Observed Differences in Zero- and First-Sound Propagation in Solid Krypton*

Howard E. Jackson,† D. Landheer,‡ and B. P. Stoicheff
Department of Physics, University of Toronto, Toronto M5S 1A7, Canada
(Received 21 May 1973)

The elastic constants of krypton single crystals near the triple point determined from neutron-scattering measurements by Skalyo and Endoh and from our Brillouin-scattering measurements differ by as much as 12%. It is suggested that these large differences arise from the different velocities of zero and first sound.

We report here differences of ~ 10% in the values of the elastic constants of solid krypton near its triple point as determined by inelastic scattering of neutrons and by Brillouin scattering. These large differences are interpreted to be due to the dissimilar propagation speeds of zero and first sound in this temperature region of large anharmonicity.

Sound waves of frequency ω such that $\omega\tau_{th} \ll 1$ (where τ_{th} is an average thermal-phonon lifetime) propagate in the adiabatic or first-sound regime; at higher frequencies such that $\omega\tau_{th} \gg 1$, propagation is termed zero sound. Cowley[1] and Maris[2] have shown that anharmonic interactions affect sound-wave propagation differently in the two regimes. The velocity of zero sound is expected to be slightly faster than that of first sound[1,2] with the largest differences occurring under conditions of maximum anharmonicity. Previous workers have detected slight differences in zero- and first-sound velocities, in qualitative agreement with theory. Svennson and Buyers[3] have observed differences in the temperature dependence of the elastic constants of KBr as deduced from neutron-scattering and ultrasonic measurements. Blinick and Maris[4] have observed such differences in the velocities of sound in quartz between 35 and 1000 MHz.

Crystals of the rare gases would appear to be ideal for such investigations since they are known to be strongly anharmonic near their melting points.[5,6] The two experiments whose results are compared here were carried out on krypton single crystals, close to the triple point. One is the careful measurement by Skalyo and Endoh[7] of phonon dispersion curves for longitudinal and transverse modes of krypton at $T = 114$ K, using inelastic neutron scattering. Their measurements were carried out at small wave vectors, and in the frequency range 25 to 250 GHz. The dispersion curves were found to be linear in this region, and their slopes were used to obtain the values of the zero-sound elastic constants given in Table I.

The second experiment is a study of the Brillouin spectrum of krypton single crystals at $T = 115.6 \pm 0.2$ K carried out in this laboratory. The techniques used have been fully described by Gornall and Stoicheff[6] and by Gewurtz *et al.*[8] in their determination of the elastic constants of xenon, argon, and neon single crystals. Two

TABLE I. Comparison of elastic constants and sound velocities for solid Kr determined by Brillouin and neutron scattering.

	c_{11}	c_{12} (10^{10} dyn/cm^2)	c_{44}	V_L (100)	V_L (110)	V_T (100)	V_{T_1} (110) (10^5 cm/sec)
Brillouin scattering[a] (at 115.6 K)	2.657 ± 0.030	1.725 ± 0.020	1.261 ± 0.015	0.974	1.11	0.671	0.408
Neutron scattering[b] (at 114 K)	2.89 ± 0.04	1.85 ± 0.04	1.44 ± 0.01	1.02	1.17	0.718	0.431
Difference $(N-B)/N$	+8.3%	+7.0%	+12.6%	+4.5%	+5.1%	+6.5%	+5.3%

[a] A refractive index $n = 1.3487$ at 4880 Å and 115.6 K [A. C. Sinnock and B. L. Smith, Phys. Rev. 181, 1297 (1969)] and density $\rho = 2.794 \pm 0.002$ g/cm^3, at 115.6 K [P. Korpiun and H. J. Coufal, Phys. Status Solidi (a) 6, 187 (1971)] were used in the determination of these constants.

[b] Ref. 7.

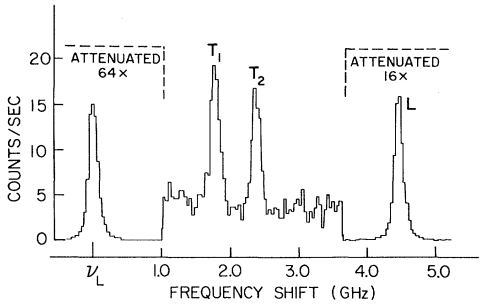

FIG. 1. Typical Brillouin spectrum of a Kr single crystal at a temperature of 115.6 K. Excitation was by 7 mW of laser radiation at 4880 Å.

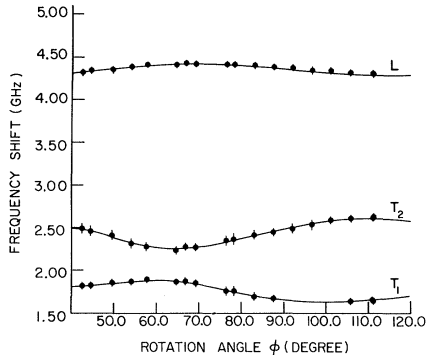

FIG. 2. Observed and calculated frequency shifts versus rotation angle ϕ (about the vertical axis of the crystal). The indicated errors on the experimental points include inaccuracies in measurements of orientation angles as well as of frequency shifts. Curves, calculated values based on elastic constants (Table I) derived from measured frequency shifts.

krypton crystals were grown from the liquid in equilibrium with the vapor. Laue x-ray transmission photographs were used to check that they were single crystals and to establish their orientation. Light (of $\lambda = 4880$ Å) scattered at 90° was analyzed with an interferometer, and the data were collected using photon-counting electronics and a multichannel analyzer.

A typical spectrum is reproduced in Fig. 1. It shows all three Brillouin components possible for a face-centered cubic crystal, one arising from scattering by longitudinal phonons (L) and two less-intense components due to transverse phonons (T_1, T_2). Such a spectrum from a cubic crystal with known orientation is sufficient to evaluate the three elastic constants c_{11}, c_{12}, c_{44}. In order to increase the precision of the constants, Brillouin spectra were recorded for eighteen different orientations (about a vertical axis) over a range of ~ 70° for each crystal. The measured values of the frequency shifts for one of the crystals are plotted against the rotation angle in Fig. 2. In all, 91 frequency shifts ranging from 1.6 to 4.4 GHz were measured for the two crystals. A least-squares analysis of these 91 independent measurements was used to obtain values of the elastic constants, resulting in the good fit to the experimental data shown by the smooth curves in Fig. 2. This analysis yielded the values

$$c_{11} = 2.657 \pm 0.030,$$

$$c_{12} = 1.725 \pm 0.020,$$

$$c_{44} = 1.261 \pm 0.015$$

(in 10^{10} dyn/cm^2) for Kr at 115.6 K.

A comparison of the two sets of elastic constants obtained from the Brillouin- and neutron-scattering measurements is made in Table I. The differences are unexpectedly large, ranging from 7% to as high as 12%. The differences in the corresponding values of phonon velocities calculated for several symmetry directions, and included in Table I, are ~ 5%. It should also be noted that the neutron-scattering values are higher than those determined from the Brillouin spectra. At the present time it is not certain that the neutron-scattering measurements by Skalyo and Endoh[7] (~ 25 to 250 GHz) are in the zero-sound regime, nor that the Brillouin scattering data (1.6 to 4.4 GHz) reported here refer to the first-sound regime. Nevertheless, the two frequency regions differ by more than a factor of 10, and the resulting differences of the elastic constants and velocities are in the direction expected for zero and first sound.[1,2]

We may compare the present results with recent theoretical estimates of differences in the elastic constants for the two frequency regimes. Goldman, Horton, and Klein[9] have calculated the respective constants for krypton using a self-consistent phonon theory, and find differences of < 4%. In a calculation on argon at the triple point, Niklasson[10] has suggested differences of the same order, ~ 4%. Since the difference in zero- and first-sound velocities depends strongly on the anharmonic part of the interaction potential, the discrepancy with the present values may arise both from anharmonic contributions and from inadequacies in the potentials used in the calculations.

In conclusion, the direct comparison of the elastic constants of solid krypton obtained from neutron- and light-scattering experiments has revealed large differences in the corresponding values. The observed differences are in the direc-

tion of the expected differences between zero- and first-sound elastic constants (and velocities) and indicate that the frequency regions of the experiments 25 to 250 GHz and 1.6 to 4.4 GHz are in or near the zero- and first-sound regimes, respectively. The large magnitude of the zero- and first-sound differences in solid krypton near the triple point, however, remains to be explained.

We wish to thank J. Skalyo, Jr., and Y. Endoh for informing us of their results prior to publication, and R. A. McLaren for assistance in this experiment.

*Research supported in part by the National Research Council of Canada, and the University of Toronto.

†Post-Doctorate Fellow 1971–1973. Present address: Department of Physics, McMaster University, Hamilton, Ontario.

‡Holder of a National Research Council of Canada "1967 Science" Scholarship, 1969–1973.

[1]R. A. Cowley, Proc. Phys. Soc., London 90, 1127 (1967).

[2]H. J. Maris, Phil. Mag. 16, 1331 (1967).

[3]E. C. Svennson and W. J. L. Buyers, Phys. Rev. 165, 1063 (1968).

[4]J. S. Blinick and H. J. Maris, Phys. Rev. B 2, 2139 (1970).

[5]See, for example, A. C. Holt, W. G. Hoover, S. G. Gray, and D. R. Shortle, Physica (Utrecht) 49, 61 (1970).

[6]W. S. Gornall and B. P. Stoicheff, Phys. Rev. B 4, 4518 (1971).

[7]J. Skalyo, Jr., and Y. Endoh, Phys. Rev. B 7, 4670 (1973).

[8]S. Gewurtz, H. Kiefte, D. Landheer, R. A. McLaren, and B. P. Stoicheff, Phys. Rev. Lett. 29, 1454, 1768 (1972).

[9]V. V. Goldman, G. K. Horton, and M. L. Klein, Phys. Rev. B 4, 567 (1971).

[10]G. Niklasson, Phys. Kondens. Mater. 14, 138 (1972).

Reflection of ^{4}He Atomic Beams from Clean LiF Surfaces*

Douglas E. Houston and D. R. Frankl

Department of Physics, The Pennsylvania State University, University Park, Pennsylvania 16802

(Received 25 May 1973)

We have studied reflection of a ^{4}He nozzle beam from a LiF crystal surface cleaved *in situ* at 10^{-9} Torr. As compared with previous results on water-covered surfaces, the selective adsorption intensity minima are pronouncedly sharpened and deepened and there is considerable fine structure. The main minima can be associated with three bound states and two positive-energy resonances. On aging the surface in the vacuum, the fine structure fades and the bound-state energy levels shift slightly.

Atomic-beam scattering from the [100] cleavage plane of LiF has been extensively studied in the past.[1-6] These studies were made at pressures of 10^{-8} Torr or higher on surfaces that had been prepared in various ways such as cleaving in air followed by heating in the vacuum,[4-6] cleaving in a stream of nitrogen,[3] and cleaving in the vacuum.[2] As O'Keefe *et al.*[3] and others have pointed out, there is strong evidence[7] that all these surfaces were covered by several monolayers of water.

We have studied the scattering of ^{4}He beams from LiF cleaved *in situ* at about 10^{-9} Torr, the background gases being mainly H_2 and CO. A schematic diagram of the apparatus is shown in Fig. 1. The beam has a velocity spread of about 10% and an angular spread of about 0.1 deg, with a diameter of $\frac{3}{4}$ mm at the sample. The crystal, a rod 5 mm×5 mm×50 mm, can be advanced along its length and cleaved repeatedly without breaking the vacuum. The crystal was initially γ

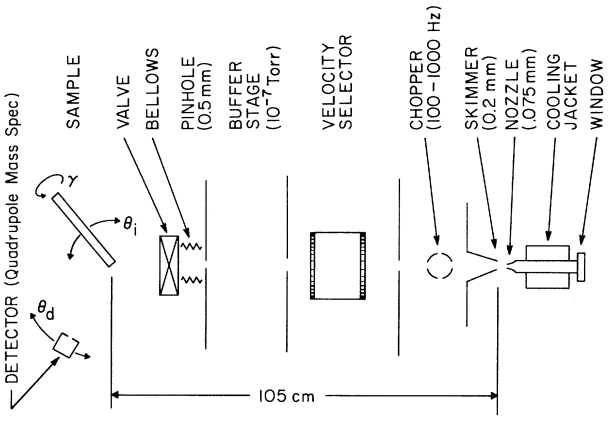

FIG. 1. Schematic diagram of apparatus. The nozzle exhaust chamber is pumped by a 600-liter/sec diffusion pump, the three subsequent chambers by 50-liter/sec diffusion pumps, and the scattering chamber by a 260-liter/sec turbomolecular pump backed by a 50-liter/sec diffusion pump.