Microwave Resonance Absorption in Gadolinium Metal*

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The microwave resonance absorption in gadolinium metal has been measured at 9000 and 24 000 Mc/sec. The g value in the paramagnetic region is found to be 1.95 ± 0.03. In the ferromagnetic region extreme line breadth prevents accurate determination of the g value. Calculations neglecting effects of line breadth give a ferromagnetic g value of 1.94.

I. INTRODUCTION

MICROWAVE resonance absorption in gadolinium metal has been measured in the 9000 and 24 000 Mc/sec ranges. This investigation was prompted by interesting properties of gadolinium metal, as emphasized by Van Vleck.[1] Gd is known to exhibit a saturation magnetization[2] corresponding closely to the value expected for an ^8S state, where the magnetic moment of each atom is due to the seven electrons in the $4f$ shell. It is therefore likely that Gd is a good representative of the atomic model of a ferromagnet, in great contrast to other ferromagnetic metals, all of which show saturation magnetizations which cannot be explained on such a simple basis. From this point of view, we wished to determine how closely the g value corresponds to the value for free electron spin, as expected for a half-filled shell. A further useful characteristic of Gd metal is that its ferromagnetic Curie point is 16°C.[2] This allows measurements to be made conveniently in both ferromagnetic and paramagnetic regions.

In the paramagnetic region we find that the g value is 1.95 ± 0.03. In the ferromagnetic region the extreme breadth of the absorption curve prevents accurate determination of the g value. However, we have made calculations of the ferromagnetic g value, ignoring possible shifts in peak positions arising from relaxation effects. These calculations lead to a ferromagnetic g value of 1.94. A limit of uncertainty cannot be assigned to this value because of the extreme and unknown breadth of the absorption curves, but it may be significant that the g value is not inconsistent with that in the paramagnetic range.

II. MEASUREMENTS

A circular disk 2.3 cm in diameter and 0.04 cm thick was used in these experiments. The sample had been annealed and contained no more than 0.1 percent of other rare earths. Absorption measurements were made by clamping the sample behind a hole of area 0.049 cm^2 drilled into the end wall of a resonant cavity. Thus the sample effectively forms a small part of this wall. Variations in the microwave power reflected from the cavity because of absorption in the sample were measured as a function of the externally applied magnetic field. The geometry was such that the external and microwave magnetic fields were in the plane of the sample and perpendicular to each other.

The measurements were made by sweeping the external magnetic field slowly through a suitable range while simultaneously measuring the microwave power reflected from the cavity. Data were obtained by placing a signal proportional to the magnetic field on the horizontal axis of a CRO and a signal proportional to the reflected power on the vertical axis. An electronic switch superimposed arbitrary field markers on the absorption pattern. Data were recorded by photographing the CRO traces.

Details of microwave and electronic circuits will not be presented here, since the techniques are relatively standard, but a few aspects of the apparatus which affect the accuracy of the results will be mentioned. The microwave power incident on the resonant cavity is amplitude—modulated at 6 kc/sec. This allows the use of a narrow band ac amplifier in the detection circuit. In the 9000 Mc/sec apparatus the 6 kc/sec modulation is imposed by the use of a microwave gyrator;[3] in the 24 000 Mc/sec apparatus modulation is imposed by a crystal placed in the microwave transmission line.

In all resonance effects there are both real and imaginary components in the absorption. The real part changes the Q of the resonant cavity and the imaginary part shifts its resonant frequency. In these experiments we measure only the real part of the absorption. Complete separation of the two aspects of magnetic absorption has been obtained by a slight modification of the technique suggested by Weidner and Whitmer.[4] The klystron is frequency modulated at a few cycles per second through a frequency range greater than any shift in the resonant frequency of the cavity. Thus at

* A brief account of the material presented in this paper will appear in *The Physical Review* under the authorship of R. Barton, A. F. Kip, C. Kittel, A. M. Portis, and F. H. Spedding.

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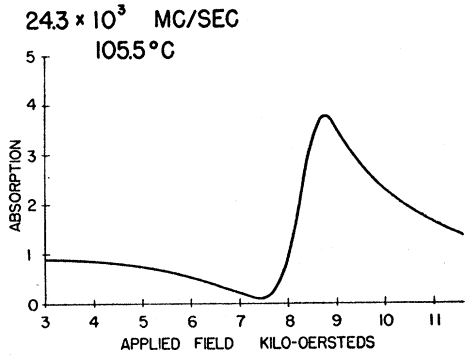

FIG. 1. Absorption curve of gadolinium metal.

some part of the FM cycle, the klystron frequency will be equal to the resonant frequency of the cavity. Since the reflection from the cavity is a minimum at resonance, it is possible to identify in the traces those points in each FM cycle for which the klystron is at the resonant frequency of the cavity. This method corresponds exactly to the "point-by-point" technique in which each point is obtained after retuning the klystron to the cavity resonant frequency.

The external magnetic field is measured by a technique similar to that used by Lamb and Retherford.[5] The emf from a rotating coil inserted in the external field is compared with the emf from a second rotating coil mounted on the same shaft. The second coil is in the field of a permanent magnet. Comparison is made in a bridge circuit in such a way that the two emf's may be made to cancel at any desired value of the applied field. By changing the bridge constants during the sweep of the external field, a series of field markers are superimposed on the trace. Absolute calibration of the field is obtained from proton resonance or from a small amount of diphenyl-trinitro phenyl hydrazyl[6] placed in the cavity. The field can be determined with an accuracy of ± 10 oersteds by this technique.

III. EXPERIMENTAL RESULTS

Figure 1 shows the experimental absorption curve of Gd taken at 105.5°C at a frequency of 24.3×10^3 Mc/sec. This temperature is in the paramagnetic region, well above the Curie point of 16°C. This curve exhibits the typical shape of resonance curves expected for metals, as found by Yager[7] and Bloembergen[8] in the case of Supermalloy and nickel. All curves taken in the paramagnetic range are similar, showing very little change in width for temperatures well above the Curie temperature.

Figure 2 summarizes the results at 24.3×10^3 Mc/sec for the positions of maximum absorption. The principal cause of the shift of the peak position in going from the paramagnetic to the ferromagnetic region is the increase in magnetization of the sample.

Figure 3 shows the variation in the width of the resonance curve as a function of temperature. The data plotted show the width of the absorption curve in oersteds, measured from the lower half-power point to the peak. The power absorbed is measured relative to the absorption of zero field. Below 0°C, in the ferromagnetic region half-widths are unobtainable because of the extreme breadth of the absorption curve.

No data are given for the 9000 Mc/sec experiments. The results at this frequency are consistent with the 24 000 Mc/sec results, but do not provide the accuracy available at the higher frequency.

IV. CALCULATIONS OF g VALUES

In order to determine the g values, it is necessary to compare our experimental curves with theoretical curves which take into account the various causes of shift in the absorption maxima. Causes of shift include demagnetizing factors due to the geometry of the sample, and relaxation effects. Furthermore, the "effective field" for the resonance condition[9] must be used. In addition, account must be taken of the effect of variation of eddy current losses in the sample, affecting the shape and position of the absorption curve.[7]

In obtaining our theoretical curves we used essentially the method of Yager, Galt, Merritt, and Wood.[10]

We assume an equation of motion of the form

$$\frac{d\mathbf{M}}{dt}=\gamma(\mathbf{M}\times\mathbf{H})-\frac{\gamma\alpha}{M}\mathbf{M}\times(\mathbf{M}\times\mathbf{H}). \qquad (1)$$

Here $\mathbf{M}$ = magnetization; $\mathbf{H}$ = internal field; $\gamma=ge/2mc$, the magnetomechanical ratio; and α = relaxation parameter.

From this equation one can calculate μ_1 and μ_2, the

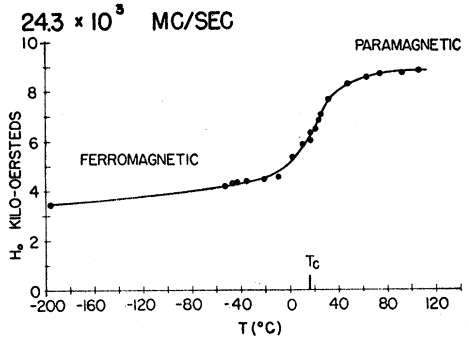

FIG. 2. Peak position of absorption curves *vs* temperature. Gadolinium metal.

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[9] C. Kittel, Phys. Rev. **73**, 155 (1948).
[10] Yager, Galt, Merritt, and Wood, Phys. Rev. **80**, 744 (1950).

real and imaginary parts of the permeability:

$$(\mu_1-1)/4\pi\chi_0=(\omega_0/\omega)^2\frac{(\omega_0/\omega)^2-1+4\delta^2/(1+\eta^2)}{[\omega_0/\omega)^2-1]^2+4(\omega_0/\omega)^2\delta^2},$$

$$\mu_2/4\pi\chi_0=[2\delta/(1+\eta^2)](\omega_0/\omega)\times\frac{1+(\omega_0/\omega)^2\eta^2}{[(\omega_0/\omega)^2-1]^2+4(\omega_0/\omega)^2\delta^2},$$

where

$$\omega_0=\frac{\eta}{\zeta}\gamma(1+\alpha^2)^{\frac{1}{2}}H_0,$$

$$\delta=\frac{\alpha}{(1+\alpha^2)^{\frac{1}{2}}}\frac{1+\eta^2}{2\eta},$$

$$\eta=(1+N_y\chi_0),$$

$$\zeta=(1+N_z\chi_0),$$

and ω=angular frequency of microwave source, H_0 =applied external field, N_z=demagnetizing factor in plane of sample, $N_y\cong 4\pi$=demagnetizing factor perpendicular to plane of sample, and χ_0=static susceptibility per unit volume.

Because of the effect of varying eddy current losses, and changes in skin depth, the quantity which we observe experimentally is $\mu_R{}^{\frac{1}{2}}$, where $\mu_R=(\mu_1{}^2+\mu_2{}^2)^{\frac{1}{2}}+\mu_2$. The theoretical curves were fitted to our experimental curves by finding the values of the relaxation parameter α which gave the best fit. This procedure is perhaps somewhat unjustified since it is not clear that all of the width of our curves is due to relaxation effects. However, this form of equation involving a relaxation parameter as given in Eq. (1), allows a good fit between experiment and theory, and presumably leads to correct g values.

Table I shows the result of these calculations at a number of temperatures in the paramagnetic region. The values of χ_0 at the various temperatures were obtained from data given by Trombe.[2]

As stated above, the large and unknown widths of the absorption curves in the ferromagnetic region preclude calculation of possible shifts due to line width. We have therefore obtained the ferromagnetic g factor by using the equation of Kittel[9] for calculating the "effective field." The g factor is then calculated from the relation $\omega=\gamma H_{\text{eff}}$. The value of the magnetization was obtained from unpublished data of Elliott, Legvold and Spedding.[11] The g value obtained at −196°C is 1.94.

TABLE I. Calculated g values of gadolinium metal in the paramagnetic region.

Temp. °C	Freq. Mc/sec	H_0 max oersteds	$4\pi\chi_0$	g
48.7	24 300	8270	0.196	1.96
64.0	24 290	8510	0.130	1.96
75.5	24 280	8690	0.103	1.95
93.7	24 280	8730	0.079	1.96
105.5	24 275	8850	0.065	1.95

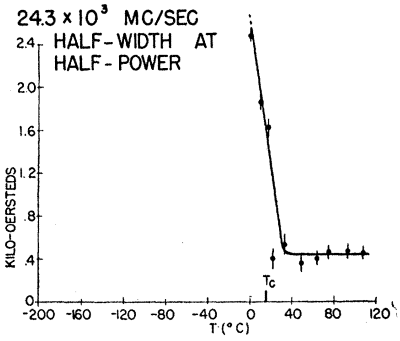

Fig. 3 Variation of absorption curve width with temperature.

V. SUMMARY

Resonance absorption curves in gadolinium taken over a wide temperature range show the expected shift due to the increase in magnetization as the sample is cooled from the paramagnetic to the ferromagnetic region. Only in the paramagnetic region can accurate g values be obtained; the extreme breadth of the absorption in the ferromagnetic region precludes accurate assignment of a g value below the Curie point. Our value, $g=1.95$, is to be compared with values available from other experiments. From the results of Trombe[2] on the value of the saturation magnetization, extrapolated to 0°K and infinite applied field, one obtains a g value of 2.03. This value may be in some doubt in view of the fact that extrapolation is necessary from 77°K to 0°K. Also the purity of Trombe's material is not accurately known. On the other hand, measurements similar to those of Trombe have been made on much purer gadolinium metal[11] down to 4°K. These measurements yield a g value of 1.96. Another value for g may be obtained from the determination of the Curie constant. Using Trombe's[2] data, we calculate $g=1.99$.

Although it seems that our value for g in the paramagnetic region is open to little question, it would be desirable to obtain and test a single crystal of gadolinium, in order to determine the g-value in the ferromagnetic region.

The author wishes to emphasize the fact that this was a cooperative project with very important contributions from R. Barton, C. Kittel, A. M. Portis, and F. H. Spedding. In addition, thanks are due to T. W. Griswold, who assisted in making many of the measurements. The research was assisted in part by the U. S. Office of Naval Research and the U. S. Atomic Energy Commission.

[11] Elliott, Legvold, and Spedding (unpublished data).