enhanced either by shining light or external injection. However, hot-electron experiments using these techniques are not yet reported. The conditions at room temperature may thus be considered marginal for a successful experiment as suggested in this paper.

At 77 °K, on the other hand, the collision frequency is increased about ten times. The condition $\omega\tau \gg 1$ may be satisfied by choosing the signal frequency one order lower. The wavelength for the signal is then required to be smaller than 370 μ. Since the Faraday rotation increases as λ^2, we find that the rotation angle is increased by a factor of 10^2. Hence, experimental requirements will be more easily satisfied at 77 °K and the suggested experiment should be considered feasible at this temperature. It is also at this temperature that BNDC has been obtained in germanium, and the results of the experiment would be of particular interest.

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Phonon Dispersion Relations in Ge at 80 °K

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Phonon dispersion relations in Ge have been measured at 80 °K for all principal symmetry directions and on some lines at the boundary of the Brillouin zone. The measurements were performed by neutron inelastic scattering using a three-axis crystal spectrometer. In order to achieve maximum accuracy, care was taken to reduce resolution widths as far as possible. In general, the estimated uncertainties of the measured phonon frequencies range from 0.3% to 0.5% for optical and from 0.3% to 1% for acoustic phonons. Frequencies of 238 phonons are tabulated and phonon linewidths are given for the symmetry directions.

INTRODUCTION

Ge is an obvious material for phonon spectrometry with neutrons since it is a good representative for the covalent bond and diamond structure and is available in excellent large crystals with quite good coherent neutron scattering properties. Moreover, its technical importance lends it a special interest, particularly as regards thermodynamic properties related to the phonon frequency distribution. In fact, it was among the first substances to be studied in the early days of neutron measurements. In 1958 Brockhouse and Iyengar[1] published phonon dispersion curves for the [100] and [110] directions obtained by crystal spectrometry, and in 1959 Ghose *et al.*[2] presented time-of-flight measurements for the [100] and [110] directions. The development of the neutron method now allows us to

achieve considerably greater accuracy and completeness. The present paper presents phonon frequencies at 80 °K for all branches along most of the symmetry lines in Ge, with an accuracy usually better than 0.5%, as well as phonon widths. The fact that the measurements are at 80 °K, while the Debye temperature is about 360 °K and the Grüneisen constant is as low as 0.4, should mean that anharmonic effects on the mean phonon frequencies are negligible.

The early measurements on Ge stimulated a considerable amount of theoretical and experimental work on the lattice dynamics of crystals possessing the diamond structure. Phonon dispersion curves have been determined experimentally in diamond, Si and α-Sn. Dolling[3] reported Si dispersion relations whose qualitative features are in near agreement with those obtained in the present experiment on Ge and with those found by Price and Rowe for α-Sn,[4] but the measurements on diamond by Warren *et al.*[5] reveal a large difference between the lattice dynamics of this crystal and the other three. In order to fit the experimental results of Brockhouse and Iyengar, Herman[6] showed that a Born-von Kármán model must include at least sixth-nearest-neighbor interactions, and that the physical significance of many of the 20 parameters of this model is doubtful. Cochran[7] applied the shell model to Ge (Dick and Overhauser[8]), and improved versions of this have successfully described phonon dispersion relations in both diamond structures and alkali halides. One of the purposes of our measurements on Ge was to perform a detailed test of the accuracy of these models, even off the symmetry directions. This will be reported in a separate paper.

EXPERIMENT

The experiment was carried out at the 30-MW research reactor in Studsvik. Our instrument, a three-axis crystal spectrometer has been described elsewhere.[9] In order to keep the reflectivity of the analyzer crystal and the efficiency of the BF_3 counter constant, the setting of the analyzer was kept fixed during the scan of a phonon resonance. All phonons were measured with neutron energy loss and using constant-κ programs.[10]

Much attention was paid to focusing and resolution.[11] The optimum conditions for a measurement were obtained by consulting a computer (a PDP-8/I). The associated wave vector, expected energy, and energy surface gradient of the phonon to be measured were fed into the computer. For every reciprocal lattice vector and collimator set chosen, the following quantities were tabulated as functions of the outgoing neutron wave vector: the monochromator and analyzer reflections and beam path for best focusing, the spectrometer angles, and the resolution in energy and momentum space. The calculated resolutions also included their individual contributions from different collimators and mosaic widths. With this information it was easy to choose the most favorable alternatives.

The monochromator used Cu (420) and (220) reflections, and the analyzer Cu (220) and (111). Their mosaic widths were 0.004 rad. The horizontal collimation was before the sample usually 0.0065 rad and after 0.0033, 0.0065, 0.013, or 0.019 rad. Our instrumental energy resolution, measured by the full width at half-maximum,[11] was in no case larger than about 0.20 THz and for acoustic phonons always better than about 0.10 THz. The radius vector of the resolution ellipsoid in momentum space was never longer than 0.03 and often shorter than 0.02 in units of $2\pi/l$, where l is the lattice constant. The mosaic width of the sample is less than 0.001 rad.

Phonon peaks often exhibit marked skewness due to curvature of the dispersion in the region covered by a measurement. The effect is especially pronounced in the vicinity of reciprocal lattice points, where the equienergy surfaces are strongly curved. We almost entirely avoided this by minimizing momentum resolution widths.

No shifts in the phonon curves were observed for different cells in reciprocal space or different scattering planes. For example, we measured the Raman frequency Γ_{25}' four times using different reciprocal lattice vectors and scattering planes and got the frequencies 9.11, 9.12, 9.12, and 9.13 THz with the estimated statistical error 0.02 THz. Figure 1 shows four examples of measured phonon resonances.

RESULTS

Phonon-dispersion relations in Ge at 80 °K have been measured for all branches in the principal symmetry directions $\Delta = [100]$, $\Sigma = [110]$, and $\Lambda = [111]$ and for most of the branches along the lines *U-W-K-L-W-X-U-L* at the surface of the Brillouin zone. Our data are presented in Fig. 2 and Table I. Notations used for classification of points and directions in reciprocal space are those introduced by Boukaert *et al.*[12] and shown in Fig. 3 and of different branches those defined in Refs. 13–15. The errors listed in Table I are based on considerations of counting statistics only; systematic errors are considered to be of no significance.

The branches of the dispersion curves are labeled by the notations of the irreducible representations of the symmetry group of the phonon wave vector. The nature of polarization of a branch is given by the transformation properties of the associated irreducible representation. For example, the modes Δ_1, Δ_2', and Λ_1, are purely longitudinal; Δ_5, Λ_3, Σ_2, and Σ_4 are purely transverse; and Σ_1 and Σ_3 nonlinearly polarized. For further information on this subject we refer to Warren.[15] For the assignment

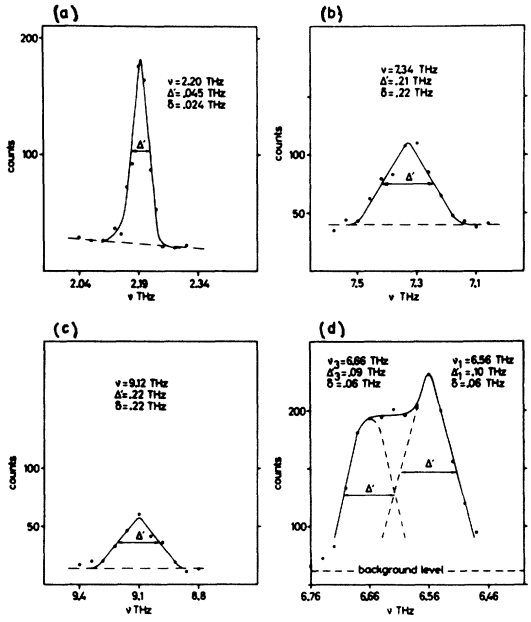

FIG. 1. Four examples of scanned resonances in order of increasing difficulty of measurement. Δ' is the observed width and δ the calculated energy resolution width (Ref. 11). (a) $\Delta_5(A)$ (0.5, 0, 0), (b) L_1, (c) Γ_{25}', (d) $\Sigma_1(A)$ and $\Sigma_3(O)$ (1.0, 0.2, 0.2).

of the labels L_1 and L_2' we have used the result of Johnson and Loudon.[14] No notations have been found in the literature for the branches on the lines L-U (L-K) and U-W (K-W).

The full phonon linewidths at half-maximum together with their estimated errors are shown in Fig. 4 for the principal symmetry directions. The extraction of widths was performed in accordance with a method described by Stedman and Weymouth.[16]

DISCUSSION

In Fig. 2 the heavy lines indicate the slopes of the respective branches when approaching Γ_{15}, as calculated from the elastic constants of McSkimin.[17] These agree with our data within the accuracy of the two experiments. Our value of the lattice constant ($l = 5.651$ Å) at 80 °K is a mean value of those given in Refs. 17–19.

The first neutron measurements on Ge were performed at room temperature. Many of these frequencies are higher than ours, contrary to expectations. There are, however, also other differences. Brockhouse and Iyengar,[1] who observed phonons on the Δ and Λ directions, did not resolve the optical modes Δ_5 and Δ_2' and reported phonon curves where the transverse branch lies consistently above the longitudinal one. The results of the time-of-flight studies by Ghose *et al.*[2] in the Δ and Σ directions show little similarity with our data. In 1963 Brockhouse and Dasannacharya[20] published measurements of nine phonons at 100 °K and 700 °K, and these frequencies, in closer agreement with ours, are consistently lower. Brockhouse[21] mentioned observation of a phonon peak of 8.5 THz at $K = U$ and at room

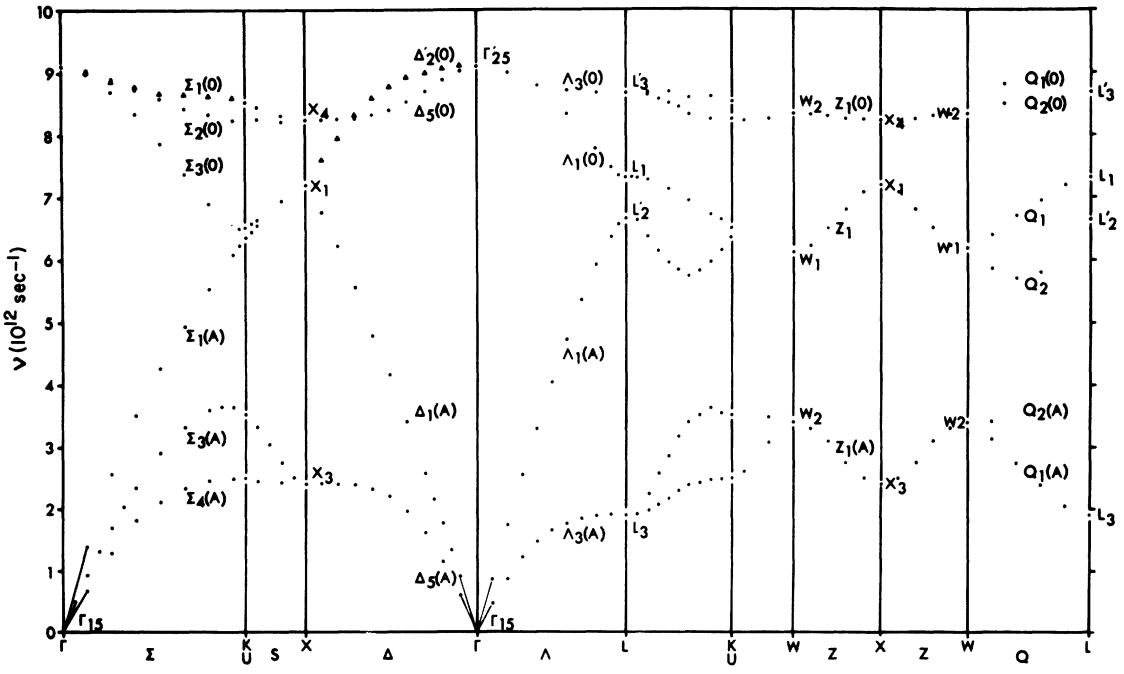

FIG. 2. Measured phonon-dispersion relations in Ge at 80 °K.

temperature, which was believed to be "an extremely close double." The present results show this to be so. The $\Sigma_1(A)$ and $\Sigma_2(O)$ modes possess a similar polarization and inelastic structure factor in the vicinity of $K = U$, where they come closer together but have not been found to cross. The closest positioned phonons we could resolve were those at (1.0, 0.2, 0.2) shown in Fig. 1(d). Neither Dolling[3] nor Price and Rowe[4] resolved the modes around $K = U$ in Si and α-Sn, respectively, but their shell-model calculations show a behavior similar to our result in the vicinity of this point.

The lifetime of a phonon is a function of anharmonic effects and equal to the inverse of the half-width of the phonon resonance expressed in angular frequency ω. The anharmonicity of Ge is very small, as evidenced, for example, by the low value of its Grüneisen constant, which is about 0.4 at 80 °K.[22] Brockhouse and Dasannacharya[20] detected no broadening of phonons even at 700 °K, in comparison with their peaks at 100 °K and did not try to extract the intrinsic linewidths. The errors in Fig. 4 reflect the difficulty in measuring the widths. The limits of error are based on the statistical uncertainties of recorded peaks and background,[16] and also include a 10% uncertainty in the resolution

TABLE I. Measured phonon frequencies in Ge at 80 °K expressed in units of 10^{12} sec^{-1}.

A.

$(l/2\pi)$ (q_x, q_y, q_z)	$\Delta_5(A)$	Δ_1	$\Delta_5(O)$	Δ_2'
0.00 0 0	• • •	• • •	9.12 ± 0.02	9.13 ± 0.02
0.10 0 0	0.60 ± 0.01	0.91 ± 0.01	9.05 ± 0.02	9.14 ± 0.02
0.15 0 0	• • •	1.34 ± 0.02	• • •	• • •
0.20 0 0	1.15 ± 0.01	1.77 ± 0.02	8.91 ± 0.02	9.09 ± 0.02
0.25 0 0	• • •	2.16 ± 0.02	• • •	• • •
0.30 0 0	1.62 ± 0.01	2.59 ± 0.02	8.73 ± 0.01	9.02 ± 0.03
0.40 0 0	1.96 ± 0.01	3.42 ± 0.01	8.56 ± 0.01	8.95 ± 0.03
0.50 0 0	2.20 ± 0.01	4.17 ± 0.01	8.42 ± 0.01	8.80 ± 0.02
0.60 0 0	2.32 ± 0.01	4.90 ± 0.01	8.35 ± 0.02	8.62 ± 0.02
0.70 0 0	2.39 ± 0.01	5.57 ± 0.01	8.29 ± 0.01	8.34 ± 0.03
0.80 0 0	2.40 ± 0.01	6.23 ± 0.01	8.28 ± 0.02	7.97 ± 0.02
0.90 0 0	2.41 ± 0.01	6.77 ± 0.01	8.27 ± 0.02	7.63 ± 0.02
1.00 0 0	2.40 ± 0.01	7.21 ± 0.01	8.26 ± 0.02	• • •

B.

$(l/2\pi)$ (q_x, q_y, q_z)	$\Lambda_3(A)$	$\Lambda_1(A)$	$\Lambda_3(O)$	$\Lambda_1(O)$
0.00 0.00 0.00	• • •	• • •	• • •	9.12 ± 0.03
0.05 0.05 0.05	0.47 ± 0.01	0.86 ± 0.01	• • •	• • •
0.10 0.10 0.10	0.86 ± 0.02	1.74 ± 0.01	9.02 ± 0.02	9.03 ± 0.03
0.15 0.15 0.15	1.22 ± 0.01	2.56 ± 0.01	• • •	• • •
0.20 0.20 0.20	1.47 ± 0.01	3.30 ± 0.01	8.82 ± 0.02	8.81 ± 0.02
0.25 0.25 0.25	1.65 ± 0.01	4.05 ± 0.01	• • •	• • •
0.30 0.30 0.30	1.76 ± 0.01	4.73 ± 0.02	8.75 ± 0.03	8.36 ± 0.02
0.35 0.35 0.35	1.84 ± 0.01	5.37 ± 0.01	• • •	• • •
0.40 0.40 0.40	1.88 ± 0.02	5.93 ± 0.02	8.70 ± 0.03	7.80 ± 0.02
0.45 0.45 0.45	1.90 ± 0.02	6.37 ± 0.03	• • •	7.50 ± 0.02
0.475 0.475 0.475	• • •	6.57 ± 0.02	• • •	7.36 ± 0.02
0.50 0.50 0.50	1.90 ± 0.02	6.66 ± 0.02	8.70 ± 0.03	7.34 ± 0.02

C.

$(l/2\pi)$ (q_x, q_y, q_z)	K-L 1	K-L 2	K-L 3	K-L 4	K-L 5	K-L 6
0.750 0.750 0.00	• • •	3.53 ± 0.03	6.37 ± 0.03	6.52 ± 0.04	8.27 ± 0.03	8.56 ± 0.03
0.725 0.725 0.05	2.48 ± 0.02	3.56 ± 0.02	6.17 ± 0.02	6.66 ± 0.01	• • •	• • •
0.700 0.700 0.10	2.47 ± 0.02	3.64 ± 0.02	5.98 ± 0.03	6.74 ± 0.02	8.28 ± 0.04	8.64 ± 0.03
0.675 0.675 0.15	2.42 ± 0.02	3.53 ± 0.02	5.84 ± 0.04	• • •	• • •	• • •
0.650 0.650 0.20	2.38 ± 0.02	3.41 ± 0.01	5.76 ± 0.05	6.96 ± 0.02	8.35 ± 0.04	8.62 ± 0.04
0.625 0.625 0.25	2.29 ± 0.02	3.18 ± 0.03	5.86 ± 0.02	• • •	8.54 ± 0.02	• • •
0.600 0.600 0.30	2.16 ± 0.02	2.86 ± 0.04	5.97 ± 0.02	7.14 ± 0.03	8.54 ± 0.03	8.72 ± 0.04
0.575 0.575 0.35	2.06 ± 0.02	2.58 ± 0.04	6.15 ± 0.03	• • •	8.60 ± 0.04	• • •
0.550 0.550 0.40	1.97 ± 0.02	2.24 ± 0.02	6.38 ± 0.02	7.29 ± 0.02	8.67 ± 0.03	8.71 ± 0.03
0.525 0.525 0.45	1.91 ± 0.02	• • •	6.64 ± 0.04	7.32 ± 0.02	• • •	• • •
0.5125 0.5125 0.475	• • •	• • •	• • •	7.34 ± 0.02	• • •	• • •
0.500 0.500 0.50	1.89 ± 0.02	• • •	• • •	• • •	8.70 ± 0.04	8.72 ± 0.03

TABLE I. (*continued*)

D.

$(l/2\pi)$ (q_x, q_y, q_z)	Σ_4	$\Sigma_3(A)$	$\Sigma_1(A)$	$\Sigma_3(O)$	$\Sigma_1(O)$	Σ_2
0.00 0.00 0.00	• • •	• • •	• • •	• • •	• • •	9.11 ± 0.02
0.05 0.05 0.00	• • •	0.49 ± 0.01	• • •	• • •	• • •	• • •
0.10 0.10 0.00	0.67 ± 0.01	0.93 ± 0.01	1.39 ± 0.01	9.01 ± 0.03	9.03 ± 0.02	9.05 ± 0.02
0.15 0.15 0.00	• • •	1.32 ± 0.01	• • •	• • •	• • •	• • •
0.20 0.20 0.00	1.29 ± 0.01	1.70 ± 0.01	2.58 ± 0.02	8.72 ± 0.04	8.89 ± 0.02	8.92 ± 0.02
0.25 0.25 0.00	• • •	2.04 ± 0.01	• • •	• • •	• • •	• • •
0.30 0.30 0.00	1.82 ± 0.01	2.35 ± 0.01	3.53 ± 0.02	8.37 ± 0.03	8.80 ± 0.03	8.76 ± 0.02
0.40 0.40 0.00	2.11 ± 0.01	2.92 ± 0.01	4.28 ± 0.03	7.89 ± 0.03	8.69 ± 0.03	8.61 ± 0.02
0.50 0.50 0.00	2.34 ± 0.01	3.34 ± 0.01	4.95 ± 0.03	7.40 ± 0.02	8.68 ± 0.03	8.45 ± 0.03
0.60 0.60 0.00	2.46 ± 0.01	3.61 ± 0.02	5.55 ± 0.01	6.91 ± 0.02	8.65 ± 0.03	8.36 ± 0.03
0.65 0.65 0.00	• • •	3.66 ± 0.05	• • •	• • •	• • •	• • •
0.70 0.70 0.00	2.49 ± 0.01	3.66 ± 0.05	6.10 ± 0.02	6.58 ± 0.02	8.63 ± 0.03	8.26 ± 0.04
0.725 0.725 0.00	• • •	• • •	6.24 ± 0.02	6.51 ± 0.04	• • •	• • •
0.75 0.75 0.00	2.50 ± 0.03	3.54 ± 0.02	6.37 ± 0.03	6.53 ± 0.02	8.55 ± 0.05	• • •
1.00 0.225 0.225	• • •	• • •	6.46 ± 0.04	6.60 ± 0.04	• • •	• • •
1.00 0.20 0.20	2.45 ± 0.02	3.34 ± 0.02	6.56 ± 0.01	6.66 ± 0.02	8.47 ± 0.04	8.28 ± 0.04
1.00 0.15 0.15	• • •	3.06 ± 0.01	• • •	• • •	• • •	• • •
1.00 0.10 0.10	2.43 ± 0.01	2.76 ± 0.02	6.96 ± 0.02	• • •	8.34 ± 0.04	8.24 ± 0.05
1.00 0.05 0.05	• • •	2.51 ± 0.01	• • •	• • •	• • •	• • •
1.00 0.00 0.00	2.41 ± 0.02	2.40 ± 0.02	• • •	• • •	8.29 ± 0.04	8.27 ± 0.05

E.

$(l/2\pi)$ (q_x, q_y, q_z)	$Z_1(A)$	Z_1	$Z_1(O)$
1 0.0 0	2.40 ± 0.02	7.21 ± 0.01	8.26 ± 0.02
1 0.1 0	2.50 ± 0.04	7.10 ± 0.02	8.27 ± 0.03
1 0.2 0	2.76 ± 0.05	6.82 ± 0.02	8.28 ± 0.03
1 0.3 0	3.10 ± 0.04	6.52 ± 0.02	8.33 ± 0.02
1 0.4 0	3.31 ± 0.04	6.24 ± 0.04	8.35 ± 0.02
1 0.5 0	3.41 ± 0.04	6.20 ± 0.05	8.36 ± 0.03

F.

$(l/2\pi)$ (q_x, q_y, q_z)	K-W 1	K-W 2	K-W 5
0.8 0.7 0.0	2.61 ± 0.04	• • •	8.25 ± 0.02
0.9 0.6 0.0	3.08 ± 0.03	3.49 ± 0.04	8.28 ± 0.03

G.

$(l/2\pi)$ (q_x, q_y, q_z)	$Q_1(A)$	$Q_2(A)$	Q_1	Q_2	$Q_1(O)$	$Q_2(O)$
0.60 0.50 0.40	2.04 ± 0.03	• • •	7.21 ± 0.03	• • •	• • •	• • •
0.70 0.50 0.30	2.38 ± 0.03	• • •	6.96 ± 0.03	5.81 ± 0.04	• • •	• • •
0.80 0.50 0.20	2.75 ± 0.04	• • •	6.72 ± 0.04	5.72 ± 0.04	8.52 ± 0.04	8.38 ± 0.04
0.90 0.50 0.10	3.14 ± 0.04	3.43 ± 0.04	6.41 ± 0.04	5.87 ± 0.04	• • •	• • •

width. It is assumed that the natural phonon line is approximately Lorentzian in shape, and the errors are standard deviations. The width of the observed peak was in most cases close to the resolution width, and the phonon widths in Ge at 80 °K are generally lower than 0.1 THz.

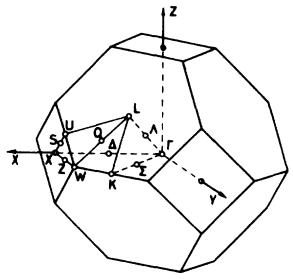

FIG. 3. Conventional notations of points and lines in the first Brillouin zone.

Parker *et al.*[23] observed first-order Raman scattering in Ge at room temperature and reported Γ_{25}' to have a frequency (9.01 ± 0.02) THz and a full width of 0.16 THz, while we have found (9.02 ± 0.02) THz at the same temperature and an extracted phonon width of (0.07 ± 0.07) THz at both 80 °K and 300 °K. Cowley[24] used a method of Maradudin and Fine[25] to calculate the frequency shift of Γ_{25}' between the two temperatures mentioned and arrived at − 0.11 THz, while we observed − (0.10 ± 0.03) THz. Using another theoretical approach, Klemens[26] ob-

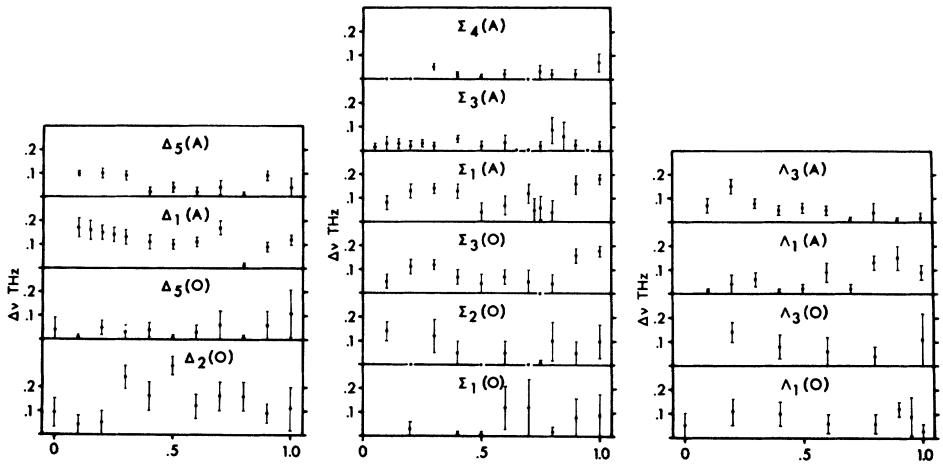

FIG. 4. Full phonon linewidths for the symmetry directions as calculated from the present data in Ge at 80 °K.

tained the phonon linewidth 0.13 THz at 300 °K, where Cowley got 0.50 THz. There is in the literature some confusion concerning the relation between lifetime and phonon width, which makes comparisons uncertain. The correct relationship[25] between lifetime τ, inverted lifetime Γ, phonon width in frequency $\Delta\nu$ and in angular frequency $\Delta\omega$ is $\tau^{-1} = \Gamma = \frac{1}{2}\Delta\omega = \pi \times \Delta\nu$, where Δ indicates the full width at half-maximum and the phonon energy $E = \hbar\omega = h\nu$. In many cases the (invalid) relation $\Gamma = \frac{1}{2}\Delta\nu$ is used.

Comparisons between neutron scattering and infrared absorption data on phonon frequencies are relatively common. Such comparisons are, however, of marginal value when only multiphonon processes are involved in the infrared absorption. The large number of closely positioned resonances affects their shapes and positions; to be able to make a correct assignment of the peaks one must have a good knowledge of the phonon dispersion curves in advance, which for the present can only be obtained by means of neutron spectrometry. This subject will be analyzed later, when we are able to use more complete results.

Our measurements have been extended to other reciprocal lattice points within the irreducible $\frac{1}{48}$ part of the first Brillouin zone. Future papers will deal with the phonon frequency distribution and the thermodynamics of Ge together with anharmonic effects and with a theoretical interpretation of the data.

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