for this compound is so low.[11] The calculations parallel the ones given above, so we only quote the results. For small **k**

$$\hbar\omega_{ac}=10J_{aa}ak \qquad \text{(C8)}$$

and the sublattice magnetization is

$$\frac{\langle\Delta S\rangle}{S}=\delta-\frac{1}{6000}\left(\frac{k_BT}{J_{aa}}\right)^2+\cdots. \qquad \text{(C9)}$$

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Influence of the Anomalous Skin Effect on the Ferromagnetic-Resonance Linewidth in Iron*

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The ferromagnetic-resonance linewidth has been measured in iron single-crystal whiskers over a temperature range from 300 to 4.2°K at frequencies of 9.2, 22.2, and 34.8 kMc/sec. The measurements of linewidth and line shape clearly indicate the importance of nonlocal conductivity effects.

I. INTRODUCTION

The first measurements of ferromagnetic resonance (FMR) in single-crystal iron whiskers at room temperature were carried out by Rodbell[1] at frequencies of 9 and 20 kMc/sec. The most important conclusion resulting from this work was that the linewidth in iron is dominated by the exchange-conductivity mechanism. Our recent measurements[2] at room temperature and frequencies of 9.2, 22.2, 34.8, and 57.8 kMc/sec confirmed Rodbell's results. In addition, with these higher frequencies, we were able to obtain a rough estimate of the Landau-Lifshitz parameter λ. However, at 20 kMc/sec, Rodbell's linewidth is some 30 Oe narrower than ours. In nickel,[2] we noted a similar discrepancy with Rodbell's results at low frequencies, but the linewidths agreed at the higher frequencies. Presumably, as was suggested for the nickel case, the discrepancy in iron is also due to the better surfaces of Rodbell's samples with a resulting reduced surface anisotropy. This difference is not expected to affect our conclusions.

Rodbell[1] also measured linewidths in iron below 300°K, but very few details were given apart from the comment that there was a distinct change in the character of the resonance on cooling; this he attributed to the onset of the anomalous-skin-effect region. On the other hand, the theory of Hirst and Prange[3] indicated that the onset of anomalous conductivity should have no pronounced effect upon the ferromagnetic resonance, and our preliminary measurements on iron[4] seemed to support this prediction. In addition, we found that the observed linewidths did not increase as rapidly as expected from normal conductivity theory, in agreement with the presence of anomalous conductivity at temperatures below 77°K. The present paper is a more detailed report of our low-temperature measurements in iron, and confirms the preliminary conclusions of Ref. 4.

II. EXPERIMENTAL METHOD

The samples used were thick iron single-crystal whiskers with axes along [100] and [111]. The [100] samples and a few of the [111] samples were given to us by Professor A. V. Gold of Iowa State University, and these whiskers were used to study FMR at 22 and 35 kMc/sec. However, at lower frequencies (9 kMc/sec), because of the large magnetocrystalline anisotropy at 300°K and below, [100] whiskers could not be used, and it became necessary to grow additional [111] whiskers for this study. Although the general techniques for iron-whisker growth have been described by Brenner,[5] a few details are included here.

The whiskers were grown by hydrogen reduction of $FeCl_2$ which had previously been prepared by baking $FeCl_2\cdot 4H_2O$ in an inert atmosphere. The $FeCl_2$ was held in a quartz boat; quartz was used to facilitate removal of whiskers. The optimum hydrogen-flow rate was determined to be about 300 ml/min, and the optimum reduction temperature was 750°C. Furthermore, better results were obtained using hydrogen saturated with water vapor at room temperature. Only about 3% of the "raw" whiskers were found to be

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[1] D. S. Rodbell, J. Appl. Phys. **30**, 187S (1959); Physics **1**, 279 (1965).

[2] S. M. Bhagat, L. L. Hirst, and J. R. Anderson, J. Appl. Phys. **37**, 194 (1966).

[3] L. L. Hirst and R. E. Prange, Phys. Rev. **139**, A892 (1965).

[4] S. M. Bhagat, J. R. Anderson, and L. L. Hirst, Phys. Rev. Letters **16**, 1099 (1966).

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

straight with relatively smooth surfaces. The remainder were either twisted, badly bent, or with highly pockmarked surfaces. The "good" whiskers were about 8 mm in length and about 0.2–0.4 mm on a side; most of them grew with the whisker axis along [111].

The samples were electropolished and the ends copper plated, as described previously.[2] The microwave setup was similar to that used in our earlier work.[2] In addition, the cavity was enclosed in a thin-walled stainless-steel can which could be evacuated. This prevented the whickers from oxidizing and provided better temperature control at intermediate temperatures between room temperature and 4°K. The temperature was monitored with a copper-constantan thermocouple between 300 and 77°K and with a platinum-resistance thermometer between 77 and 4.2°K; the temperature values are good to about ±2°K. However, the thermocouple was cemented to the outside of the cavity and there may have been some lag between the sample temperature and the thermocouple reading. This error was reduced by making observations during both heating and cooling cycles. It was also found that use of a very undercoupled cavity helped to minimize this temperature difference.

The magnetic field was measured in the higher temperature range with a rotating-coil Gaussmeter, while for low temperature work the magnet current, previously calibrated against the Gaussmeter, was monitored.

One of the major problems in measurement of FMR over a wide temperature range is to obtain a proper mounting in which the whisker is not strained but, at the same time, is not free to move. In our room-temperature work the samples were cemented in place with GE 7031 adhesive; at low temperatures it became apparent that the samples were strained with this mounting. A more successful approach was to hold the whisker inside a quartz capillary (~1 mm i.d.) with a small amount of Apiezon grease. The quartz capillary was held inside the cavity in a Styrofoam block. As described in Ref. 6, a good indication of proper mounting is the absence of anomalously large signals at low magnetic fields.

In order to interpret the linewidths on the basis of normal or anomalous conductivity theory, it is necessary to measure the resistivity of an iron whisker from 300 to 4.2°K. This was done on a representative whisker in a longitudinal field of 2kG using conventional techniques. Figure 1 shows a plot for this sample of the resistivity ratio ρ_{300}/ρ_T versus temperature down to 77°K. The experimental setup used for the resistivity measurement did not permit measurements at intermediate temperatures between 77 and 4.2°K, but the resistivity ratio at 4.2°K was found to be about 600.

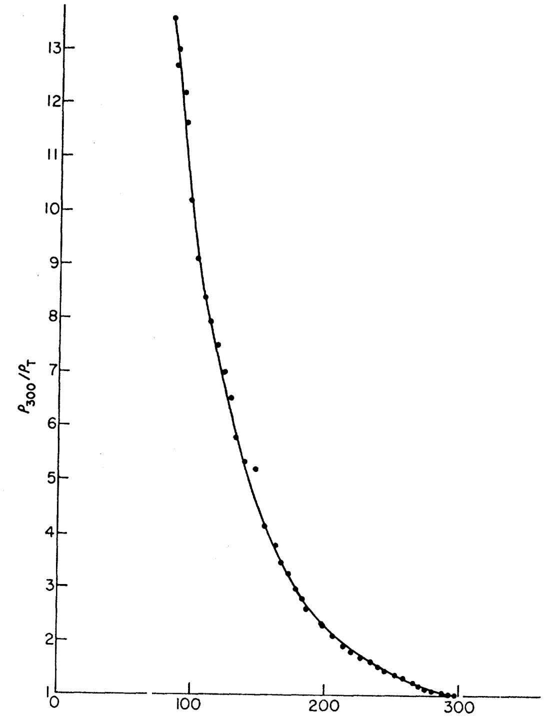

FIG. 1. Resistivity ratio versus temperature for a representative iron whisker.

III. DISCUSSION AND RESULTS

The FMR linewidth Γ_{pp} can be written roughly as

$$\Gamma_{pp}=\Gamma_{pp}^{\lambda}+\Gamma_{pp}^{\text{ex}}, \tag{1}$$

where[7]

$$\Gamma_{pp}^{\lambda}=1.45\omega\lambda/M_s \tag{2}$$

is the Landau-Lifshitz relaxation damping term, and, for normal conductivity,

$$\Gamma_{pp}^{\text{ex}}=CA^{1/2}\omega^{1/2}/\rho^{1/2} \tag{3}$$

is the exchange-conductivity contribution.[8] The microwave frequency is given by ω, while λ is the Landau-Lifshitz damping parameter, M_s is the saturation magnetization, ρ is the resistivity, and A is the exchange stiffness constant. The constant C depends upon the relative values of λ and A. In addition, there is a surface-anisotropy contribution[2] to the linewidth which is closely related to Γ_{pp}^{ex} and has the same frequency and resistivity dependence.[8]

In the low-temperature region, considerably below the Curie temperature, M_s varies only slowly with temperature, and one therefore expects Γ_{pp}^{λ} to be essentially constant if λ is independent of temperature.[9]

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[6] S. M. Bhagat and L. L. Hirst, Phys. Rev. **151**, 401 (1966).

[7] L. Landau and E. Lifshitz, Physik. Z. Sowjetunion **8**, 153 (1935).

[8] W. S. Ament and G. T. Rado, Phys. Rev. **97**, 1558 (1955).

[9] Although a temperature-dependent λ has been suggested as an explanation of the low-temperature FMR linewidth in nickel (Ref. 6), we have no evidence for a temperature-dependent λ in iron.

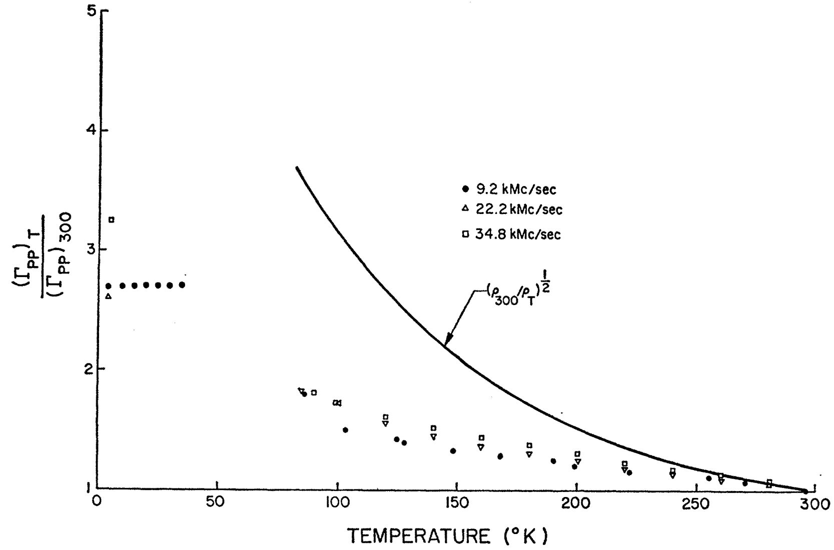

FIG. 2. Relative linewidth $\Gamma_{pp}(T)/\Gamma_{pp}(300)$ versus temperature for iron whiskers at 9.2, 22.2, and 34.8 kMc/sec. The full curve is obtained from normal-conductivity theory.

However, Γ_{pp}^{ex} is a strong function of temperature varying approximately as $M_s/\rho^{1/2}$. In Fig. 2 are plotted the relative linewidths $\Gamma_{pp}(T)/\Gamma_{pp}$ (300°K) for frequencies 9.2, 22.2, and 34.8 kMc/sec. These values all lie on approximately the same temperature curve and are thought to be good to approximately 10%. The experimental linewidths can be compared with the normal-conductivity exchange term $(\rho_{300}/\rho_T)^{1/2}$ which is plotted as the solid curve in Fig. 2. It is clear that at about 270°K the observed values begin to deviate from the normal-conductivity curve. In addition, the measured widths saturate at about 50°K, and at 4°K the observed width is nearly a factor of 7 less than normal-conductivity theory would predict. We have attributed this difference to the onset of the anomalous-conductivity regime in iron.

As is well known, the anomalous-skin-effect situation arises when the mean-free-path of the conduction electrons is much greater than some relevant skin depth. In ferromagnetic materials, this is expected to occur at relatively high temperatures, because the dynamic permeability at resonance is large. In this case, the conductivity becomes wave number (q)-dependent and, according to the nonlocal conductivity theory, the conductivity decreases with increasing q becoming inversely proportional to q in the extreme anomalous limit. This result is described in terms of an additional parameter C_F, where C_F/q is the conductivity in the extreme anomalous limit.[10] Thus, as the temperature is reduced and the electron mean-free-path becomes longer than the skin depth, the linewidth should increase much more slowly than predicted by normal-conductivity theory and approach a constant value at very low temperatures.[3]

The results shown in Fig. 2 suggest the qualitative correctness of the Hirst-Prange theory. A quantitative comparison is somewhat hampered because C_F is not known. However, in Ref. 4, a value of 1.5×10^{24} cm^{-1} sec^{-1} was chosen for C_F in order to fit the linewidths at 22.2 and 34.8 kMc/sec at temperatures of 300, 80, and 4°K. This value has been used here to compute the 9.2-kMc/sec linewidth as a function of resistivity ratio as shown by the smooth curve in Fig. 3.[11] The temperature dependence of the dc resistivity has been obtained from Fig. 1. The points in Fig. 3 are experimental results for four [111] samples.[12] As can be seen, the agreement between the nonlocal theory and the experimental linewidths is quite good.

Concomitant with the exchange broadening, there is also an exchange shift, but the temperature dependence of the anisotropy constants K_1 and K_2 is not well enough known[13] for a direct comparison of experiment with theory. However, in Table I we show, for 9.2 kMc/sec, the computed exchange shift ΔH with respect to the center of the line at 300°K; these values might be useful if better anisotropy data become available.

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[10] A. B. Pippard, Rept. Progr. Phys. **23**, 176 (1960).

[11] For this computation, the parameter values of Ref. 4 have been used: surface anisotropy energy $K_s(=0.4$ erg/cm$^2)$, Landau-Lifshitz parameter $\lambda(=0.7\times10^8$ sec$^{-1})$, and the exchange-stiffness parameter $A(=1.9\times10^{-6}$ erg/cm). The value for C_F is in rough agreement with estimates based upon the electronic specific heat (Ref. 4).

[12] There are two points to be noted regarding the room-temperature data on the [111] whiskers grown here. First, in the present set of samples we were unable to find any with linewidth less than about 95 Oe; this is about 5 Oe larger than reported in Ref. 2. Second, no unexplained weak resonance on the high-field side of the main resonance, such as we reported in Ref. 2, was observed in present [111] samples. It is possible that the present batch of whiskers has poorer surfaces, but a microscopic examination did not show this.

[13] C. D. Graham, Phys. Rev. **112**, 1117 (1958); R. M. Bozorth, *Ferromagnetism* (D. Van Nostrand Co., Inc., New York, 1951), p. 568.

TABLE I. Relative exchange shift at 9 kMc/sec.

ρ_{300}/ρ_T	Temperature (°K)	ΔH (Oe)
1	300	0
2	211	−20
3.125	174	−40
6.25	130	−80
9.375	103	−100
12.5	90	−130
25		−180
50		−210
100		−260
300		−285
600	4	−295

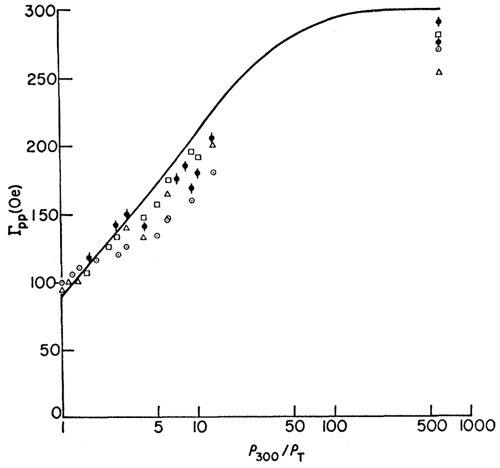

FIG. 3. Linewidth of FMR versus resistivity ratio for iron whiskers at 9.2 kMc/sec. The squares, triangles, and circles represent results for different samples. The smooth curve is calculated using the nonlocal theory of Hirst and Prange (Ref. 3).

We may note that although the calculated linewidth saturates at about 50°K, no such effect appears in the exchange shift. Experimentally, for [111] whiskers at 9.2 kMc/sec, the line center shifts from 840±20 Oe at room temperature to 730±20 Oe at 4.2°K. Since K_1 is expected to increase with decreasing temperature,[13] a shift of the line center to higher fields would be expected if the exchange effect were small. The observations suggest that the exchange shift is of the same order of magnitude as that given in Table I.

IV. CONCLUSIONS

We have shown that the FMR linewidths in iron single-crystal whiskers do not increase as rapidly with decreasing temperature as predicted by normal-conductivity theory. In fact, the nonlocal conductivity theory of Hirst and Prange,[3] with a C_F value of 1.5×10^{24} cm^{-1} sec^{-1}, predicts the experimental results quite closely.

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