IMAGE OF THE FERMI SURFACE IN THE LATTICE VIBRATIONS OF LEAD

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On theoretical grounds Kohn[1] has proposed that there should exist positions in reciprocal space at which the slopes of the dispersion curves of the lattice vibrations in metals become infinite. The positions are related to the Fermi surface of the electrons, and the infinite slope to the sharpness of the Fermi surface. These Kohn anomalies have not previously been observed experimentally for certain, and recent calculations[2] suggest that they may be quite small, at least for some materials. We believe that we have observed the anomalies in a single cyrstal of lead.

Previous experiments[3] on lead at this laboratory yielded dispersion curves which could be interpreted on the Born-von Kármán theory of lattice dynamics only if it were assumed that there exist long-range forces between ions in the crystal, the forces often varying in sign. These results were consistent with the existence of Kohn anomalies, but direct evidence was weak.

The new experiments were carried out using the triple-axis crystal spectrometer in such a way that, for each energy distribution, the momentum transfer, $\hbar\vec{Q}=\hbar(\vec{k}_0-\vec{k}')$, remained fixed.[4] The centers of the neutron groups observed give the frequencies ν of the lattice vibrations (of wave vector $\vec{Q}$ in the extended zone scheme) through the relation $h\nu=E_0-E'$. In the expressions $\vec{k}_0$ and $\vec{k}'$ and E_0 and E' are the incoming and outgoing neutron wave vectors and energies, respectively.

Figure 1(a) shows a series of neutron groups obtained as the terminus of $\vec{Q}$ moves along the line $\vec{Q}=(2\pi/a)[\zeta,\zeta,\zeta]$ between the reciprocal lattice points (1, 1, 1) and (2, 2, 2) in steps of 0.025 in ζ. (a is the lattice constant.) The neutron groups observed correspond to longitudinal phonons whose reduced wave vector $\vec{q}$ is in the [111] direction of the Brillouin zone. The centers of the groups are shown plotted in Fig. 1(b) as a function of $(a/2\pi)\vec{Q}$. Near the zone boundary a sharp anomaly occurs on both sides of the zone boundary, as it should, and three independent series of measurements at different energies gave almost identical results.

For a spherical Fermi surface the positions of the anomalies are given[1] by the equation

$$2K_F=|2\pi\vec{\tau}+\vec{q}|,$$

where $\vec{\tau}$ is a vector of the reciprocal lattice and K_F is the radius of the Fermi surface. In Fig. 2 the $(1\bar{1}0)$ plane of reciprocal space is shown, each reciprocal lattice point, $a\vec{\tau}$, surrounded by its zone. A spherical Fermi surface containing four

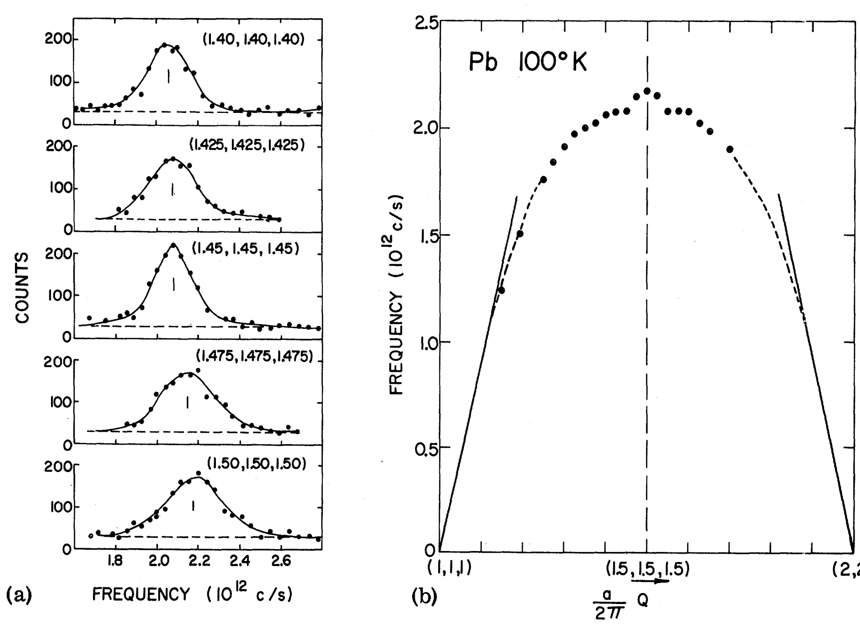

FIG. 1. (a) A series of neutron groups (actual counts) obtained for values of $\vec{Q}$ along the line $\vec{Q}=(2\pi/a)[\zeta,\zeta,\zeta]$ from $\zeta=1.4$ to $\zeta=1.5$, for $E'=0.01228$ ev. The background is indicated by the dashed line. (b) The frequency corresponding to neutron groups observed along the line $\vec{Q}=(2\pi/a)\times[\zeta,\zeta,\zeta]$. The dashed line corresponds to the previously measured dispersion curve for the longitudinal [111] branch. The solid lines have the slope of the velocity of sound as calculated from the elastic constants.

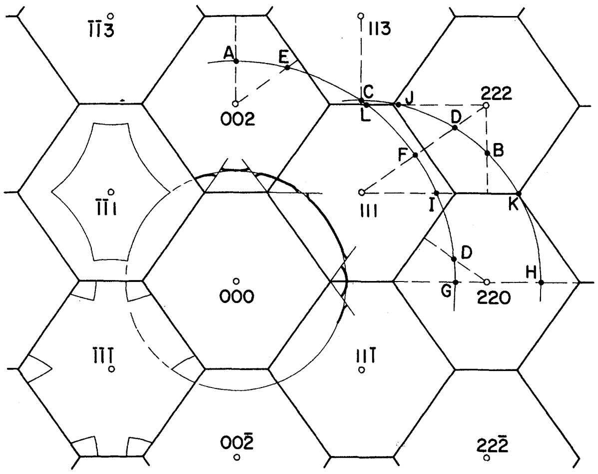

FIG. 2 The $(1\bar{1}0)$ plane of the reciprocal lattice of (face-centered cubic) lead, each point surrounded by its zone. A spherical Fermi surface containing four free electrons (extended zone scheme) intersects the plane as shown by the circle about the point (0,0,0). In the reduced zone scheme this spherical surface gives rise to the surface shown about $(\bar{1},\bar{1},1)$ in the second zone, and to the surfaces shown about $(\bar{1},\bar{1},\bar{1})$ in the third zone. Possible effects of gap formation on the extended zone surface are indicated by bold lines. The Kohn construction for free electrons is shown by the circular arcs *AECLFIDG* and *CJDBKH*. The points *A*, *B*$\cdots$*L* indicate positions of Kohn anomalies for free electrons in the symmetric directions [100], [110], and [111], shown by dashed lines.

electrons[5,6] is drawn around the point $a\vec{\tau} = (0, 0, 0)$. In the reduced zone scheme this Fermi surface gives a full first zone, a second zone with a pocket of holes as shown around the point $(\bar{1}, \bar{1}, 1)$, and a complicated structure in the third zone shown in cross section around the point $(\bar{1}, \bar{1}, \bar{1})$. The locus of the Kohn anomalies connected with the point (0, 0, 0) is shown as the circular arc *AECLFIDG*. Other types of anomalies also occur, as indicated by the arc *CJDBKH* which is connected with the point (2, 0, 0) [a point not in the $(1, \bar{1}, 0)$ plane]. The positions designated by the letters *A*, *B*, $\cdots$, *L* are those for which anomalies in the symmetric [100], [110], and [111] directions of the reduced zone occur for a spherical Fermi surface. Some of the anomalies have a multiplicity greater than unity, because they can be produced from several reciprocal lattice vectors (e.g., *E* with multiplicity 3, *C* with 4, *H* with 2, etc.). Some of the anomalies can occur only in longitudinal modes (e.g., *A*, *F*, *G*); these have unit multiplicity. The signs of the anomalies (i.e., $\mathrm{grad}_{\vec{q}}\,\nu = +\infty$ or $-\infty$) can be assigned by the plausible rule that the frequency increases anomalously when $\vec{q}$ passes from the interior to the exterior of the Kohn sphere since the electron gas then "stiffens." Thus *A*, *E*, *F*, and *I* have positive signs, while *C*, *D*, and *G* have negative signs.

The spherical Fermi surface applies to ideally free electrons. In fact energy gaps will be produced at Bragg reflection planes, as illustrated by the modifications to the spherical Fermi surface shown as bold lines in Fig. 2. These gaps round off the Fermi surfaces in the reduced zone, and render the Kohn construction[1] more complicated. In particular, only points *A*, *F*, *G*, and possibly *I*, are certain to be unchanged in a qualitative sense by the passage to the real Fermi surface.

It will be observed that point *F*, expected to occur only in the longitudinal mode, corresponds very closely to the anomaly in Fig. 1(b). However, this assignment is ambiguous because of possible interference with point *E*.

In Fig. 3 the longitudinal branch for the [110] direction is shown. These measurements were made along three separate lines through reciprocal space; from (2, 2, 0) to (1, 1, 0), from (2, 2, 0) to (3, 3, 0), and from (1, 1, 1) to (2, 2, 1). Two different monochromator planes were used from (2, 2, 0) to (1, 1, 0). An anomaly, which can be identified with point *G*, appears at $aq/2\pi \simeq 0.40$. The position of the anomaly yields the result that the radius of the Fermi surface in the [110] direction (extended zone scheme) is about $1.21(2\pi/a)$, instead of the value $1.24(2\pi/a)$ holding for free electrons. Thus the width of the arms in the third zone (reduced zone scheme) is smaller by about 15% than for free electrons, in qualitative agreement with deductions by Gold[5] from his measurements on the de Haas–van Alphen effect. [Note added: From inspection of Fig. 3, Kohn and Woll (private communication) would place the anomaly at ~0.47, and thus make the width about one third smaller than for free electrons.]

Many other features of the unusual dispersion curves of lead[3] may be interpreted in terms of the Kohn effect. Of particular interest is the spectacular drop in frequency found for the longitudinal branch in the [100] direction near the zone boundary, which may be connected with anomaly *C*. This anomaly has fourfold multiplicity and therefore should be exceptionally strong. However, details are not clear since this point is strongly influenced by gap formation at the (002) Bragg plane, as indicated in Fig. 2.

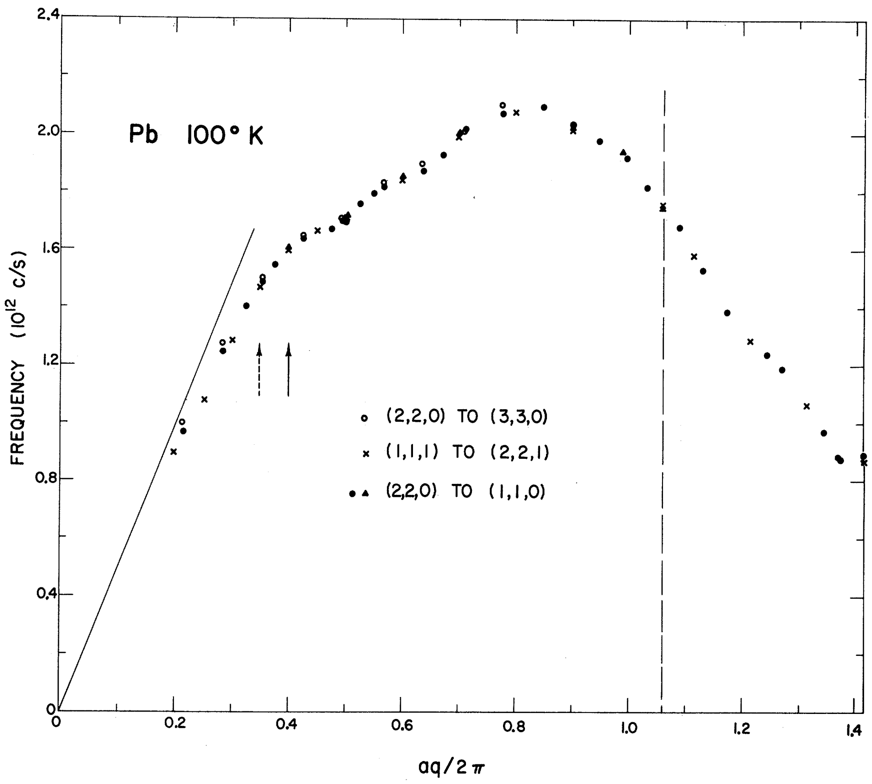

FIG. 3. The longitudinal branch in the [110] direction of the reduced zone. The branch is continued beyond the zone boundary to the point (1,1,0). The continuation, lying on the square face of the Brillouin zone, has the same symmetry as the [110] direction within the zone. The anomaly assigned as G is indicated by the solid vertical arrow at $qa/2\pi \simeq 0.40$. For free electrons it would appear at 0.35 as indicated by the dashed arrow. Other possible anomalies can also be seen. Measurements in a single series are thought to have relative errors about the size of the points. The scatter in points belonging to different series may be indicative of the absolute errors.

As mentioned earlier, recent calculations[2] have indicated that the anomalies should be quite small. The strength of the effect in lead is probably connected with the strong phonon-electron interaction in this material, as indicated for example by the comparatively high superconducting transition temperature. This is supported by the fact that the Kohn effect has not been detected in similar measurements on a single crystal of sodium.[7]

The anomalies are, of course, smeared out by resolution in $\vec{q}$ space. (Indeed, Fermi surfaces may be more easily delineated by means of x-ray intensity measurements than by neutron measurements, because of the better resolution in $\vec{q}$ space available with x rays.) However, even under good resolution the anomalies will appear to be smeared because of the logarithmic character of the infinity in the slope, which makes exceedingly small the range in ν over which $|\mathrm{grad}_{\vec{q}}\,\nu|$ is large.

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