On the Ferromagnetic Resonance in Nickel and Supermalloy*

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Both the real and imaginary part of the high frequency permeability of flat specimens of nickel and supermalloy have been investigated at 9030 mc/sec. and 24400 mc/sec., with a constant field H_0 parallel to the plane of the specimen. For the higher frequency, experiments were extended beyond the Curie point (358°C for nickel and about 400°C for supermalloy). The decrease in saturation magnetization causes the maximum and the minimum of ferromagnetic absorption to shift toward higher field strengths. The g factor is independent of temperature within the limit of error (2 percent); $g=2.12$ for supermalloy and $g=2.20$ for nickel.

The line width in nickel increases first slowly, but above 200°C more rapidly from 250 gauss at room temperature to 700 gauss at the Curie point, the width being defined as half the width between the points where the imaginary part of the susceptibility has dropped to half its maximum value. In supermalloy the width is 110 gauss between 20°C and 300°C and then rises sharply to 350 gauss at 420°C. There is no discontinuity at the Curie point, but the intensity of the absorption drops very rapidly beyond that temperature. At the lower frequency the width is reduced by approximately a factor 1.6.

The observed data are compared with existing theories. Only interactions of the ferromagnetic spins with the lattice vibrations or the conduction electrons could probably account for the observed order of magnitude of the width. It is not clear, however, how the width could be independent over a large temperature interval, if these interactions were the only broadening agent. A pseudo-dipolar interaction, introduced by Van Vleck, might give an explanation for the line width at lower temperatures.

I. INTRODUCTION

As early as 1935, Landau and Lifshitz[1] wrote a fundamental paper on ferromagnetic permeabilities and predicted the existence of resonance effects. The first experimental observation was made by Griffiths[2] in 1946. Kittel[3] extended Landau's theory and showed how the ratio of the frequency and the magnetic field at which resonance occurs depends critically on the demagnetizing factors, and therefore on the geometry of the sample. The classical equation of motion for the magnetic moment of a ferromagnetic body expresses the balance between the exerted torque and the rate of change of angular momentum

$$(dM_{x,y}/dt)=\gamma[\mathbf{M}\times\mathbf{H}_{\text{eff}}]_{x,y}-M_{x,y}/T_2, \quad (1a)$$

$$(dM_z/dt)=\gamma[\mathbf{M}\times\mathbf{H}_{\text{eff}}]_z-(M_z-M_0)/T_1 \quad (1b)$$

with

$$\gamma=ge/2mc, \quad (1c)$$

where g is the Landé factor and M_0 is the static saturation magnetization. These equations have the same form as those describing the nuclear magnetic resonance and the notation is the same as in Bloch's paper.[4] In the nuclear case H_{eff} can usually be put equal to the externally applied magnetic field H^{ext}, consisting of a constant field H_0 and a radiofrequency component which are mutually perpendicular,

$$H_x^{\text{ext}}=H_1e^{i\omega t}, \quad H_y^{\text{ext}}=0, \quad H_z^{\text{ext}}=H_0. \quad (2)$$

In the ferromagnetic case we have to add three terms to the external field in order to get the effective field:

$$\mathbf{H}_{\text{eff}}=\mathbf{H}^{\text{ext}}-N_{x,y,z}\mathbf{M}+\mathbf{H}^{\text{anis}}+\alpha\nabla^2\mathbf{M}. \quad (3)$$

The terms $-N_xM_x$, $-N_yM_y$, $-N_zM_z$, where N is the demagnetizing factor, represent the internal field produced by the magnetization of the sample. In the ferromagnetic case these terms have the same order of magnitude as the applied field.

The anisotropy energy of a ferromagnetic sample adds the torque $\partial f/\partial\vartheta_{x,y,z}$ to the equations of motion, where $\partial\vartheta_{x,y,z}$ denotes a rotation around the x, y or z axis, respectively. Kittel[3] showed that this torque can in many cases be written as $\mathbf{M}\times\mathbf{H}^{\text{anis}}$. Finally an inhomogeneous distribution in the magnetization gives rise to an increase in the exchange energy of the spin system, the effect of which may be described[1] by a contribution $\alpha\nabla^2\mathbf{M}$ to the effective field with

$$\alpha=4JS^2/aM_0^2, \quad (4)$$

where J is the exchange energy and a the distance between two neighboring spins.

The damping terms in Eqs. (1) have a simpler form than the corresponding expression (21) in Landau's paper.[1] It should be pointed out that T_1 and T_2 may be arbitrary functions, usually depending on the frequency of the applied field and on the temperature, and that in general T_1 and T_2 are not equal, but that

$$1/T_2=1/(2T_1)+1/T_2^*. \quad (5)$$

Interactions between the spin system and the lattice vibrations determine the quantity T_1 and magnetic and exchange interactions within the system of spins determine T_2^*. A discussion of these interactions will be postponed to Section IV. Presently we shall consider T_1 and T_2 only from the phenomenological point of view.

We shall now omit the last two terms in Eq. (3). The

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† Society of Fellows.
[1] L. Landau and E. Lifshitz, Physik. Zeits. Sowjetunion **8**, 153 (1935).
[2] J. H. E. Griffiths, Nature **158**, 670 (1946).
[3] C. Kittel, Phys. Rev. **73**, 155 (1948).
[4] F. Bloch, Phys. Rev. **70**, 460 (1946).

influence of these terms will be discussed in Section IV and will be shown to be small. A stationary solution of Eqs. (1) can then be obtained by putting $M_{x,y}=M_{x,y}e^{i\omega t}$. Combining (1), (2), and (3) we get the solution

$$M_x=\frac{\gamma^2 M_z\{H_0+(N_y-N_z)M_z\}}{[i\omega+(1/T_2)]^2+\gamma^2\{H_0+(N_x-N_z)M_z\}\{H_0+(N_y-N_z)M_z\}}H_1, \tag{6}$$

$$M_y=\frac{\gamma M_z(i\omega+1/T_2)}{(i\omega+1/T_2)^2+\gamma^2\{H_0+(N_x-N_z)M_z\}\{H_0+(N_y-N_z)M_z\}}H_1, \tag{7}$$

while M_z can be obtained by substituting these expressions into

$$(M_z-M_0)/T_1=-\gamma\langle H_1M_y\rangle_{\text{Av}}+\gamma(N_x-N_y)\langle M_xM_y\rangle_{\text{Av}}. \tag{8}$$

The average is taken over the time, and we neglect small periodic variations in M_z. For small values of the radiofrequency field H_1, when

$$\gamma^2H_1^2T_1T_2\ll 1,$$

the right-hand side of Eq. (8) may be put equal to zero and we have $M_z=M_0$, which is valid for all experiments described in the present paper. At a very high power level saturation phenomena will occur which play such an important role in nuclear resonance. It may be pointed out that experiments at high power levels, as have been carried out by Slichter[5] in paramagnetic salts, would give information on T_1 in ferromagnetic materials. Using Eq. (6) with $M_z=M_0$ and the well-known relation for the susceptibility

$$\mu-1=\mu_1-1-i\mu_2=4\pi\chi=4\pi M_x/H_1, \tag{9}$$

we obtain the following expressions for the real and imaginary parts of the ferromagnetic high frequency permeability,

$$\mu_1=\frac{4\pi\gamma^2M_0\{H_0+(N_y-N_z)M_0\}(\omega_0^2-\omega^2)}{(-\omega^2+\omega_0^2)^2+4\omega^2/T_2^2}+1, \tag{10}$$

$$\mu_2=\frac{4\pi\gamma^2M_0\{H_0+(N_y-N_z)M_0\}2\omega/T_2}{(-\omega^2+\omega_0^2)^2+4\omega^2/T_2^2} \tag{11}$$

with a resonant frequency ω_0 given by

$$\omega_0^2=\gamma^2\{H_0+(N_x-N_z)M_0\}\times\{H_0+(N_y-N_z)M_0\}+1/T_2^2. \tag{12}$$

We shall now discuss how these permeabilities are obtained from experiment. This research was originally undertaken to get an understanding of the quantity T_2 in ferromagnetic materials. Experiments around the Curie temperature seemed therefore advisable, since the causes which determine T_2 are best understood for paramagnetic substances.

II. EXPERIMENTAL METHOD

A standard method of determining the high frequency magnetic susceptibility of an insulator is to fill part of the space—preferably where the electric field is zero—inside a resonance circuit with the material to be investigated.[6] The real part of the susceptibility is then determined by the shift in resonant frequency and the imaginary part by the change in the Q of the resonant circuit.

In the case of a ferromagnetic metal the important electric losses, caused by the finite resistance of the material, cannot be separated physically from the magnetic losses. The sample is now conveniently made one of the walls of the resonant circuit. If we extend a well-known formula for the effect of the finite skin depth on the losses and the resonant frequency of a cavity[7] to the case where the permeability of the wall is different from unity, we find in an approximation to the first order in $1/Q$

$$1/Q_{\text{fer}}=\mu_R^{\frac{1}{2}}/Q_0, \tag{13}$$

$$-2\Delta\omega/\omega_0=\pm\mu_L^{\frac{1}{2}}/Q_0, \tag{14}$$

with

$$\mu_R=(\mu_1^2+\mu_2^2)^{\frac{1}{2}}+\mu_2, \tag{15}$$

$$\mu_L=(\mu_1^2+\mu_2^2)^{\frac{1}{2}}-\mu_2, \tag{16}$$

and

$$Q_0=(2/\delta_0)\int\int H^2dS\Big/\int\int\int H^2dV. \tag{17}$$

The surface integral extends over the ferromagnetic surface, δ_0 is the skin depth and Q_0 the quality factor of the wall, if its permeability were unity. The + sign in Eq. (14) holds if μ_1, as given by Eq. (10), is positive, the − sign, if μ_1 is negative. In this latter case of *anomalous dispersion* the skin depth losses are accompanied by a decrease of the effective length of the cavity

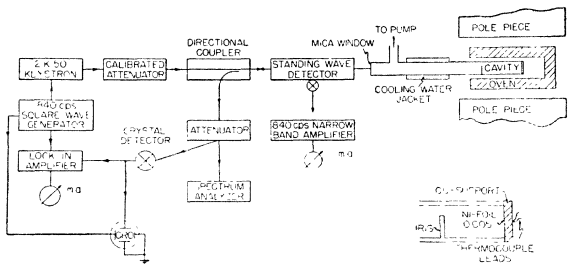

FIG. 1. Block diagram of the apparatus.

[5] C. P. Slichter, thesis, Harvard University (1949).

[6] H. G. Beljers, Physica **14**, 629 (1949).

[7] See, e.g., J. C. Slater, Rev. Mod. Phys. **18**, 441 (1946), especially the derivation of formula (III-59); also J. Bernier, *Onde Electrique* **26**, 305 (1946).

and an increase of its resonant frequency. The quantities on the left-hand side of Eqs. (13) and (14) can be measured experimentally by any of the usual methods.[8] Our particular experimental arrangement is shown in the block diagram of Fig. 1. It is essentially the same as one used by Yager.[9] Also the technique of evaluation of the data is at many points the same as indicated in his papers.

A 2K50 klystron generates power at about 24,400 mc/sec. All voltages for the klystron, including the heater voltage, are derived from an electronically regulated power supply. A square wave of 840 c.p.s. is applied to the repeller, which provides an off-on modulation. The microwave power which can be varied by a calibrated attenuator is partly reflected by the cavity system which terminates the end of the wave guide. The reflected power is fed into a crystal detector by means of a directional coupler and the rectified signal passes through an 840 c.p.s. narrow band amplifier and lock-in detector. The standing wave pattern set up by the reflections from the cavity can be measured by a standing wave detector also followed by an 840 c.p.s. amplifier. The over-all response of the crystal-amplifier combinations was determined with the calibrated attenuator, and deviated slightly from a square law. The microwave power could alternatively be fed into a spectrum analyzer which had a calibrated wave meter for frequency measurements.

The rectangular cavity consists of a section of guide, one guide wave-length long, shorted at one end by a slab of ferromagnetic material. On the other side it is coupled by an asymmetrical window through which it is excited in the E_{102} mode. The size of the diaphragm is so chosen that the cavity is undercoupled to such an extent that the reflected power in zero external field is between 10 and 20 percent of the incident power. The cavity is placed between the pole pieces of an electromagnet. If we call the plane of the ferromagnetic wall the xz plane, then the microwave field at the wall is in the x direction and the constant field in the z direction. The pole pieces were 7 inches in diameter and 2 inches apart. A current of 10 amp. at 200 volts through the excitation coils provided a field of about 10,000 gauss. This current could be maintained for several minutes without a special cooling arrangement for the coils. Since the ferromagnetic resonance curves are wide, there are no special requirements on the homogeneity or regulation of the field. The field strength was measured with less than 0.5 percent error with a flip coil and fluxmeter combination, which was calibrated at one point with a proton magnetic resonance. The width of the gap is ample so that a small oven can be put around the cavity. The oven consists of 50 turns of non-magnetic wire (total resistance 20Ω), sandwiched between two asbestos cylinders, $1\frac{3}{4}$ inches outside diameter and 6 inches long. A current of 4 amp. was sufficient to heat the cavity to 450°C. The temperature was measured with a platinum-platinum-rhodium thermocouple. One of the joints was clamped against the end wall of the cavity. To prevent oxidation of the end wall and copper side walls of the cavity, the pressure in it was kept below 0.01 mm Hg with a small roughing pump. Microscopic examination of the inside walls showed no trace of oxidation after completion of the runs. The choke flange with sealed mica window (0.0015-inch thick) was 15 inches away from the cavity. In between, a cooling jacket was soldered around the guide through which a convection current of water was circulated. A dummy mica window was placed a quarter wave-length away from the one which kept the pressure to minimize microwave reflections.

Great care should be taken in the preparation of the samples. They should be kept free from strains and surface contaminations, since the microwaves penetrate only 10^{-4} to 10^{-5} cm. The surface should also be as smooth as possible. Electrolytic nickel foil** (0.005-inch thick) was used, spectroscopically analyzed to be 99.8 percent pure, the main contaminant being 0.17 percent of cobalt. Supermalloy (0.004-inch thick) was generously put at our disposal by Dr. Bozorth of the Bell Telephone Laboratories. The samples were cut to size between two brass disks, carefully avoiding shearing forces. They were soldered with gold to a copper disk which served as support and annealed for one hour at 850°C, then slowly cooled to room temperature. They were electropolished in a bath of 90 percent phosphoric and 10 percent chromic acid, kept at 90°C. The current of 5 amp./cm^2 was maintained for 20 sec. Finally, they were silver-soldered to the end of the wave-guide where care was taken that the solder would not flow over the inside wall. The soldering operations were all carried out in an atmosphere of purified hydrogen. To check whether they had any spurious effect on the ferromagnetic resonance, data were also taken at room temperature with a cavity which could not be evacuated and where the sample foil was pressed tightly against a narrow edge of the guide wall. The contact resistance did not appreciably lower the value of Q. No change in the resonance curve was observed for nickel. The value of $\mu_R{}^{\max}$ for supermalloy was 25 percent lower for the soldered cavity, probably due to a slight strain in the material. Data reproduced well after each run at higher temperature.

The demagnetizing factors would be $N_x=0$, $N_y=4\pi$, $N_z=0$, if the foils were infinitely thin. This assumption is valid for the x and y direction, since the foil is only one skin-depth thick for the microwave components. The z component requires a small correction:[10] $N_z=-\pi^2(d/a)\sim 0.08$, where a and d are the axes of an

[8] G. C. Montgomery, *Technique of Microwave Measurements* (McGraw-Hill Book Company, Inc., 1947).

[9] W. A. Yager, Phys. Rev. **75**, 316 (1949).

** The nickel was made available by the courtesy of the Superior Tube Company.

[10] J. C. Maxwell, *Electricity and Magnetism* (Clarendon Press, Oxford, 1881), Volume 2, p. 69.

oblate spheroid, d corresponding to the thickness of the sample, and a to its diameter. Most conveniently this correction amounting to about 50 gauss is directly applied to the value of the external field H_0.

The experiments at X band were carried out only at room temperature with a similar arrangement of the electric circuits. The power at 9030 mc/sec. was generated by a 2K25 klystron.

The procedure is now to vary the frequency of the klystron slowly by changing the repeller voltage, or the thermal tuning grid voltage in the case of the 2K50, until minimum reflection into the directional coupler indicates resonance in the cavity. The voltage standing-wave ratio at resonance, β, is equal to the ratio of the unloaded Q of the cavity, Q_u, and the external or radiation Q_e,

$$\beta=Q_e/Q_u=Q_e/Q_c+Q_e/Q_{\text{fer}}, \tag{18}$$

where $1/Q_c$ and $1/Q_{\text{fer}}$ represent the losses in the copper and ferromagnetic walls, respectively. A correction has been made for the attenuation in the guide between the detector and the cavity. The value of β is measured as a function of the constant magnetic field H_0 at various temperatures. Actually in each run β was measured with the standing wave detector only for one or two values of H_0. In other points the reflected power P_r was measured with the directional coupler output and β was determined from the formula

$$\frac{P_r}{P_i}=\frac{(1-\beta)^2}{(1+\beta)^2}.$$

This procedure was not only much faster, but also gave more accurate results near maximum absorption, where the VSWR becomes very high and P_r/P_i may be as large as 0.6. In addition we determine the total Q by varying the frequency through the resonance of the cavity and measuring the VSWR as a function of frequency. From Eq. (18) and the relation

$$1/Q=1/Q_u+1/Q_e,$$

Q_e and Q_u are known. Q_c could be calculated from the geometry of the cavity and the conductivity of copper. Then Q_{fer} would be known from Eq. (18) and μ_R from (13). The y component of the magnetization does not contribute to Q_{fer}, since the cavity is only excited in a mode with a vanishing y component of the magnetic field at the sample. In practice this procedure does not work very well and gives erroneous values for μ_R. It is a common phenomenon that the theoretical values for Q are usually 10 to 30 percent higher than the experimental values. Causes for this discrepancy are scratches and roughness of the surface[11] and perhaps also spurious losses in soldered or clamped joints. An error of 30 percent in Q_c would cause an error of a factor 3 or more in μ_R. The following procedure would therefore be preferable. Let μ_R and $'\mu_R$ be two values of the permeability, belonging to two values of the magnetic field H_0 and $'H_0$, with measured SWR β and $'\beta$. From Eqs. (13) and (18) we then obtain an expression from which Q_c is eliminated,

$$\mu_R=\{'\mu_R^{\frac{1}{2}}+(\beta-\beta')(Q_0/Q_e)\}^2. \tag{19}$$

The quantity Q_0 has still to be computed from Eq. (17). Values for the d.c. conductivity, which enter into δ_0, have been taken from the *Physikalisch Chemische Tabellen* in the case of nickel. For supermalloy[12] only the value at room temperature is known $1/\sigma=65\times10^{-6}$ Ω/cm^{-1}. The assumption, verified in similar alloys, that the resistance is equal to that of nickel at the same temperature plus a temperature independent part, makes it possible to calculate δ_0 for supermalloy at higher temperatures. It is true that the calculated value of Q_0 may again be somewhat too high. But now we have only one wall with a well-polished, well-defined surface, so that the systematic error in μ_R is probably less than 10 percent too high. Typical Q values at K band are $Q_u=2440$, $Q_e=6730$, $Q_0=25{,}000$ for nickel at 24°C and $Q_u=1750$, $Q_e=6650$, $Q_0=12{,}000$ for nickel at 350°C.

As the reference value $'\mu_R$ in Eq. (19) the limiting value of the permeability for very high fields H_0 may be used. From (10), (11) and (15) we see that $'\mu_R$ then approaches unity. At 9100 mc/sec. this procedure has been used. But at 24,000 mc/sec. the available field strength was unfortunately not sufficiently larger than

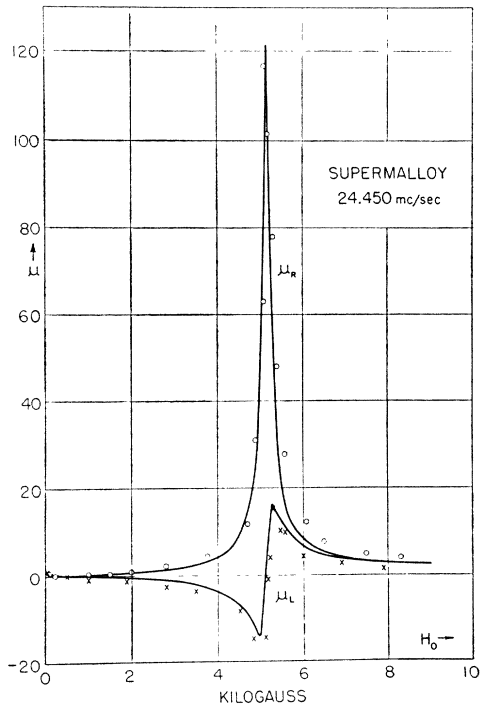

FIG. 2. The real and imaginary parts of the effective permeability in supermalloy at 24400 mc/sec.

[11] S. P. Morgan, J. App. Phys. **20**, 352 (1949).

[12] O. L. Boothby and R. M. Bozorth, J. App. Phys. **18**, 173 (1947).

the resonance field to warrant an extrapolation of β' to its value for infinitely high fields. Here we assumed $'\mu_R$ to be near unity in zero external field. It should be kept in mind that for very low values of H_0 the domains will not be aligned and the magnetization M_0 will tend to a very small value. Inspection of (10), (11), and (15) shows that μ_R will be close to unity in that case. Actually, better agreement between theory and experiment is obtained when $'\mu_R$ is taken to be slightly smaller, between 0.8 and 0.9. The value is adjusted for a good fit at the minimum of the curves (Figs. 3 and 5) which will be discussed in the next section. The maximum value of the absorption near resonance is very insensitive to small variations in $'\mu_R$. Nevertheless, the error in the absolute value of μ_R is rather high and could be as large as 25 percent. It is subject to systematic errors, one of which, the determination of Q_0, has already been mentioned. Another error arises from spurious reflections from choke flanges, pumping slot and mica window. The SWR from these causes was measured to be less than 1.05, by putting a tapered load in the guide. The directivity of the coupler was better than 30 db. The phases

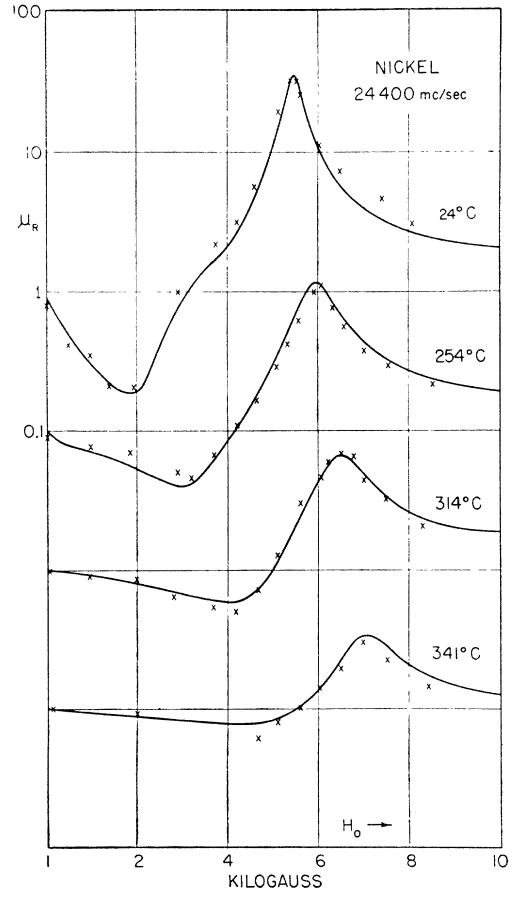

FIG. 3. The effective permeability of nickel at 24400 mc/sec. for various temperatures. Each curve is lowered by one decade with respect to the one above it. The numbers along the ordinate axis apply to the top curve. The drawn lines have been calculated theoretically.

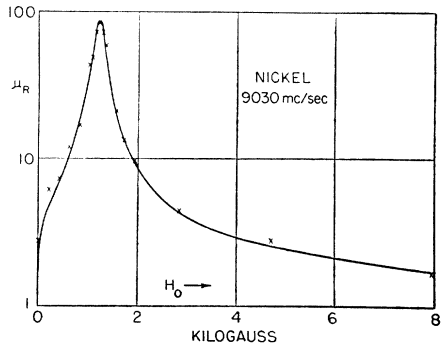

FIG. 4. The effective permeability of nickel at 9030 mc/sec. at room temperature.

with which these spurious reflections add to the signal reflected from the cavity vary largely, as the effective length of the guide is enlarged by heating. If the cavity were well matched, intolerable errors would result in the measured reflected power. Therefore the coupling was chosen such that the reflected power was always more than 10 percent of the incident power. The error is smaller near the maximum, where the reflection is larger, and cancels to a considerable extent when the difference $\beta-'\beta$ is taken in Eq. (19) for the variation in μ_R. But the error in μ_R is twice as large as that in the power measurement because the square must be taken on the right side of (19). Adding an uncertainty of about 5 percent in the calibration of the attenuator and 5 percent in the measurement of Q_e, one obtains a rather large possible error in μ_R of 30 percent in extreme cases. More precise measurements should be carried out with a transmission type of cavity and different couplings should be used in the range of low and very high values of μ_R.

III. EXPERIMENTAL RESULTS

In Fig. 2 the real and imaginary parts of the *effective* permeability are plotted, calculated from Eqs. (10), (11), (15), and (16), together with some experimental points for supermalloy at 24,400 mc/sec. It is seen that the theory is generally confirmed. The ratio $|\mu_R/\mu_L|$ is much larger than the more familiar quantity $|\mu_2/\mu_1|$. The detuning of the cavity was 8 mc/sec. from the maximum to the minimum in μ_L and the anomalous dispersion is demonstrated quantitatively by this ex-

TABLE I. Nickel.

Temp.	Frequency mc/sec.	H_0^{max} gauss	$4\pi M_s$ gauss	g	μ_R^{max}	$1/T_2$
24°C	24410	5400	6000	2.24	34	$5.0^5\times10^9$ sec.$^{-1}$
114	24380	5550	5700	2.20	32	5.0^5
190	24350	5800	5100	2.19	21	6.4^5
254	24330	5950	4500	2.20	12	9.7
314	24320	6600	3300	2.16	7	11.0
341	24310	7000	2200	2.18	3.5	13.8
358	24280	7200	1000	2.26	1.5	14.5
24	9050	1160	6000	2.23	85	3.3^5

TABLE II. Supermalloy.

Temp. °C	Freq. mc/sec.	H_0^{max} gauss	H_0^{min} gauss	$4\pi M_s$ gauss	g	μ_R^{max}	$1/T_2$ sec.$^{-1}$
28	24310	5150	300	7900	2.13	110	2.1×10^9
126	24290	5400	1000	7250	2.11	79	2.6
179	24280	5550	1350	7000	2.10	71	2.7
246	24250	5700	2000	6250	2.11	71	2.4
311	24230	6150	3000	5450	2.07	46	3.0
355	24210	6300	3600	4680	2.10	38	3.0
407	24200	7100	5300	3280	2.06	8.7	8.1
				(2500)	(2.11)		(6.2)
420	24190	7400	6100	2250	2.05	4.5	10.5
				(1500)	(2.15)		(7.0)
440	24180	8200	?	——		2.0	
24	9050	1040	——	7800	2.10	300	1.3

periment. We shall now proceed with a more systematic discussion and plot μ_R on a logarithmic scale to bring out more detail in the wings of the absorption curve.

(a) Nickel

The experimental values for μ_R at 24,400 mc/sec. together with the theoretical curves, are shown in Fig. 3 for various temperatures. The three parameters, γ, T_2, and M_0, occurring in (10) and (11) have to be determined. Note that ω is a constant and ω_0 is a variable, changing with H_0 in our experiment. The values for M_0 have been taken from the static magnetization experiments of Weiss and Forster.[13] At low temperatures the magnetization is practically constant but drops off rapidly in very small fields. For temperatures close to the Curie temperature, M_0 increases continually with increasing H_0. Then γ and T_2 are determined from the position and the value of the maximum absorption according to the relations

$$\omega=\gamma\{H_0^{max}(H_0^{max}+4\pi M_0)\}^{\frac{1}{2}} \tag{20}$$

and

$$(1/T_2)=4\pi\gamma^2 M_0(H_0^{max}+4\pi M_0)/\omega\mu_R^{max}. \tag{21}$$

The main uncertainty in g, which is proportional to γ according to Eq. (1c), comes from inaccurate knowledge of M_0; H_0^{max} was measured with an error of 0.5 percent, except at the highest temperatures, where the line becomes broad and the intensity small. The error in g is probably between 1 and 2 percent. The values of g are listed in Table I; $g=2.20$ remains constant over the whole temperature range, within the limits of error. The uncertainty in T_2 is much larger, the same as in μ_R, as discussed in the preceding section. Nevertheless, it can be established beyond doubt from the listed values that the half-width between the points where μ_2 has dropped to half its maximum value increases from 250 gauss at room temperature, first slowly and then above 200°C more rapidly, to about 750 gauss at 358°C. At the Curie point no discontinuity occurs in the absorption curve, but at higher temperatures the signal soon becomes unobservably small because of the rapid decrease in magnetization M_0. It is furthermore seen that the shape of the absorption curve is also well represented by the theoretical formulas (10) and (11). The minimum is brought out clearly and shifts more rapidly than the maximum toward higher field strengths as the temperature is increased and the magnetization decreased.

The results for room temperature at 9030 mc/sec. are shown in Fig. 4, and are also included in the table. The g factor is the same, but the line width $\Delta\omega$ calculated from the maximum is decreased by a factor 1.5.

(b) Supermalloy

Unfortunately no values for the saturation magnetization have been published, except at room temperature.[12] In order to determine M_0 we use in addition to Eq. (20) the relation

$$\omega=\gamma(H_0^{min}+4\pi M_0), \tag{22}$$

where H_0^{min} is the field at the minimum in the absorption curve. Then γ and M_0 are solved from (20) and

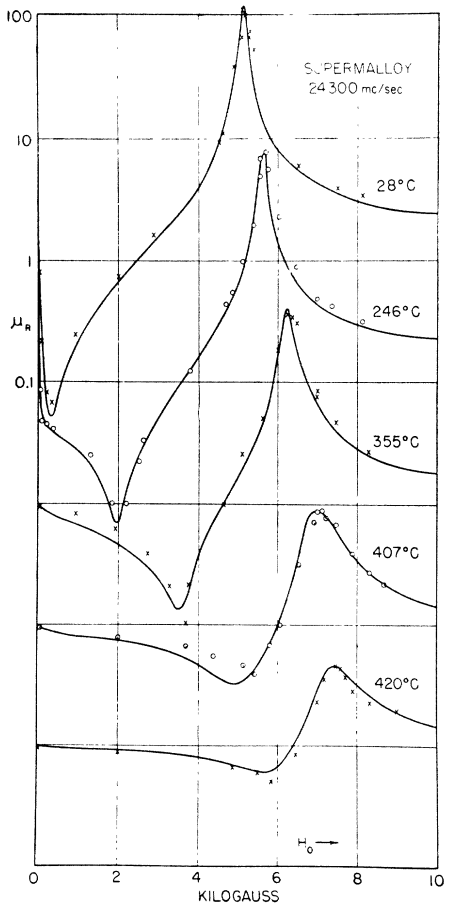

FIG. 5. The effective permeability of supermalloy at 24300 mc/sec. for various temperatures. Each curve is lowered by one decade with respect to the one above it. The numbers along the ordinate axis apply to the top curve. The drawn lines have been calculated theoretically.

[13] See, e.g., R. Becker and W. Döring, *Ferromagnetismus*, (Julius Springer, Berlin, 1939), p. 25.

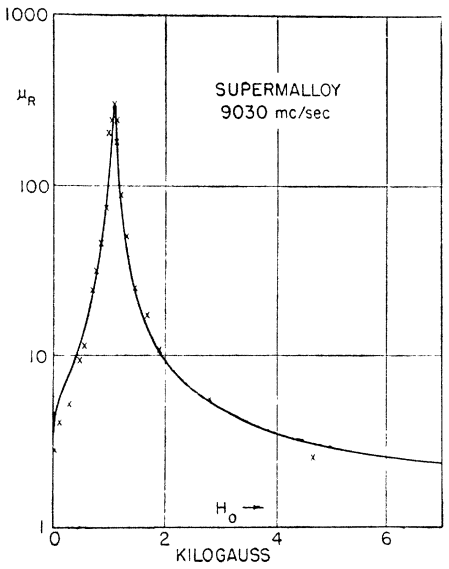

FIG. 6. The effective permeability of supermalloy at 9030 mc/sec. at room temperature.

(22). Equation (22) was first derived by Kittel,[3] and is based on the assumptions that the line-width is small and that the variation in μ_2 may be neglected at the position of the minimum, and also that M_0 is independent of H_0. This procedure can therefore not be applied at higher temperatures where the line gets broader, the maximum and minimum are close together and M_0 is not constant. In general the calculated values for M_0 listed in Table II will be too high. At room temperature there is good agreement, but comparison with preliminary unpublished data from the Bell Telephone Laboratories shows that our values between 200°C and 400°C are a few hundred oersteds too high. Much larger deviations must be expected near 400°C. To show the influence on g and T_2 of lower values of M_0, more probable values of M_0 have tentatively been added to the table between brackets for the highest temperatures. Within this uncertainty in M_0 the value of $g=2.12$ is constant over the whole temperature range. Yager reported earlier $g=2.17$, but informed the author that this value is not final. Experimental points and theoretical curves for μ_R in supermalloy have been plotted in Fig. 5. There is fair agreement even far out in the wings of the curve. The half-width at room temperature is 105 gauss, 2.5 times smaller than for nickel. It remains practically constant up to 300°C, then rises rather abruptly by a factor of 3 near the Curie point, which is at about 400°C. Again no discontinuities occur at this point, but the intensity decreases rapidly proportional to the magnetization. In Fig. 7 the line-widths in supermalloy and nickel are plotted as functions of temperature.

The values of μ_R at 9050 mc/sec. are shown in Fig. 6. The g-factor is independent of the frequency, but the width $\Delta\omega$ is decreased by a factor 1.6. The minimum does not appear, since even in zero field the condition (22) cannot be satisfied at this frequency.

IV. COMPARISON WITH THEORY

The anomalous g values have been discussed by Kittel.[14] New and more precise measurements seem to converge to the lower end of the range of experimental values quoted in Kittel's paper. So do ours, but the experimental g values are still too high to fit the relation $g-2=2-g'$, where the magneto-mechanical ratio $g'=1.92$ for nickel. A quantum mechanical derivation of this relation has been given recently by Van Vleck.[15] No reason can be given for the discrepancy. The effect of the finite skin depth[16]—the term with $\nabla^2\mathbf{M}$ in Eq. (3)—would lower g by only 0.1 percent. The term with the anisotropy energy will change g, depending on the orientation of the crystalline sample with respect to the magnetic field. Since we work with a polycrystalline material, this effect will tend to broaden the line, but the center position will remain unaffected. A unidirectional stress on the sample, however, might produce a shift of the whole line. This effect did not occur, as

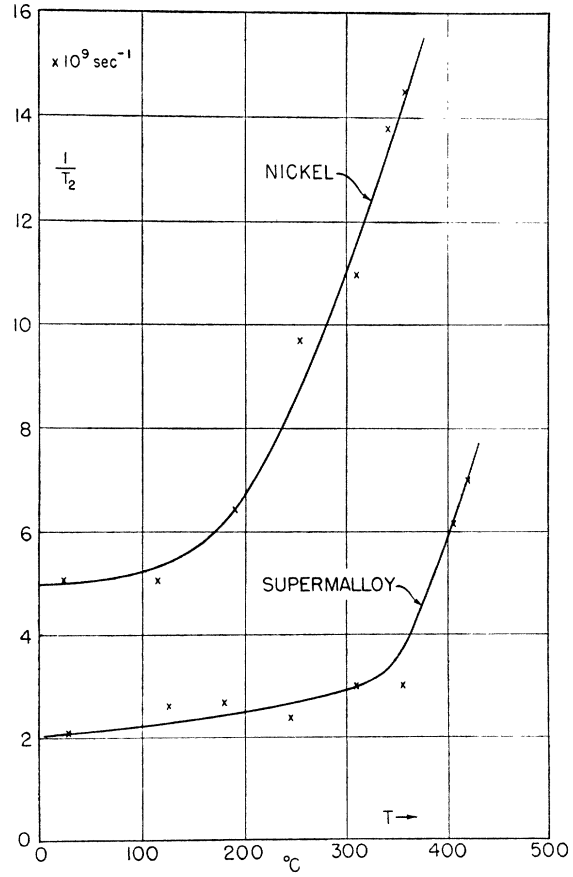

FIG. 7. Line-widths in nickel and supermalloy as functions of temperature at 24400 mc/sec.

[14] C. Kittel, Phys. Rev. **76**, 743 (1949).
[15] J. H. Van Vleck, Phys. Rev. **78**, 266 (1950).
[16] C. Herring and C. Kittel, Phys. Rev. **77**, 725 (1950).

could be demonstrated by rotating the whole sample; no shift of the resonance line occurred. It seems reasonable that the g value does not change with temperature, because the very small variation in lattice configuration by heating is not expected to alter the contribution of the orbital motion to the g value.

Various causes which contribute to the line width can be listed in the following way:

(a) The dipolar interaction between the spins has been discussed by Van Vleck.[15] At the absolute zero the contribution to the second moment from the magnetic interaction between neighboring spins vanishes. This may be understood directly from the physical fact that all spins are perfectly aligned and so the enrivonment of every spin is exactly the same. As the temperature is increased more and more spins turn antiparallel, causing a decrease in M_s and contributing to a dispersion of the local field values for the different spins. Finally, above the Curie temperature about as many spins are parallel as antiparallel and the second moment is then given by Van Vleck's formula for paramagnetic salts.[17] There should then be a continuous increase of the second moment from zero to the paramagnetic value, which is of the order of 2000 gauss. But the exchange forces, which enhance the fourth moment,[17] will narrow the line by a large factor, approximately given by $h\Delta\omega/J=10^4$, where J is the exchange energy and $\Delta\omega$ is the observed width. Therefore, the contribution to the effective width from the dipolar interaction should be less than 1 gauss even above the Curie temperature. No rigorous calculation of the fourth moment is available and the estimate is admittedly very rough. But in some organic paramagnetic salts, where the exchange integral is a hundred times smaller, line widths of only a few gauss have been reported.[18] Since, moreover, the temperature dependence of the dipolar broadening should be different, we can be certain that the observed widths must be explained in a different way. Akhieser[19] calculated the interaction within the spin system by considering collisions between spin waves. He gives a value of $T_2{}^*=10^{-7}$ sec. at 10°K. If we use his formula for the temperature dependence, we find $T_2=10^{-9}$ sec. at room temperature, corresponding roughly to 10 gauss. This is much too small, in agreement with our previous remark and Akhieser's observation that the spin-lattice interactions play a more important role at 300°K.

(b) Van Vleck introduced anisotropic quasi-dipolar interactions to explain the magnetic anisotropy in a cubic crystal like nickel. It has already been pointed out that the anisotropy energy will have an influence on the resonance condition. The shift in the resonance field is of the order of $2K_1/M_s$, where K_1 is the anisotropy constant decreasing rapidly with increasing temperature for nickel. In a polycrystalline sample of nickel we would expect the line to be broadened by about 40 gauss at 20°C and much less at higher temperatures. But apparently this effect is very much smaller than the observed widths in nickel. In supermalloy the anisotropy constant is still much smaller than in nickel.

Van Vleck points out, however, that the anisotropic spin interactions, in the presence of impurities, may give a broadening of the line. This contribution, dependent on the density of crystalline imperfections, would not vanish at the absolute zero.

(c) The effect of the finite skin depth on the line width can be estimated in different ways. Using the picture of Bloch waves, the microwave field will create a spin wave near the surface. The time it takes for the excited wave to travel one interatomic distance a is h/J, and to diffuse outside the skin depth $T_2{}^*\sim\delta^2h/Ja^2$. When we take $J/h=10^{13}$, $\delta=2\times10^{-5}$ cm, $a=4\times10^{-8}$ cm, this corresponds to a width of 0.1 gauss. The same order of magnitude is obtained if we put $c\nabla^2M=4\pi i\mu\omega M$ in Eq. (3) and go through the steps of the derivation of (10) with this term included. A term $\delta(y)\partial M/\partial y$ should be added to account for the discontinuity in $\partial M/\partial y$ at the boundary.

All causes discussed so far determined $T_2{}^*$. We shall now turn to the interaction of the spin system with the outside world, which determines T_1.

(d) The interaction of the spin waves with the lattice vibrations has been treated by Akhieser. He obtains $T_1=10^{-6}$ sec. at 10°K. Unfortunately his formula is not valid at higher temperatures and does not include the case in which an external constant field H_0 is present. We should have $T_1=1.5\times10^{-10}$ sec. at 300°K in order to explain the observed width in nickel. This does not seem unreasonable. But Polder[20] claims that Akhieser's theory yields too small relaxation times, because only spin waves with long wave-lengths should be taken into account for ferromagnetic resonance. The width in nickel changes approximately proportionally to the square of the absolute temperature at 300°K and more nearly directly proportionally to T at room temperature. But in supermalloy there seems to be a large temperature range with little variation in the width, which would be hard to explain with spin-lattice interaction.

(e) Finally, another mechanism of relaxation is suggested: the interaction of the ferromagnetic spins with the conduction electrons. No theory is available for the collision process of the spin waves with the conduction electrons. However, if we describe the process as one in which the magnetic quantum number m of the ferromagnetic spin system changes by unity and the corresponding magnetic energy quantum $h\nu_0$ is taken up by the conduction electron, we can obtain an estimate of the probability by using the formula which Heitler and Teller derived in their theory of nuclear magnetic

[17] J. H. Van Vleck, Phys. Rev. **74**, 1168 (1948).
[18] Holden, Kittel, Merritt, and Yager, Phys. Rev. **77**, 147 (1950).
[19] A. Akhieser, J. Phys. U.S.S.R. **10**, 217 (1946).
[20] D. Polder, Phil. Mag. **40**, 99 (1949).

relaxation in metals, and which has been verified experimentally.[21] One finds

$$1/2T_1=\frac{|V_{ij}|^2}{E_0^2}\frac{kT}{h}, \tag{23}$$

where E_0 is the Fermi energy and V_{ij} is the matrix element of interaction between a ferromagnetic and conduction electron, connecting states with $\Delta m=\pm 1$. This interaction consists of a magnetic dipolar term of the order of $\beta^2 a^{-3}\sim 1$ cm^{-1}, where β is the Bohr magneton and a the atomic radius, and an exchange interaction. Only if we take for the latter a large value so that $V_{ij}\approx 100$ cm^{-1}, would we find the right order of magnitude $T_1\sim 5\times 10^{-10}$ sec. to explain the observed width, which should vary proportionally to the absolute temperature.

V. CONCLUSION

The discussion of the preceding section may be summarized in the following conclusions. The increase in width, approximately as T^2, for temperatures higher than 300°C can be understood as a decrease in spin-lattice relaxation time. The absence of a discontinuity at the Curie point is in agreement with this explanation. Above that point, where the substance is paramagnetic, the causes of line broadening are well known and spin-lattice interactions alone can account for the observed width. Collisions with lattice phonons seem to be a more likely cause than collisions with conduction electrons, although a theoretical treatment for the latter process is wanting.

The very slight, if any, variation of the width in supermalloy below 350°C remains unexplained. The dipolar interaction and the finite skin depth exchange effect do not give the right order of magnitude. Only Van Vleck's suggestion of a combination of anisotropic forces and imperfections of the lattice remains a possibility. However, the variation of T_2 with frequency, approximately as $\omega^{-\frac{1}{2}}$ at room temperature, is not to be expected from any of the causes (a), (b), or (c), but could be reconciled with a spin-lattice mechanism.

There is no reason to expect perfect agreement of the *shape* of the experimental curve with theory. The shape given in Eqs. (10) and (11) is merely a consequence of the phenomenological introduction of damping terms in Eq. (1). The broadening by dipolar interaction alone would, for example, produce a Gaussian-like distribution, but the exchange interaction would make the intensity in the wings larger and produce more resemblance with the resonance curve of a damped harmonic oscillator. This curve one should expect for the spin-lattice relaxation mechanism and it is therefore gratifying that the experimental data fit this type of curve rather well. It is not warranted to assume the damping term $1/T_2$ to be proportional to H_0, if one uses the Bloch equation; $1/T_2$ is a function of the frequency and temperature only.

More experimental data are needed, first of all measurements on the width at low temperatures. A single crystal of nickel should be used to avoid dispersion of the resonance frequencies by increasing anisotropy. In the second place, experiments on insulators like the ferrites at low temperatures would be useful. The effect of the conduction electrons is then eliminated. Finally, saturation experiments at a high level of the microwave power will give definite information about T_1 separately, so that the spin-lattice theories may be tested directly.

The author is greatly indebted to Professor Van Vleck and Dr. Kittel for illuminating discussions on the theoretical aspects, to Mr. Yager for discussion and communication of his experimental results before publication, and to Mr. Cooke and the staff of the Electronics Research Laboratory for advice and aid in microwave techniques.

[21] N. Bloembergen, Physica **15**, 588 (1949).