Galvanomagnetic Effects in Iron Whiskers*

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(Received 25 July 1966)

Measurements have been made of the temperature dependence of the Hall effect in the range 1–300°K, and of the transverse and longitudinal magnetoresistance at low temperatures, in [100] and [111] iron whiskers of high purity (resistance ratios extrapolated to $B=0$ ranged from 242 to 852). Analysis of the Hall data shows that both the "ordinary" Hall coefficient R_0 and the "extraordinary" Hall coefficient R_s vary enormously with temperature, and these variations cannot be explained by existing theories. The theoretical relation $R_s \propto \rho^2$, where ρ is the resistivity, does not hold for temperatures below that of liquid nitrogen, and around this temperature R_0, which is positive at room temperature (hole conduction), becomes negative. R_0 and R_s appear to be isotropic, but at low temperatures these coefficients are extremely sensitive to impurities. The longitudinal magnetoresistance at 4.2°K shows a large effect of the domain structure on the electrical resistance, which decreases appreciably (46% for the purest specimen) on magnetizing the specimens to saturation. Small negative magnetoresistance (~10%) has also been observed for some specimens in weak transverse fields.

I. INTRODUCTION

IN the description of the galvanomagnetic effects in ferromagnetic metals it is necessary to distinguish between the effects which depend on the total magnetic induction B and the effects which are explicitly related to the intrinsic magnetization M. Thus the Hall effect in a single domain of a ferromagnetic metal is usually described by the equation[1]

$$e_H = R_0 B + R_s 4\pi M\,, \qquad (1)$$

where e_H is the Hall resistivity or the Hall field per unit current density; the "ordinary" Hall coefficient R_0 describes the effect of the macroscopic average field in a manner similar to that for nonferromagnetic metals, whereas the "extraordinary" Hall coefficient R_s (also referred to as the spontaneous or anomalous Hall coefficient) is characteristic of the ferromagnet. As might be expected, the absolute values of R_0 are usually found to be of the same order of magnitude as those of the corresponding Hall coefficient for nonferromagnetic metals, but R_s is usually found to be much greater than R_0 except in very pure metals at low temperatures.

The anomalous Hall effect in ferromagnets has been subject to extensive theoretical and experimental investigation. On the theoretical side, there seems to be general agreement that the effect arises because of the influence of the spin-orbit interaction on the motion of the conduction electrons. Most of the existing theories[2–5] give qualitative results and only a few attempts[6,7] seem to have been made to evaluate the strength of the effect numerically and to account for the observed sign (positive for iron and negative for nickel). The theoretical predictions for R_s are intimately related to the ordinary electrical resistivity ρ and can be summarized as follows:

$$R_s = a\rho + b\rho^2 \quad \text{(impurity scattering)}\,, \qquad (2a)$$

$$R_s = c\rho^2 \quad \text{(phonon scattering)}\,, \qquad (2b)$$

where a, b, and c are constants.

The most detailed measurements of the temperature variation of the Hall effect in polycrystalline ferromagnetic metals seem to have been carried out by Volkenshtein, Fedorov, and Vonsovskii[8] and by Volkenshtein and Fedorov[9] on iron, nickel, and cobalt in the temperature range 4.2–300°K. For all three metals, both R_0 and R_s showed a pronounced temperature dependence with that of R_s being greater. The high-temperature results obtained for iron were in fair agreement with the theoretical result [Eq. (2b)], but the low-temperature behavior of R_s could not be explained. For nickel, on the other hand, the experimental results at high temperatures gave $R_s \propto \rho^{1.02}$, and the theory completely fails to account for the results for cobalt, for which it was found that R_s is positive for $T > 215$°K and negative at lower temperatures.

In order to achieve a better understanding of the nature of the Hall effect in ferromagnets, it was evidently highly desirable to carry out measurements on single-crystal specimens of maximum purity. Thus far the only attempt at detailed measurements on single

* Work was performed at the Ames Laboratory of the U. S. Atomic Energy Commission. Contribution No. 1818.
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[1] J.-P. Jan, in *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic Press Inc., New York, 1957), Vol. 5, p. 1.
[2] R. Karplus and J. M. Luttinger, Phys. Rev. **95**, 1154 (1954).
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[4] J. Smit, Physica **21**, 877 (1955); **24**, 39 (1958).
[5] J. Kondo, Progr. Theoret. Phys. (Kyoto) **27**, 772 (1962).
[6] C. Strachan and A. M. Murray, Proc. Phys. Soc. (London) **73**, 433 (1959).
[7] H. R. Leribaux, Phys. Rev. **150**, 384 (1966).
[8] N. V. Volkenshtein, G. V. Fedorov, and S. V. Vonsovskii, Zh. Eksperim. i Teor. Fiz. **35**, 85 (1958) [English transl.: Soviet Phys.—JETP **8**, 61 (1959)].
[9] N. V. Volkenshtein and G. V. Fedorov, Zh. Eksperim. i Teor. Fiz. **38**, 64 (1960) [English transl.: Soviet Phys.—JETP **11**, 48 (1960)]. These authors give no explicit mention of their treatment of demagnetizing effects.

crystals seems to be that on nickel and cobalt by Volkenshtein, Fedorov, and Shirokovskii.[10] These authors found both R_0 and R_s to be anisotropic for cobalt; for nickel R_s was found to be isotropic, but R_0 showed a small anisotropy which gradually disappeared as the temperature approached 0°K.

Here we report measurements of the Hall effect in single-crystal iron whiskers of high quality in the temperature range 1–300°K. Values of the resistance ratio $\gamma_e = \rho_{298°K}/\rho_{4.2°K}$, as measured in the earth's magnetic field, ranged from 180 to 450 for the various specimens; the whiskers are thus at least an order of magnitude better in purity than the polycrystalline iron with $\gamma_e = 11.4$ used by Volkenshtein and Fedorov.[9] While the anomalous Hall effect has been our major concern, we have also made measurements of the longitudinal and transverse magnetoresistance in the same specimens at 4.2°K and in the fields of up to 50 kG and 20 kG, respectively. Preliminary results of this work have already been reported.[11]

II. EXPERIMENTAL DETAILS

Iron whiskers were grown by hydrogen reduction of ferrous chloride following Brenner's method.[12] As has been pointed out by Wayman,[13] the size of the whiskers was found to depend not only on the reducing temperature and the hydrogen flow rate but also on the amount of the salt available for the reduction process, the size increasing with the amount of the salt. Whiskers about $\frac{1}{2}$ mm in width and up to 20 mm in length could be produced by reducing about 100 g of ferrous chloride.[14] The salt was placed in an iron boat and the reduction was carried out in a quartz tube 65 mm in diam. For a fixed reducing temperature and hydrogen flow rate, the quality and size of the whiskers varied considerably in different runs; the best results were obtained at 730°C and with a hydrogen flow rate of about 250 cc/min. Whiskers having good surfaces and the most uniform cross sections were selected and sections about 12 mm long were cut using a Servomet spark-erosion apparatus. The surface layer of each specimen was then removed and the tips were rounded off by electropolishing. The orientation was determined by means of back-reflexion x-ray photographs; the whisker axis was always found to be within 1° of one of the symmetry axes.

The specimens were mounted in good thermal contact with a thick copper plate (thus minimizing temperature gradients and the associated thermomagnetic effects) and electrical leads were carefully attached using small quantities of pure indium solder. Two pairs of potential probes were used, one pair to measure the magnetoresistance and the other to measure the Hall voltage, and the potential leads were connected sufficiently far from the current contacts so that the end effects[15] had a negligible influence on the resistance measurements. The leads were of fine copper wire and were arranged in such a way that there were no strains on the specimens. The temperature was varied by immersing the specimens in baths of liquid helium, liquid hyrogen, liquid nitrogen, and a mixture of dry ice and acetone. Unusually large thermal emf's were encountered when the specimen was in direct physical contact with liquid nitorgen, presumably due to local temperature fluctuations in the bath. This difficulty was overcome by enclosing the specimen and its mounting in a nonmagnetic jacket and by using helium exchange gas to establish thermal contact with the bath. The temperature of each bath could be varied in the usual manner by pumping off the vapor.

The potential measurements were made using a Kapitza-Milner bridge[16] and a dc photocell galvanometer amplifier. The sensitivity of the apparatus was usually better than 10^{-9} V, and under favorable conditions voltage changes of about 0.5×10^{-9} V could be detected. Magnetoresistive-type voltages between the Hall probes (due to the unavoidable misalignment of the probes) were eliminated by the conventional method of field reversal, achieved in this work by rotating the specimen through 180°. The measurements of importance were those carried out in applied fields in excess of about 14 kG; no hysteresis was ever observed at such field strengths, and careful spot checks were made to confirm that the quicker method of achieving field reversal by sample rotation did in fact yield the same results at those obtained by gradually reducing, then reversing and subsequently increasing the current in the electromagnet. For each field direction, the bridge was balanced for reversal of the specimen current, thereby eliminating any possible thermal effects. In contrast to the findings of Semenenko and Sudovtsov,[17] the galvanomagnetic coefficients were found to be independent of the strength of the measuring current (typically 470 mA for the Hall measurements and 65 mA for the magnetoresistance measurements).

Since the experimental geometry corresponds closely to a long cylinder transverse to the applied magnetic field H, the demagnetizing field can be taken to be $2\pi M$ and Eq. (1) can be written as

$$e_H = R_0 H + (R_0 + 2R_s)2\pi M. \quad (3)$$

[10] N. V. Volkenshtein, G. V. Fedorov, and V. P. Shirokovskii, Fiz. Metal i Metalloved 11, 152 (1961).

[11] P. N. Dheer, Bull. Am. Phys. Soc. 9, 550 (1964).

[12] S. S. Brenner, Acta. Met. 4, 62 (1956).

[13] C. M. Wayman, J. Appl. Phys. 32, 1844 (1961).

[14] $FeCl_2 \cdot 4H_2O$, reagent grade (Amend Drug and Chemical Company, and Fisher Scientific Company, both of New York).

[15] N. E. Alekseevskii, N. B. Brandt, and T. I. Kostina, Zh. Eksperim. i Teor. Fiz. 34, 1339 (1958) [English transl.: Soviet Phys.—JETP 7, 924 (1958)].

[16] P. Kapitza and C. J. Milner, J. Sci. Instr. 14, 165 (1937).

[17] E. E. Semenenko and A. I. Sudovtsov, Zh. Eksperim. i Teor. Fiz. 47, 486 (1964) [English transl.: Soviet Phys.—JETP 20, 323 (1965)]. These authors report a dependence of the resistance on the strength of the measuring current, the resistance increasing by 20% when the measuring current was increased from 0.1 to 1000 mA. We have not observed any change in resistance when the current was increased from 65 to 680 mA.

TABLE I. Specimen characteristics. $\gamma=\rho_{298°K}/\rho_{4.2°K}$.

Specimen and symbol in figures	Whisker axis and current direction	Direction of applied field H in Secs. IIIA, IIIB	Direction of Hall probes	γ_e[a]	γ_d[b]	Transverse shape and/or dimensions (mm)
Fe 2 ×	[111]	[$\bar{1}$10]	[11$\bar{2}$]	196	316	hex. side 0.32
Fe 3 △	[111]	[$\bar{1}$10]	[11$\bar{2}$]	213	391	hex. side 0.21
Fe 5 ○	[100]	[010]	[001]	213	301	0.40×0.40
Fe 7 □	[100]	[010]	[001]	452	852	0.35×0.53
Fe 12 ▽	[100]	[010]	[001]	180	242	0.34×0.26

[a] The values γ_e are from measurements made in the presence of only the earth's magnetic field and thus refer to the unmagnetized state (domains present).
[b] More meaningful, however, are the ratios γ_d obtained by extrapolating the slowly-varying longitudinal magnetoresistance data for $H>\sim 1$ kG down to $B=0$. (See also Ref. 17).

Above magnetic saturation M has a constant value M_s, and thus e_H varies linearly with H; the slope of the line gives R_0, and extrapolation of the linear variation above saturation down to $H=0$ yields the intercept $(R_0+2R_s)2\pi M_s$, from which R_s can be determined. Thus R_0 and R_s can be determined entirely from measurements made above magnetic saturation, so that a knowledge of the details of the magnetization curves for the specimens is not necessary. The same apparatus was used to measure the electrical resistivity, and the transverse and longitudinal magnetoresistance at low temperatures. The resistance ratios γ and the directions of the primary current, magnetic field, and the Hall probes for the various specimens are given in Table I.

III. RESULTS

A. Hall Effect

Typical curves showing the variation of the Hall resistance (Hall voltage per unit current) with the applied magnetic field H are shown in Fig. 1. For all specimens and at all temperatures, the Hall resistance was found to vary linearly with the field for $H>\sim 14$ kG, and this linearity is taken to imply that the specimens were in the state of magnetic saturation. While most of the measurements were made in fields below 21 kG, a few measurements were carried out in a 31 kG electromagnet in order to verify that the saturation was indeed complete. It is difficult to calculate exactly the lowest field at which the specimens are magnetized to saturation, but the experimental value of $H\sim 14$ kG is physically reasonable since the applied field must be large enough to overcome the demagnetizing field of the specimen. We have taken the demagnetizing field to be that appropriate to a long circular cylinder, namely, $2\pi M_s=10.9$ kG, whereas it could be slightly higher for the actual specimen shapes; the cross sections were found to be square or slightly rectangular for [100] whiskers and hexagonal for [111] whiskers but the sharp corners had been rounded off by the electropolishing.

As can be seen from Fig. 1, the onset of saturation at $H\sim 14$ kG is more pronounced at high temperatures; at low temperatures the linear portions of the curves appear to extend to much lower values of the field, and this observation is presumably a consequence of the smaller values of R_s which are found as the temperature is reduced. Another feature which was found for all whiskers is the extremely small magnitude of the Hall resistance at liquid-nitrogen temperatures and its change of sign in this temperature region (See Fig. 1). The Hall coefficients R_0 and R_s were found from the linear regions above 14 kG by application of Eq. (3), and we shall now discuss their temperature dependences in turn.

The temperature variations of R_0 for the five whiskers are presented in Fig. 2, which also shows for comparison the variation found by Volkenshtein and Fedorov[9] for polycrystalline iron of less purity. The probable error in R_0, as estimated from a least-squares analysis of the data, is usually less than 5% at high temperatures and

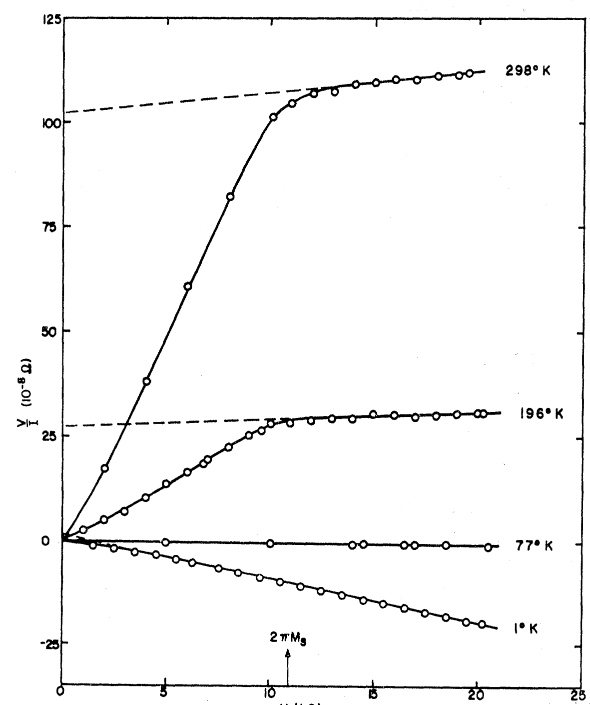

FIG. 1. Variation of the Hall resistance with applied magnetic field for whisker Fe 5. (See Table I for the experimental geometry and for specimen details.)

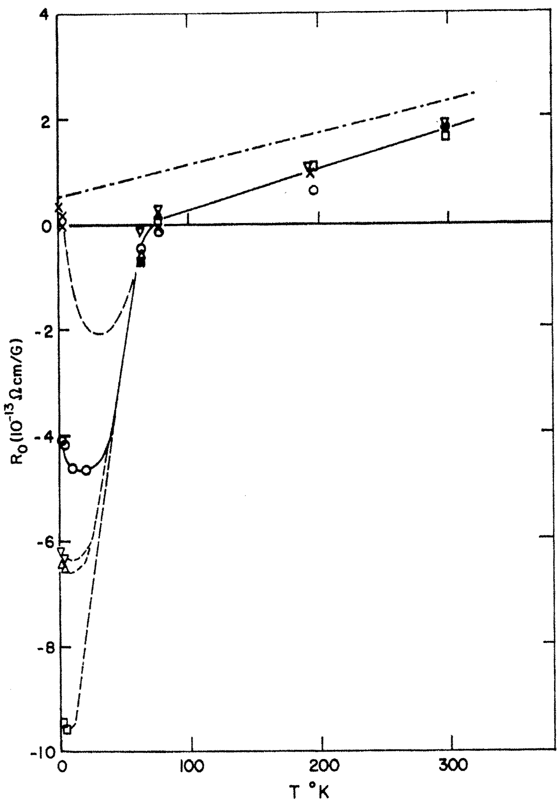

FIG. 2. Temperature dependence of the "ordinary" Hall coefficient R_0 for iron whiskers (see Table I for experimental geometry and for specimen details). Dot-dash, results of Volkenshtein and Fedorov (Ref. 9) for polycrystalline iron.

about 1% at low temperatures. In addition, there could be systematic errors in all the measurements because of slight irregularities in the cross sections of the specimens, but these errors are probably not larger than 10%. We therefore conclude that the spread of the results from the different whiskers above liquid-nitrogen temperatures is probably within the limits of experimental accuracy, and thus there does not seem to be any evidence for a dependence of R_0 on crystal orientation at high temperatures. The ordinary Hall coefficient shows a marked (roughly linear) increase at high temperatures; this trend is quite similar to that found for polycrystalline iron by Volkenshtein and Fedorov,[9] although their values of R_0 are considerably larger than ours.

At low temperatures, the experimental situation with regard to the behavior of R_0 is not so clearly defined. Unlike the results for polycrystalline iron,[9] R_0 for the whiskers is found to exhibit a marked negative anomaly below liquid-nitrogen temperatures, and the absolute value of R_0 as $T\rightarrow 0$°K is in most cases several times the value at room temperature. The present data are not sufficient to investigate possible anisotropy effects or to relate R_0 to the residual-resistance ratio γ, although we note that it is for the purest whisker [Fe 7; see Table I] that the low-temperature anomaly is strongest. In the simplest multiband model,[18] the various positive and negative contributions to the Hall constant are weighted by the squares of the partial conductivities, whose relative strengths depend on the scattering mechanisms involved. However, it is not at all clear why the results for the various specimens should differ so greatly in the impurity scattering region as $T\rightarrow 0$°K.

The temperature variation of R_s for the various specimens is shown in Fig. 3. With the exception of the results for specimen Fe 2, the values for R_s are unlike those found for R_0 in that they are found to remain positive throughout the entire range 1–300°K and the variation of R_s shows a distinct minimum near 70°K. The values of R_s at liquid-helium temperatures are again very sensitive to the specimen purity, and nothing definite can be inferred about possible anisotropy effects. At high temperatures, R_s appears to be isotropic and independent of sample purity, but we have not been able to find a satisfactory explanation for the fact that the results for specimen Fe 5 differ significantly from those obtained for the other four whiskers. Just as was found for R_0, the values of R_s for the whiskers are on the whole smaller than those reported by Volkenshtein and Fedorov[9] for polycrystalline material, with the differ-

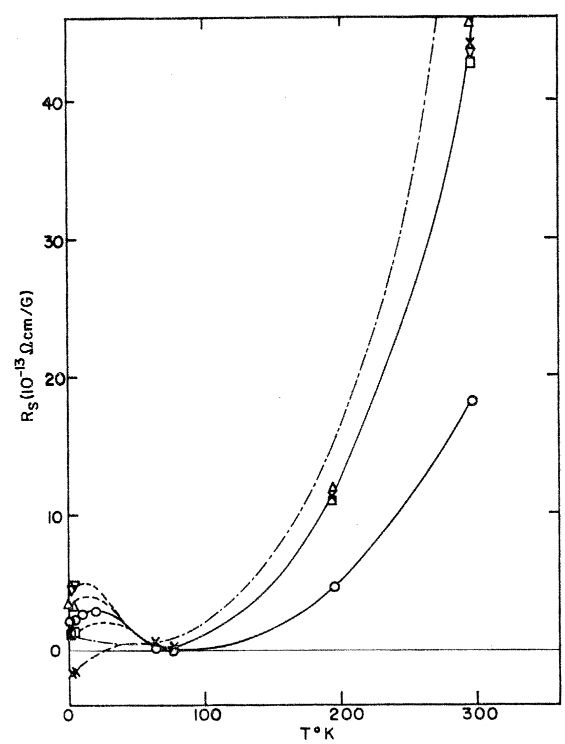

FIG. 3. Temperature dependence of the "extraordinary" Hall coefficient R_s for iron whiskers (see Table I for experimental geometry and specimen details). Dot-dash, results of Volkenstein and Fedorov (Ref. 9) for polycrystalline iron.

[18] A. H. Wilson, *Theory of Metals* (Cambridge University Press, New York, 1954), 2nd ed., p. 213.

ence increasing to nearly a factor of two at room temperature.

Above 70°K the whisker results were found to be more or less in accordance with the theoretical prediction $R_s \propto \rho^2$ for phonon scattering, with the exponent of the resistivity ranging from 1.9 to 2.2 for the various specimens. However, it does not seem possible to explain the low-temperature behavior of R_s in terms of any existing theories.

B. Magnetoresistance

In the absence of any external field, a bulk ferromagnet is magnetically inhomogeneous consisting of a number of Weiss domains oriented in such a way that there is no net magnetization. The presence of domains has a profound effect on the electrical resistivity, and this effect should be of greatest importance at low temperatures where the other contributions to the resistivity are small. For this reason the resistance ratio γ_e, as determined in the weak earth's field, it is not expected to be a good criterion for the purity of a ferromagnetic metal. In view of the large variations in the Hall coefficients R_0 and R_s from sample to sample at low temperatures, measurements of the electrical resistance were carried out in both longitudinal and transverse magnetic fields; since the application of a magnetic field modifies the domain structure, such measurements should enable the contribution to γ from domain effects to be ascertained.

The longitudinal magnetoresistance was measured at 4.2°K in fields up to 47.5 kG produced by a superconducting solenoid. The results are shown in Figs. 4 and 5, in which ρ_H is the resistance in the applied field H and ρ_0 is the zero-field resistance. (No attempt was made to compensate for the earth's field.)

In the low-field region of technical magnetization in which $H < \sim 1$ kG, application of a longitudinal field causes a sharp decrease in the electrical resistance (Fig. 4) and the magnitude of the decrease is greatest (46%) for specimen Fe 7 which has the highest resistance ratio γ_e. These results are similar to those obtained by Seme-

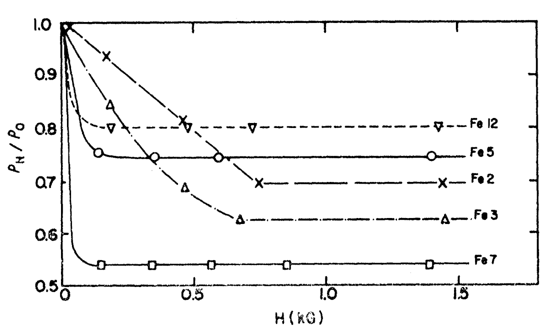

FIG. 4. Longitudinal magnetoresistance for iron whiskers at 4.2°K: the low-field region. For specimens Fe 5, Fe 7, and Fe 12 the applied field is directed along the easy axis of magnetization (see Table I).

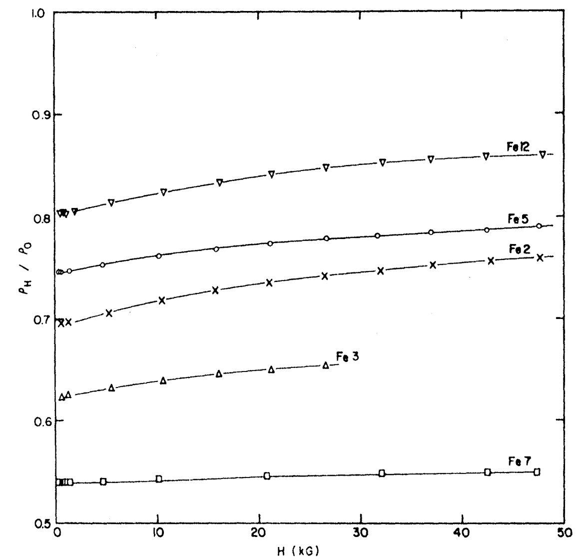

FIG. 5. Longitudinal magnetoresistance for iron whiskers at 4.2°K and in applied fields up to 47.5 kG (see Table I for specimen details). The behavior for $H < 1.5$ kG is shown in detail in Fig. 4.

nenko and Sudovtsov,[17] who have observed a 30% decrease in polycrystalline material, and are in general consistent with the observations of Isin and Coleman,[19] who have carried out a detailed investigation of the longitudinal and transverse magnetoresistance in iron whiskers. In addition to the sharp decrease in the resistance, which is the main effect, Isin and Coleman[19] have also observed strong hysterisis effects in the low-field region which indicate that the low-field magnetoresistance is indeed influenced by the domain development during magnetization. The main effect, however, seems to depend on the bulk magnetization of the specimen rather than on the detailed domain structure, and is not yet understood.

At fields beyond saturation (Fig. 5), the resistance increases gently with the field, presumably because of the ordinary longitudinal magnetoresistance in metals. (The increase in resistance observed by Isin and Coleman[19] is more pronounced than that observed by us.) To eliminate the effects of the domains, the slowly-varying high-field data were, therefore, extrapolated to $B = 0$, and the resistance ratios γ_d determined in this manner are given in Table I; these ratios should provide a more meaningful index of specimen purity.

The observed changes in resistance upon application of transverse fields up to 20.5 kG and at 4.2°K are shown in Fig. 6; similar results were obtained at 1.0°K. The low-field behavior differs somewhat from sample to sample, but an initial decrease in resistance of about 10% was found for most of the whiskers. Isin and Cole-

[19] A. Isin and R. V. Coleman, Phys. Rev. **137**, A1609 (1965); **142**, 372 (1966); R. V. Coleman and A. Isin, J. Appl. Phys. **37**, 1028 (1966).

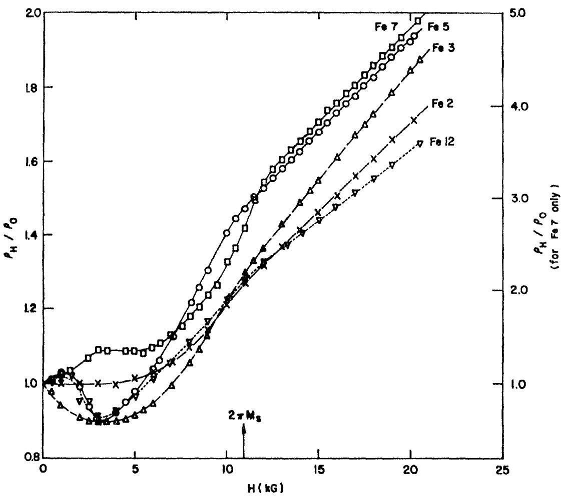

FIG. 6. Transverse magnetoresistance for iron whiskers at 4.2°K (see Table I for specimen details). The scale on the right refers only to specimen Fe 7.

man[19] and Reed and Fawcett[20] have reported similar and even more pronounced effects; for example, the decrease observed by Reed and Fawcett[20] for one of their samples was about 94% of the zero-field resistance. This pronounced decrease appears to be due more to better specimen perfection than to the mechanism suggested by Berger and deVroomen,[21] according to which a decrease in the initial transverse magnetoresistance would be expected if the specimen axis and the field were not exactly perpendicular. The magnitude of the decrease has also been found to depend on the diameter of the whisker[19] and the orientation of the field.[19,20] Isin and Coleman[19] have also observed hysteresis effects similar to, but less pronounced than, those observed in the longitudinal fields.

As is to be expected, the magnitude of the transverse magnetoresistance is much larger than that found for longitudinal fields, and above the initial decrease our data are in general agreement with the results of Refs. 19 and 20. In the range 10–20 kG the resistance changes almost linearly with the field, but this linearity over a limited field should not be interpreted as foreshadowing the onset of magnetoresistive saturation. Indeed, when the field range was extended to 30.7 kG for one specimen (Fe 5) at 4.2°K, the resistance was found to increase very nearly as B^2. This type of variation is in agreement with the results found at much higher fields[19,20]; however, the magnetic fields used in the present study are not high enough to obtain further information about the topology of the Fermi surface.

C. Field-Rotation Effects

Figures 7 and 8 illustrate the variations of the resistance and the Hall voltage, respectively, when a transverse magnetic field is kept constant at about 20 kG and the specimen is rotated about its axis; the measurements were made at 4.2°K, and θ is the angle between the magnetic field and the normal to the line joining the Hall probes.

Any interpretation of the observed anisotropy of the low-field magnetoresistance in terms of the coefficients ρ_i in Döring's[22] relation for a cubic ferromagnet,

$$\frac{\Delta\rho}{\rho}=\rho_0+\rho_1\sum_i \alpha_i^2\beta_i^2+\rho_2\sum_{ij}\alpha_i\alpha_j\beta_i\beta_j+\cdots, \qquad (4)$$

where α_i and β_i are the direction cosines of the magnetization and the measuring current, is complicated by the fact that the specimens were not perfect circular cylinders. The rather small observed anisotropy in the magnetoresistance would first have to be corrected for shape anisotropy (i.e., the effect of the variation of the demagnetizing field with the direction θ) and it is not possible to calculate this correction for all field direc-

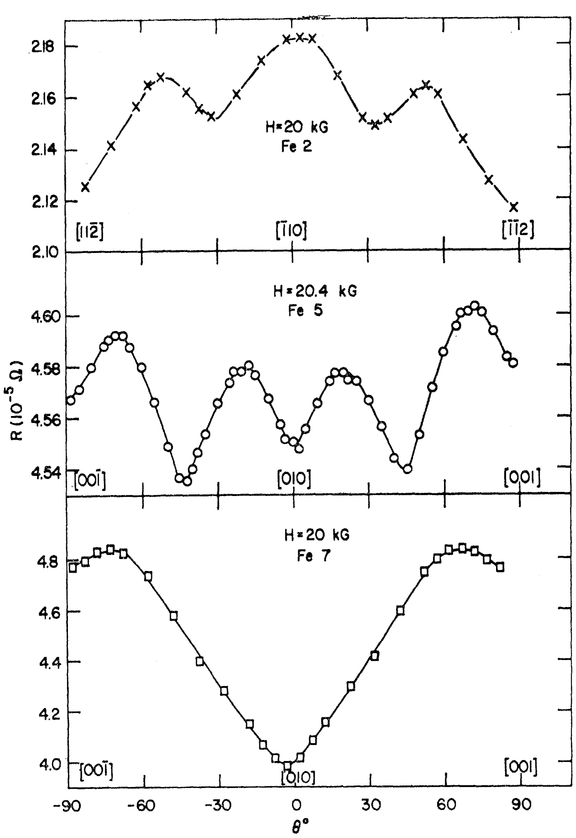

FIG. 7. Anisotropy of the transverse magnetoresistance for three iron whiskers at 4.2°K and at 20 kG (see Table I for specimen details).

[20] W. A. Reed and E. Fawcett, Phys. Rev. **136**, A422 (1964); in *Proceedings of the International Conference on Magnetism, Nottingham, 1964* (The Institute of Physics and The Physical Society, London, 1965), p. 120.

[21] L. Berger and A. R. deVroomen, J. Appl. Phys. **36**, 2777 (1965).

[22] W. Döring, Ann. Physik **32**, 259 (1938).

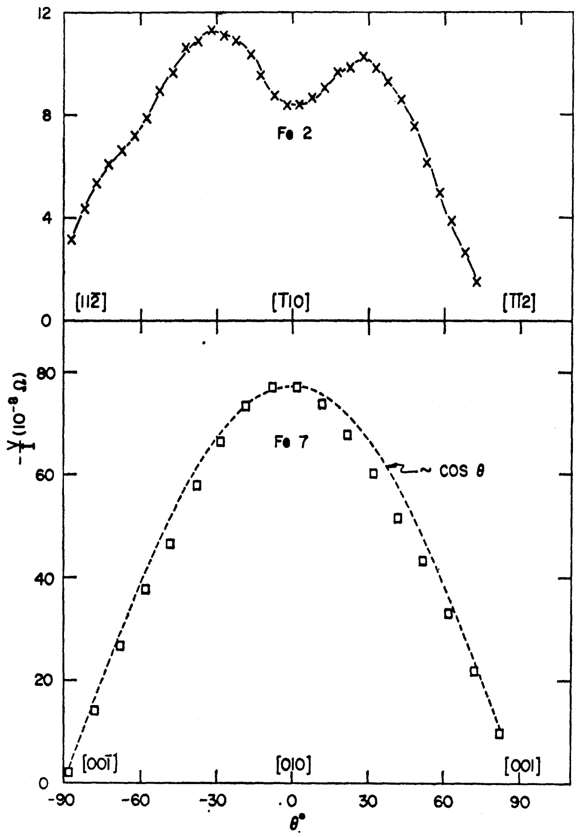

FIG. 8. Anisotropy of the Hall voltage for two iron whiskers at 4.2°K and at 20 kG (see Table I for specimen details).

tions. The effects of shape anisotropy should be largest for specimens with pronounced rectangular cross sections, and thus the shape anisotropy would seem to account for the absence of four-fold symmetry in the data for the [100] specimen Fe 7 (see Table I). To check this hypothesis, the demangetizing fields for this specimen and for the directions $\theta=0°$ and $\theta=90°$ were estimated from the tables of demagnetizing coefficients published by Brown[23] for infinite rectangular parallelepipeds. These results and the observed resistance at $\theta=0°$ were then used to calculate the expected resistance at $\theta=90°$ by suitable extrapolation of the relevant magnetoresistance data in Fig. 6. It is satisfactory that the calculated value was within 2% of the measured resistance at $\theta=90°$.

The rotation curves for the low-temperature Hall voltage show deviations from a $\cos\theta$ dependence, and the two rotation curves shown in Fig. 8 are typical results obtained at 4.2°K. As the field is tilted away from the normal ($\theta=0°$) position, the Hall voltage for [100] specimens decreases more rapidly than $\cos\theta$. For [111] specimens, on the other hand, there exists a range of field directions around $\theta=0°$ in which the Hall voltage actually *increases* with increasing $|\theta|$. This behavior is shown in Fig. 8 for specimen Fe 2; similar results were obtained for the other [111] specimen Fe 3, with the difference that the maximum Hall voltage was observed at about $\theta=\pm15°$ and was only 3% greater than the value at $\theta=0°$. Rotation curves were also measured at room temperature, but here they were found to follow a $\cos\theta$ variation quite closely. This result is consistent with the apparent isotropy of R_0 and R_s at high temperatures (Figs. 2 and 3) and it also suggests that, unlike the magnetoresistance effects, the measurements of the low-temperature Hall effect have not been much influenced by the shape anisotropies of the specimens. The deviations from the $\cos\theta$ behavior at 4.2°K thus indicate that at low temperatures at least one of the coefficients R_0 and R_s is intrinsically anisotropic. We have not, however, attempted to determine the anisotropy of R_0 and R_s separately by a systematic study of the field rotation curves, though some measurements at 4.2°K on specimen Fe 2 with the field in the direction $\theta=30°$ gave the values $R_0=-1.95\times10^{-13}$ Ω cm/G and $R_s=0.55\times10^{-13}$ Ω cm/G, which are quite different from those results obtained at 4.2°K with the field in the direction $\theta=0°$ (Figs. 2 and 3).

[23] W. F. Brown, Jr., *Magnetostatic Principles in Ferromagnetism* (North-Holland Publishing Company, Amsterdam, 1962).

IV. DISCUSSION

Although the measurements on the whisker specimens were undertaken primarily for the purpose of obtaining a better understanding of the extraordinary Hall effect in iron, the final outcome of this investigation is to introduce yet further unsolved problems rather than to resolve existing difficulties; this is true for both the ordinary and extraordinary Hall coefficients.

The fact that iron is a compensated metal[20] implies that R_0 is not simply related to the number of carriers in the metal; R_0 does not depend solely upon the geometrical features of the Fermi surface and thus this quantity cannot be regarded as a true 'constant' of a compensated metal. Nevertheless, certain features of the results for the ordinary Hall coefficient were quite unexpected. These are the strong temperature dependence of R_0, especially the dramatic variation at low temperatures, and the observation that the limiting value of R_0 as $T\rightarrow0°K$ (i.e., when phonon and spin-wave scattering should be "frozen out," leaving only the scattering by physical and chemical defects) is not the same even for similarly-oriented specimens (Fig. 2).

It is also very difficult to understand why the rapid transition from positive to negative values of R_0 should take place at a temperature as high as about 70°K, i.e., well above the residual-resistance region. One would indeed expect R_0 to show some temperature dependence at high temperatures because the exchange splitting depends on temperature, and so will therefore the sizes of the various portions of the Fermi surface. The exchange splitting should be roughly proportional to the magnetization, but since the latter changes by only 5%

between 0°K and room temperature, it seems unlikely that changing Fermi surface is the cause of the much greater temperature variation of R_0. The high-field susceptibility which is observed above the normal magnetic saturation should, strictly speaking, also be included in Eq. (3), i.e., M should become $M_s+\chi H$, but from the low value of χ determined recently by Freeman *et al.*[24] it seems that any effects on either R_0 or R_s should be negligibly small.

An alternative hypothesis which might have some bearing on the temperature dependence of R_0 is to suppose that Eq. (1) is not a complete description of the Hall effect in pure ferromagnets and that third-order interaction terms might play an important role; these terms consist of third-degree products of the fields H and M and are therefore odd with respect to reversal of the applied field.[1] (Possibly these interaction effects might have some bearing on the similarity between the temperature dependence of R_0 and that of the magnetoresistance coefficient ρ_1 in Eq. (4), as measured by Tatsumoto.[25]) On the other hand, appreciable temperature dependences have been also reported for the high-temperature Hall coefficients of other compensated, *but nonferromagnetic*, transition metals,[26] and thus the temperature dependence of R_0 for the whiskers may be related to effects such as anisotropies in relaxation times,[26] rather than specifically to the presence of the spontaneous magnetization in iron.

Although the high-temperature variation of the extraordinary Hall coefficient R_s appears to be consistent with the theoretical relation (2b), the actual numerical values are not yet well understood. At these temperatures the main contributions to R_s in pure ferromagnets should arise from the scattering of electrons by phonons and by spin waves. Leribaux,[7] assuming the electron-phonon interaction to be the only scattering mechanism and Wood's[27] calculations for the band structure of bcc iron, has obtained the following expression for R_s for a single crystal of iron in which the magnetization and the current flow are along {100} directions:

$$R_s=[20.9/4\pi M_s(T)]\rho^2 \quad \Omega\ \mathrm{cm/G}. \qquad (5)$$

At room temperature the mean value of ρ is found to be 9.5×10^{-6} Ω cm, and if we take $4\pi\ M_s=21.6$ kG, then Eq. (5) predicts $R_s=0.9\times10^{-13}$ Ω cm/G. This result is an order of magnitude smaller than the average experimental value $R_s=43.1\times10^{-13}$ Ω cm/G for the [100] whiskers Fe 7 and Fe 12.[28] However, Leribaux has had to make many simplifying assumptions concerning the wave functions, Fermi surface, etc., in iron in order to arrive at his final result [Eq. (5)], and thus any conclusions drawn from this comparison of numerical results are probably premature. Nevertheless, the discrepancy between theory and experiment seems to be in line with the assumption of Kagan and Maksimov[29] that there is no temperature region in which the phonon scattering makes an important contribution to R_s; at very low temperatures the principal contribution should arise from impurity scattering and at higher temperatures from spin waves. Unfortunately, a quantitative comparison of the experimental results with the theory of Kagan and Maksimov[29] cannot be made at present because of the difficulties associated with the numerical evaluation of various parameters of the theory.

Existing theories completely fail to explain the results below liquid-nitrogen temperatures, where R_s increases with decreasing temperature (Fig. 3). Similar results have been obtained for polycrystalline specimens of iron by Volkenshtein and Fedorov[9] and by Jan and Gijsman,[30] but these authors find the minimum to occur at lower temperatures and the increase in R_s as $T\rightarrow 0$°K to be less pronounced.

ACKNOWLEDGMENTS

This problem was suggested by Dr. A. V. Gold and I am indebted to him for many useful ideas. I have also benefited from discussions with Dr. H. R. Leribaux, Dr. S. H. Liu, Dr. L. Berger, and Dr. H. Juretschke. It is a pleasure to thank D. R. Stone for his help with the measurements in the superconducting magnet.

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[28] For reasons which we do not understand, the room-temperature value for Fe 5, the third [100] whisker, is considerably lower, with $R_s=18.2\times10^{-13}$ Ω cm/G (see Fig. 3). We are inclined to regard this result as unreliable, but only because it seems inconsistent with the results obtained from the other [100] and [111] whiskers. *Note added in proof.* Dr. H. R. Leribaux (see Ref. 7) seems to have obtained a better agreement between the observed and calculated values of R_s than that indicated in this paper. He was, however, not aware of our results on the specimens Fe 7 and Fe 12. He has also used too high a value of ρ for calculating R_s.

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