High-Field Magnetoresistance of bcc Iron

W. A. REED AND E. FAWCETT
Bell Telephone Laboratories, Murray Hill, New Jersey
(Received 16 April 1964; revised manuscript received 17 June 1964)

The transverse magnetoresistance of single crystals of bcc iron of the highest available purity, measured at a temperature of 4.2°K in applied magnetic fields up to 100 kOe, has a magnitude and field dependence which show that this metal is compensated. The curves showing the magnetoresistance as a function of the direction of the magnetic field exhibit narrow minima when the field lies in a {110} plane near a <001> axis and in a {001} plane near a <110> axis. These minima are attributed to sets of periodic open orbits directed along the <110> and <001> axes, respectively. The absence of complete saturation of the magnetoresistance, when the magnetic field is at any one of the minima, suggests either that the number of open orbits is very small, or that they result from magnetic breakdown. To distinguish experimentally between these two possibilities will require the use of higher magnetic fields and/or purer samples.

WE have measured the magnetoresistance of several single crystals of iron in the bcc phase at a temperature of 4.2°K in applied magnetic fields up to 100 kOe. The samples, approximately 1 mm in diameter and 15 mm long, were prepared by Materials Research, Inc., by a strain-annealing technique, and three were selected having their axes as close as possible to the three symmetry axes ⟨001⟩, ⟨110⟩, and ⟨111⟩. The residual resistivity ratios of these samples, measured between room temperature and 4.2°K in zero applied field, are 172, 206, and 220, respectively.[1] In the experimental apparatus used, the resistance is continuously recorded as the sample is rotated about its own axis, which can be tilted at any desired angle to the field of the solenoidal magnet.[2] The field dependence of the magnetoresistance can likewise be measured for any required direction of the field relative to the crystal axes.

The magnetoresistance ratio, $\rho(B)/\rho(0)$ of the three samples in an applied transverse field, $H=100$ kOe, is between 10 and 20. The transverse magnetoresistance is nearly isotropic, varying by less than $\pm 10\%$ as each sample is rotated about its axis. When the transverse magnetoresistance ratio is plotted against the magnetic induction[3] B, the exponent m in the expression

$$\rho(B)/\rho(0)=\text{const}\, B^m, \qquad (1)$$

which best fits the curve at the higher fields, varies for the three samples from 1.60 to 1.93 for general field directions not coincident with symmetry planes or symmetry axes.

All metals are either compensated, having a net number n_A of carriers per unit cell equal to zero (i.e., having equal numbers of electrons and holes), or uncompensated, having n_A equal to a positive or negative integer.[4] The theory of Lifshitz *et al.*[5] shows that, for general field directions, the transverse magnetoresistance for a compensated metal has a field dependence which approaches a quadratic power law ($m=2$) in the high-field region, whereas for an uncompensated metal it approaches saturation ($m=0$). The high-field region is defined formally by the inequality $\omega_c\bar{\tau}\gg 1$ for *all* carriers, ω_c being the cyclotron frequency and $\bar{\tau}$ the relaxation time for scattering of the carrier

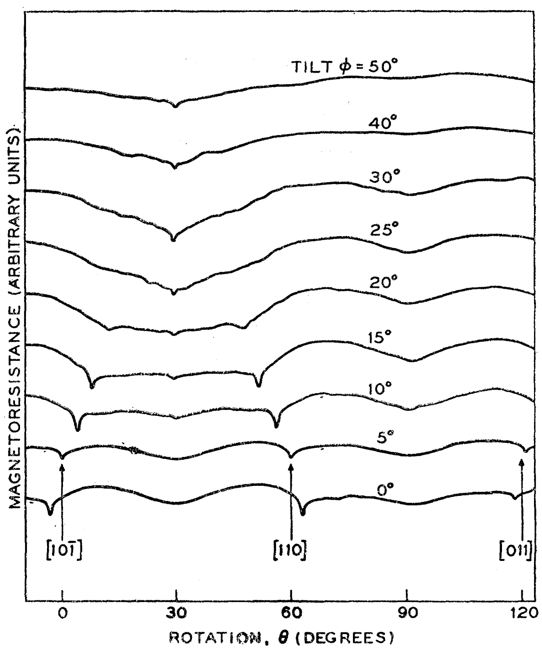

FIG. 1. Magnetoresistance of iron. The axis of the ⟨111⟩ sample is about 5° from the [1$\bar{1}$1] axis in the stereogram of Fig. 2. The resistance is plotted as the sample is rotated about its own axis (angle θ), which is tilted at various angles φ to the applied field, $H=90$ kOe (the dotted lines in Fig. 2 show the locus of **H** for $\varphi=0°$, 20° and 40°). Each curve is displaced vertically by an equal interval at the left-hand ordinate axis from the curves for neighboring values of φ, so that the zero of each curve, as well as the ordinate scale, is arbitrary.

[1] The field dependence of the transverse magnetoresistance at a temperature of 4.2°K in applied magnetic fields up to 80 kOe of an iron single crystal of random orientation having a residual resistivity ratio of 191 was described previously (see Ref. 4).

[2] This apparatus will be described in a paper now in preparation [G. F. Brennert, W. A. Reed, and E. Fawcett (to be published)].

[3] We use the expression $B=\mu_0(H+2\pi M)$, with a demagnetization factor, $2\pi M\sim 11$ kOe, for iron, appropriate to the cylindrical geometry of the sample.

[4] E. Fawcett and W. A. Reed, Phys. Rev. **131**, 2463 (1963).

[5] 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)].

averaged around its cyclotron orbit. In practice,[6] the behavior of a compensated metal in the high-field region is recognized by the conditions $\rho(B)/\rho(0)\gg 1$ and $m\lesssim 2$, whereas for an uncompensated metal $\rho(B)/\rho(0)\sim 1$ and $0<m<1$. Using these criteria, we conclude that iron is a compensated metal. Since *all* carriers must be in the high-field region to observe the effects of compensation, we conclude further that there are *only* itinerant electrons and holes in iron.[4]

A particularly interesting feature of the data is that the curves showing the magnetoresistance ratio of the ⟨111⟩ sample (i.e., the sample with its axis near [1$\bar{1}$1]) as a function of the rotation angle θ for various tilt angles φ exhibit symmetric sets of narrow minima (Fig. 1). For $\varphi\leq 20°$ the positions of these minima on a stereogram (Fig. 2) occur when the field **H** lies in a {001} symmetry plane near a ⟨110⟩ symmetry axis. For $\varphi\geq 20°$ these minima are replaced by a shallower, but still quite distinct, minimum when **H** lies in the (011) plane near the [100] axis. The latter belongs to the {110}-⟨001⟩ set, this notation indicating that minima of the set occur when **H** lies in a {110} plane near a ⟨001⟩ axis. Thus the minima belonging to this set in the (10$\bar{1}$) plane are absent in Figs. 1 and 2, since the closest approach of **H** to [010] is an angle of 30°, when $\varphi=0°$.

Minima belonging to the former set, i.e., the {001}-⟨110⟩ set, occur also in the ⟨110⟩ sample. They are not seen in the ⟨001⟩ sample since **H** lies in the (100) and (010) planes near the [011] and [101] axes only when the tilt angle is large ($\varphi\sim 45°$, see Fig. 2), and these shallow minima cannot be distinguished above noise at such a close approach to the longitudinal orientation ($\varphi=90°$). Conversely, minima belonging to the {110}-⟨001⟩ set are not seen in the ⟨110⟩ sample at large tilt angles near the [100] axis, but they do appear at small tilt angles in the (110) plane near the [001] axis. The minima at [100] and [010] in the ⟨001⟩ sample are special cases of the {110}-⟨001⟩ set, which lie at the intersection of two {110} planes. No minima are resolved in this sample at other tilt angles, perhaps because it has a lower purity than the other samples, as its lower residual resistivity ratio suggests.

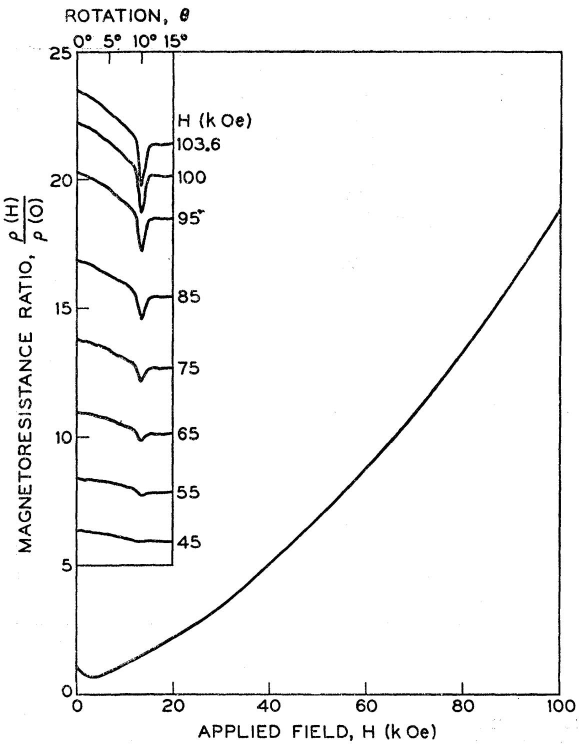

FIG. 3. Magnetoresistance of iron. The main diagram shows the field dependence of the magnetoresistance up to an applied field, $H=100$ kOe, when **H** is directed along the minimum near [10$\bar{1}$] of the curve $\varphi=10°$ of Fig. 1. The insert shows the latter curve in the neighborhood of the minimum for several values of H.

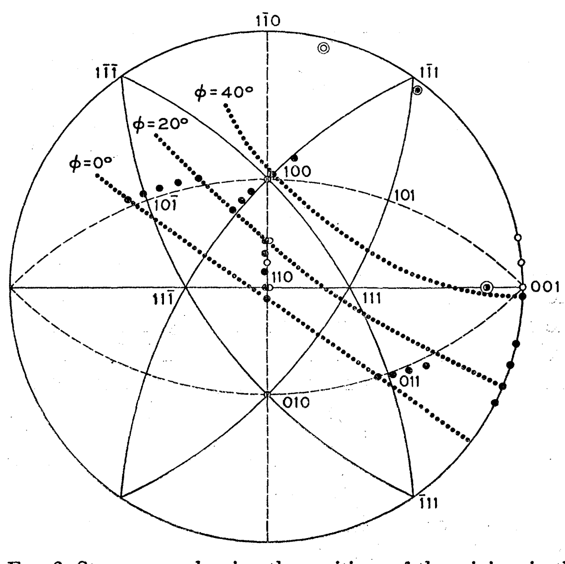

FIG. 2. Stereogram showing the positions of the minima in the magnetoresistance rotation curves for iron. The symbols ●, ○, and ◎ show the positions of the minima for the samples whose axes are shown by the same symbols circled. The continuous and dashed lines show the {110} and {001} planes, respectively. The dotted lines show the locus of **H** as the ⟨111⟩ sample is rotated about its axis at various tilt angles φ (see Fig. 1).

The curve showing the field dependence of the magnetoresistance is rather similar for all field directions measured in the three samples. It always exhibits a minimum at low fields ($H<10$ kOe) followed by a region of monotonically increasing slope up to the highest field. Presumably this minimum is a ferromagnetic phenomenon resulting from a combination of effects such as domain-wall scattering, spin-wave scattering, and magnetocrystalline anisotropy. Similar behavior is observed in nickel below the ferromagnetic saturation field.[7]

In Fig. 3 we show the field dependence curve at a

[6] E. Fawcett, Advan. Phys. (to be published, 1964).

[7] E. Fawcett and W. A. Reed, Phys. Rev. Letters 9, 336 (1962).

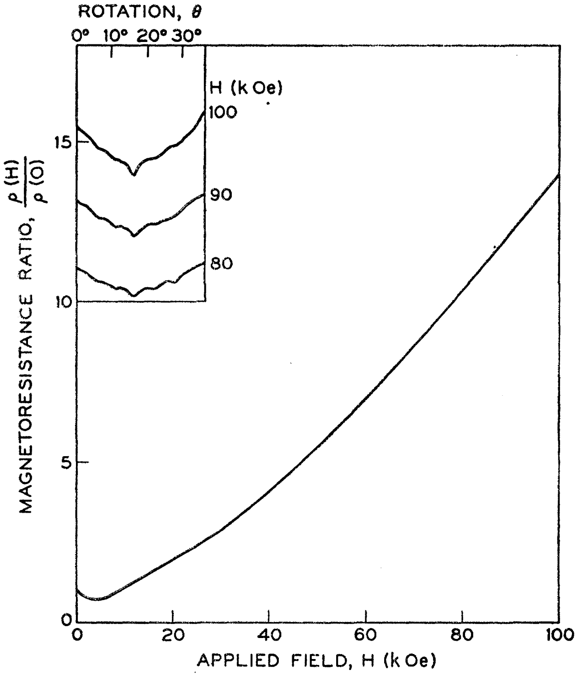

FIG. 4. Magnetoresistance of iron. The main diagram shows the field dependence of the magnetoresistance up to an applied field, $H=100$ kOe, when **H** is directed along the minimum at [001] for the ⟨110⟩ sample. The insert shows the variation of the magnetoresistance with the rotation angle θ in the neighborhood of the minimum for several values of H.

minimum of the {001}-⟨110⟩ set. The exponent m in Eq. (1) which best fits the curve has the value 1.71. At a neighboring orientation, near but not coincident with the minimum, the value of m is 1.93. The corresponding values of m at and near the minimum of the {110}-⟨001⟩ set shown in Fig. 4 are 1.56 and 1.60, respectively.

We attribute the two sets of narrow minima to the effects of two sets of periodic open orbits along the ⟨001⟩ and ⟨110⟩ axes. The fact that, in the ⟨111⟩ sample, as the tilt angle is changed, minima of one set disappear as minima of the other set appear (Fig. 2) suggests that both sets exist on the same open sheet of the Fermi surface.

According to theory,[5] the magnetoresistance should approach saturation when open orbits exist in a direction $\mathbf{\Omega}$ perpendicular to the current direction **J**. When **J** makes an angle $\alpha<\pi/2$ with $\mathbf{\Omega}$, the magnetoresistance approaches

$$\rho(B)/\rho(0)=\mu_0^2B^2\cos^2\alpha\,, \tag{2}$$

where μ_0 is a mobility characteristic of the open orbits and we consider only the high-field region for which $\mu B\gg 1$ for all carriers. In a compensated metal the magnetoresistance also has a quadratic field dependence at a neighboring field direction where there are no open orbits, and in the high-field region approaches

$$\rho(B)/\rho(0)=\mathcal{C}\langle\omega_c\bar{\tau}\rangle_{\text{av}}^2=\mathcal{C}\bar{\mu}^2B^2\,, \tag{3}$$

where $\bar{\mu}$ is an average mobility over the whole Fermi surface, and $\mathcal{C}$ is a field-independent coefficient which is of order unity when **B** is approximately transverse to **J**. We usually expect μ_0 to be of the same order of magnitude as $\bar{\mu}$, so that from Eqs. (2) and (3) the open orbits produce a minimum in the magnetoresistance curve when $\alpha\lesssim\pi/2$, although it would be possible for the open orbits to produce a peak if $\mu_0^2\cos^2\alpha>\mathcal{C}\bar{\mu}^2$. However, in either case, the feature of the magnetoresistance curve characteristic of a one-dimensional set of periodic open orbits in the narrowness of the associated set of minima or peaks which occur when **B** passes through the symmetry planes.

The fact that the magnetoresistance does not saturate at the bottom of the minimum shown in Fig. 3 ($m=1.71$) is consistent with the explanation that it results from open orbits along the [010] axis, since α is rather less than $\pi/2$ ($\alpha=55°$, see Fig. 2). But saturation of the magnetoresistance is also not observed in the ⟨110⟩ sample at the minimum near the [001] axis shown in Fig. 4 ($m=1.56$) and at the [110] axis ($m=1.50$), for which the corresponding values of α are close to $\pi/2$.

A possible explanation of this behavior is that the ratio D of the area of the Fermi surface traversed by open orbits to its total area is small. The magnetoresistance for $\alpha=\pi/2$ then saturates at a large value of the order $1/D$ only when $\mu_0B\gg 1/D$.[8] According to this explanation, the present measurements are in the intermediate range where $\mu_0B\sim 1/D$, and complete saturation at the minima will be achieved by employing higher magnetic fields, or purer samples to increase the mobilities.

An alternative explanation is that the open orbits result from magnetic breakdown, so that we can regard D as a function of B, starting from zero at low values of B and increasing rapidly at a critical field B_b toward a value D_∞ when magnetic breakdown is complete. B_b is given by an expression of the form[9]

$$(\epsilon_F\hbar\omega_b)^{1/2}=\Delta\epsilon\,, \tag{4}$$

where ϵ_F is the Fermi energy, $\Delta\epsilon$ is the energy gap where the magnetic breakdown occurs, and $\omega_b=eB_b/m^*c$ is the cyclotron frequency in the field B_b of the carriers of cyclotron mass m^* in the orbits which break down. According to this explanation, the present measurements are in the range of magnetic fields where magnetic breakdown is just beginning, i.e., $B_b\sim 100$ kG.

Note added in proof. After the measurements on the strain-annealed samples were completed, some single crystals of bcc iron in the form of whiskers were measured and found to have much higher magnetoresistance with deep saturating minima. These additional results provide clearer evidence for the existence of open orbits than the earlier work, without essentially changing the description of the high-field magnetoresistance of bcc

---

[8] E. Fawcett and W. A. Reed, Phys. Rev. **134**, A723 (1964).
[9] E. I. Blount, Phys. Rev. **126**, 1636 (1962).

iron. The iron whiskers were prepared by Dr. T. Turnbull and Mrs. J. Button of Lincoln Laboratory and were kindly supplied by Dr. S. Foner of National Magnet Laboratory.

Three of the largest whiskers were selected, having approximate dimensions 0.2 mm diam and 5 mm length, and 0.1-mm copper potential leads and heavier gauge current leads were attached with 60-40 solder. These samples were found to have similar anisotropy of their magnetoresistance in an applied field of 100 kOe. Their orientations determined by x-ray were found to be similar, the axis of each whisker being near a {100} plane but not coincident with either a ⟨001⟩ or ⟨011⟩ axis.

The curves showing the anisotropy of the magnetoresistance exhibit sets of narrow minima in the same positions as those plotted in Fig. 2. But now the minima in the {001} and {110} planes appear for *all* directions of **H** in these planes, though the former still dominate near the ⟨110⟩ axes and the latter near the ⟨001⟩ axes.

In Fig. 5, when **H** is directed along one of these minima (curve a) the magnetoresistance clearly approaches saturation. The other minima show a similar field dependence when the angle α between the axis perpendicular to the plane in which the minimum occurs and the current direction has a value $\alpha \lesssim \pi/2$ ($\alpha = 68°$ for the minimum at the [100] axis in Fig. 5). As for the strain-annealed samples (Fig. 2), no minimum is observed when **H** lies in a {001} or {110} plane for which $\alpha \gtrsim 0$, which shows that the observed minima are not due to discompensation (resulting for example from magnetic breakdown of electron orbits to form hole orbits, or vice versa, when **H** lies in these planes). These results confirm the conclusion that the minima result from open orbits along the ⟨001⟩ and ⟨110⟩ axes.

The magnetoresistance ratio referred to the resistance at room temperature is shown in the right-hand ordinate scale of Fig. 5, since from the field dependence of the magnetoresistance in applied fields, $\mathbf{H} < 1$ kOe, it appears that the resistance $\rho(0)$ in zero applied field at 4.2°K is not a suitable reference value. As H increases from zero the resistance falls to a minimum value $\rho_{\min}$ about a factor ten smaller than $\rho(0)$, when $H \lesssim 1$ kOe, and then rises until the field dependence obeys a quadratic power law between 10 kOe $\lesssim H \lesssim 40$ kOe (Fig. 5). The resultant broad minimum in the field dependence curves is similar to the much shallower minimum seen in the strain-annealed samples (Figs. 3 and 4), and presumably has the same origin.

To compare the high-field magnetoresistance of the whiskers with that of the strain-annealed samples, it would be desirable to plot the field dependence data on a Kohler diagram, in which the magnetoresistance ratio referred to the resistance ρ_0 when the induction B is zero is regarded as a function of B/ρ_0. This is not possible in a ferromagnet, since even when the macroscopic value of B is zero the microscopic value in each domain is equal to the saturation magnetization.

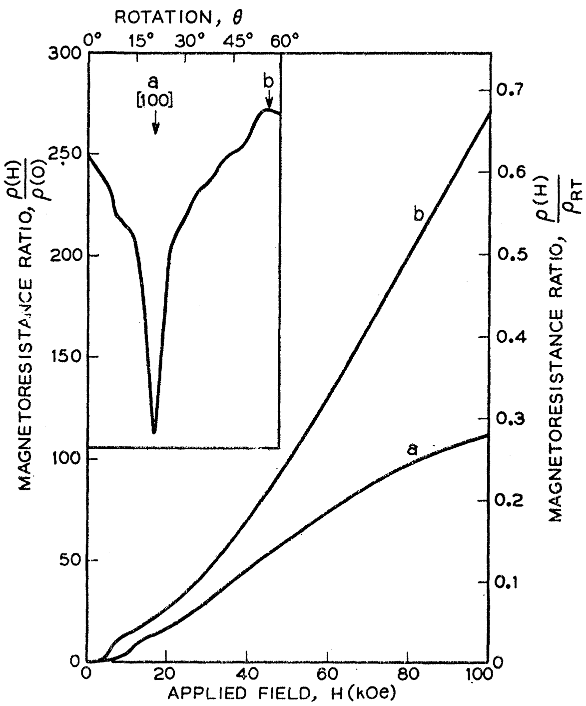

FIG. 5. Magnetoresistance of an iron whisker. The axis of the whisker is 6° from the (100) plane and 22° from the [001] axis. The main diagram shows the field dependence of the magnetoresistance up to an applied field, $H = 100$ kOe, when **H** is directed along the narrow minimum at the [100] axis (curve a) and at the neighboring broad maximum (curve b). The insert shows the variation of the magnetoresistance in an applied field, $H = 100$ kOe, for a tilt angle $\varphi = 5°$, as **H** passes through these directions. The left-hand and right-hand ordinate scales show the magnetoresistance ratio referred, respectively, to the resistance $\rho(0)$ in zero applied field at 4.2°K (as in Figs. 3 and 4), and to the resistance ρ_{RT} at room temperature (295°K).

Furthermore, the validity of Kohler's rule, that for a given metal and field direction the magnetoresistance is a function only of B/ρ_0 (and therefore $\langle\omega_c\bar{\tau}\rangle_{\text{av}}$), depends upon the assumption that the scattering processes remain unchanged by the application of the field. This assumption is not warranted in a ferromagnet, since one expects domain-wall scattering to vary with the applied field. However, it appears that $\rho_{\min}$, which presumably corresponds to the least domain-wall scattering, is a better approximation to ρ_0 than $\rho(0)$, since Kohler's rule is obeyed fairly well with this substitution when the field dependence data for a nonsymmetry field direction are compared.

With $\rho_{\min}$ as the reference value of the resistance, the magnetoresistance ratio at 100 kOe in the direction b of Fig. 5 is 2800, which shows on substitution into Eq. (3) that $\langle\omega_c\bar{\tau}\rangle_{\text{av}} \sim 50$. From Figs. 3 and 4, the corresponding value of $\langle\omega_c\bar{\tau}\rangle_{\text{av}}$ for the strain-annealed samples is about 5, so that the impurity scattering at 4.2°K in the whiskers is about one order of magnitude smaller.

Despite this considerable improvement in the purity, we are still unable to determine unequivocally whether the open orbits result from magnetic breakdown or from very small necks on a multiply-connected surface. The saturation value of the magnetoresistance ratio extrapolated from curve a of Fig. 5 and referred to $\rho_{\min}$ is about 1000. According to Falicov and Sievert,[10] the saturation value due to orbits produced by magnetic breakdown is

$$\rho_{\text{sat}}/\rho_0 = 1 + C\omega_b\tau, \tag{5}$$

where ω_b is the breakdown cyclotron frequency from Eq. (4) and C is a constant of order unity. If the open orbits in iron result from magnetic breakdown of closed orbits, we may therefore attribute the high value of the saturation value to a high value of $\omega_b\tau$, and write

$$\rho_{\text{sat}}/\rho_{\min} \sim C\omega_b\tau/D_\infty, \tag{6}$$

where D_∞ is the fraction of the Fermi surface traversed by open orbits.

Alternatively, if the open orbits are associated with very small necks, corresponding to a fraction $D \sim 10^{-3}$, then the saturation value of the magnetoresistance ratio is independent of $\bar{\tau}$. Unfortunately, it is not possible to make a reliable estimate of the saturation value of the magnetoresistance by extrapolating the field dependence curves in Figs. 3 and 4. Thus, the final conclusion as to the origin of the open orbits awaits further investigation of still higher purity samples.

[10] L. M. Falicov and P. R. Sievert, Phys. Rev. Letters 12, 558 (1964). In this paper, Eq. (5) is given for the cases when open orbits break down to form closed orbits, and when a compensated metal becomes uncompensated due to magnetic breakdown of hole orbits for form electron orbits. But the expression is also correct for the case of interest where closed orbits break down to form open orbits [L. M. Falicov (private communication)].

ACKNOWLEDGMENTS

We thank G. F. Brennert and R. D. Nafziger for orienting and mounting the samples for measurement.

PHYSICAL REVIEW VOLUME 136, NUMBER 2A 19 OCTOBER 1964

Spin-Wave Thermal Conductivity of Ferromagnetic EuS*

D. C. McCollum, R. L. Wild, and J. Callaway
University of California, Riverside, California
(Received 7 May 1964)

Experimental and theoretical investigations have been made of the spin-wave thermal conductivity of the ferromagnet europium sulfide. Measurements of the thermal conductivity from 1.5–4°K have been made on compressed powders. The theoretical temperature dependence of the spin-wave thermal conductivity below 4°K has been calculated for several external magnetic fields, assuming boundary scattering and considering first- and second-neighbor interactions. With no external magnetic field, the experimental conductivity can be approximately represented by $\mathcal{K} = 1.2T^2$ mW/cm(°K) which is reasonably close to the theoretical temperature dependence. However, an external field of 9 kG produced a drop in conductivity ranging from 2 or 3% at 4°K to 25% at 1.5°K; according to theory the conductivity should decrease by 11% at 4°K and 31% at 1.5°K. This disagreement between experiment and theory is quite marked at higher temperatures and will require further study for its resolution.

I. INTRODUCTION

HEAT transport in ferromagnetic insulators might be expected to occur by means of lattice vibrations and spin waves. A spin-wave thermal conductivity has recently been observed in ferrimagnetic yttrium iron garnet by Douglass[1] and by Lüthi.[2] Scattering of phonons by spin waves has been assumed by Douthett and Friedberg[3] to explain their measurements of ferrite thermal conductivities. Apparently no observations of spin-wave thermal conductivity have been reported for a ferromagnet. Assuming that magnons and phonons have the same constant scattering length, a rough estimate of the thermal conductivity of europium sulfide (ferromagnetic below about 16°K) in the liquid-helium temperature range shows that the spin-wave part should be much larger than the phonon part, as is the case with the specific heat.[4] The experiments discussed below were done in an attempt to discover whether or not spin waves do contribute appreciably to the thermal conductivity of EuS at these temperatures.

II. EXPERIMENTAL PROCEDURE

All of the rectangular specimens were cut from pellets compressed at 10 kbar. Single crystals have not been grown, as yet, due to the high temperatures required for growth by sublimation techniques. Earlier measurements of thermal conductivity in the liquid-helium temperature range were made of pressed pellets receiving no heat treatment after compression.[5] The

* Supported by the U. S. Air Force Office of Scientific Research.
[1] R. L. Douglass, Phys. Rev. 129, 1132 (1963).
[2] B. Lüthi, Phys. Chem. Solids 23, 35 (1962).
[3] D. Douthett and S. A. Friedberg, Phys. Rev. 121, 1662 (1961).
[4] D. C. McCollum, Jr. and J. Callaway, Phys. Rev. Letters 9, 377 (1962).
[5] R. L. Wild and D. C. McCollum, Jr., Bull. Am. Phys. Soc. 8, 208 (1963).