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Phonon Dispersion Measurements on a Krypton Single Crystal*

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ABSTRACT

Phonon dispersion relations for the symmetric [100], [110], and [111] branches in fcc krypton have been measured by triple-axis neutron spectrometry. Measurements were carried out at 79°K on a single crystal sample grown from the melt at a pressure of 2.31 kbar.

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The phonon dispersion relations in fcc krypton have been measured on the triple-axis spectrometer at the Brookhaven High Flux Beam Reactor. The single crystal used for the experiment was grown from the melt at a pressure of 2.31 kilobars in an aluminum alloy pressure cell incorporating a nucleation tip at the bottom. The cell was cylindrically shaped with an inside diameter of 12 mm and an outside diameter of 47 mm. The growth process was carried out in a temperature-controlled dewar with heaters appropriately placed so as to prevent blockage of the high pressure tubing and to maintain a suitable temperature gradient along the sample holder. After complete solidification the sample was annealed for three weeks at 166^{o}K, i.e. about 10^{o} below the 2.31 kbar melting temperature, and then was cooled to 79^{o}K, the temperature at which the dispersion curve data were taken. This process yielded a crystal with a measured lattice parameter of 5.725 ± 0.010 Å. On the basis of this lattice parameter, compressibility data, and previously reported values of the lattice parameter at 79^{o}K and 1 atmosphere,[1] it is estimated that the sample pressure decreased to about 0.3 kbars in cooling to 79^{o}K.

The size of the krypton crystal, estimated by beam masking experiments, was approximately 8 x 8 x 11 mm^3. The small size of the crystal, for an inelastic neutron scattering experiment, has somewhat limited the accuracy obtainable in the present investigation. Th quality of the crystal was very good, however, as indicated by the measurement of a Bragg rocking curve with a full width at half maximum of only 7'. This high degree of perfection, which is of some interest in itself, is probably due not only to the annealing treatment, but also to the high pressure growth technique itself. Note that in this method of crystal preparation, in which the solidified rare gas fills an entire cell at a high pressure, the high vapor pressure exhibited by such crystals near their melting temperatures generates no additional problem in containment of the crystal during a long term high temperature annealing process. Moreover, it is felt that the reason the crystal was not damaged on cooling slowly from 166°K down to 79°K is that the pressure on the sample never becomes negative in that interval.

This apparent absence of crystal damage on cooling a solidified rare gas is consistent with the observations of Peterson, Batchelder and Simmons[2] on crystals of argon grown and handled in an essentially stress-free manner within a thin-walled mylar tube, but is in contrast with the behavior of crystals of solidified rare gases grown in inconel tubes by White and Woods[3] at pressures near 1 atmosphere. In the latter case, thermal conductivity data indicated the crystals to be quite imperfect. This could well have been due to crystal damage in pulling away from the tube, because of the large thermal contraction of the solidified gas relative to inconel. A positive external pressure at the solidus eliminates this effect, however, unless the sample is cooled down to a temperature at which the sample pressure passes through zero. It should be possible to grow krypton crystals at some higher pressure and cool them at constant density to $0^{o}K$ with no danger of pressure reversal.

The measurements reported here were carried out at several incident neutron energies between 18 and 35 meV. Typical neutron groups corresponding to the momentum transfer at the zone boundaries gave neutron counts of approximately 3/min above a backround of 2/min. Better signals were obtained for the neutron groups at smaller q values, however. The points in Figs. 1 and 2 contain the experimental dispersion relations obtained for the [100], [110] and [111] branches at a temperature of $79^{o}K$. The [110] axis of the crystal lay about 15^{o} away from the dewar axis, and this somewhat inconvenient orientation rendered it impossible to observe the transverse branch propagating along [110] polarized in the $[1\bar{1}0]$ direction. Since no data exist at present for the elastic stiffnesses of krypton, it is not possible to compare initial slopes of the dispersion curves with those associated with velocity of long wavelength acoustic disturbances.

The experimental work reported on krypton in this letter is being continued with the following objectives: (1) to obtain improved values of the phonon energies proper, (2) to measure at constant volume the anharmonic shift of phonon energies with temperature, and (3) to measure the quasi-harmonic volume dependencies of phonon energies. In carrying out (3), inelastic neutron scattering studies are planned on crystals grown at a series of pressure points, exploiting thereby the change of density of the solid along the melting line. To achieve an acceptable level of accuracy in a reasonable length of experimental time, it will first be necessary to prepare larger single crystals than the one used in the present work.

A detailed analysis of this experiment and its results will be published later.

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References

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2. O.G. Peterson, D.N. Batchelder, and R.O. Simmons, J. Appl. Phys. 36, 2682 (1965).

3. G.K. White and S.B. Woods, Phil. Mag. 3, 785 (1958).

Figure Captions

Fig. 1. Phonon dispersion relations of Kr at $79^{\circ}K$ along [001] and [111] directions. The conversion factor from meV to the frequency (10^{12} cps) is 0.242.

Fig. 2. Phonon dispersion relations of Kr at $79^{\circ}K$ along [110] direction. The transverse mode shown has its polarization vector in the[100] direction.

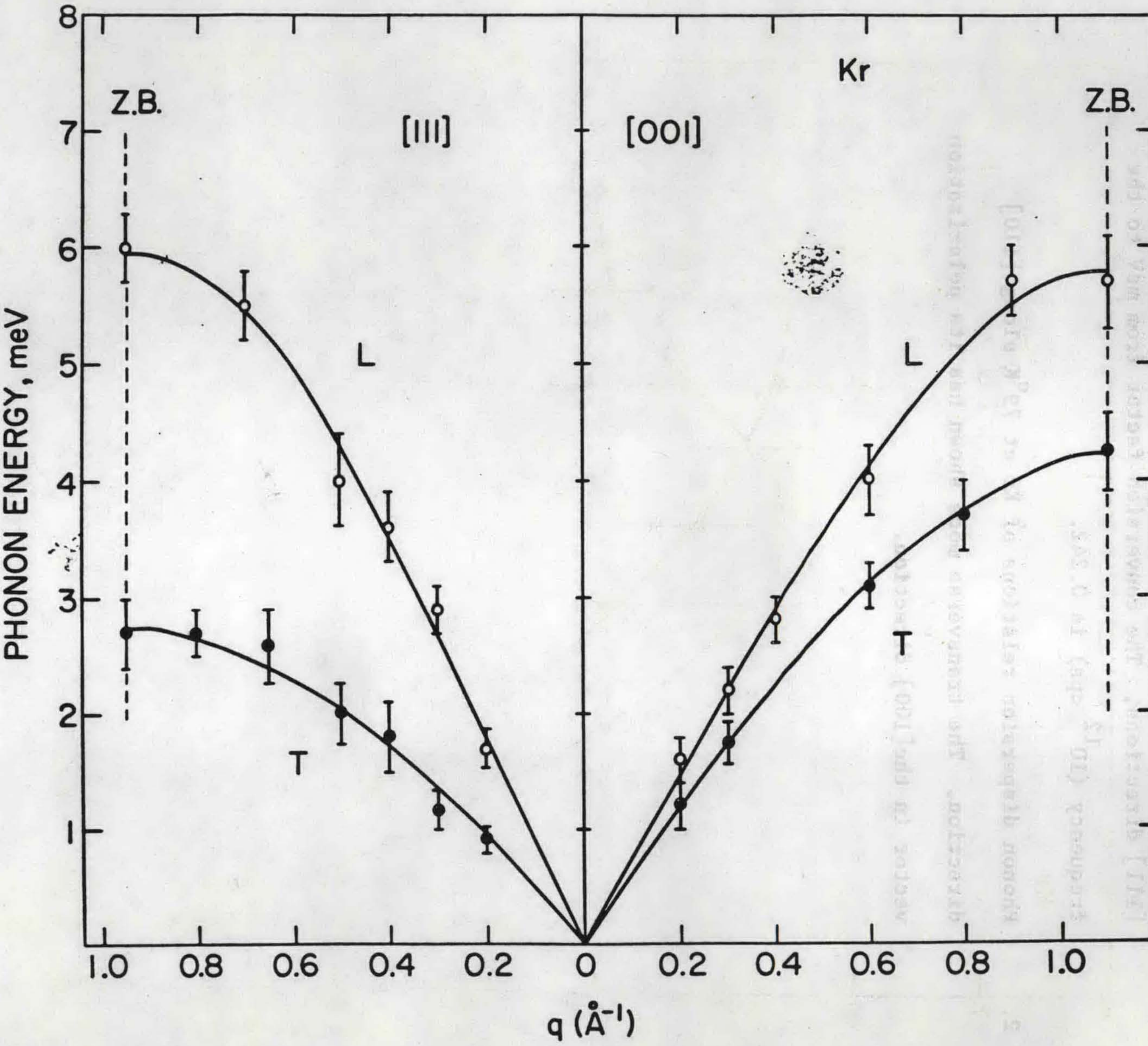
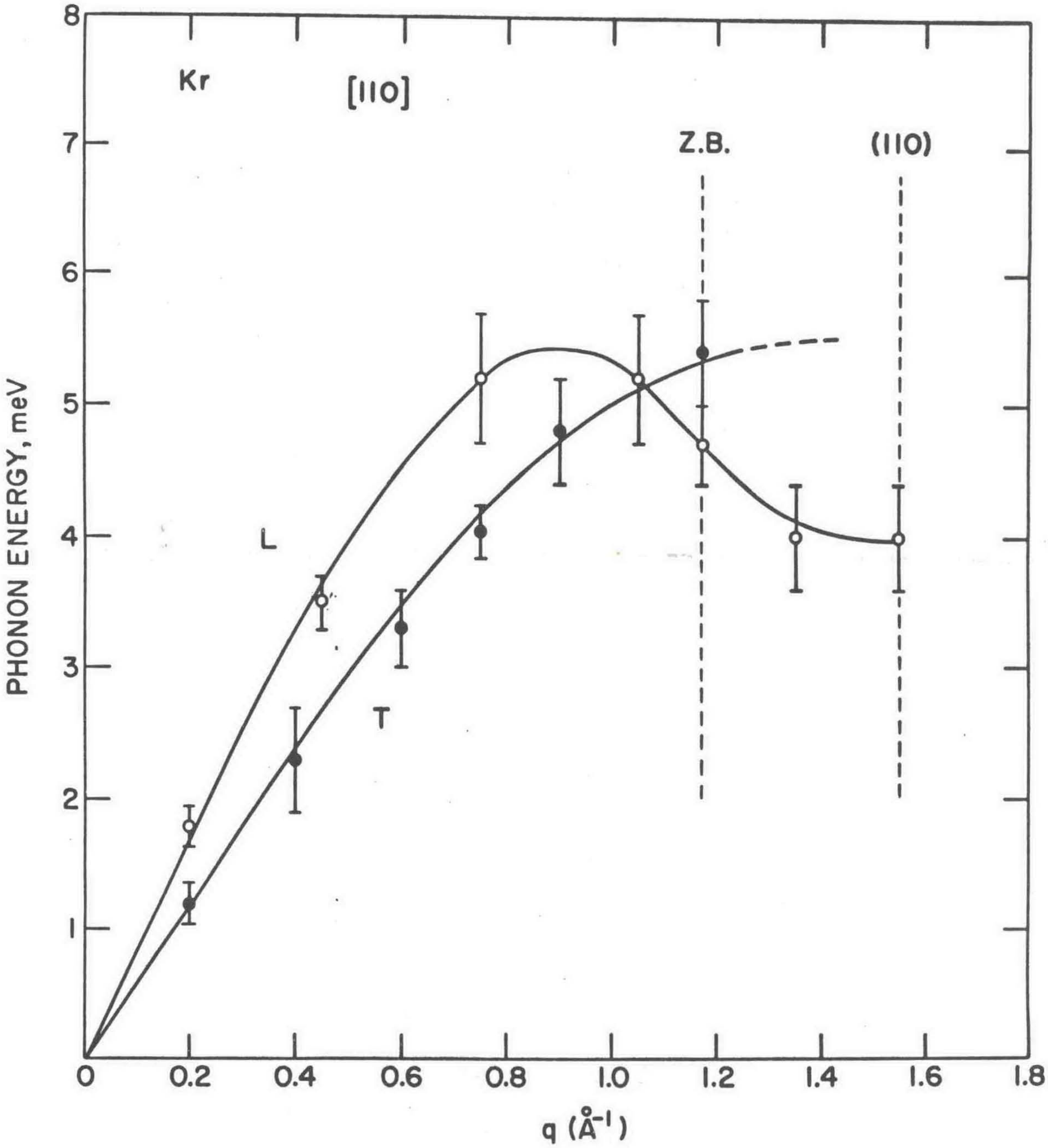


FIGURE 1



Kr
[110]
Z.B.
(110)
L
T
PHONON ENERGY, meV
q (Å^{-1})
0
0.2
0.4
0.6
0.8
1.0
1.2
1.4
1.6
1.8
1
2
3
4
5
6
7
8

FIGURE 2