as Ge. As we mentioned above, our resonant mechanism scatters longitudinal phonons more strongly than transverse phonons and, hence, the depression in Q_p is smaller for the latter. In addition, the "transverse phonon" depression will occur at a higher temperature than that found for longitudinal phonons [mainly due to the substitution of $\bar{v}_t^2$ for $\bar{v}_l^2$ in (5.22)]. Since phonon-phonon scattering is dominant at these temperatures, the bound electron-transverse phonon interaction is negligible for a second reason.

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Theory of Kohn Anomalies in the Phonon Spectra of Metals

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An expression is derived for the change in frequency of the lattice vibrations in a metal caused by the interaction of the phonons with the conduction electrons. The various factors affecting the magnitude of these Kohn anomalies are considered, and a connection is made with the value of the electrical resistivity of the pure metal. The valence of the metal is found to be the most important factor determining whether such anomalies should be observable. The results of the calculations are applied to Pb, and give good agreement with experiment.

INTRODUCTION

IT has been pointed out by Kohn[1] that the interaction of the conduction electrons in a metal with each other and with the vibrations of the crystal lattice should cause anomalies in the phonon spectra of metals. In particular, it has been suggested that the group velocity of phonons of wave number $\mathbf{q}$ will exhibit a logarithmic singularity whenever

$$\mathbf{q}=2\mathbf{k}_f+\mathbf{g}, \tag{1}$$

where $\mathbf{k}_f$ is the wave number of an electron at the Fermi surface, and $\mathbf{g}$ is a vector of the reciprocal lattice. There is, at the time of writing of this paper, some controversy as to whether these anomalies should be observable experimentally. While evidence for such behavior has been found by Brockhouse *et al.*[2] in their investigation of the lattice vibrations of lead, Harrison[3] has suggested that this reflects the form of the electron dispersion relations rather than being directly due to the electron-phonon interaction in the way envisaged by Kohn. It is the purpose of the present work to calculate the expected magnitude of the Kohn anomalies in various metals in a more quantitative way than has hitherto been attempted.

A previous calculation of the magnitude of this effect is due to Woll and Kohn[4] who adopted a semiclassical approach to the problem of calculating the vibration frequencies of a lattice of point charges in a sea of interacting electrons. Their method is a simplification of Nakajima's[5] quantum-mechanical calculation and is essentially a Hartree method which considers only terms of first order in both the electron-electron and electron-lattice interactions. However, while it is very satisfying to be able to calculate the required quantities from first principles, one cannot have complete confidence in a calculation that neglects electron correlation. A more serious criticism arises when it is pointed out that the result given by these authors contains terms that involve the product of parameters describing both electron-electron and electron-lattice interactions. Because the Hamiltonian is diagonalized only to first order in each, the validity of such terms is clearly in doubt, and in fact does, as we shall see, lead to contradictions.

The effect of the interaction of the electrons is to attenuate the Kohn anomalies. In the Hartree approximation, this attenuation is quite large; this is, however, an overestimate of the significance of these interactions, and is considerably reduced by the effect of the lattice potential on the electron wave functions.

We can separate the amplitude of the Kohn anomalies into two parts: The first part is due to the second-order corrections to the phonon energies caused by the electron-phonon interaction; the second part, which we describe by an attenuation coefficient, α, arises from the presence of infinities in the derivative of the matrix element for the interaction itself. No attempt is made in this paper to calculate α, although some of the factors affecting its magnitude will be discussed. The calculation of the direct effects of the interaction is a matter for greater confidence, for here we may have recourse to experiment. The electrical resistivity of pure metals is

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[1] W. Kohn, Phys. Rev. Letters **2**, 393 (1959).
[2] B. N. Brockhouse, K. R. Rao, and A. D. B. Woods, Phys. Rev. Letters **7**, 93 (1961); B. N. Brockhouse, T. Arase, G. Caglioti, K. R. Rao, and A. D. B. Woods, Phys. Rev. **128**, 1099 (1962).
[3] W. A. Harrison, Phys. Rev. **129**, 2512 (1963).
[4] E. J. Woll, Jr., and W. Kohn, Phys. Rev. **126**, 1693 (1962).
[5] S. Nakajima, Busseiron Kenkyu, **65**, 116 (1953). More accessible are the summaries of his work given by G. V. Chester, Phil. Mag. Suppl. **10**, 357 (1961) and in Ref. 10.

calculated directly from the strength of the electron-phonon interaction, and it is possible to invert these formulas to obtain an average value for the relevant matrix elements. These matrix elements of course contain all the correlation effects.

GENERAL FORMULA

We shall now derive an expression for the lowest order term in the change in frequency due to the electron-phonon interaction. We start by considering a gas of independent quasiparticles (we shall refer to them as electrons) interacting with the phonon field of a crystal lattice, and calculate the change in energy of the phonons by elementary perturbation theory in a similar procedure to that of Fröhlich.[6] Here is a catalog of some of the quantities we shall need: $\mathbf{k}$ crystal momentum of an electron; $\mathcal{E}_\mathbf{k}$ energy of an electron; $\mathbf{v}_\mathbf{k}=\partial\mathcal{E}/\hbar\partial\mathbf{k}$ electron velocity; $f_\mathbf{k}$ occupation number of state $\mathbf{k}$; $\mathbf{k}_f$ wave number on the Fermi surface; m electron mass; e electron charge; k Boltzmann constant; $\mathbf{g}$ reciprocal lattice vector; $\mathbf{q}$ phonon wave number; $n_\mathbf{q}$ average phonon occupation number; T absolute temperature; Θ Debye temperature; p valence; ρ electrical resistivity; σ density; Q Debye cutoff wave number; N_0 Avogadro's number; A atomic weight; δ Kronecker delta-function; ω phonon frequency; $M(\mathbf{k},\mathbf{k}',\mathbf{q})$ electron-phonon matrix element; V volume of crystal; $\Delta\omega$ change in phonon energy; ρ_1, ρ_2 radii of curvature of the Fermi surface; v_s velocity of sound; $\mathcal{E}_f$ Fermi energy.

The matrix element for scattering of an electron from $\mathbf{k}$ to $\mathbf{k}'$ by the absorption of a phonon $\mathbf{q}$ is[7]

$$M(\mathbf{k},\mathbf{k}')=(\hbar n_\mathbf{q}/2MNV\omega)^{1,2}\delta(\mathbf{k}',\mathbf{k}+\mathbf{q}+\mathbf{g})J(\mathbf{k},\mathbf{k}'),$$

where for any given metal the function $J(\mathbf{k},\mathbf{k}')$ depends only on the wave functions of the electrons. The change in energy of the phonon state $\mathbf{q}$ due to this interaction is

$$n_\mathbf{q}\hbar\Delta\omega=\sum_{\mathbf{k},\mathbf{k}'}\frac{|M(\mathbf{k},\mathbf{k}')|^2}{\mathcal{E}_\mathbf{k}-\mathcal{E}_{\mathbf{k}'}-\hbar\omega}f_\mathbf{k}\delta(\mathbf{k}',\mathbf{k}+\mathbf{q}+\mathbf{g}). \quad (2)$$

The gradient of this quantity with respect to $\mathbf{q}$ displays a logarithmic infinity at values of $\mathbf{q}$ close to those given by Eq. (1), when for certain terms in the summand the energy denominator becomes small. We note that so far we have not treated the electron interactions explicitly; they have simply been absorbed into the matrix element for the effective interaction.

At zero temperature, the occupation function $f_\mathbf{k}$ is equal to unity inside the Fermi surface and zero outside. The summation is then simply performed for phonon wave numbers near those at which anomalies occur. We consider two points, A and B, on the Fermi surface where the electron velocities, $\mathbf{v}_a$ and $\mathbf{v}_b$, are parallel, and where the principal radii of curvature of the surface

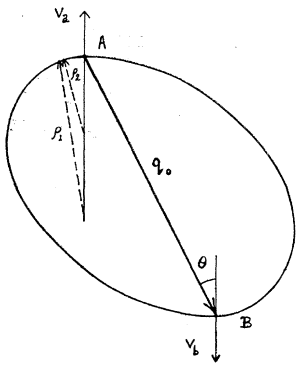

FIG. 1. Any two points on the Fermi surface where the electron velocities are parallel may give rise to an anomaly.

at A are ρ_1 and ρ_2 (Fig. 1). Let the vector AB be $\mathbf{q}_0$, the angle between $\mathbf{v}$ and $\mathbf{q}_0$ be θ, and consider the value of expression (2) for a phonon of wave number $\mathbf{q}=\mathbf{q}_0+\boldsymbol{\delta}\mathbf{q}$, where $\boldsymbol{\delta}\mathbf{q}$ and $\mathbf{q}_0$ are parallel. When $\boldsymbol{\delta}\mathbf{q}$ is small, we may write

$$\mathcal{E}(\mathbf{k}_b)=\mathcal{E}(\mathbf{k}_a+\mathbf{q}_0)=\mathcal{E}(\mathbf{k}_a),$$

$$\mathcal{E}(\mathbf{k}_a+\boldsymbol{\delta}\mathbf{k})-\mathcal{E}(\mathbf{k}_b+\boldsymbol{\delta}\mathbf{q}+\boldsymbol{\delta}\mathbf{k})=\hbar(\mathbf{v}_a-\mathbf{v}_b)\cdot\boldsymbol{\delta}\mathbf{k}-\hbar\mathbf{v}_b\cdot\boldsymbol{\delta}\mathbf{q}.$$

Then,

$$\sum_\mathbf{k}\frac{f_\mathbf{k}}{\mathcal{E}_\mathbf{k}-\mathcal{E}_{\mathbf{k}+\mathbf{q}}\pm\hbar\omega}=\sum_{\delta\mathbf{k}}\frac{\hbar^{-1}f_\mathbf{k}}{(\mathbf{v}_a-\mathbf{v}_b)\cdot\boldsymbol{\delta}\mathbf{k}-\mathbf{v}_b\cdot\boldsymbol{\delta}\mathbf{q}\pm\omega_\mathbf{q}}. \quad (3)$$

Assuming that the matrix element and electron velocities are slowly varying, we can find the contribution of states near A to the summation by integration over a paraboloid with vertex at A. It is found to be

$$\frac{-V}{2\pi^2\hbar}(\rho_1\rho_2)^{1/2}\frac{v_b}{(\mathbf{v}_a-\mathbf{v}_b)^2}\cos\theta\delta q\ln\left|\delta q\pm\frac{q_0v_s\sec\theta}{v_b}\right|. \quad (4)$$

This expression applies to all shapes of electron Fermi surface; for a surface containing holes the sign is reversed, since v_b must be counted negative. It is valid provided $|\mathbf{v}_a-\mathbf{v}_b|$, ρ_1^{-1}, ρ_2^{-1} differ from zero; otherwise the order of the infinity in the group velocity of sound is raised.[8]

At a finite temperature T, the occupation function $f_\mathbf{k}$ is described by Fermi-Dirac statistics. The integral leading to expression (4) is then modified, and the logarithmic term becomes approximately

$$\ln\left\{\left|\delta q\pm\frac{q_0v_s\sec\theta}{v_b}\right|+\frac{kT\sec\theta}{\hbar}\left(\frac{1}{v_a}+\frac{1}{v_b}\right)\right\}.$$

The group velocity of sound no longer tends to infinity as q tends to either of its critical values, but to $\ln T$. It is roughly constant over a range of q of $2kT/\hbar v\cos\theta$. Finally, we add the contributions of the terms with positive and negative signs—that is, we add the anomalies due to phonon absorption and emission.

[6] H. Fröhlich, Proc. Roy. Soc. (London) **A215**, 291 (1952).
[7] J. M. Ziman, *Electrons and Phonons* (Clarendon Press, Oxford, England, 1960), pp. 364, 182, 358.
[8] This has also been shown by different methods by A. M. Afanas'ev and Yu. Kagan, Zh. Eksperim. i Teor. Fiz. **43**, 1456 (1962) [translation: Soviet Phys.—JETP **16**, 1030 (1963)].

When the phonon energy is of the order of $2kT$, the two kinks in the dispersion curve merge to give a single region over which the group velocity is proportional to $\ln T$. Since most experimental determinations of phonon spectra by neutron or x-ray diffraction are performed at room or liquid-air temperatures, which are of the order of Θ, the two anomalies will not generally be resolvable.

MAGNITUDE OF THE ANOMALIES

As we remarked in the Introduction to this paper, it is the same matrix element as occurs in the theory of the electrical resistivity of pure metals that appears in Eq. (2). For an isotropic metal, the Bloch theory of electrical conduction gives the following relationship at high temperatures between ρ and a certain average of the matrix element[7]:

$$\left(\frac{|M(\mathbf{k},\mathbf{k}')|^2}{n_q\hbar\omega}\right)_{\rm av}=\frac{8e^2\hbar^3k_f{}^6}{3\pi m^2VQ^4k}\frac{\rho}{T}. \tag{5}$$

Let us define a number C as the ratio of the actual value of expression (4) to its value in a free-electron model—that is, we put

$$(4\hbar/m)(\rho_1\rho_2)^{1/2}[v_b/(v_a-v_b)^2]\cos\theta=C.$$

Expression (4) is then equal to approximately

$$-(CVm/8\pi^2\hbar^2)\delta q\ln|\delta q|, \tag{6}$$

and we can combine relations (2), (5), and (6) to give

$$\frac{\Delta\omega}{\omega}=\frac{e^2\hbar N_0}{\pi mk}\frac{p^3\sigma\rho}{AT}\frac{\delta q}{Q}C\ln|\delta q|, \tag{7}$$

where an extra factor of 2 is included to allow for both phonon emission and absorption. This tells us what the relative shift of the phonon frequency in a metal would be if (a) the Fermi surface were spherical, so that C was equal to unity, (b) the matrix element were independent of $\mathbf{q}$, and (c) electron interactions could be ignored. In a real metal, none of these will be true. We introduce a coefficient L to describe the ratio of the actual matrix element $M(\mathbf{q})$—assumed now to be independent of $\mathbf{k}$ and $\mathbf{k}'$—to the average $\bar{M}$ used in expression (5), that is

$$M(q_0)=L\bar{M}.$$

A further coefficient, α, to be discussed in the next section, is also introduced to describe the effects of electron interactions not already included in M. Swallowing the remaining constants of Eq. (7) into a dimensionless factor D, i.e., putting

$$D=(e^2\hbar N_0/\pi mk)(p^3\sigma\rho/AT),$$

we have finally

$$\Delta\omega/\omega=DCL\alpha(\delta q/Q)\ln|\delta q|.$$

The order of magnitude of D gives us a very good idea of whether a kink will be observable in any given metal. Values of this parameter for some common metals are given in Table I. It is to be seen that the valence, entering as it does to the third power in the definition of D, is the dominating factor. We also note how much greater the coefficient is for Pb than for Al.

TABLE I. Values of the quantities p, σ, and D for various metals.

	Na	Cu	Ag	Al	Pb	Sn
p	1	1	1	3	4	4
σ	0.97	9.0	10.5	2.7	11.3	5.8
D	0.28	0.38	0.20	13	115	55

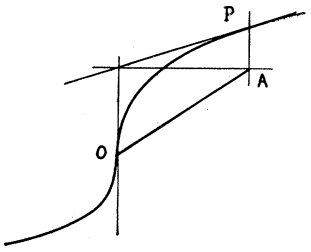

FIG. 2. The construction to find $DCL\alpha$. O is the estimated center of the kink and P is any point on the curve.

The meaning of the set of coefficients $DCL\alpha$ in the interpretation of experimental results may be illustrated very simply. It is a property of functions of the form

$$y(x)=ax\ln(bx)+cx,$$

that

$$x^{-1}(y-xdy/dx)=a,$$

whence it is clear that in the construction of Fig. 2 the gradient of OA is equal to a and is independent of the position of P. This provides a simple means of testing whether a kink in an experimental dispersion curve has the appropriate form to be considered a Kohn anomaly, and, if so, of evaluating the factor $DCL\alpha$.

EFFECTS OF ELECTRON INTERACTIONS

Equation (2) describes the phonon frequencies in terms of the matrix element for scattering of independent quasiparticles. If we were to start with a set of interacting electrons in Bloch states, we should need to known the transformation by which the matrix element $M(\mathbf{q})$ of the quasiparticle gas could be derived from that for the Bloch electrons $M_0(\mathbf{q})$. Such an expression has been given by Bardeen,[9] and an essentially similar one by Nakajima,[5] who finds

$$M(\mathbf{q})=M_0(\mathbf{q})\left(1+F(\mathbf{q})\sum_{\mathbf{k}}\frac{f_{\mathbf{k}}}{\mathcal{E}_{\mathbf{k}}-\mathcal{E}_{\mathbf{k}+\mathbf{q}}\pm\hbar\omega}\right)^{-1},$$

where $F(\mathbf{q})$ describes the electron interaction. We write this as

$$M(\mathbf{q})=M_0(\mathbf{q})(1+\beta)^{-1}.$$

Because the summation is the same as that of expression

[9] J. Bardeen, Phys. Rev. **52**, 688 (1937).

(3), the matrix element for the quasiparticle gas has the same rapid variation near $\mathbf{q}_0$ as has the frequency shift itself, and the magnitude of the kink is correspondingly diminished. In the limit of small interaction constant, this term enters the frequency shift as a factor $(1-2\beta)$.

This result is inconsistent with that of Nakajima, who calculates that the frequency shift contains a term of only $(1-\beta)$ in the same limit. It is simply seen—by examining for instance, the steps from Eqs. (2.4) to (2.5) in Ref. 4—that this difficulty is due to neglect of terms of order βM at all steps in the calculation save the last. It is thus difficult to estimate the magnitude of the coefficient α which describes these effects, because of uncertainty in both the validity of the Hartree approximation and the accuracy of the formalism in which it is used. Bardeen and Pines[10] have estimated using a collective description of the interactions that Nakajima's method is valid whenever $q<k_c$, the cutoff wave number for plasma oscillations. At greater wave numbers exchange terms may enter, reducing the effect of the electron interactions. The presence of the lattice potential also reduces the difference[9] between the long- and short-wavelength values of $M(\mathbf{q})$.

A simple form for $M_0(\mathbf{q})$ is given by the rigid-ion approximation,[11] which predicts that $M_0(\mathbf{q})$ is initially constant but begins to decrease rapidly as q becomes greater than Q. Electron interactions reduce its value for small q to something which depends on the Bloch character of the electrons and which is zero for free electrons. Presumably the Bloch character is most pronounced, and, hence, the effect of electron interactions least observable, in those metals in which the electron-phonon interaction is strongest, and in which the Kohn anomalies are consequently most likely to be observed. With this justification, and because we know so little about the magnitudes involved, we shall, in what follows, ignore the effects of electron interactions, putting α equal to unity.

GEOMETRICAL CONSIDERATIONS

We can distinguish three separate cases in which Eq. (2) predicts that in principle an anomaly will occur:

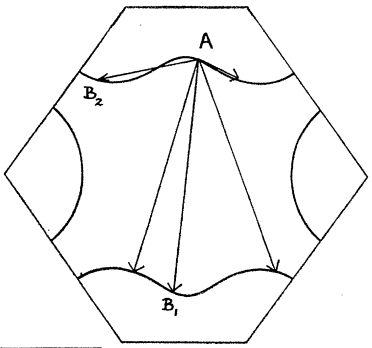

FIG. 3. For a multiply connected Fermi surface, there may be many different situations in which anomalies can occur.

(i) *A and B equivalent points.* In this case expression (4) simplifies, since $\mathbf{v}_a=-\mathbf{v}_b$ and θ is zero. This is the situation envisaged by Kohn in which the effect occurs for a phonon that spans the Fermi surface;

$$\mathbf{q}\simeq 2\mathbf{k}_f+\mathbf{g}.$$

(ii) *A and B nonequivalent points in the same band.* When the Fermi surface is everywhere convex, there can clearly be no two nonequivalent points where the electron velocities are parallel. For a multiply connected Fermi surface, however, this is not the case, and there may be many different points on the Fermi surface where the electron velocities are parallel (Fig. 3). When there is a minimal amount of symmetry in the crystal structure, such points occur in pairs and there may be some cancellation between them.

An important point about such transitions as these is that the electron velocities on the two parts of the Fermi surface may be parallel rather than antiparallel. In the schematic diagram of a section of the Fermi surface of Cu shown in Fig. 3 for instance, the point A is on the "belly" of the surface while B_2 is a saddlepoint situated on one of the "necks." In this case, $(\mathbf{v}_b-\mathbf{v}_a)^{-2}$ will be larger than either v_b^{-2} or v_a^{-2} and the factor C may be much greater than unity. In the model of nearly free electrons[12] which was used elsewhere,[13] C for Cu was found to be roughly equal to 4. This is insufficient to counteract the effects of the low value of D given in Table I, and to render the anomaly observable.

(iii) *A and B in different bands.* The geometrical factor C for such transitions may be large for the same reasons as in case (ii). However, anomalies may not be present in certain crystal directions if the matrix element connecting these states falls to zero or is small. It is commonly true, especially in divalent metals, that when electron and hole surfaces coexist they lie very near to the boundaries of the Brillouin zone. As is well known, states that are actually on the zone boundary possess definite symmetry characteristics. The electron-phonon interaction is consequently incapable of causing transitions between states that are on the same zone boundary but in different bands. Since phonon spectra are normally measured along symmetry directions, anomalies may frequently be absent where they might otherwise have been expected.

There is one further situation in which the effect vanishes for reasons of symmetry. That is when the wave number at the Fermi surface happens to be equal to one quarter of a reciprocal lattice vector. There is then a positive anomaly superimposed upon a negative one of equal magnitude, and no net kink is observable.

APPLICATION TO Pb

Lead is of especial interest, both because the coefficient D is so large for this metal and because the phonon

[10] J. Bardeen and D. Pines, Phys. Rev. **99**, 1140 (1955).
[11] Ref. 7, p. 185.
[12] N. F. Mott and H. Jones, *Metals and Alloys* (Clarendon Press, Oxford, England, 1936), p. 59.
[13] P. L. Taylor (to be published).

spectrum and Fermi surface have been the subject of careful experimental investigation. It is thought to contain four conduction electrons per atom, forming a Fermi surface consisting of a simple hole surface in the second zone and a multiply connected electron surface in the third zone.[14–17] A section through a (110) plane is shown in Fig. 4, based on the application of the model of nearly free electrons to Gold's data[14] on the de Haas–van Alphen effect. We shall attempt to show that such a surface is fully compatible with the phonon spectrum in the [110] direction determined by Brockhouse *et al.*[2] from neutron scattering experiments and, hence, also with Paskin and Weiss' results from x-ray experiments.[18]

On a surface as complex as that proposed by Gold, there will be many different pairs of points which might cause anomalies. However, it has been estimated by

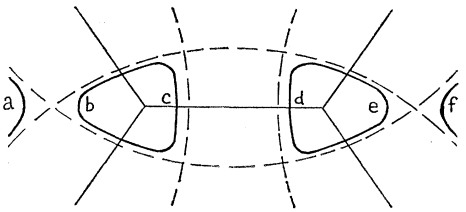

FIG. 4. A section in a (110) plane through the Fermi surface of lead. The broken lines are the free-electron surface and the solid lines a proposed surface.

that author that the portions of the surface of which Fig. 4 is a section are close to being cylindrical in shape. One of the radii of curvature appearing in the expression for the coefficient C will then be large, and we may expect these regions to dominate the effect. Figure 5 shows on the same scale the lengths of wave vector in the [110] direction at which anomalies may be expected on the basis of this model together with a rough estimate of their relative magnitudes, calculated using a nearly free electron model. For the reasons of symmetry discussed earlier, transitions cannot occur from regions a and f to b, c, d, or e, and the transitions from b to d are largely cancelled by those from c to e. Because a and f are a large distance from any zone boundary, umklapp processes are unlikely to be able to cause any transitions between them. The large distance between these points also means that $M(q)$ will be small, and so the value of L for these transitions is correspondingly small.

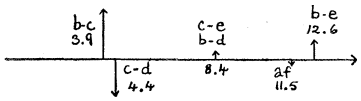

FIG. 5. The positions at which kinks in the phonon spectrum in the [110] direction are predicted, with a rough indication of their magnitude. The positions are marked in units of $0.1(2\pi/a)$.

A comparison of Fig. 5 with the experimental phonon dispersion curve (Fig. 3 of first Ref. 2) shows that a satisfactory agreement has been obtained. There is a large kink at $q\simeq 0.4(2\pi/a)$ which can be accounted for by the combined effects of the transitions $b-c$, $c-d$, and $d-e$, and an upward anomaly at $q\simeq 1.24(2\pi/a)$ which coincides with the position predicted for transitions $b-d$.[19] The uncertainty with which the radii of curvature in the plane perpendicular to that of Fig. 4 are known makes a comparison of the magnitude of the kink with the theoretical prediction difficult; there is, however, no obvious inconsistency.

It thus seems reasonable to suppose that Kohn anomalies are experimentally observable, in certain metals at least, and that the drastic reduction in the effect, suggested by Woll and Kohn, need not always occur. It is perhaps also worth noting that this effect is rare among Fermi surface tools in being capable of distinguishing between electron and hole surfaces.

ACKNOWLEDGMENTS

Thanks are due to the Magnavox Company for their generous support of this work, and to Dr. Paskin for some comments on his experimental results.

[14] A. V. Gold, Phil. Trans. Roy. Soc. (London) **A251**, 85 (1958) and Phil. Mag. **5**, 70 (1960).
[15] W. A. Harrison, Phys. Rev. **118**, 1190 (1960).
[16] A. R. Mackintosh, Proc. Roy. Soc. (London) **A271**, 88 (1963).
[17] J. A. Rayne, Phys. Rev. **129**, 652 (1963).
[18] A. Paskin and R. J. Weiss, Phys. Rev. Letters **9**, 199 (1962).

[19] B. N. Brockhouse *et al.*, (first Ref. 2), in a paper which makes some general comments similar to those presented here on the magnitude of anomalies, has attributed this kink to transitions $a-f$. This cannot be correct, since such transitions could lead only to a downward anomaly.