Phonon dispersion of indium along [111]

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The phonon spectrum of indium along [111], measured by inelastic neutron scattering, is reported. The two shear modes at the zone-boundary point $(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$ are split slightly (on account of a 7.5% tetragonal distortion). They have very low frequencies, ~0.7 and 1.0 THz, compared to the longitudinal mode, ~3.4 THz. These measurements verify the theoretical dispersion predicted by the dynamic pseudopotential theory of phonons for free-electron-like metals.

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Measurement of the phonon spectrum of indium by inelastic neutron scattering is difficult because of the large thermal-neutron absorption cross section (194 b). Nevertheless, data along [100], [001], [110], and [101] have been presented.[1] (The crystal structure of In is face-centered tetragonal; i.e., were it not for a 7.5% tetragonal extension along its c axis, In would be fcc.) Finding the experimental dispersion along [111] is the purpose of this study. It is of special interest on account of the recent discovery of anomalous x-ray diffraction peaks at half-integral (hkl) points in reciprocal space.[2]

The thermal expansion of In along its c axis is extremely anomalous[3] (i.e., it is negative above 280 K). A search for a

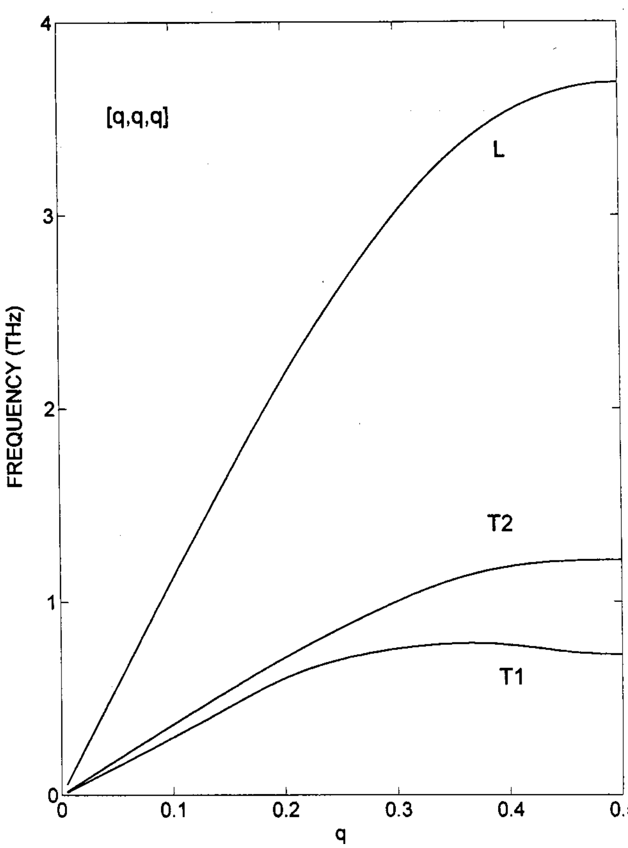

FIG. 1. Predicted phonon dispersion in In along [qqq], calculated in Ref. 2 and based on the dynamic pseudopotential model (Ref. 4), applied to In (Ref. 6). (1 THz = 4.136 meV/h).

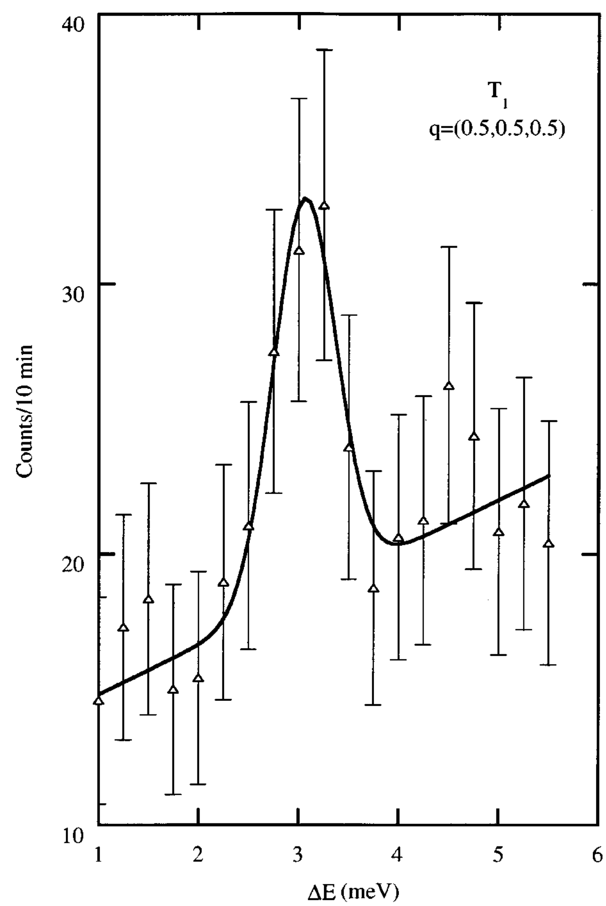

FIG. 2. Inelastic scattering caused by the T_1 phonon at $(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$, measured at $(-\frac{3}{2}, \frac{5}{2}, \frac{1}{2})$.

TABLE I. Crystal geometry and spectrometer characteristics. Monochrometers and analyzers were vertically focusing. Scan range: $[0<q\leqslant 0.5]$. Collimation for the first two points of the T_1 branch was (40-20-20-40).

	Surface	Scan	Monochrometer	Analyzer	Collimation (min)	E_f (meV)	T (K)
T_1	[−1, 1, 0]	$(-2+q,2+q,q)$	PG(002)	PG(002)	40-40-40-40	14.56	150
T_2	[0, 0, 1]	$(q,q,2+q)$	Be(101)	PG(002)	48-60-80-70	14.8	150
L	[1, 1, 1]	$(2+q,2+q,2+q)$	Be(101)	PG(002)	48-60-40-240	30.5	75

possible broken symmetry was inconclusive because the intensities of the half-integral x-ray peaks were found to be proportional to *T*. Consequently they arise from thermal-diffuse scattering (TDS) by phonons. The theory of such a satellitelike structure, based on the dynamic pseudopotential theory of phonons in free-electron-like metals,[4,5] agrees with the observed structure.[2] There was no need to readjust the three parameters of the lattice-dynamics model[6] that were chosen to fit the published phonon data.[1]

The predicted phonon dispersion[2] along [111] is shown in Fig. 1. The splitting of the two shear modes, T_1 and T_2 (which would be degenerate in a fcc crystal) is, of course, caused by the tetragonal distortion. What is unexpected is the large ratio, ~4, of the *L* mode to the *T* modes at $(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$. This ratio is ~2 in many fcc cubic metals:[5] Cu, Ag, Au, Ca, Sr, Yb, and Al. It is clear from Fig. 1 that x-ray scans along [111] near $(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$ cannot give rise to sharp TDS peaks; and none was found.[2] Only scans along paths through half-integral (*h*,*k*,*l*), nearly parallel to the hexagonal zone face, centered at (*h*,*k*,*l*), display sharp TDS peaks. These are caused by a sharp minimum of the T_1 mode along such a path.[7] [TDS is proportional to $\omega(q)^{-2}$; so it exhibits a sharp maximum where the phonon frequency $\omega(q)$ has a sharp minimum.] It is important, of course, to verify experimentally the predicted $\omega(q)$ along [111], given in Fig. 1.

Separate crystals of In were cut (using a South Bay acid saw and a chromic acid gentle etch) for measuring each phonon branch (T_1, T_2, L) in order to minimize neutron absorp-

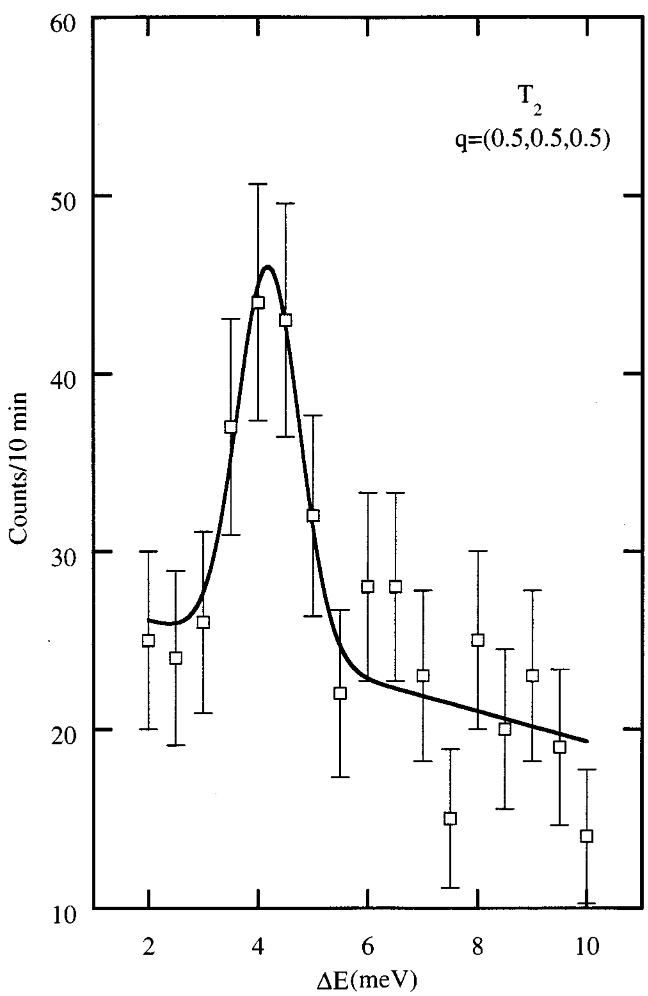

FIG. 3. Inelastic scattering caused by the T_2 phonon at $(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$, measured at $(\frac{1}{2}, \frac{1}{2}, \frac{5}{2})$.

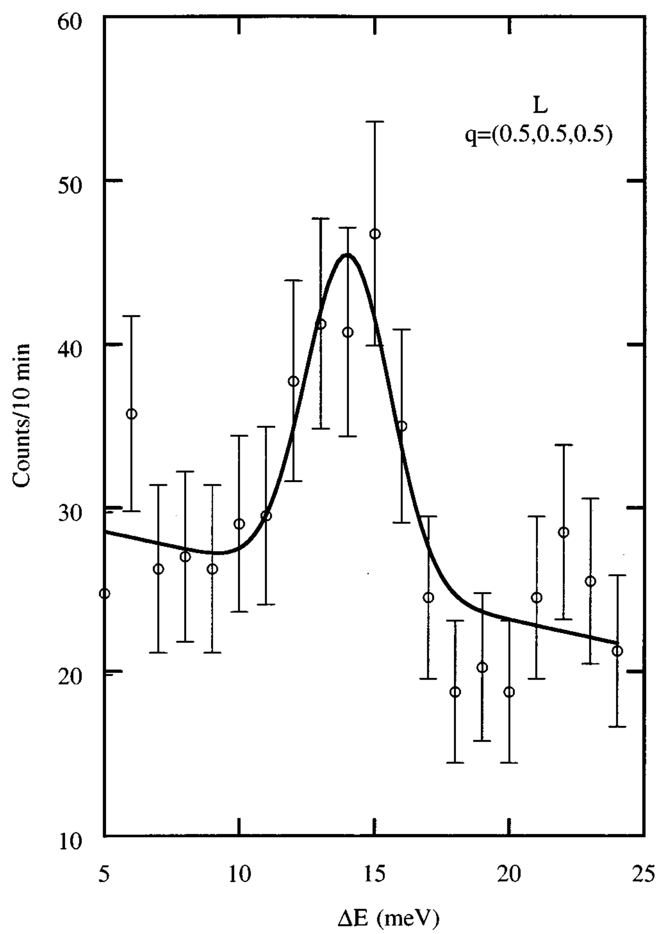

FIG. 4. Inelastic scattering caused by the *L* phonon at $(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$, measured at $(\frac{5}{2}, \frac{5}{2}, \frac{5}{2})$.

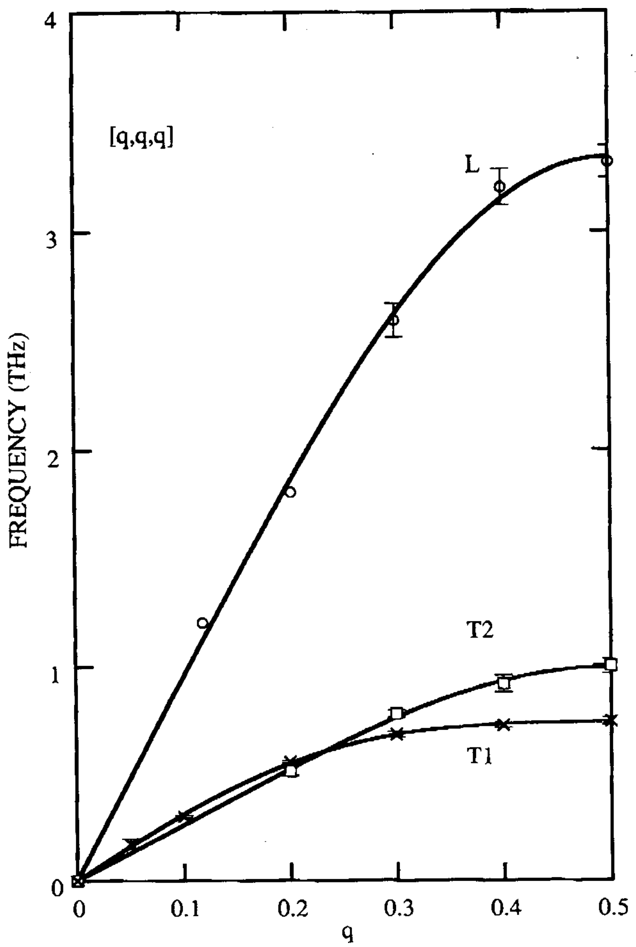

FIG. 5. Measured phonon dispersion of In along [*qqq*]. Technical details of the measurements are given in Table I. (1 THz $=4.136$ meV/h).

tion and to isolate each branch by taking advantage of the $(\hat{\epsilon}\cdot\mathbf{Q})^2$ factor in the scattering intensity. ($\hat{\epsilon}$ is the phonon polarization vector, and **Q** is the neutron-scattering vector.) The T_1 branch was measured with the triple-axis spectrometer at the University of Missouri Research Reactor. The T_2 and L branches were measured with the Oak Ridge National Laboratory HB-2 spectrometer. Sample geometry and spectrometer settings are given in Table I. Constant **Q** scans were employed except for the first two points of the L branch (which were constant ΔE).

TABLE II. Phonon frequencies, $\hbar\omega$ (in meV), of the T_1, T_2, and L branches along [*qqq*]. ($4.136\ \mathrm{meV}/h = 1$ THz).

q	$\hbar\omega$, T_1	$\hbar\omega$, T_2	$\hbar\omega$, L
0.05	0.54		
0.1	1.249		
0.114			5.00
0.2	2.302	2.302	
0.203			7.50
0.3	2.855	3.276	10.786
0.4	2.999	3.865	13.399
0.5	3.049	4.192	14.072

The inelastic scattering peaks at $q=(\frac{1}{2},\frac{1}{2},\frac{1}{2})$, the center of the Brillouin zone's hexagonal face, are shown in Figs. 2, 3, and 4. The solid curves are optimized fits for a single gaussian on a sloping background. The dispersion data for all three branches, tabulated in Table II, are displayed in Fig. 5. The curves through the data are fits to

$$\hbar\omega(q)=A\sin(\pi q)+B\sin(3\pi q).$$

The A's are 3.37, 3.95, and 13.55; and the B's are 0.32, -0.16, and -0.31 (for the T_1, T_2, and L modes, respectively, all in meV).

Comparison of Fig. 1 to Fig. 5 shows that the dynamic pseudopotential model[2,6] successfully predicts the main features of the phonon dispersion along [111]. In particular the unusual large ratio ($\sim$4) of the longitudinal to the transverse frequencies at ($\frac{1}{2}$, $\frac{1}{2}$, $\frac{1}{2}$) is confirmed.

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[7] Reference 2, Fig. 5.