High field antiferro-, ferri-and paramagnetic resonance at millimeter wavelengths

S. Foner

To cite this version:
HIGH FIELD ANTIFERRO-, FERRI- AND PARAMAGNETIC RESONANCE AT MILLIMETER WAVELENGTHS (1)

By S. FONER (2),
Lincoln Laboratory, Massachusetts Institute of Technology, Lexington, U. S. A.

Abstract. — Pulsed magnetic fields have been employed to "tune" the high frequency magnetic interactions of antiferro-ferri- and paramagnetic systems to 4 mm and 8 mm wavelengths. Examples of resonance experiments for each of these magnetic systems are given, the nature of the information obtained is reviewed, and results of these experiments are summarized. Related magnetic measurements are also described; a detailed summary of susceptibility measurements for single crystal MnF₂, CoF₂ and Cr₂O₃ is given. Applications to high frequency devices are indicated.

Pulsed magnetic fields up to 750 kilogauss, produced by capacitor discharge through suitably designed coils [1], permit a variety of investigations [2], [3] which heretofore were not possible. In particular, magnetic resonance phenomena which involve large effective internal fields often occur in the high-frequency ranges not yet attained with present day technology. However, the high intensity pulsed fields may be used to "tune" the resonance to relatively low frequencies (35 to 70 kMcps) dictated by source power, pulse detection sensitivity and space limitations. In this paper, several examples of such resonances in antiferro-ferri- and paramagnetic materials are briefly described in order to demonstrate the scope and limitations of the technique. In order to interpret the resonance experiments additional detailed magnetic data are often required. A summary of these magnetic measurements is also presented. Because present information is most limited for antiferromagnets, more details of both the resonance and magnetic measurements are presented for this class of materials. Finally, some brief remarks concerning possible high-frequency applications of these materials and the pulsed field technique are also presented. The details of these various investigations are to be published elsewhere.

1. Antiferromagnetic Resonance. — The resonance condition for an uniaxial antiferromagnetic single crystal, derived by Kittel [4], Keffer and Kittel [5] and others [6] for \( H_\theta < (2H_EH_\Lambda)^{1/2} \) is given by

\[
\omega f_\gamma = \left[ 2H_EH_\Lambda + \left( \frac{\pi H_\theta}{\alpha} \right)^2 \right]^{1/2} \pm \left( 1 - \frac{\alpha}{2} \right) H_\theta
\]

where \( \omega \) is the angular frequency, \( \gamma \) is \( g\mu_B/2mc \), \( H_E \) is the exchange field, \( H_\Lambda \) is the anisotropy field, \( \alpha = \gamma H_\Lambda / H_E \) (obtained from independent magnetic data), and \( H_\theta \) is the applied field parallel to the easy axis. Estimates show that for most antiferromagnets \( (H_EH_\Lambda) \) is so large that low field observation requires one-to-one-tenth millimeter wavelength radiation. The high field mode can be observed at much lower wavelengths if \( H_\theta \) is sufficiently large [4], [5]. The present capacitor discharge system [1], (2 000 \( \mu \)F, 3 000 V) and coils permit measurements at 70 kMcps with fields up to 600 kilogauss at room temperature and up to 350 kilogauss at 4.2 K when a suitable Dewar assembly is inserted into the pulsed field coil. This range of fields permits a survey of many antiferromagnets. To date single-crystal MnF₂, Cr₂O₃, FeTiO₃, MnO, \( \alpha \)-Fe₂O₃, FeF₂, CoF₂, CoO, and NiO have been examined; the results for the first two are summarized below, the next three appear to be more complex and are still being investigated, and no resonance has yet been observed in the last four. The important quantity, \( H_\Lambda \), is obtained as a function of temperature from the resonance data — it is difficult to estimate or measure by other means. The normalized angular dependence of this resonance for \( \alpha = 0 \), obtained by numerical methods, and shown in figure 1, indicates that for low \( \omega f_\gamma \) accurate alignment of the crystal is neces-
sary for this resonance mode to be observed. A second resonance for $H_0 > (2H_n H_A)^{1/2}$, just after "spin-flop", should be observed when $H_0 = [2H_n H_A + (\omega / \gamma)]^{1/2}$. The angular dependence of this mode, shown in figure 2, indicates that for our parameters some good fortune is essential in order to observe this resonance. (This behavior is borne out by many experiments.)

The results for $\text{MnF}_2$ [7], [8], summarized in figure 3, demonstrated that the molecular field approximations and the dipolar source of $H_A$ predicted by Keffner [9] were satisfied in magnitude and in temperature dependence. Most recent results indicate that $(2H_n H_A)^{1/2}$ is closer to 92 kilogauss, but well within the experimental error. Since the various interactions in $\text{MnF}_2$ now appear to be well understood, this particular antiferromagnet warrants a detailed analysis. Line-widths as narrow as 250 to 500 gauss at 36 kMcps were observed at 4.2 °K. These measurements, corresponding to small differences measured near 80 kilogauss, are limited by various problems inherent with pulse techniques.

Examples of the resonance data obtained for $\text{Cr}_2\text{O}_3$, again with $H_0$ parallel to the easy axis, are shown in figure 4. A constant value of $(2H_n H_A)^{1/2} = 60$ kilogauss from 4.2 °K to 200 °K is calculated from this data if $g = 2.00$ is assumed. In contrast with the observations in $\text{MnF}_2$, the...
temperature dependence of $(2HEH_A)^{1/2}$ does not fit a Brillouin function for spin $3/2$. For Cr$_2$O$_3$, $H_F$ is about $2.1 \times 10^6$ gauss, (about four times that of MnF$_2$), $T_N = 308 \text{ oK}$ ($T_N = 68 \text{ oK}$ for MnF$_2$), and $H_A$ at low temperatures is 900 gauss ($H_A = 8.500$ gauss in MnF$_2$). Although the resonance data near $T_N$ agrees with Dayhoff's results [10], the extrapolated temperature dependence and absolute magnitude of $(2HEH_A)^{1/2}$ are quite different. The low temperature value of $H_A$ is about $1/3$ that estimated by Dayhoff and $1/100$ to $1/1000$ of earlier estimates [11]. Apparently the lattice distortions [12] which extend over twice the temperature range investigated by Dayhoff, are important [3]. This is one of several reasons which argue that low-frequency data near $T_N$ are not sufficient and that either high frequency or high field experiments are necessary. These results, as well as recent detailed magnetic data, indicate that Cr$_2$O$_3$ is not as simple as MnF$_2$. This is not surprising because crystal field interactions are expected to be important for the Cr$^{3+}$ ion, whereas for the Mn$^{2+}$ ion, theory and experiments indicate this effect is small.

Angular data taken at 4.2 oK and 77 oK for $\nu \approx 0.2$ qualitatively agree with the results in figure 1; an additional high field resonance appears for $\theta > 5^o$ and tends toward $k_\theta = 1$, and all resonances disappear for $\theta$ greater than about $12^o$. A broad very low field resonance was also observed for large $\theta$. This is not predicted by the simple theory, and is being investigated further.

2. Ferrimagnetic Resonance. — For the simplest case $(g_1 = g_2)$, the high frequency or "exchange" resonance for a two sublattice ferrimagnetic is given by [4]

$$\omega I_\gamma = \left[ \left( \frac{nH_B}{2} \right)^2 + (2 - n) H_B H_A + H_A^2 \right]^{1/2}$$

$$\pm \left( H_0 - \frac{nH_B}{2} \right),$$

where $M_1 + M_2 = nM_1$ and $H_B = \lambda M_1$. Here again $H_0$ can be used to "tune" the resonance to low frequencies, so that $H_B$ may be determined. In this case, $H_A$ and $n$ are determined from magnetic data. Even for our pulsed fields, the range of experiments is limited to materials of relatively small $H_B$ or small $n$. When $g_1 \neq g_2$ the usual resonance is given by $\omega = \frac{M_1 + M_2}{M_1 + M_2} H_0 (g_1 - g_2)$ and the "exchange" resonance is correspondingly more complicated. In particular, for large $H_0$, a large term proportional to $H_0 H_B (g_1 - g_2)$ must also be considered in [4].

Evaluation of the antiferro- and ferrimagnetic resonance data also requires accurate magnetic data over a wide temperature range. Detailed single-crystal susceptibility and magnetization data are easily obtained in a uniform magnetic field with the vibrating-sample magnetometer [18]. Since such data are of general interest, some of the results are summarized below.
As an example, the perpendicular susceptibility of a small single crystal of MnF$_2$, plotted on an expanded scale, is shown in figure 5. The results of such measurements on MnF$_2$ are: 1) $\chi_{\perp}$ varies by less than 1% from 4.2 °K to 50 °K; of the numerous spin-wave theories, Ziman's [19] predicts that $\chi_{\perp}$ should be independent of temperature; 2) if the break in $\chi_{\perp}$ or the $\chi_{\parallel}$ curve is used as a criterion, $T_N = 68$ °K, just at the specific heat anomaly; 3) data from 300 °K to 350 °K lead to $\chi_{\parallel} \propto T^{1.5}$, where as below 20 °K, $\chi_{\parallel} \propto T^{1.8}$. On the other hand, similar measurements on CoF$_2$ indicate $\chi_{\parallel}$ increases with increasing temperature, $\chi_{\parallel} \propto T^{1.5}$ above 15 °K, and $\chi \propto T^{3.8}$ below 15 °K, but does not follow a simple power law at lowest temperatures. Furthermore, $\chi_{\parallel}$ does not approach a zero value at low temperatures. Finally, the susceptibility measurements on Cr$_2$O$_3$ show that $\chi_{\perp}$ is constant to within 1% from 4.2 °K to 100 °K, and then increases with increasing temperature, and that $\chi_{\parallel}$ reaches a small constant value below 20 °K, and does not show a simple power dependence below 20 °K.

Recently, $^{19}$F nuclear resonance data [20] of various antiferromagnetic fluorides have been used to examine the temperature dependence of the sublattice magnetization, $M$. It should be realized that susceptibility measurements permit evaluation of $H_B$, do not depend on the presence of appropriate nuclear spins species, and may compare in accuracy to the resonance results. The low temperature $^{19}$F resonance data indicate that $\Delta M \propto T^{3.5}$ for MnF$_2$, and $\Delta M \propto T^{3.6}$ for CoF$_2$, where $\Delta M = M(0) - M(T)$. If $M$ varies as an appropriate Brillouin function, $\chi_{\parallel}$ should show a temperature dependence one power lower than given by the $M$ data. This is almost the case — the deviations and the particular temperature dependence must be explained by spin-wave theory. Magnetization measurements from 1.5 to 4.2 °K are being made to study this problem in greater detail.

Magnetization measurements of various single-crystal rare-earth iron garnets generally agreed with earlier polycrystalline data [21] in both magnitude and temperature dependence.

5. Applications to high frequency devices. — The three magnetic classes discussed above all have the general feature of very high frequency response because of large effective internal fields. Conversely, when millimeter and sub-millimeter wavelength sources become available, one would expect that these materials will be useful in practical devices. Since it is not economically feasible to produce continuous fields of the required magnitude, magnetic systems with the appropriate "built in" internal field will be required. Tuning could be accomplished by varying the crystal orientation, field $H_0$, and/or temperature. For example, one could make an antiferromagnetic millimeter wave modulator. One advantage of antiferromagnetic devices would be that demagnetizing field effects would be negligible.

It was pointed out earlier that suitable high frequency sources are not yet available. Recently we have been investigating the possibilities of generation and amplification of millimeter waves with a pulsed-field Maser [22]. One proposed scheme involves a three-energy-level paramagnetic system to which a pulsed field is applied in order to generate an output frequency much higher than the pumping frequency. Tests of this device are being made.

The author wishes to thank Dr. H. J. Zeiger for valuable discussions, Dr. H. H. Kolm for his valuable contributions to the development of the pulsed field system, Mr. B. Feldman for assistance with the experiments, and the numerous interested scientists [23] from various institutions, who so kindly furnished the single crystals essential for this work.

REFERENCES

DISCUSSION

Mr. Wolf. — I was privileged to see Dr. Foner's results for Cr₂O₃ a few days ago and I have made a rough estimate of the contribution of the crystal field effect to the anisotropy, using the measured D values for ruby. It seems that one would expect quite a large contribution from this cause, of the order of 1 300 oersteds at \( T = 0 \), and I think that this can explain qualitatively, and may be even quantitatively why his curve of \( k/2H_a \) was so different from a Brillouin function.

Mr. Kittel. — It would be interesting to see the details of this explanation.

Mr. Clogston. — By the use of nuclear resonance, Jaccarino has observed that the sub-lattice magnetization in various antiferromagnetic materials departs widely as a function of temperature from the standard Brillouin curve.

Mr. Foner. — If this is the case, the appropriate corrections must be made for the temperature dependence of the sublattice magnetization before \( H_a \) can be evaluated as a function of temperature, but \( K \) versus \( T \) does not depend directly on \( M \).

Mr. Nagamiya (Remark). — The quantity \( (2H_e H_a)^{1/2} \) for Cr₂O₃ deduced from Foner's curve for 36 GHz in figure 4 appears as shown in the accompanying figure. In deducing this, the measured values due to McGuire et al. (Phys. Rev., 102, 1000), were used. Since \( (2H_e H_a)^{1/2} \) is equal to \( (2AK)^{1/2} \), where \( A \) is the molecular field constant and is equal to \( 1/\chi_L \) and \( K \) is the anisotropy constant, one can deduce the value of \( K \) at absolute zero with the use of the same \( \chi_L \) and Foner's value of \( (2H_e H_a)^{1/2} \) at absolute zero : \( K = 4.45 \times 10^4 \) erg/gram. The dipolar anisotropy constant \( K_{dip} \) calculated by Tachiki and Nagamiya (Phys. Soc. Japan, 13, 452, 1958), is \( 9.28 \times 10^4 \) erg/gram, so one may take the crystalline field anisotropy constant to be

\[
K_{cryst} = -4.83 \times 10^4 \text{ erg/gram},
\]

which is one tenth of the value deduced from optical measurements in ruby and opposite in sign. Assuming these values of \( K_{dip} \) and \( K_{cryst} \), the temperature dependence of the quantity \( (2H_e H_a)^{1/2} \) was calculated and is shown also in the accompanying figure. The discrepancy between these two curves above 130 °K might be due to the inappropriate assumption of the molecular field in the calculated curve; short range order and crystalline deformation accompanying the growth of the antiferromagnetic ordering might play an important role here.