

teraction in iron contributes significantly to the magnon energies.

Our data also contain information on the form factor associated with the spin waves via the observed intensities. As seen in the upper plot in Fig. 1, the magnon intensity falls off slightly, but significantly, with increasing 2θ . This behavior is even more pronounced in the data for larger misset angles. This angular dependence is approximately what one expects for a $3d$ -type form factor. Thus in the range of q values for which data are presented here, we are dealing with fluctuations whose spin density is comparable to that of $3d$ unpaired electrons.

A few measurements on a single crystal of pure iron were also made; however, the inclusion of small misoriented crystallites inherent in the nature of the growth process precluded the possibility of taking all the data from this crystal. The results shown in Fig. 2 for the two points that were measured are in excellent agreement with the iron-silicon data. Also drawn for comparison purposes is the quadratic law with the value $D = 286 \text{ meV \AA}^2$.

The exact values of β and γ are subject to change as further data at higher energies are included. Measurements are currently underway to extend the data into this region. It is also planned to determine the entire dispersion curve by direct energy analysis utilizing polarized neutrons.

We are very grateful to Dr. T. Riste, Dr. M. Blume, and Dr. R. D. Lowde for many stimulating discussions, and to Dr. H. J. Williams and Dr. A. J. Williams for the loan of an excellent single crystal.

*Work performed under the auspices of the U. S. Atomic Energy Commission.

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¹T. Riste, K. Blinowski, and J. Janik, *J. Phys. Chem. Solids* **9**, 153 (1959); E. Frikke and T. Riste, Proceedings of the International Conference on Magnetism, Nottingham, England, 1964 (The Institute of Physics and the Physical Society, University of Reading, Berkshire, England, 1965).

²G. A. Ferguson and A. W. Saenz, *J. Phys. Chem. Solids* **23**, 117 (1962); E. J. Samuelson, T. Riste, and O. Steinsvoll, *Phys. Letters* **6**, 47 (1963); T. Riste, G. Shirane, H. A. Alperin, and S. J. Pickart, *J. Appl. Phys.* **36**, 1076 (1965).

³R. D. Lowde and N. Umakantha, *Phys. Rev. Letters* **9**, 452 (1960). M. Hatherly, K. Hirakawa, R. D. Lowde, J. F. Mallett, M. W. Stringfellow, and B. H. Torrie, *Proc. Phys. Soc. (London)* **84**, 55 (1964). The exact value of D quoted here was kindly supplied by R. D. Lowde, private communication, 1965.

⁴H. A. Alperin, O. Steinsvoll, R. Nathans, and G. Shirane, to be published.

⁵B. N. Brockhouse and H. Watanabe, Inelastic Scattering of Neutrons in Solids and Liquids (International Atomic Energy Agency, Vienna, 1963), Vol. II, p. 297.

⁶M. L. Glasser and F. J. Milford, *Phys. Rev.* **130**, 1783 (1963).

OBSERVATION OF ELECTRON SPIN RESONANCE IN COPPER*

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(Received 16 June 1965)

We have observed electron spin resonance in single-crystal copper at a frequency of 9400 Mc/sec, over a temperature range from 1.4 to 60°K, utilizing the "selective-transmission" technique.¹⁻⁴ In this method there is a dc magnetic field and a perpendicular rf magnetic field in the usual manner, and the electrons absorb power from an rf field at their resonant frequency during the time they are in the skin depth. However, if the spin relaxation time is longer than the electron collision time, some of the electrons diffusing over to the other side of the sample will still be in a nonequilibrium state. That is, there will be a net precessing

magnetization at the Larmor frequency. This precessing magnetization will set up eddy currents and radiate power. It is this "transmitted" power that is measured as a function of frequency (or dc magnetic field). Under suitable experimental conditions this power can be made larger than the power transmitted via the normal skin-effect damping and spurious leakages and, hence, can be a very sensitive unambiguous test for electron spin resonance.

The samples were 2.5 cm in diameter, 0.0038 (I), 0.0127 (II), and 0.0441 (III) cm thick and formed part of the common wall between a pair

of TE_{101} cavities tuned to the same frequency. The exposed sample area was 0.4×0.4 in. The microwave power into the "transmit" cavity was 1000 cycle modulated and was ≈ 100 mW. The "receive" cavity was coupled to a superheterodyne detector incorporating a 60-Mc/sec i.f., phase-sensitive detection, which had a sensitivity of $\approx 10^{-19}$ W for a one-second time constant. By careful construction and sample mounting the leakage power was down 170 db under normal skin-effect conditions. Additional unmodulated microwave power was fed through a phase shifter to the mixer crystal which operated as a linear detector. This system thus allows the detection of that component of the transmitted power which is in phase with the reference microwave field. The dc magnetic field was always perpendicular to the rf field and could be rotated in the plane normal to the sample. Temperatures were determined utilizing a calibrated carbon resistor, and temperatures intermediate to liquid helium and hydrogen were obtained using a cold-finger in conjunction with a heater, controlled via a sensing device and suitable electronics. Samples I and II were cut with an acid-string saw, chemically polished and extensively electropolished with special precautions taken to insure a uniformly thick sample. Their orientations were (110) and (111), respectively. Sample III was spark-discharge⁵ cut and then treated as the other samples. Its orientation was (111).

Typical traces of transmission signal versus magnetic field are shown in Fig. 1. The phase shifter was adjusted to give a symmetric "absorption"-type signal for all cases. In the case of the usual reflection measurements on samples thicker than the skin depth, an asymmetric absorption (or its derivative) is observed,^{6,7} and the location of the resonant field must be determined from a knowledge of the parameters affecting the over-all line shape. In this experiment, inasmuch as we are at liberty to adjust the admixture of χ' and χ'' , we can always obtain symmetric lines. The validity of this approach was checked experimentally by measuring the transmission signal for lithium and comparing with a reference g marker of phosphorus-doped silicon for the temperature range from 1.5 to 300°K. The g value for copper as determined from the center of the line is 2.031 ± 0.003 and is independent of orientation, temperature, and

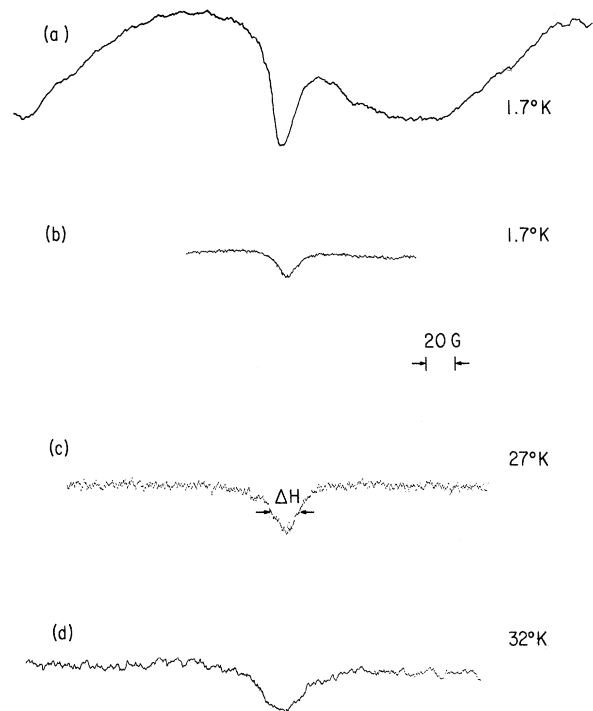


FIG. 1. Typical recorder traces of transmission microwave signal versus dc magnetic field for sample II. Transmitted power is proportional to the square of the signal. The frequency is ≈ 9400 Mc/sec, and the g value is 2.031 ± 0.003 . In (a), using a broad sweep at low temperatures, the "geometric cyclotron-resonance" signals are present in addition to the electron spin resonance. (b) is the same as (a), but adjusted for optimum observation of the electron spin resonance signal. For temperatures above $\approx 25^\circ\text{K}$, the baseline is straight and the line broadens [(c) and (d)].

sample to the accuracy stated. We know of no theoretical calculations for the g value of copper but note that it has a g shift opposite to the alkalis, and in the same direction as beryllium.⁶

We take the linewidth, ΔH , to be the width of the line at half the peak height. These data are plotted in Fig. 2. It is important to realize that the ΔH as plotted are subject to correction if one attempts to convert them in a consistent way to a spin-relaxation time T_2 . This is because the signals are observed over a range of conditions for which it is not easy to determine the theoretical line shapes. That is, as the temperature is lowered, the skin depth changes from normal to strongly anomalous, the electron mean free path becomes comparable to the size of the sample, and the electron diffusion length in a time T_2 ranges

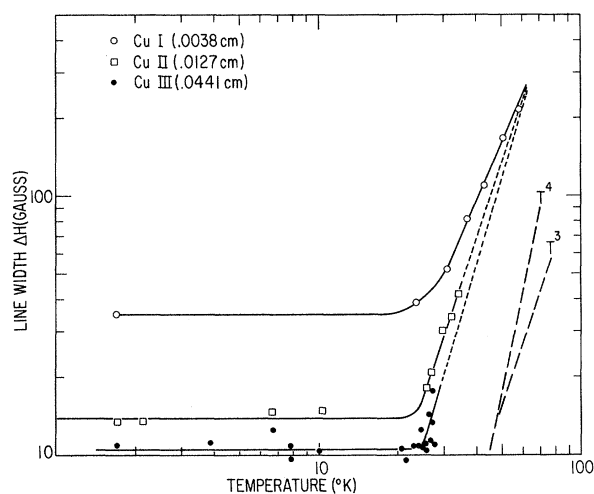


FIG. 2. Linewidth (ΔH) versus temperature. Estimated errors for I and II at all temperatures and for III below 10°K are $\pm 10\%$. For I and II the dc field was parallel to the sample surface. For III it was at an angle of 40° .

from less than to much greater than the sample thickness. Again, by comparison with lithium one can establish that the relation $\gamma\Delta HT_2 \sim 1.5$.

One of the advantages of the transmission technique is that there is no signal (barring leakage) in the absence of resonant transmission. Therefore, one can expect field-independent baselines, without the complications of spurious absorptions commonly seen in reflection spectroscopy. This is indeed found to be true to within the noise level of the equipment under normal skin-effect conditions. However, when the electron collision time τ is long enough such that $\omega\tau > 1$, and if the sample is comparable in thickness to a cyclotron radius, it is conceivable that one can observe transmission cyclotron-resonance signals. We have observed signals which we term "geometric cyclotron resonance" in many metals under the conditions mentioned above. The observed signal is dependent on the orientation of the magnetic field and the thickness of the sample, and will be treated in a separate paper. For our present experiment it presented experimental problems at low temperatures, as one had to extract the electron spin resonance line shape from a background signal often orders of magnitude larger. This constitutes the major source of uncertainty in the low-temperature measurements. At

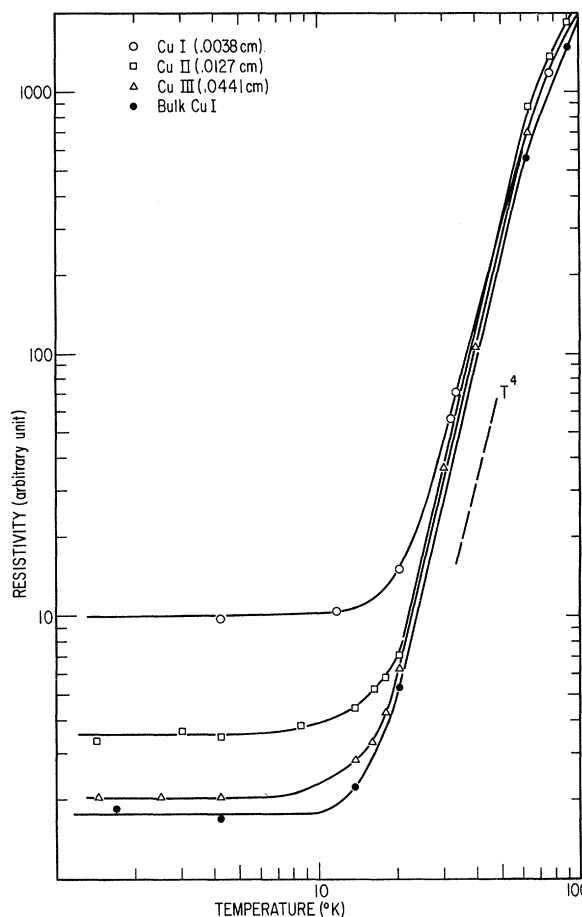


FIG. 3. Relative resistivity versus temperature. A resistance ratio of $\rho_{R.T.}/\rho_{4.2^\circ\text{K}} = 5000$ is 2 in the units shown. The estimated errors are $\pm 5\%$.

the higher temperatures, as expected, these effects disappeared, since $\omega\tau$ is < 1 , but due to the increasing linewidth and a diminishing transmission due to a shorter electron-spin-diffusion length, the signal-to-noise ultimately becomes a problem.

In Fig. 3 we have plotted the relative resistivity of the samples measured after the spin-resonance runs. The data for samples II and III and bulk I were obtained using the eddy-current method,⁸ while those for sample I were obtained by standard measurements of current and voltage. If one takes into account the changes of observed resistivity due to surface scattering when the mean free path is comparable to the dimensions of the sample,⁹ the data are consistent with the bulk-resistivity ratio $\rho_{R.T.}/\rho_{4.2^\circ\text{K}} \approx 5500$, and where the fraction of electrons specularly reflected is $0 < p < 0.5$.

A comparison of the temperature dependence

of the linewidths for the different samples in Fig. 2 in their linear region underscores the danger of interpreting the intrinsic temperature dependence as a function like T^n unless a wide temperature range is covered and unless it is invariant over the relevant sample parameters. The linewidth data below 20°K and the resistivity data below 8°K are essentially temperature independent. A comparison of Fig. 2 and Fig. 3 suggests that the linewidths at the lowest temperatures can be written as the sum of a surface-relaxation term (ΔH_s), and a constant term due to impurity scattering (ΔH_i). Since the mean free path is greater than the thickness of the sample, we will assume ΔH_s is inversely proportional to the thickness. A consistent fit of the three equations in the two unknowns can be made for linewidths chosen within the limits of error yielding $\Delta H_i = 7$ G. We expect the curve drawn for sample III to be the closest to the intrinsic behavior of copper, and after subtracting the correction of ΔH_i we find the linewidth is proportional to the resistivity from 60°K (taken as the limit of the extrapolated curve) down to 27°K.

The only bulk metal to exhibit a temperature-dependent linewidth before this work was sodium.⁶ Various attempts have been made to determine the temperature dependence using small particles.^{10,11} A theoretical analysis of the dependence of T_1 has been covered in a review by Yafet,¹² who concludes that the linewidth should vary as the resistivity. If one plots the observed data for sodium in a manner similar to Fig. 2, one finds good agreement between the various reported measurements, and that the curve is similar to that for sample I over the same temperature range. At high temperatures the linewidth is propor-

tional to the resistivity. This suggests that surface effects and other temperature-independent relaxation processes have probably been the determining factor in the linewidths in sodium at low temperatures, and that further work to determine quantitatively their separate contributions must be done before conclusions about low-temperature spin-relaxation processes in metals can be made.

We wish to thank Professor G. Feher for continuing advice and Dr. F. W. Young for generously supplying us with the copper crystals and helpful suggestions. We would also like to thank Dr. J. Kunzler for supplying us with a copper single crystal. One of us (S.S.) would like to thank Professor A. Ron for many helpful discussions.

*Work supported by the National Science Foundation.

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¹M. Ya. Azbel', V. I. Gerasimenko, and I. M. Lifshitz, *Zh. Eksperim. i Teor. Fiz.* **32**, 1212 (1957) [translation: *Soviet Phys.-JETP* **5**, 986 (1957)].

²M. Ya. Azbel' and I. M. Lifshitz, *Progress in Low Temperature Physics*, edited by C. J. Gorter (North-Holland Publishing Company, Amsterdam, 1961), Vol. III.

³R. B. Lewis and T. R. Carver, *Phys. Rev. Letters* **12**, 693 (1964).

⁴N. S. VanderVen and R. T. Schumacher, *Phys. Rev. Letters* **12**, 695 (1964).

⁵Servomet Spark Machine, manufactured by Metals Research Ltd.

⁶G. Feher and A. F. Kip, *Phys. Rev.* **98**, 337 (1955).

⁷Freeman J. Dyson, *Phys. Rev.* **98**, 349 (1955).

⁸C. P. Bean, R. W. DeBlois, and L. B. Nesbitt, *J. Appl. Phys.* **30**, 1976 (1959).

⁹E. H. Sondheimer, *Advan. Phys.* **1**, 1 (1952).

¹⁰R. A. Levy, *Phys. Rev.* **102**, 31 (1956).

¹¹F. Vescial, N. S. VanderVen, and R. T. Schumacher, *Phys. Rev.* **134**, A1286 (1964).

¹²Y. Yafet, *Solid State Phys.* **14**, 1 (1963).