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Differing values of the elastic constants of xenon determined by Brillouin and neutron scattering*

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New Brillouin-scattering experiments using three Xe single crystals confirm earlier light-scattering values of the adiabatic elastic constants near the triple point. The values for C_{11} and C_{12} are found to be *larger* than the corresponding values determined by neutron scattering, in contrast to results obtained for Kr, Ar, and Ne.

A recent comparison[1] of values of the elastic constants of krypton near its triple point, determined by Brillouin[2] and neutron[3] scattering exhibited differences of approximately 10% in C_{11}, C_{12}, and C_{44} (Table I). Since the neutron values were larger than the corresponding Brillouin values, and since the frequency regime probed by the neutron scattering experiments was higher by factor of 10–100, the observed differences were explained as arising from differences in zero and first sound. The signs and magnitudes of these differences were in agreement with theoretical expectations.[4] Earlier calculations by Goldman *et al.*[5] and Niklasson[6] suggested differences of ~5%. Further calculations by Goldman and Klein[7] have produced differences as high as 15% depending on the phonon self-energy.

More recently, values of the elastic constants for xenon,[8] argon,[9] and neon[10] were determined by neutron scattering at temperatures near their triple points, and comparisons with the Brillouin-scattering values[11-13] became possible. For argon and neon, the neutron scattering values were found to be ~5% larger (Table I) and therefore in reasonably good agreement with theory. However, an unexpected result was obtained for xenon; namely, that the light-scattering values were *larger* than those obtained by neutron scattering (by ~5% and 9% in C_{11} and C_{12}, respectively). Since the direction of these differences is at variance with available theory, and with the results for solid Kr, Ar, and Ne (Table I), the experimental values were suspect.

Since there has been considerable improvement in apparatus from the time of the first Brillouin-scattering experiments with xenon by Gornall and Stoicheff[11] in 1971, it was thought worthwhile to repeat the experiment with the new apparatus de-

TABLE I. Comparison of elastic constants for Ne, Ar, and Kr obtained by Brillouin and neutron scattering.

Rare gas	Method	Temperature	Elastic constants (kbar) C_{11}	C_{12}	C_{44}
Neon	Brillouin [a]	24.3	11.70 ± 0.21	7.31 ± 0.17	6.02 ± 0.15
	Neutron [b]	23.7	12.06 ± 0.15	7.32 ± 0.15	6.33 ± 0.09
	$(N-B)/N$ [c]		+3.0%	0.0%	+4.9%
Argon	Brillouin [d]	82.3	23.8 ± 0.4	15.6 ± 0.3	11.2 ± 0.3
	Neutron [e]	82	24.8 ± 0.6	15.3 ± 0.5	12.4 ± 0.4
	$(N-B)/N$		+4.0%	−2.0%	+9.7%
Krypton	Brillouin [f]	115.6	26.57 ± 0.30	17.25 ± 0.20	12.61 ± 0.15
	Neutron [g]	114	28.9 ± 0.4	18.5 ± 0.4	14.4 ± 0.1
	$(N-B)/N$		+8.1%	+6.7%	+12.6%

[a] Reference 13.
[b] Reference 10.
[c] Percentage differences between values obtained by neutron and Brillouin scattering experiments.
[d] Reference 12.
[e] Reference 9.
[f] References 1 and 2.
[g] Reference 3.

TABLE II. Brillouin frequency shifts (in GHz) for three Xe crystals at $T = 160.5$ K.

Crystal orientation[a]			Frequency shifts		
θ	ϕ	χ	L	T_1	T_2
Crystal 1					
−46.3	61.5	148.0	3.897	2.307	⋯
−49.2	82.6	147.1	4.123	1.932	⋯
−46.9	52.3	147.3	3.769	2.489	⋯
−47.7	89.6	147.5	4.232	1.834	⋯
−46.3	98.0	147.2	4.284	1.748	⋯
−46.6	108.5	146.6	4.309	⋯	⋯
−47.0	94.5	147.6	4.238	1.825	⋯
−49.4	84.9	147.0	4.163	1.920	⋯
−45.3	74.9	148.0	4.056	2.045	⋯
−46.4	65.9	147.8	3.965	2.222	⋯
−46.9	58.4	147.5	3.851	2.368	⋯
−47.8	102.6	147.9	4.296	1.796	⋯
Crystal 2					
−81.4	80.9	125.3	3.798	2.403	⋯
−80.9	85.9	125.4	3.803	2.429	⋯
−80.4	92.0	125.3	3.829	2.397	⋯
−83.1	97.5	125.4	3.881	2.330	⋯
−79.6	68.7	126.0	3.892	2.310	⋯
−85.1	60.9	123.7	4.012	2.153	⋯
Crystal 3					
−79.5	−120.8	−55.8	4.158	1.816	⋯
−78.9	−96.0	−57.0	3.955	2.229	⋯
−79.3	−86.2	−57.0	3.859	2.330	⋯
−77.2	−76.6	−57.2	3.802	2.423	⋯
−75.5	−67.4	−57.8	3.790	2.469	⋯
−12.2	5.4	57.8	4.295	1.761	2.263
−12.4	11.4	61.5	4.242	1.735	2.378

[a] θ, ϕ, χ are the Euler angles defining crystal orientation with respect to laboratory axes.

scribed by McLaren *et al.*[13] Three single crystals of Xe at a temperature of 160.5 K were grown from the liquid (in equilibrium with the vapor). As before, Laue x-ray diffraction in transmission was used to check that the crystals were single, and to establish their orientations. Light from a single-frequency Ar^+ laser operating at $\lambda = 5145$ Å was used for excitation, and light scattered at an angle of $89.7 \pm 0.3°$ was analyzed with a piezoelectrically scanned Fabry-Perot interferometer having a free spectral range of 9.479 ± 0.004 GHz.

Brillouin spectra were obtained for several orientations of each crystal, and the Brillouin shifts are listed in Table II. The usual analysis[13] of the combined data from all three crystals, including correlations amongst the elastic constants and their errors, yielded the values of C_{11}, C_{12}, and C_{44} given in Table III.

These values are compared with the earlier results of Gornall and Stoicheff[11] in Table III. It is seen that they agree within the quoted errors. Moreover, when the temperature difference of 5 K is taken into account, calculations by Holt *et al.*[14] (given in the form of graphs) lead to decreases of ~2.5% for C_{11}, ~1% for C_{12}, and ~3.5% for C_{44} at 160.5 K, and the corrected values are shown in parentheses in Table III. It is seen that the results of the two Brillouin-scattering experiments give identical values of the elastic constants.

In Table IV are given values of the speed of sound calculated for high-symmetry directions, using the elastic constants determined here. Included in Tables III and IV, respectively, are values of

TABLE III. Elastic constants and associated parameters for Xe single crystals near the triple point.

Method	Temperature (K)	Elastic constants[f] (kbar) C_{11}	C_{12}	C_{44}	Anisotropy A	Bulk modulus B (kbar)
Brillouin	160.5[a]	29.3 ± 0.4	18.9 ± 0.4	14.1 ± 0.2	2.69 ± 0.09	22.3 ± 0.2
Scattering	156.0[b]	29.8 ± 0.5	19.0 ± 0.4	14.8 ± 0.4	2.74 ± 0.30	22.6 ± 0.4
	(160.5)[c]	(29.1)	(18.8)	(14.3)		
Neutron Scattering	159.6[d]	28.3 ± 0.5	17.3 ± 0.5	15.0 ± 0.2	2.73 ± 0.05	21.0 ± 0.5
$(N-B)/N$[e]		−3.5%	−9.2	+6.0		

[a] Present investigation.
[b] Reference 11.
[c] Values of Ref. 11 corrected to 160.5 K using temperature dependence given in Ref. 14.
[d] Reference 8.
[e] Percentage differences between values obtained by neutron and Brillouin scattering.
[f] At 160.6 K, density = 3.406 g/cm³ (Ref. 15), and refractive index at 5145 Å = 1.4536 (Ref. 16).

TABLE IV. Comparison of sound speeds in high-symmetry directions. The Brillouin scattering values are calculated from the elastic constants given in Table III, and the neutron scattering values are given by the slopes of measured phonon dispersion.

Speed and direction	Brillouin values m/sec	Neutron values m/sec
$v_{100L} = (C_{11}/\rho)^{1/2}$	927 ± 6	...
$v_{100T} = (C_{44}/\rho)^{1/2}$	643 ± 5	666 ± 8
$v_{110L} = [(C_{11} + C_{12} + 2C_{44})/2\rho]^{1/2}$	1059 ± 11	1060 ± 26
$v_{110T_1} = [(C_{11} - C_{12})/2\rho]^{1/2}$	391 ± 14	407 ± 4
$v_{110T_2} = (C_{44}/\rho)^{1/2}$	643 ± 5	...
$v_{111L} = [(C_{11} + 2C_{12} + 4C_{44})/3\rho]^{1/2}$	1099 ± 9	1089 ± 4
$v_{111T} = [(C_{11} - C_{12} + C_{44})/3\rho]^{1/2}$	490 ± 10	494 ± 8

the elastic constants and sound speeds of Xe (at 159.6 K) obtained by Lurie, Shirane, and Skalyo[8] from neutron-scattering experiments. These values were determined by measurements of phonon dispersion curves for transverse and longitudinal modes whose frequencies ranged from 0.12 to 1.1 meV, and 0.45 to 2.4 meV, respectively. Over the lower frequency regions, the dispersion curves were linear, and their slopes were used to derive the longitudinal and transverse velocities, and elastic constants. As in the neutron experiments with Kr, Ar, and Ne, the lowest frequencies probed were considerably higher than those in the Brillouin experiments, and in this case by at least a factor of 15.

A comparison of the present results with the neutron-scattering values gives differences $(N-B)/N$ of -3.5%, -9.2%, and $+6.0\%$ for C_{11}, C_{12}, and C_{44}, respectively. The fact that the anisotropy parameters $A = 2C_{44}/(C_{11} - C_{12})$ for all three studies are uniformly 2.7 is an indication that the crystals were single. Also, the quoted temperatures and vapor pressures are almost the same. Nevertheless, the discrepancy of larger values for C_{11} and C_{12} obtained by Brillouin scattering in comparison with those obtained by neutron scattering, persists, and is not understood at the present time.

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