The n_{eff} plot corresponding to Eq. (13) approaches the value 2, the number of valence electrons in Zn, but it is not an asymptotic approach as, e.g., in the case of Al.[15] The n_{eff} plot corresponding to Eq. (14) beings to increase only near the plasma frequency. The absence of saturation in these cases is perhaps related to the presence of d states within a few eV of the valence band. Even so, the curve corresponding to Eq. (13) is quite simple.

Note added in proof. The reflectance was measured at 100 Å intervals for $\lambda > 1600$ Å, and at 25 Å intervals at shorter wavelengths. The curves in Fig. 3 are drawn exactly through the measured reflectances. The rms deviation between the calculated optical constants and the smooth curves shown in Figs. 4 and 5 is about 5% for $\lambda > 1400$ Å; it is about 10% for $\lambda < 1400$ Å.

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Magnetoresistance of Potassium

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The linear magnetoresistance of single-crystal specimens of potassium, which is observed far into the high-magnetic-field regime, is not in accord with generally accepted ideas concerning the bands structure of potassium. The linear dependence of resistivity on field is, however, in accord with a charge-density-wave model for the metal. The charge-density wave modifies the Fermi surface by introducing many energy gaps that slice the Fermi surface. These heterodyne gaps undergo progressive magnetic breakdown. The model predicts the largest magnetoresistance for crystals in which the magnetic field is oriented along the [100] or [111] directions, and the smallest effect for crystals in which **H** is oriented parallel to [110]; these predictions are in agreement with observation.

I. INTRODUCTION

ONE of the most puzzling and unexplained observations on the alkali metals is that of their magnetoresistance: first, that they show mangetoresistance at all, and secondly, that the magnetoresistivity is linear in field to the highest magnetic fields measured.[1,2] Attempts to explain away the magnetoresistance as due to probe effects have not succeeded since probeless techniques also yield a linear change in resistance with field.[3] Recently, Penz and Bowers[4] used the helicon method to determine the magnetoresistivity of single-crystal specimens of high-purity potassium to fields of 55 kG and of polycrystalline potassium to 110 kG. Although the results show the magnetoresistivity to vary somewhat with crystallographic direction, it again is linear in field over the range studied.

It is well known that a degenerate electron gas with a spherical Fermi surface shows no magnetoresistance, and with a closed (although not necessarily spherical) Fermi surface shows constant magnetoresistance in large magnetic fields. It is generally believed that the alkali metals can be characterized by such a model and that their Fermi surfaces (at least those of sodium and potassium) deviate only very slightly from sphericity.[5] Even if the Fermi surfaces are more complicated than is generally believed, it is difficult to understand the *linear* magnetoresistance; theory predicts[6] that the transverse magnetoresistance should saturate in the high-field regime if all orbits are closed, but should vary as H^2 for certain crystallographic directions if open orbits are present. (The situation is somewhat different in compensated metals, but here again the only predicted behavior at high fields is an H^2 dependence or saturation.) Once open orbits are admitted, an approximately linear magnetoresistance over a restricted range of magnetic fields might be obtained through an averaging over various pieces of the Fermi surface. But to achieve this result, a complicated Fermi surface would be required. Since the Fermi surfaces of the alkali metals are generally thought to be simple (and closed) the resistance versus field ought to saturate for $\omega_c\tau > 1$, whereas experimentally it remains linear to $\omega_c\tau \approx 100$. (Here ω_c is the cyclotron frequency and τ is the relaxation time for scattering of electrons.)

[1] E. Justi, Ann. Physik **3**, 183 (1948).
[2] D. K. C. MacDonald, in *Handbuch der Physik*, edited by S. Flügge (Springer-Verlag, Berlin, 1956), Vol. 14, p. 137.
[3] F. E. Rose, Ph.D. thesis, Cornell University, 1964 (unpublished).
[4] P. A. Penz and R. Bowers, Solid State Commun. **5**, 341 (1967); P. A. Penz, Ph.D. thesis, Cornell University, 1967 (unpublished).
[5] D. Shoenberg and P. J. Stiles, Proc. Roy. Soc. (London) **A281**, 62 (1964); M. J. G. Lee, *ibid.* **A295**, 440 (1966).
[6] I. M. Lifshitz, M. Ya. Azbel, and M. I. Kaganov, Zh. Eksperim. i Teor. Fiz. **31**, 63 (1956) [English transl.: Soviet Phys.—JETP **4**, 41 (1957)].

The purpose of this paper is to show that one model of the Fermi surface of potassium does in fact lead to a linear magnetoresistivity over a large range of magnetic fields, although it would eventually saturate at very high fields. The model requires the existence of either a charge-density wave (CDW) or a spin-density wave (SDW), which is orientable by the field. A CDW (or SDW) state has not been established conclusively for any of the alkali metals, but the existence of such a state has been postulated to explain other anomalies which have not been explained on other bases.[7–9] The CDW (or SDW) modifies the Fermi surface by introducing many additional energy gaps which are progressively *broken down* by the magnetic field through the phenomenon of magnetic breakdown.[10] The transverse magnetoresistance shows a linear dependence upon field throughout the breakdown region.

In Sec. II we review the experimental situation for single crystal potassium. In Sec. III we discuss several model calculations of magnetoresistance in which magnetic breakdown plays a role. Finally, in Sec. IV we discuss the CDW model of potassium and calculate its magnetoresistance.

II. SINGLE-CRYSTAL MAGNETORESISTIVITY OF POTASSIUM

Penz and Bowers[4] measured the transverse magnetoresistance of single crystals prepared from high purity potassium with residual resistance ratios (RRR) at 4.2°K in the range 1000 to 4000. These were measured to 55 kG, which for the purity mentioned corresponds to a $\omega_c\tau$ of the order of 100. Penz and Bowers used a probeless technique, namely, the helicon method; with the z direction being defined as that of the magnetic field, this technique measures $\frac{1}{2}(\rho_{xx}+\rho_{yy})$.

Their results are summarized in Table I in which the data are presented in terms of the parameter S, defined as

$$S=\frac{\rho(H)-\rho(0)}{\rho(0)\omega_c\tau}\times 100\%. \quad (1)$$

S is essentially the normalized slope of the linear variation in resistivity ρ as a function of field H. One interesting result is that S apparently depends upon the orientation of magnetic field relative to the crystallographic axes. When the field is oriented along [100] or [111] the magnetoresistance (averaged over the samples measured) appears to be two and one half times larger than when the field is oriented along [110]. This is an important point for the present paper, since the model to be discussed in Sec. IV predicts strong directional dependences. For fields oriented along [123] the situation is less clear since the spread in S values is quite large; however, the average S is less than that for [100] or [111].

TABLE I. Magnetoresistivity of single-crystal potassium.[a]

Sample	[hkl] field direction	$\omega_c\tau$ at 55 kG	S(%)	10^{-3} RRR
1	100	133	0.34	3.1
2	100	146	0.42	3.4
3	100	143	0.26	3.4
4	100	56	0.54	1.3
5	100	46	0.54	1.1
6	100	64	0.65	1.5
7	100	63	0.43	1.5
8	100	61	0.25	1.4
9	110	143	0.20	3.4
10	110	39	0.18	0.9
11	110	54	0.18	1.3
12	110	61	0.20	1.4
13	110	74	0.26	1.7
14	110	50	0.10	1.2
15	110	60	0.10	1.4
16	111	165	0.55	3.9
17	111	127	0.31	3.0
18	111	136	0.33	3.2
19	123	92	0.55	2.2
20	123	139	0.22	3.3
21	123	166	0.46	3.9
22	123	59	0.22	1.4
23	123	49	0.10	1.2
24	123	100	0.10	2.4

[a] Reference 4.

The samples used to obtain the data in Table I were prepared in such a way that they were subjected to a minimum amount of strain. By deliberately straining the crystal Penz was able to increase the slope S by as much as a factor of two. Presumably the spread in S values for various specimens with the same field orientation is due to small residual strains in these crystals.

III. MODEL CALCULATIONS OF MAGNETORESISTANCE IN METALS WITH MAGNETIC BREAKDOWN

Magnetoresistance calculations for metals with specified Fermi-surface topology are usually made by a semiclassical approach in which the time dependence of the electron's group velocity $\mathbf{v}$ is determined from the equations

$$\hbar\dot{\mathbf{k}}=-(e/c)\mathbf{v}\times\mathbf{H}, \quad (2)$$

$$\mathbf{v}(\mathbf{k})=\hbar^{-1}[\partial\epsilon(\mathbf{k})/\partial\mathbf{k}]. \quad (3)$$

The conductivity tensor σ_{ij} as a function of magnetic field may be calculated by the path integral method[11]

[7] A. W. Overhauser, Phys. Rev. Letters **13**, 190 (1964).
[8] D. R. Gustafson and G. T. Barnes, Phys. Rev. Letters **18**, 3 (1967).
[9] A. W. Overhauser, Phys. Rev. **167**, 691 (1968). This paper introduces the concept of the charge-density wave and shows that the earlier discussed anomalies can be explained in terms of either the CDW or the SDW. As is further shown in this paper, the CDW appears to be a more likely candidate for the alkali metals.
[10] M. H. Cohen and L. M. Falicov, Phys. Rev. Letters **8**, 231 (1961); E. I. Blount, Phys. Rev. **126**, 1636 (1962).
[11] R. G. Chambers, Proc. Phys. Soc. (London) **A65**, 458 (1952); **A238**, 334 (1956).

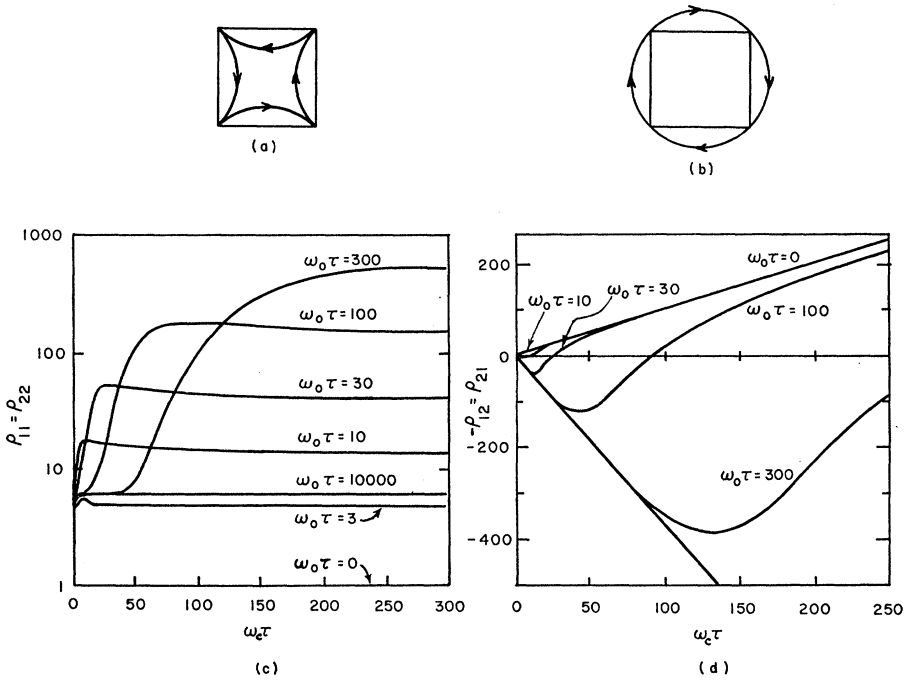

FIG. 1. Magnetoresistance and Hall resistance for a transition via magnetic breakdown from a closed-hole to a closed-electron orbit [from Falicov and Sievert (Ref. 12)]. (a) The low-field orbit (as $H\rightarrow 0$); (b) the high-field orbit (as $H\rightarrow\infty$); (c) the transverse magnetoresistance; (d) the Hall resistance.

yielding

$$\sigma_{ij}=-(e^2/4\pi^3)\int_{\text{all k}} v_i(\mathbf{k})\frac{df_0}{d\epsilon}d^3k\int_{-\infty}^{t(k)} v_j(s)\times\exp\left[\frac{s-t}{\tau}\right]ds. \quad (4)$$

Here f_0 is the electron distribution function, $\mathbf{k}$ is the electron's wave number, and $\mathbf{H}$ is the magnetic field. This method does not include quantum oscillatory effects (de Hass–Shubnikov oscillations) but does reproduce the gross features of the field-dependent conductivity.

When magnetic breakdown effects the important, the connectivity of the various electron orbits is changed as a function of field, and Eq. (4) cannot be used directly. Falicov and Sievert[12] devised a suitable matrix generalization of (4) which allowed them to include magnetic breakdown, and, as a result, they were able to investigate a large number of models in which the connectivity of the Fermi surface is modified in a prescribed way by the magnetic field. Thus, for example, they studied the transition from open to closed orbits, the transition from extended orbits to circular closed orbits, and transitions from closed hole-like orbits to closed electronlike orbits. Each model calculation was investigated as a function of the following parameters: ω_c, ω_0, and τ. $\omega_c\equiv eH/mc$ is the cyclotron frequency, τ is the relaxation time, and ω_0 is a frequency related to the probability of magnetic breakdown; in fact, the probability of an energy gap being broken down by the field is $\exp(-\omega_0/\omega_c)$ per passage through the gap region.

[12] L. M. Falicov and P. R. Sievert, Phys. Rev. **138**, A88 (1965).

An alternative definition of ω_0 is the following: at high magnetic fields, when the orbit is almost completely broken down, ω_0 is the probability per unit time of Bragg reflection into the low-field topology by the energy gap under consideration. Thus, the effective scattering time at high magnetic field is

$$\tau_{\text{eff}}^{-1}=\tau^{-1}+\omega_0. \quad (5)$$

ω_0 depends upon the magnitude of the energy gap Δ. Falicov and Sievert write $\omega_0=C\Delta^2/\epsilon_F\hbar$ with C a constant of order unity, and ϵ_F is the Fermi energy. Actually, as has been pointed out by many authors,[13–15] the "constant" C depends on the geometry of the orbits, and ω_0 may be written as

$$\omega_0=\frac{\Delta^2 k_F^2}{8\hbar\epsilon_F\mathbf{K}\cdot(\mathbf{k}\times\mathbf{b})}, \quad (6)$$

where $\mathbf{K}$ is the wave vector corresponding to the energy gap Δ and $\mathbf{b}$ is a unit vector in the direction of the magnetic field.

Of the various idealized calculations considered by Falicov and Sievert, the only ones that appear to apply to the potassium case (to be discussed in Sec. IV) are transitions from extended closed orbits to circular closed orbits, and transitions from closed (hole) orbits to closed (electron) orbits. Only the latter model calculation seems capable of explaining the *linear* magnetoresistivity of the metal. The results of Falicov and Sieverts calculation for this model are presented in Fig. 1.

[13] A. S. Joseph, W. L. Gordon, J. R. Reitz, and T. G. Eck, Phys. Rev. Letters **8**, 334 (1961).
[14] W. A. Harrison, Phys. Rev. **126**, 504 (1962).
[15] J. R. Reitz, J. Phys. Chem. Solids **25**, 53 (1964).

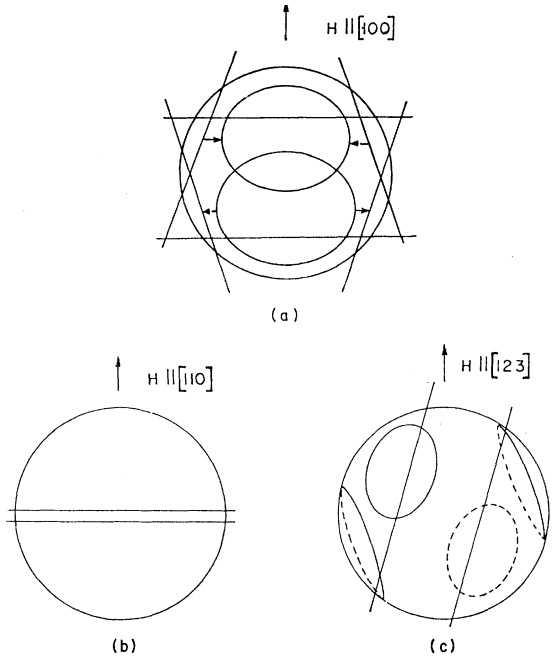

FIG. 2. Structure of the Fermi surface of potassium as modified by the heterodyne periods. The **Q** vector is assumed parallel to the magnetic field **H**. (a) **H**‖[100]; (b) **H**‖[110]; (c) **H**‖[123]. The deformation of the Fermi surface at the heterodyne gaps and the conical points of the surface (due to the **Q** vector, itself) are neglected. A low-field hole orbit is marked out by the arrows in (a); the broken arrows correspond to paths on the back side of the sphere.

IV. FERMI SURFACE OF POTASSIUM

We are now in a position to discuss the Fermi surface of potassium and its effect on the magnetoresistance of this metal. The Fermi surface to be discussed derives from a charge-density-wave model with a single charge-density wave of wave vector **Q** which is orientable by magnetic fields of 10 kG or more. The orientation effect is such that **Q** aligns approximately parallel to the field.[16] Stability arguments demand that Q be slightly larger than the diameter of the Fermi surface, and calculations[7] for potassium indicate that $Q=1.33(2\pi/a)$, where a is the lattice constant.

Since $Q>2k_F$, the charge-density periodicity does not change the connectivity of the Fermi surface. There are, however, other periodicities in the structure; there are, for example, periodic potentials associated with the reciprocal-lattice vectors $\mathbf{G}_{hkl}$. But gaps arising from these do not intersect the Fermi surface either. Further, there are beat periods or heterodyne periods arising from interaction of the charge-density and lattice periods.[17]

[16] An SDW model is equally acceptable. The **Q** vector does not have to align exactly parallel to the field, but can align approximately along the field, picking a compromise position determined by crystal strain and magnetic field.

[17] The importance of subsidiary periods and their effect on galvanomagnetic properties has been studied previously for the case of Cr: W. M. Lomer, in *Proceedings of the International Conference on Magnetism, Nottingham, 1964* (The Institute of Physics and the Physical Society, London, 1965), p. 127; L. M. Falicov and M. J. Zuckermann, Phys. Rev. **160**, 372 (1967).

The most pronounced heterodyne periods are those governed by the relation

$$\mathbf{K}=2\pi\mathbf{G}_{khl}\pm\mathbf{Q} \tag{7}$$

and these give rise to energy gaps which we denote by Δ. We thus have an energy-band structure which depends on the orientation of the magnetic field.

Figures 2 and 3 show *all* of the periodicities (7) whose **K** is less in magnitude than the diameter of the Fermi surface for magnetic fields (and **Q**) oriented along [100], [110], [123], and [111]. These give rise to energy gaps and an associated zone structure. The magnitude of the energy gaps Δ is unknown; this will be treated as a parameter to be determined from fitting to the magnetoresistivity. For two of the orientations shown there is a group of hole-like orbits at low fields. For $H\|[100]$ the orbit marked with arrows is such an orbit (solid line represents that part of the orbit on the front surface of the sphere and broken line that part on the rear surface). For $H\|[111]$, a remapping of the orbit on the midplane shows one of the hole orbits. These hole orbits are converted into electron orbits at high fields as a result of magnetic breakdown. Magnetic breakdown causes other connectivity changes also—small electron orbits being converted into large electron orbits,—but the largest effects and those which contribute to the linear magnetoresistivity are those caused by the change from hole orbits to electron orbits. For **H** strictly parallel to [110] there is no change in connectivity as a func-

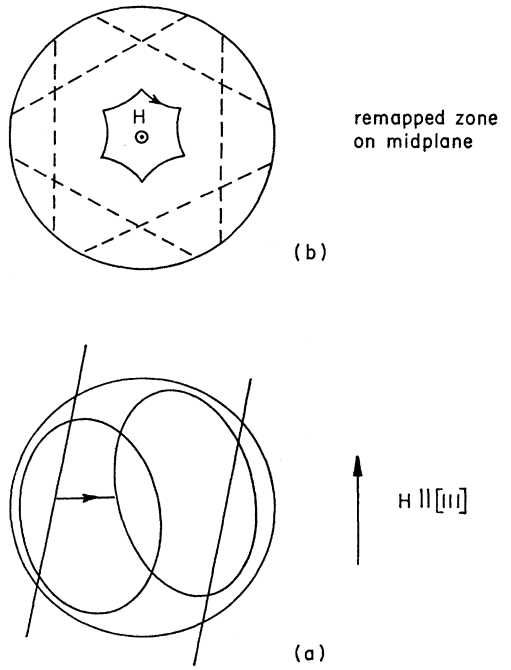

FIG. 3. Same as Fig. 2. (a) **H**‖[111]. The arrow marks part of a low-field-hole orbit; (b) A cut through the midplane of the Fermi surface shown in (a). Parts of the Fermi surface are remapped inside the zone formed by the heterodyne planes in order to show the hole orbit.

tion of field. If **H** (and hence **Q**) is misaligned slightly from the [110] direction, there will be a few extended double orbits at low field which can be broken down. This effect does not lead to a linear magnetoresistivity. For **H**||[123] we again have only changes from extended orbits to circular orbits.

We return now to the [100] and [111] orientations, where we have hole orbits at low field. We have a group of orbits and hence a range of ω_0. [For the geometry considered the $\mathbf{K}\cdot(\mathbf{k}\times\mathbf{b})$ element gives rise to a $\cos\Theta$ factor, where Θ is the angle between the orbit tangent at the zone intersection and **K**.] At large $\omega_c\tau$ we thus have a magnetoconductivity which is made up predominantly from circular orbits but with a small admixture of orbits which are transforming (via magnetic breakdown) from hole orbits to electron orbits. These orbits are characterized by a range in the parameter, ω_0. The contributions to the conductivity from various parts of the Fermi surface add together.

In order to test this model to see if it produces a linear magnetoresistance, the calculations of Falicov and Sievert (Fig. 1) were inverted to obtain the components of their conductivity tensor as a function of $\omega_c\tau$. The conductivity of a small group of these orbits, with $\omega_0\tau$'s ranging from 50 to 150, was added to that of circular orbits without breakdown. The result (transformed back again to resistivity) is an essentially linear magnetoresistance over the range of interest: to $\omega_c\tau=150$. With breakdown orbits making up about 1% of the total orbits, the increase in resistivity over this range is 60%.

V. CONCLUSIONS

A charge-density-wave model for potassium in which the Fermi surface is modified by heterodyne gaps and varies with orientation of the magnetic field gives rise to a linear magnetoresistance for single crystals when the magnetic field is oriented either along the [100] or [111] direction. The resistance will eventually saturate at very high magnetic field. The intrinsic energy gap (corresponding to $\cos\Theta=1$), obtained by fitting the model to Penz and Bower's data, corresponds to $\omega_0\tau \approx 50$; this gives $\Delta=0.07$ eV. Such a value appears to be reasonable since the energy gap associated with **Q** has been estimated at 0.6 eV.

There is no linear magnetoresistance predicted for **H**||[110] and **H**||[123]; in fact, if the field and **Q** are oriented precisely along [110] in a strain-free specimen, we would predict no magnetoresistance at all. In a strained crystal the charge-density wave vector **Q** will pick a compromise alignment depending on the strain field and the magnetic field. This will produce a different zone structure. Since low-field hole orbits do not appear to be uncommon, orbits of this type may be mixed in, even though they do not appear in the "ideal" orientation. In order to test this possibility we have looked at the band structure resulting from small misalignments of **Q** and **H**. With **H**||[110], but **Q** misaligned 11° (toward [100]), three new pairs of heterodyne gaps intersect the Fermi surface. At a 13° misalignment a well developed hole orbit appears.

The Fermi-surface model described here predicts a number of small low-field orbits which can possible be observed in de Haas–van Alphen studies of the alkali metals. There are, however, reasons why these might not have been seen before: (1) the curvature of the Fermi surface in the vicinity of the extremal orbits may be so large that not enough orbits contribute in phase with one another to be observed; (2) since strain effects are more important at low magnetic fields there is the likelihood of competing zone structures at low field which would again make individual orbits more difficult to observe; and (3) low-field de Haas–van Alphen studies in strain-free alkali metal crystals have perhaps not been adequately investigated. One observation which lends support to the Fermi-surface model proposed here is that of Okumura and Templeton[18] concerning high-field de Haas–van Alphen studies in cesium. They found that the quality of de Haas–van Alphen signals depended upon crystal orientation in the magnetic field, the best signals being obtained when **H**||[110]. For other field directions, e.g., **H**||[111], it was difficult to obtain a signal at all.

Note added in proof. The model discussed in this paper predicts that the Hall coefficient of potassium should decrease with increasing H. This effect arises because the high-field Hall coefficient essentially measures the difference between electron and hole concentrations. P. A. Penz [Phys. Rev. Letters **20**, 725 (1968)] has indeed observed this phenomenon on both single-crystal and polycrystalline material. The largest effect was for a polycrystalline specimen, which had a reduction in Hall coefficient of 7% at $\omega_c\tau=300$.

[18] K. Okumura and I. M. Templeton, Proc. Roy. Soc. London **A287**, 89 (1965).