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FERROMAGNETIC RESONANCE IN Ni-Cd FERRITE SYSTEM AT MICROWAVE FREQUENCIES*

BY

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ABSTRACT

Ferromagnetic resonance experiments on spherical polycrystalline specimens of Ni-Cd ferrite system $Ni_{1-x}Cd_xFe_2O_4$ at frequencies of 9700 MC and 24000 MC were performed at room temperature.

The purpose of this paper is to give a brief description of the experimental method and to discuss the variation of the g-factor (g), the line width (ΔH), the damping constant (λ), and the relaxation time (T), as functions of composition. The results show that the values of the effective g-factor are from 2.15 to 2.00 and those of the internal field are from 260 oersteds to about zero, and it was also found that in the magnetic region $0 \leqslant x \leqslant 0.7$, $3.0 \times 10^{-10} < T < 10 \times 10^{-10}$ sec, $0.3 \times 10^8 < \lambda < 1.4 \times 10^8$ rad/sec, and $130 < \Delta H < 330$ oersteds at 9700 MC; and $1.9 \times 10^{-10} < T < 5.0 \times 10^{-10}$ sec, $0.20 \times 10^8 < \lambda < 1.1 \times 10^8$ rad/sec, and $230 < \Delta H < 600$ oersteds at 24000 MC.

1. Introduction

The phenomenon of ferromagnetic resonance was first observed by Griffiths (**1**) in 1946 and many theoretical and experimental investigations have since been performed on this subject. It has been much used as a powerful means for investigating the magnetic properties of magnetic materials. We can determine important ferromagnetic quantities such as the crystalline anisotropy constant, the spectroscopic splitting factor or g-factor, the line width of resonance, and the relaxation time by using the ferromagnetic resonance method.

The resonance experiments on polycrystalline binary ferrite specimens $Ni_{1-x}Cd_xFe_2O_4$ ($x=0, 0.1, 0.2, \cdots, 1.0$) were performed at frequencies of 9700 MC and 24000 MC at room temperature, and a discussion was made on the results of measurement concerning the g-factor (g), the damping constant (λ), the line width (ΔH), and the relaxation time (T) at each Cd-concentration.

The points which have important relations to the ferromagnteic resonance in polycrystalline specimens will be considered in the discussion. A new field of the

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solid state electronics up to microwave frequencies was opened by the development of the ferrites among materials with spinel structure and recently of ferromagnetic semiconductors with garnet structure which are transparent and have extremely narrow line width.

At present various commmercial ferrites are in most cases made of polycrystalline ferrites and it is very important in practice to investigate various electric and magnetic properties of polycrystalline ferrites and to compare the properties among them.

Particularly it is of much interest from the theoretical and practical points of view that the material is utilized at or near ferromagnetic resonance. We discuss the domain rotation in a sintered polycrystalline ferrite, whose principal differences from a crystalline are summed up in three respects as follows (**2**). First, sintered crystallites must be considered to be randomly oriented; second, they will interact magnetically with each other somewhat in the manner of coupled oscillators; and third, the magnetic properties of the polycrystalline are different from grain to grain according to chemical purity, shape, dimensions, porosity and internal stress. Owing to these complicated circumstances the theory of resonance phenomena in polycrystalline ferrite at microwave frequencies have not yet completely settled.

2. Theory of experiment

The power transmission coefficient T of the cavity is (**3, 4**):

$$T = \frac{4Q_L^2}{Q_1 Q_2} \cdot \frac{\left(\frac{1}{2Q_L}\right)^2}{\left(\frac{\omega-\omega_0}{\omega_0}\right)^2 - \left(\frac{1}{2Q_L}\right)^2}, \tag{1}$$

where Q_1, Q_2 and Q_L are the input, the output, and the loaded Q, respectively, ω is the angular frequency of the incident microwave, and ω_0 the resonant frequency of the cavity.

By definition,

$$\frac{1}{Q} = \frac{1}{Q_w} + \frac{1}{Q_1} + \frac{1}{Q_2} + \frac{1}{Q_d} + \frac{1}{Q_m}, \tag{2}$$

where Q_w is the intrinsic Q due to Joule's heat in the cavity wall, Q_d due to the dielectric loss in the cavity space and the sample, and Q_m concerning the magetic resonance absorption in the sample.

When the cavity is completely at resonance, the transmission coefficient T becomes

$$T = \frac{4Q_L^2}{Q_1 Q_2}. \tag{3}$$

We choose a reference point at very large magnetic field H_z where the loss consists of lossess other than that due to the resonance absorption, since the ferromagnetic resonance absorption cannot occur.

If $Q_{L(0)}$ is used to denote the loaded Q at the reference point and $Q_{L(H)}$ is the loaded Q at the resonance field, the magnetic absorption loss becomes, from Eq. (2),

$$\frac{1}{Q_m}=\frac{1}{Q_{L(H)}}-\frac{1}{Q_{L(0)}}=\frac{1}{Q_{L(0)}}\left[\left(\frac{Q_{L(0)}}{Q_{L(H)}}\right)-1\right],$$

and further by Eq. (3) it becomes

$$\frac{1}{Q_m}=\frac{1}{Q_{L(0)}}\left[\left(\frac{T_0}{T}\right)^{1/2}-1\right], \tag{4}$$

where T_0 and T denote the power transmission coefficient at the reference point and that at the resonance respectively.

When the crystal detector shows square-law character, the detection current I is proportional to the power transmission coefficient T. As $Q_{L(0)}$ is independent of the dc magnetic field, Eq. (4) becomes

$$\frac{1}{Q_m}=\text{const}\cdot\left[\left(\frac{I_0}{I}\right)^{1/2}-1\right], \tag{5}$$

where I_0 and I denote the detection current at the reference point and that at the resonance respectively. Consequently, we can determine the relative magnetic loss and the line shape at resonance by calculating $\left[\left(\frac{I_0}{I}\right)^{1/2}-1\right]$ as functions of the dc magnetic field.

In the above theory, we assume that the microwave power incident to the input window of the cavity is always constant, whether the resonance absorption exists or not. But we should be careful of the fact that the frequency and the output of the microwave oscillator are generally affected seriously by the state of the load.

This tendency is particularly remarkable at shorter wavelengths.

The first two attenuators in Fig. 1 are inserted in order to prevent the oscillator from the fluctuation due to the reaction from the load.

3. Experimental method

The 97000 MC and 24000 MC microwave apparatuses used for the measurenments are shown schematically in Fig. 1.

A small ferrite specimen is placed on the bottom wall of a rectangular transmission resonance cavity which is excited in the NE_{10n} mode, n being varied from 6 to 10. The resonance of the cavity is attained by driving the choke piston which composes the upper wall of the cavity by screw micrometer.

The signal oscillator consists of a low-power (~30 mW) reflex klystron, which is a 2K25 at 9700 MC and a 2K33 at 24000 MC.

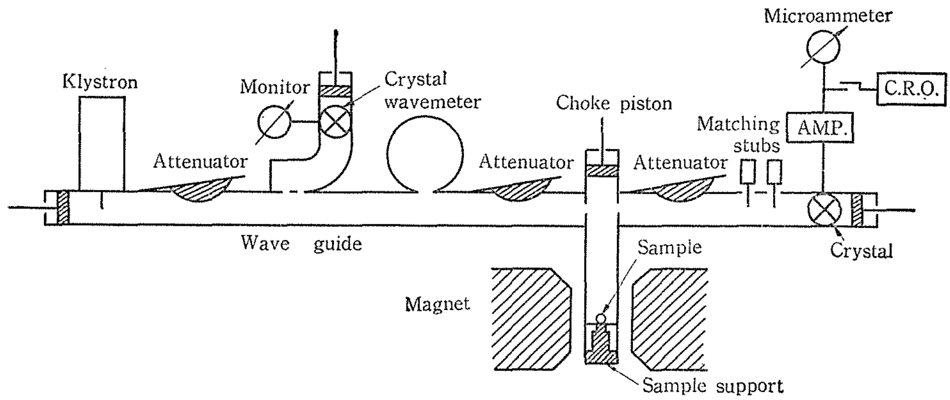

Fig. 1. Schematic diagram of microwave apparatus used at 9700MC and 24000MC.

The oscillator is amplitude modulated by a 6 KC rectangular wave and the output is always monitored by a precision wavemeter in order to keep its intensity and frequency constant.

All measurements are made, of course, with the cavity tuned to the resonance. The cavity is placed between the pole pieces of an electromagnet (10 cm dia.) in such a way that the rf magnetic field is orthogonal to the external dc magnetic field H_z.

The electromagnet used is one designed for investigating the nuclear magnetic resonance. The magnetic field at the sample position was measured by proton resonance method.

The field can be determined with an accuracy of 1% by this method.

A current of 10 amp. through the field coils provided a field of about 14000 oersteds. As shown in Fig. 1, we measured the variation in the signal transmitted through the cavity as the dc magnetic field was changed.

After detection, the signal was amplified by a narrow-band amplifier which was tuned to 6 KC modulation frequency.

Since the fluctuation of frequency and output of the oscillator limits the sensitivity and accuracy of experiment, we should carefully design the voltage regulated power supply to the klystron. In order to achieve greater stability and sensitivity of the present experiment, it is not sufficient only to stabilize the dc voltage supplied to the oscillator, but it is necessary that the very frequency of the oscillator is stabilized by the use of a feed-back circuit system, for example, Pound frequency stabilizer. The polycrystalline specimens made by a sintering and pressing process were prepared spherical by a method developed by Bond (**6**).

In order to obtain the result free from the size effect, the measurements should be done, though it is a tedious work, on very many samples of different diameters which were chosen between 0.3 mm and 1.5 mm.

When the sample diameter becomes larger than about 1 mm, several subsidiary absorption peaks generally appear on the true absorption curve (**7, 8**), due to the dimensional resonance and the resonance caused by the non-uniformity of rf magnetic field in the sample, making it difficult to analyze the resonance curve.

4. Results and discussion

Since the line width, g-value, relaxation time and damping constant of ferrite are in close relation with each other, they cannot be treated independently. However, we shall here discuss the experimental results, dividing them into the following four parts.

A) *The g-factor and the internal field*

The g-values in polycrystalline nickel ferrites have hitherto been found to be from 2.12 to 2.25. This fact shows the presence of spin-orbit coupling. In mixed ferrites the g-value depends remarkably upon composition and temperature.

Although the g-value should be independent of both the applied dc field and the frequency used, it shows, as is well known, conspicuous size effect and frequency dependence according to the results of microwave resonance experiments (**9, 10, 11**). The size effect is avoided by using the dc resonance field H_z which is determined by extrapolating the sample diameter to zero in the resonance experiments. However, the apparent g-factor, g_{app} calculated from Kittel's resonance formula (**12**) :

$$\omega_0 = \gamma H_z , \qquad (6)$$

for a spherical sample, shows marked frequency dependency, as seen in Fig. 3.

To avoid the discrepancy between curves (a) and (b) in the figure, T. Okamura (**13**) proposed the corrected resonance condition:

$$\omega_0 = \gamma (H_z + H_i) , \qquad (7)$$

where H_i is the ad hoc internal field which is a material constant independent of frequency, and H_z the same as in Eq. (6). Therefore, we can determine the effective g-factor (true g-factor) g_{eff} and the internal field H_i from Eq. (7) by the resonance experiments performed at two or more different frequencies. The experimental values of the internal field H_i and the effective g-factor of Ni-Cd ferrite system are given in Figs. 2 and 3 respectively.

The theoretical formula of the effective g-factor in ferrimagnetics was derived by Tuya (**14**) and Wangsness (**15**) and it is found to be in good agreement with expesiments (**10, 16**). The size effect has generally been considered to depend upon the dimensional resonance, the non-uniformity of the rf magnetic field in the sample, and the skin effect. The value of internal field H_i is about 260 oersteds at the composition of $x=0$ (nickel ferrite) and it gradually decreases with richer Cd content, becoming almost 0 in the non-magnetic region ($x > 0.7$).

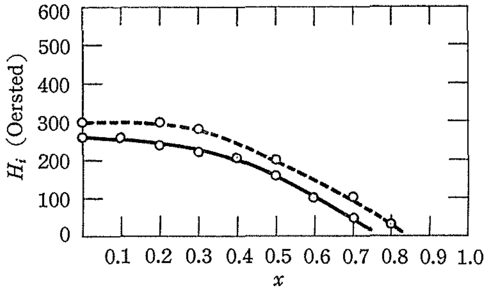

Fig. 2. Solid line: internal field in Ni-Cd ferrite system *versus* Cd content. Dotted line: that in Ni-Zn ferrite system *versus* Zn content.

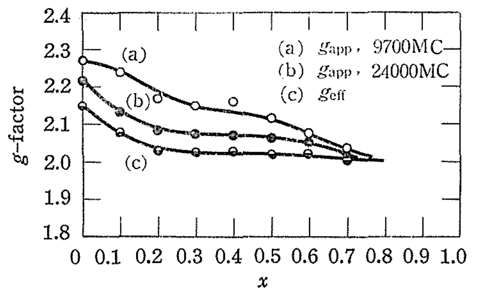

Fig. 3. Apparent g-factor, g_{app} and effective g-factor, g_{eff} of Ni-Cd ferrite system as functions of composition observed at 9700MC and 24000MC.

The dotted line in Fig. 2 shows the internal field in Ni-Zn binary ferrites as a function of Zn concentration. Though many theoretical and experimental investigations (**17**, **18**) have been carried out, the theory of mechanism of the internal field in ferrite has still been unsatisfactory because of the complicated circumstances in polycrystalline material. However, it has been found experimentally (**10**) that the internal field depends upon the porosity or density in the material and decreases with it.

B) *Relaxation time*

The rf magnetic field in the cavity at resonance decays exponentially with time according to exp $(-\omega_0 t/2Q)$, and consequently the magnetization changes in the same manner. Therefore, the relaxation time associated with the resonance is given by

$$T=\frac{2Q}{\omega_0}=\frac{2H_{res}}{\omega_0 \Delta H}, \qquad (9)$$

where H_{res} denotes the resonance field, Q is the Q-value of the cavity and ΔH the full width between half power points of the absorption curve.

The variation of the relaxation time as a function of Cd content is seen in Fig. 4.

The value of T is of the order of 10^{-10} sec, and is larger at 9700 MC than at 24000 MC, showing frequency dependence. The relaxation time of a single crystal of nickel ferrite is found to be 1.5×10^{-10} sec (**8**), a larger value than that of the present polycrystalline sample.

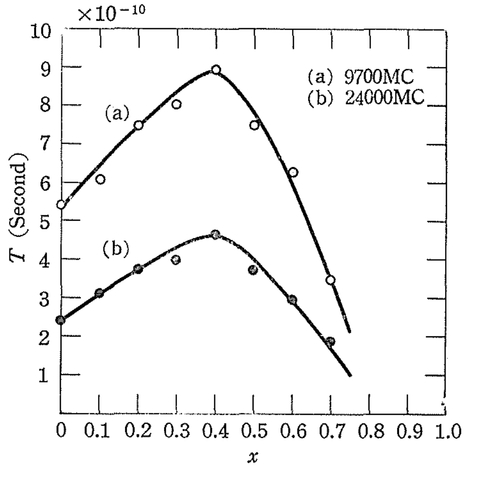

Fig. 4. Variation with composition of relaxation time of Ni-Cd ferrite system observed at 9700MC and 24000MC.

The reason for this seems to originate in the increase of the line width due to the increase of the anisotropy energy in polycrystalline specimen. The relaxation time T is maximum at $x=0.4$ and shows a remarkable reduction at $x=0.7$ at both frequencies, as shown in Fig. 4.

This fact is also considered to be due to the variation of line width with composition. The relaxation time T of commercial ferrite relates to the figure of merit F as $F=\omega T$ (**19**), which is an important quantity describing the quality of the microwave gyrator.

C) *Line width*

It is known that the line width of the resonance which has hitherto been measured is as broad as several hundreds oersteds and tends to a constant value at very low temperatures.

The physical mechanisms which can explain the two experimental results mentioned above are not well understood. As mentioned in the previous section, the true line width is superposed by the spurious line width owing to the dispersion of the resonance frequency in the individual crystal grain in polycrystalline specimen.

The latter broadening is expected to be about one fourth of the anisotropy field $H_a = \frac{2K_1}{M_s}$, provided that $H_a \ll H_{res}$.

The chemical impurities, vacancies, and lattice defects seem to contribute to the line width, but this effect will probably be small.

As the magnetic ions on the octahedral site are randomly distributed in ferrites with inverted spinel structure, the fluctuation of the magnetic interaction in the lattice as a whole occurs and is also effective to the line width. Eddy currents are rather unimportant to the line width in ferrites.

Fig. 5 shows the variation of the line width of Ni-Cd ferrite system as a function of composition.

It is of interest that the line width at 97000 MC is about two times larger than that at 24000 MC. It is experimentally known that the line width varies linearly with frequency.

Although many experimental and theoretical studies have been continued,

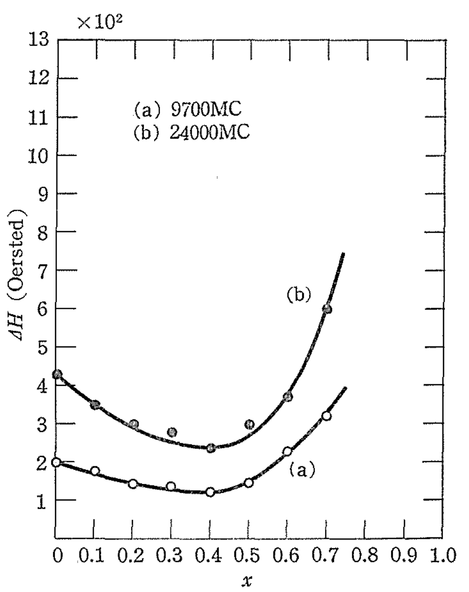

Fig. 5. Line width of Ni-Cd ferrite system as function of composition observed at 9700 MC and 24000 MC.

the microscopic mechanism of the line width of the resonance line in the ferrimagnetics such as ferrite is still not clear.

However, the line width theory developed by Clogston *et al.* (**20, 21**) will hereafter be a strong clue to the source of line width in ferrimagnetics.

D) *Damping constant*

To account for the line width acutually observed, it is necessary to correct the Kittel's equation of motion by adding a term representing dissipation of energy. The shape of resonance line can be estimated by this damping term which is determined by the line width.

Since the microscopic mechanism of line width has still been obscure, the damping term is also doubtful. Several damping terms (**22, 23**) have been introduced in a phenomenological way.

We used the equation of motion for the magnetization vector $\boldsymbol{M}$ of the ferromagnetic material in the form, first introduced by Landau-Lifshitz (**22**) :

$$\frac{d\boldsymbol{M}}{dt} = \gamma[\boldsymbol{M}\times\boldsymbol{H}] - \frac{\lambda}{\boldsymbol{M}^2}[\boldsymbol{M}\times(\boldsymbol{M}\times\boldsymbol{H})], \tag{10}$$

where $\boldsymbol{H}$ denotes the effective magnetic field at resonance, γ is the effective gyromagnetic ratio for the two-lattice system, and λ the damping constant responsible for the damping of magnetic energy, which has a dimension of frequency and is called the damping frequency or the damping constant.

The second term of the right-hand side of Eq. (10) is a vector expressing the damping and rotates the vector $\boldsymbol{M}$ toward the field vector $\boldsymbol{H}$ and leaves the magnitude of $\boldsymbol{M}$ a constant of the motion. Hence, Eq. (10) is valid only when the small amplitude of the rf field is used.

The other form of damping is the Bloch-Bloembergen form (**23, 24**) which has two characteristic parameters but does not conserve M. It has been shown experimentally that we should use the damping form of the Bloch-Boembergen type in the case using the microwave of high energy. The value of damping constant λ is determined by measuring the line width ΔH.

The relation between λ and ΔH is given by (8)

$$\lambda = \frac{\gamma M_s \Delta H}{2H_{\mathrm{res}}}, \tag{11}$$

where M_s denotes the saturation magnetization and H_{res} is the magnetic field required for resonance.

In the case of polycrystalline material λ in Eq. (11) is considered to express not the intrinsic damping constant but the effective damping constant which includes the spurious damping as mentioned above.

Fig. 6 gives the results of measurement of λ.

The value of λ seems to depend somewhat upon the frequency, taking larger value as the frequency decreases and is of the order of about 10^8.

It is remarkable in these expriments that first, the relaxation time T appears to be more dependent upon the frequency than the damping constant λ is; second, at the composition of $x=0.4$, the saturation magnetization M_s (**25**) and the relaxation time T have both maximum values and λ becomes minimum, and third, at $x=0.7$ the line width ΔH shows sharp increase and λ sharp decrease on the contrary.

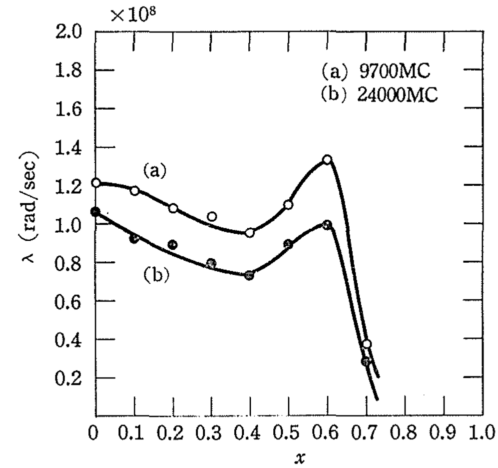

Fig. 6. L–L type damping constant as function of composition of Ni–Cd ferrite system observed at 9700MC and 24000 MC.

It is reasonable that at the composition of $x=0.4$, the dissipation of energy takes a minimum value which just coincides with the maximum relaxation time.

The increase of ΔH at $x=0.7$ seems to be mainly due to the decrease of exchange force and the increase of the anisotropy energy, and the decrease of λ at the same composition is due more to the large decrease of M_s than to the increase of ΔH.

On the other hand, the damping constant is determined by the experiment of domain wall motion (**26**).

It is difficult, however, to determine the damping constant of metals accurately owing to the effect of eddy currents.

The values of λ of various ferrites are listed in Table I below.

Table I. Measured values of damping constant λ of various ferrites.

Material	Frequency (MC)	λ (ferrom. res.)	λ (wall motion)
Fe_3O_4 (single crystal)	24000	9×10^8 rad/sec*	3.5×10^8 rad/sec*
$NiFe_2O_4$ (single crystal)	24000	2.1×10^7 " *	2.2×10^7 " *
	9000	7.2×10^7 " *	—
$NiFe_2O_4$ (polycryst.)	24000	1.0×10^8 " †	—
	9700	1.2×10^8 " †	—
$Ni_{0.5}Cd_{0.5}Fe_2O_4$ (polycryst.)	24000	0.90×10^8 " †	—
	9700	1.1×10^8 " †	—

* The values by J. K. Galt, J. Andrus, and H. G. Hopper, reference **23**.
† The values by the author.

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