determination of the limiting slope, thus determining whether K_{33} vanishes at the singular points on the effective zones. Additional information on the nature of K_{ij} would be expected from measurements using shear waves. The attenuation for the four pure-mode shear waves depends on the representations of K_{12} and K_{33} discussed above, but it also depends on the three representations of K_{12} which are odd under the reflection σ_{db}. Therefore, such measurements would give information on the unknown part of K_{12} but would not give enough conditions to improve the over-all determination of the deformation parameters. These results reinforce the previous conclusion. It is necessary to have a rather complete model (that is, fewer than six or seven parameters) for K_{ij} before electronic acoustic attenuation measurements can be used to "measure" the deformation.

Further studies of this type would require accurate attenuation measurements over a very wide frequency range, and this is especially difficult for metals showing strong dislocation effects. Some reduction of these effects might be obtained by irradiating the sample with neutrons or γ rays. The irradiation produces vacancies and interstitial defects which act to pin the dislocations in place and diminish their interaction with acoustic waves. However, the defects also scatter the electrons, thus reducing the mean free path and restricting the ql range obtainable with convenient frequencies. These difficulties notwithstanding, it is believed that the results of this study demonstrate that electronic acoustic attenuation measurements provide a powerful tool for studying the nature of the electron-phonon interaction at low wave numbers.

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Dispersion Relations for Phonons in Lead at 80 and 300°K

R. STEDMAN, L. ALMQVIST,* G. NILSSON, AND G. RAUNIO*
AB Atomenergi, Studsvik, Nykoping, Sweden
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Using a three-axis neutron spectrometer, phonon frequencies have been determined for lead for wave vectors in the principal symmetry directions and at many other points in reciprocal space. The results are presented as dispersion curves and as contour maps of frequency on the surface of an element of reciprocal space. The measurements were mainly at 80°K, but also at 300°K in some selected cases. Data on frequency shifts from 80 to 300°K are given, as well as on frequency widths at 300°K.

1. INTRODUCTION

COMPREHENSIVE measurements on lead at 100°K were made a few years ago by Brockhouse *et al.*.[1] Our original purpose in making further measurements was to try to achieve higher accuracy in the examination of the Kohn anomalies observed by Brockhouse *et al.*,[1] and to extend the measurements in order to trace out the Fermi surface as completely as possible. In order to interpret anomalies, however, we found it necessary to delve deeper than we had anticipated, and we therefore postpone that topic until later, confining ourselves at present to dispersion relations, phonon widths, and shifts of frequencies between 80 and 300°K.

The measurements at 80°K were at small intervals in the principle symmetry directions and at many asymmetric positions in reciprocal space also, so that a complete and quite accurate mapping of the dispersion relations can be made by simple interpolation. At 300°K, on the other hand, only some 40 one-phonon resonances were recorded, compared with 300–400 at 80°K, the object being to obtain representative values of frequency shifts and widths.

Some measurements were made on transverse branches at particularly low frequencies, using fine collimators to achieve good momentum resolution. Such measurements are difficult because of the close proximity to a position where Bragg scattering of relatively enormous intensity occurs, and because even small errors in zero positions become significant. High accuracy is essential if anything of interest is to be seen in the shape of dispersion curves or the temperature shifts of phonons at these low frequencies. Our fine

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* Permanent address: Chalmers Institute of Technology, Gothenburg, Sweden.

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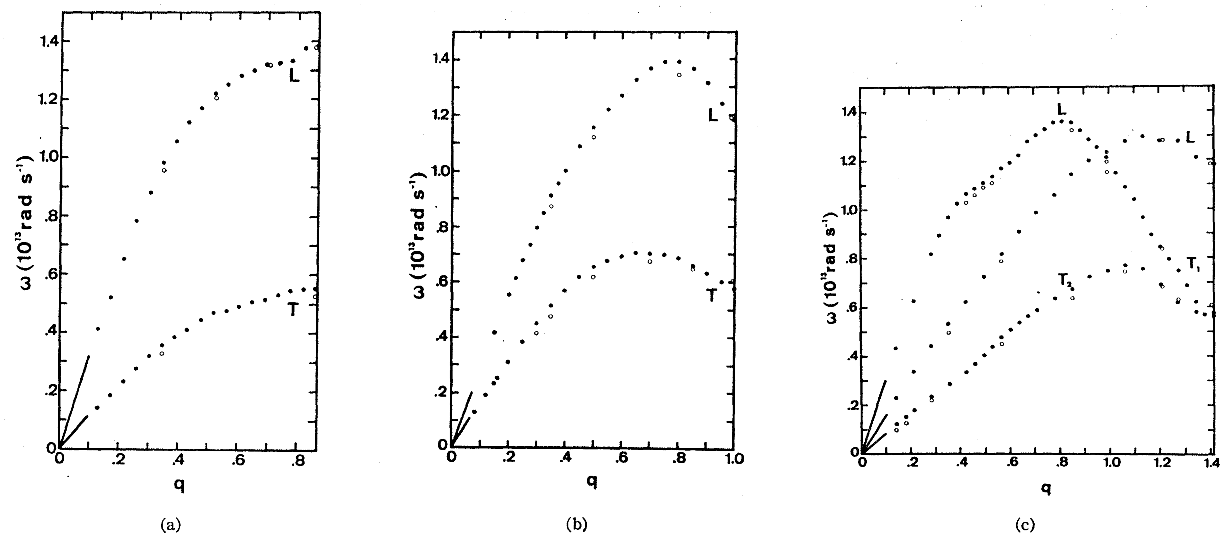

FIG. 1. Dispersion curves for lead at 80°K (filled circles), with some phonon frequencies at 300°K (open circles): (a) (1,1,1) direction, (b) (2,0,0) direction, (c) (2,2,0) direction.

measurements in this case actually revealed nothing of note and may be regarded as primarily a trial of apparatus and method. The lowest frequency for a useful measurement was 10^{11} Hz, but further improvements should reduce this limit somewhat.

An account of the apparatus and method was given in a previous article,[2] so here we may proceed directly to results.

2. RESULTS

Figures 1(a), 1(b), and 1(c) show dispersion curves for the (1,1,1), (2,0,0), and (2,2,0) directions at 80°K, together with several phonon frequencies at 300°K. The probable error in the phonon frequencies at 80°K because of counting statistics is on an average 0.006×10^{13} rad sec^{-1}, rising to twice this value at some of the higher frequencies, while no more than half as large at low frequencies. Errors associated with calibration of the spectrometer are considered to be smaller than 0.003×10^{13} rad sec^{-1}. At 300°K the uncertainty in phonon frequencies is about twice as large as at 80°K, with values of 0.03 or 0.04 at the highest frequencies, because of particularly broad peaks and high background. The lines through the origin have slopes corresponding to the velocity of sound in each case.[3]

Contour maps of phonon frequencies on the inner

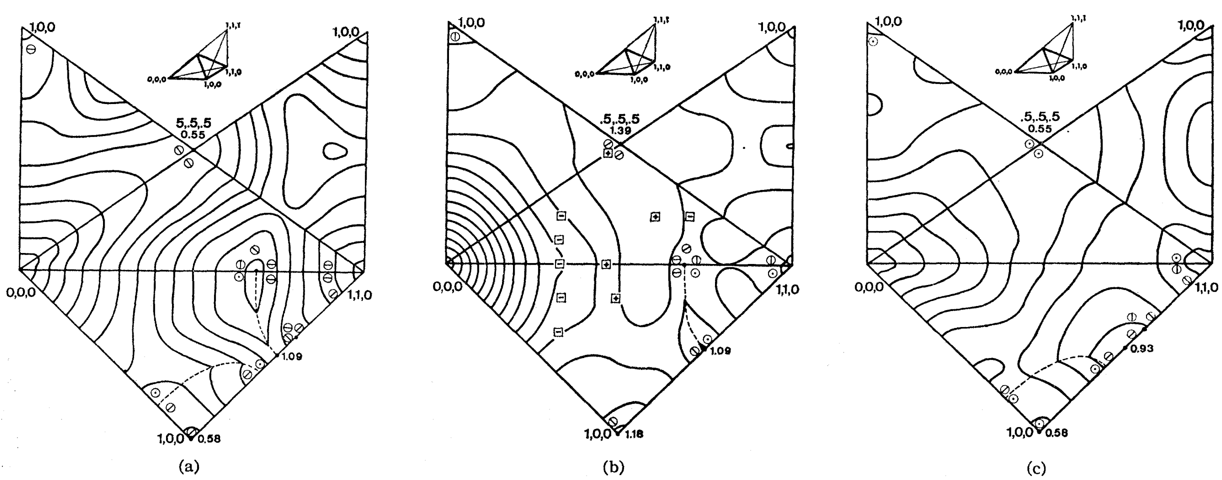

FIG. 2. Contour maps of phonon frequencies for the inner surface of the tetrahedron shown in the inset. 80°K. (a) L branch, (b) T_1 branch, (c) T_2 branch. The interval between curves is 0.1×10^{13} rad sec^{-1}. The unit for frequencies given at some boundary points is 10^{13} rad sec^{-1}. Symbols in circles indicate polarization, while boxes indicate the sites of anomalies.

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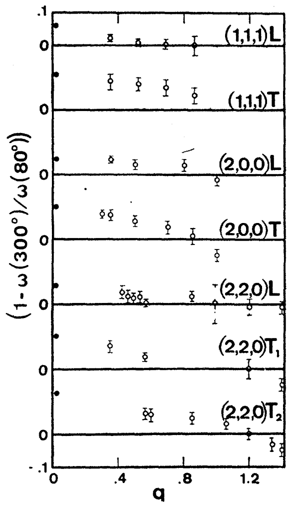

FIG. 3. Relative shifts in phonon frequencies from 80 to 300°K.

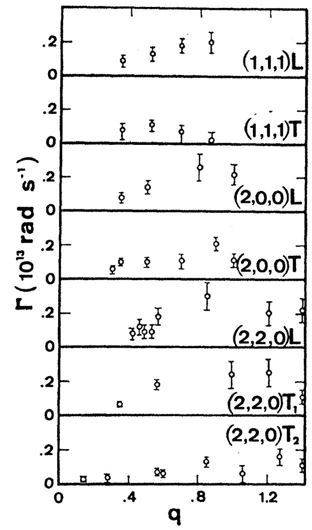

FIG. 4. Phonon frequency widths at 300°K.

surface of an elemental tetrahedron are shown in Figs. 2(a), 2(b), and 2(c). The tetrahedron corresponds to 1/48 of the first Brillouin zone, with a simplified boundary opposite the origin. The maps are based on results for the symmetry directions and for a large number of other positions. Anomalies indicated in the L map are some of those observed in local detailed profiles; a plus sign indicates an upward kink, travelling from the origin, and a minus sign indicates a downward one.

Figure 3 is of relative phonon shifts from 80 to 300°K. It is a little difficult to indicate the vertical scale clearly in such a compressed figure, so it should perhaps be pointed out that each zero is 0.2 from the one above it. The filled circles near $q=0$ are from ultrasound data.[3]

The widths of observed one-phonon resonances at 80°K were often not much greater than the resolution widths, so the errors in the derived phonon widths are large. An average general magnitude is 0.05 of the corresponding frequency, though the variation about this average is considerable. At 300°K the resolution was adequate, but peaks were often poorly defined. Results are shown in Fig. 4.

3. COMMENTS

A. Dispersion Relations

Brockhouse *et al.* have published tables of phonon frequencies for lead at 100°K. Our results at 80°K agree with theirs, almost everywhere well within the assigned errors. The slight temperature difference is of no significance, since shifts of frequency from 80 to 100°K are expected to be less than 0.1 of the shifts from 80 to 300°K, and therefore (cf. Fig. 3) negligible. The only differences between the two sets of results that are systematically greater than the margins of error are at the maxima of the (2,0,0) L and (2,2,0) L branches, where our values are 2% higher. Our results indicate a few more irregular details, e.g., an upward anomaly in the (2,2,0) L curve at $q=0.65$, a double kink in the (2,2,0) L curve between $q=1.1$ and 1.4 (also in the T_1 branch), and a hump in the (2,2,0) T_2 curve between $q=0.45$ and 0.6.

Measurements with particularly fine collimators, corresponding to a resolution in $\mathbf{q}$ space not exceeding 0.02 in any direction, showed that previous measurements with moderately fine collimators on the (2,2,0) T_2 branch at small q had given results which were too high. The reason is apparent from Fig. 2(c): The region concerned lies in a deep valley, and particularly good momentum resolution is required to resolve this. The same effect may be expected to occur elsewhere—e.g., at the sharp minimum in the T branches at $\mathbf{q}=(1,1,0)$ or (1,0,0), where the intensity did not permit the finer type of measurement—but the required correction is probably small, and we have not bothered to make it.

In the plane $q_3=0$ of Fig. 2 there are two crossover singularities, i.e., curves of sharp contact between the L and T_1 branches, and between the T_1 and T_2 branches. They are marked by dashed lines. The manner in which the polarization rotates around these singularities is also indicated in the figure.

It is only to be expected that dispersion relations with so many turning points and irregularities cannot be accounted for by any relatively simple model of the interatomic forces, and this surmise is borne out by the results of various theoretical analyses. Brockhouse *et al.*[1] carried out a Fourier analysis of their dispersion curves to obtain force constants in a Born–von Kármán model, and found that even an approximate fit required many Fourier terms, corresponding to appreciable forces out to at least eighth neighbors. As they indicated, such analysis is not very meaningful physically, and it would be of much greater interest to extract from the experimental results the information they undoubtedly contain about the role of the conduction electrons. Since then, attempts have been made to apply to lead the theory of the metal lattice bond which has developed in recent years, where the attraction between ions via the conduction electrons is described in terms of an ion-electron interaction and the screening reaction of the electron gas. While the theory gives a good account of the observed dispersion curves for phonons in sodium[4,5] and potassium,[6] and to some extent aluminium,[4,7] it has hitherto not been successful for lead, as may be seen from the diagrammatic comparison of theoretical and experimental curves in papers by Toya[8] and Vosko *et al.*[4] These latter authors attempted to keep as close as possible to first principles in their calculations, but another way of comparing theory and experiment, originally applied to sodium by Cochran,[9] is to fit the potential which in some form or other is at the center of the theory to the neutron data. This semiempirical potential may then be judged in other connections, e.g., with regard to some theoretical model of the potential, to data on the resistivity of the liquid metal, or to the shape of the Fermi surface. Schneider and Stoll[10] have used this approach and six adjustable parameters to obtain a bare-ion potential for lead, which, however, gives an incorrect result when applied to a calculation of the resistivity of liquid lead (a failure the authors attribute to something else, however).

B. Frequency Shifts and Widths

Shifts of frequency with temperature are, of course, small effects and not easy to measure, but nevertheless our results and those of Brockhouse and his collaborators[11] are in close agreement. The upward shift (negative values in Fig. 3) in all three branches at and near (1,0,0) is confirmed in our measurements, which include the (2,2,0) direction as well. The (2,2,0) L results near the Kohn anomaly at $q=0.46$ suggest that the anomaly moves or changes shape slightly, but unfortunately the accuracy is not sufficient to establish whether the observation is significant or not.

In stating that the widths at 80°K are roughly 0.05 of the corresponding frequencies, it is not implied that there is a direct proportionality between the two quantities, but merely that this is a simple approximate description of the approximate data. Brockhouse *et al.*[12] have given some data on widths at 425°K, for the (2,0,0) direction and the point (0.5,0.5,0.5), and these results may be compared with ours at 300°K, though with some minor reservations. In extracting frequency widths from observed resonances, they assumed that the phonon line shape is Lorentzian, while we merely assume a bell-shaped line and neglect the wings[2]. This difference of procedure has, however, little effect on the comparison because our resolution widths for the measurements concerned were nowhere greater than 0.5 of the width of the observed resonance and would have yielded almost the same phonon widths even if we had used their procedure. They took resolution widths to be 0.9 of the widths of the corresponding resonances as observed at 100°K; if an error arises from this assumption it is almost certain to entail too large a value for the resolution width, i.e., too small a value for γ (our Γ) at 425°K. Bearing this in mind, as well as the circumstance that widths at 425°K are expected to be roughly 50% greater than at 300°K, our results are in fair agreement with theirs.

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