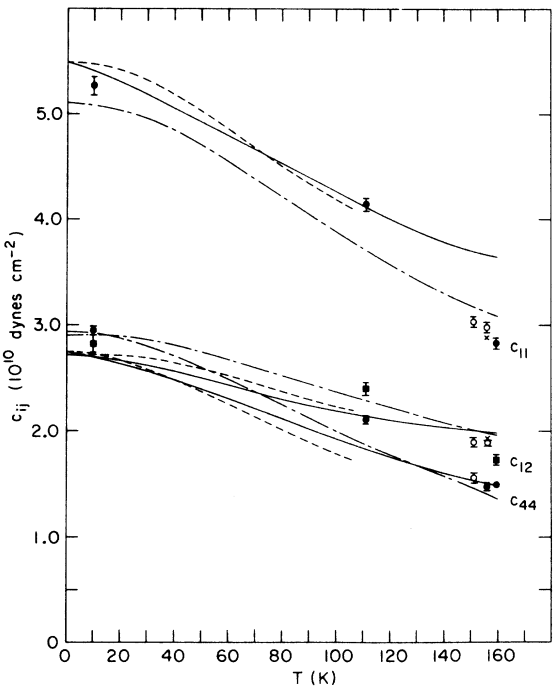

FIG. 3. Temperature dependence of the elastic constants. Solid points are the present measurements; open circles are the Brillouin-scattering measurements (Ref. 1). All the theories use a Lennard-Jones (6-12) potential. The solid line is the near-neighbor self-consistent calculation (Ref. 4); the dashed line is a near-neighbor quasiharmonic calculation (Ref. 3); the dash dot curve is an all-neighbor quasiharmonic result (Ref. 5). The crosses are a Monte Carlo calculation due to Klein and Murphy (Ref. 20).

TABLE IV. Comparison of experimental bulk modulus values (units are 10^8 dyn cm^{-2}).

Temperature (K)	This expert.	Brillouin-light scattering[a]	Ultrasonic (polycryst.)[b]	*PVT* meas.[c]
159.6	210±5	•••	209±13	•••
156	•••	226±4	213±11	•••
111	298±6	•••	273±14	•••
10	364±8	•••	•••	355±20

[a]W. S. Gornall and B. P. Stoicheff (Ref. 1).
[b]P. A. Bezuglyi *et al.* (Ref. 2).
[c]J. R. Packard and C. A. Swenson (Ref. 14).

in the table the light-scattering results and the results of two other measurements, the ultrasonic attenuation of Bezuglyi *et al.*,[2] and the molar volume work of Packard and Swenson.[14] The latter reported isothermal values; thus only their low-temperature results, where the differences between isothermal and adiabatic quantities vanish, may be compared with the other measurements. The ultrasonic measurements were not extended below 40 K and therefore only the higher-temperature values are quoted here. Both other determinations are in good agreement with our results. We made no attempt to compare our high-temperature values to the isothermal measurements of Packard and Swenson[14] since the correction term involves some quantities which are not well known. The light-scattering result for B_s is higher than our value and in the wrong direction to be attributed to differences between zero and first sound.

A measure of the anisotropy of the elasticity is given by the parameter

$$A = 2c_{44}/(c_{11} - c_{12}).$$

In the absence of anisotropy, $A = 1$. Our measurements of the anisotropy (given in Table III) show a slight increase in going from low temperatures to the melting point. The high-temperature result is in very good agreement with the light-scattering result. Remarkably similar numerical values obtain for the other rare-gas crystals near their melting points and at low temperatures. A summary of these is shown in Table V. These experimental values agree well with recent Monte Carlo calculations on argon and krypton.[20]

In the last few years there has been a considerable amount of interest in determining the importance of three-body forces in rare-gas crystals.[24-28] Most of this effort has centered around inclusion of a triple dipolar term of the Axilrod-Teller type[29] in the potential. Its effect on the elastic constants at 0 K has been studied as manifested in the quantity $\delta = (c_{44} - c_{12})/c_{12}$.

Following the suggestion of Lucas,[30] Hüller[31] has recently evaluated c_{ij} and δ including all orders of dipole interactions. His result was that the third-order term appears to be the most important, and δ with all orders included was not very different from δ with only triplet forces, probably because of some cancellations among the higher-order terms. Hüller found, however, that no conclusion could be drawn from a comparison of the existing widely ranging experimental determinations of δ for argon and the theoretical calculations. Another recent calculation by Zucker and Doran[32] has taken into account third-order higher multipolar interactions. They found that the multipolar terms make a significant contribution to the elastic constants and to δ.

However, the difficulty facing all these studies is that in order to compare the results with experimental elastic constants or δ, the many-body portion must be added to the contribution from two-body interactions, and the latter is not precisely calculable. Indeed, different choices for the two-body part lead to very different values for the c_{ij} and only slightly different values for δ. Two-body forces affect the latter only through the zero-point motion and hence the effect of an uncertain two-body potential is minimized in xenon. The results of some of the calculations at 0 K with and without three-body and many-body forces are listed in Table VI. The experimental value of δ reported here suggests that the many-body effects are smaller than the theories propose.

In conclusion, we have presented accurate measurements of the sound velocities and elastic con-

TABLE V. Anisotropy of the elasticity in rare-gas crystals.

Crystal	Reference	*A* Near melting	*B* Low temperature
neon	a	•••	2.78±0.38(4.7 K)
neon	b	•••	2.33±0.08(5.0 K)
neon	c	2.74±0.25(24.3 K)	•••
argon	d	•••	1.90 (4 K)
argon	e	•••	2.42±0.30(4 K)
argon	c	2.80±0.60(82.0 K)	•••
krypton	f	2.76±0.05(114 K)	•••
krypton	g	•••	2.34±0.05(10 K)
xenon	h	2.74±0.30(156 K)	•••
xenon	This work	2.73±0.05(159.6 K)	2.41±0.04(10 K)

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TABLE VI. $T=0$ K Elastic constants, bulk modulus, anisotropy, and δ for xenon.

$\times 10^8$ dyn/cm^2	Experiment (10 K)	Near-neighbor two body[a]	All-neighbor two body[a]	Three body with L-J two body[b]	Three body with exp-6, two body[b]	All-order dipole with L-J two body[c]	All-order dipole with exp-6, two body[c]	Three-body multipolar[d]
c_{11}	527±9	550	511	449	527	472	551	···
c_{12}	282±8	271	289	265	313	278	326	···
c_{44}	295±4	275	293	242	290	259	307	···
A	2.41±0.04	1.97	2.64	2.63	2.71	2.67	2.73	···
B	364±8	364	363	327	385	343	401	···
δ	0.05±0.04	0.015	0.014	−0.092	−0.075	−0.068	−0.059	−0.114

[a]T. H. K. Barron and M. L. Klein (Ref. 18).
[b]A. Hüller, W. Götze, and H. Schmidt (Ref. 27).
[c]A. Hüller (Ref. 31).
[d]I. J. Zucker and M. B. Doran (Ref. 32).

stants in the zero-sound regime for solid xenon. Limited measurements as a function of temperature point out the inadequacy of the theories in describing the temperature dependence. Our results near the melting point when compared against the Brillouin-scattering measurements do not show an expected zero-sound–first-sound difference as reported for krypton. In fact, where the neutron scattering and light-scattering results disagree, the discrepancies are in the opposite direction expected from estimates of this phenomenon. We find our results are consistent with other measurements of the bulk modulus and anisotropy. Furthermore, we find that the experimental departure from the Cauchy relation, δ, at low temperature suggests that previous calculations of many-body effects probably overestimate their importance for this material. Measurements of the complete phonon dispersion relations at 10 K have been completed and will be reported elsewhere.

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