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Temperature Dependence of the Line Width of Ferrimagnetic Resonance in Polycrystalline Nickel Cadmium Ferrite*

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竹本　將：多結晶ニッケル・カドミューム・フェライトの磁気共鳴に於ける半値巾の温度特性

ABSTRACT

Ferrimagnetic resonance experiments were carried out on polycrystalline nickel cadmium ferrite ($Ni_{0.7}Cd_{0.3}Fe_2O_4$) in the temperature range from −180°C to 100°C at 9500MC and 24000MC in order to investigate the temperature dependence of the line width ΔH and the g-value.

The writer used a standard electron spin resonace equipment at 9500MC, because of the high fidelity in measurements. As a result, it was clarified that the line width ΔH is affected to a great extent by both the temperature and the frequency, in such a way that it increases with decreasing temperature and with increasing frequency. The measured line width was 400 oersted at −180°C and 140 oersted at 100°C, showing a tendency to increase markedly with decreasing temperature below O°C. The g-value was 2.13 at −180°C and 2.03 at 100°C, gradually increasing with decreasing temperature. It was further known that the resonance field and the line width were remarkably affected by a slight deviation of the sample shape from sphericity.

1. Introduction.

The writer intended to investigate the electric and magnetic properties of the binary ferrite at microwave frequencies and to find some relations between them. With this end in view, the writer has hitherto measured the complex dielectric constant and the complex permeability[(1)] of the Ni-Cd ferrite system $Ni_{1-x}Cd_xFe_2O_4$ (x=0, 0.1, ……, 1.0) at room temperature at both 9700MC and 24000MC, and later the line width, the g-value and the damping constant[(2)] of the sample by the method of ferromagnetic resonance at the same temperature and frequencies. The present experiment which is the continuation of the above mentioned experiments was performed to determine the temperature dependence of the line width and the g-value of the sample.

Now, the causes of the additional line broadening and the theories of the line width of polycrystalline ferrite which have been considered so far are outlined as follows. As is well known, the ferromagnetic resonance observation plays an important role in determining the line width, the g-factor and the anisotropy constant, which are, as a rule, refered to the uniform mode.

However, unfortunately the mode of the ferrite is far from uniform, being complicated by various causes as described below.

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The complication is far more remarkable in polycrystals than in single crystals, and as is generally known, the polycrystalline material shows the broader line width than the single crystal. It was first pointed out by Van Vleck[3] and later by Kittel and Abraham[4] that the additional line broadening arises mainly from the crystalline anisotropy caused by the random orientations of the easy axes of crystallites in ferrite. Afterwards, it was made clear that the field (dc and rf) inhomogeneity gives rise to the additional line broadening of the absorption curve, that is to say, the spin waves of higher orders excited by the field inhomogeneity cause the additional absorption peaks which are superposed on the intrinsic line width. This problem was first treated by Clogston, Suhl, Walker and Anderson[5], and later by Geschwind and Clogston. Geschwind and Clogston[6] discussed this effect on the basis of the quantum mechanical spin wave theory at very low temperatures. They showed that exchange fields are not effective in narrowing the lines broadened by the field inhomogeneities in the sample but that the substantial narrowing is possibly produced by the mutual interaction of long range dipole fields. Therefore, the strength of the coupling is proportional to the saturation magnetization M_s. On the other hand, the anisotropy field H_a can be regarded as a measure to show the tendency of the grains to behave independently.

Recently Schlömann[7] discussed the ferromagnetic resonance phenomena in polycrystalline ferrite using the spin wave theory in two cases in which $M_s \gg H_a$ (strong coupling) and $M_s \ll H_a$ (weak coupling), and he confirmed that his theory agreed with the experimental results of a few special specimens.

Anderson and Suhl[8] have proposed a theory concerning the line width of the absorption curve of nickel ferrite on the basis of the spin wave idea and the randomness of divalent nickel ions and ferric ions in a sub-lattice of the inverse spinel structure.

This Anderson-Suhl's theory has recently been adopted as a theory of the line width of ferromagnetic oxides, but this theory is used seldom for the analysis of the observed line width because the spin wave theory is correct only at very low temperatures, that is, in the case of small oscillations of the spins. However, yttrium iron garnet[9] which has recently been discovered is one of the most interesting materials to which the spin wave theory is effectively applied and hence the spin wave theory is hoped to develop as a fundamental theory of the ferromagnetic relaxation and the line width of ferromagnetic oxides such as ferrite.

In addition to the above mentioned causes of the spurious line broadening, the line width of polycrystalline material is, as is generally known, remarkably affected by the dimensions[10] of the sample and by the concentration[11] of ferrous ions in it.

Most of the phenomena of ferromagnetic resonance are at present fairly well understood in case of the single crystal of ferrite, but the line width of the absorption curve has not yet been satisfactorily explained except in a very few cases. One of the most important reasons for this seems to be as follows.

The mensurements of the temperature dependence of the line width of ferromagnetic resonance have not been preformed enough and hence the experimental conclusions have not yet been obtained. On the other hand, the polycrystalline materials are far more used for practical purposes than single crystals and it is strongly demanded to investigate the characteristics of the former.

2. Measurements.

The ferrimagnetic resonance experiment was performed on nickel cadmium ferrite $Ni_{0.7}Cd_{0.3}Fe_2O_4$ at the microwave frequencies of 9500MC and 24000MC. A standard electron spin resonance equipment was used in the experiment at 9500MC, but it was necessary that the scanning field was broadened to about 2000 oersted because of the large line width of ferromagnets. At 24000MC, however, an ordinary apparatus[(2)(12)] equiped with a transmission type cavity containing the sample was used and the signal from the cavity was detected, amplified and indicated by a micro-ammeter.

On the other hand, the writer adopted the following method in order to improve the sensitivity and the accuracy of measurements at 9500MC. The spherical sample prepared by the method developed by Bond[(13)] is located in a reflection cavity of magic T. The opposite arm is terminated with an adjustable absorption load. The essential components of the apparatus are shown in Fig. 1.

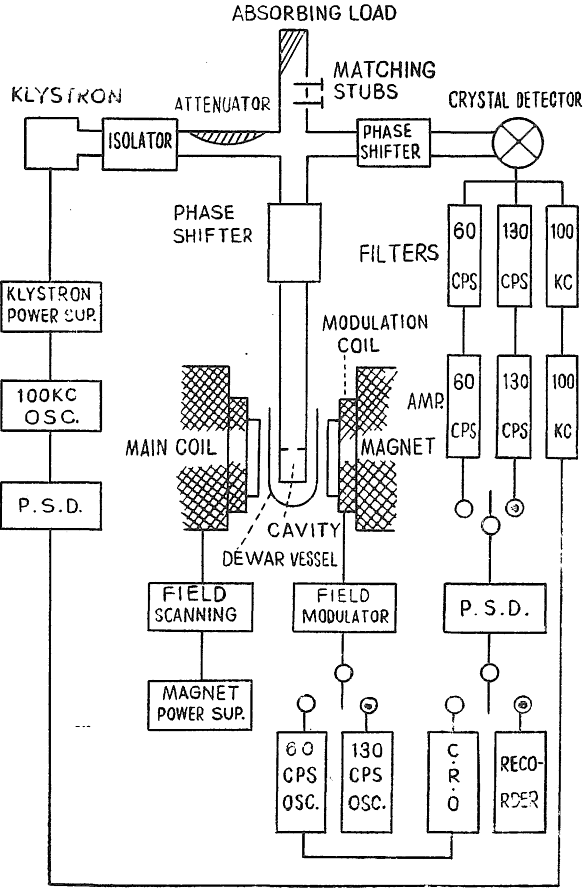

Fig. 1 Block diagram of the appratus at 9500 MC.

The applied dc magnetic field was modulated at 130 cps with an amplitude of 3.4 oersted. The 130 cps component in the output was detected by a phase-sensitive detector and recorded as a function of scanning magnetic field on a chart recorder.

A feed back system was employed to stabilize the frequency of the klystron from the effect caused by the absorption at resonance. The line width was taken as the distance between the points of maximum slope of the absorption curve.

After confirming that the abscissa on the chart was proportional to the current in the dc magnetic field coil, the dc magnetic fields were calibrated by observing the resonance absorption of a small sample of powdered DPPH simultaneously with the resonance absorption of a ferrite sample. It was further necessary that the sample diamter should be less than 0.30mm in order to sufficiently avoid the frequency shift of the klystron due to the absorption at resonance.

The cavity used was that of the cylindrical H_{111} mode and was inserted into a temperature controled Dewar vessel at low temperature measurements, liquid oxygen being used as low temperature agents.

We can study the accurate structure of the absorption curve accompanied by subsidiary peaks. Any subsidiary peaks could not be found in the range of the dc magnetic field swept by scanning field.

3. Preparation of Sample.

The Ni-Cd ferrite $Ni_{0.7}Cd_{0.3}Fe_2O_4$ was prepared by the flowing process. The raw materials NiO, CdO and Fe_2O_3 of high purity were milled in steel ballmills for about 24 hours, and the mixture of very fine particles obtained was dried and prefired in order to obtain a homogenous sample. After prefiring, the material was milled. Then, the powder was pressed in steel dies at the pressure of 1 ton/cm^2 into a shape. The final sintering was performed at about 1300°C in the air atomosphere, and the end-product was annealed slowly in the air. It was found in the present experiment using a ESR equipment, that the resonance field and the line width were markedly affected by a slight deviation of the sample from sphericity.

Not a few times, the differences of several tens oersted were observed in the magnitudes of both the resonance field and the line width according to the orientations of the sample (even in case it could be regarded as commonly sperical) in the dc magnetic field.

The spericity of sample has not been rigorously an issue so far, but it was confirmed that it is very important in the resonance experiment.

The sample used in the present experiment was selected, by using a microscope, out of scores of spherical samples which were prepared by the Bond method and the diameter 0.25mm was accurate to two significant figures.

4. Results and discussion.

A theoretically least understood property, though a most important, of the single crystal ferrite, not to speak of the polycrystalline ferrite, is the line width of the ferromagnetic resonance line.

This circumstance seems to be due to very few experiments being perfomed on the line width, especially on its temperature dependence, and consequently the definite experimental conclusions

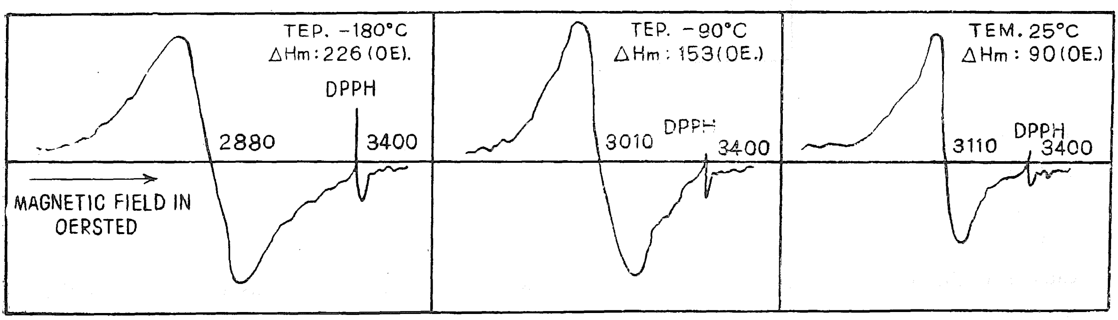

Fig. 2 Temperature variations of the derivatives of the resonance absorption curves in $Ni_{0.7}Cd_{0.3}Fe_2O_4$ at 9500 MC.

having not yet been obtained. The writer would be happy if the results of measurements described below should contribute to explaining the mechanism of the line width.

The derivatives of absorption curves at various temperatures are shown in Fig. 2.
These figures are tracings of the actual experimental records. The resonance field of absorption and the line shape were found to change strikingly as the temperature was lowered. It seems to be mainly due to the temperature dependence of the internal field H_i.

The resonance field of DPPH can be regarded as constant through the present experiments, since the g-value of DPPH is nearly constant in the temperature range from 77°K to 330°K. The full line width ΔH as a function of temperature at 9500MC and 24000MC is plotted in Fig. 3.

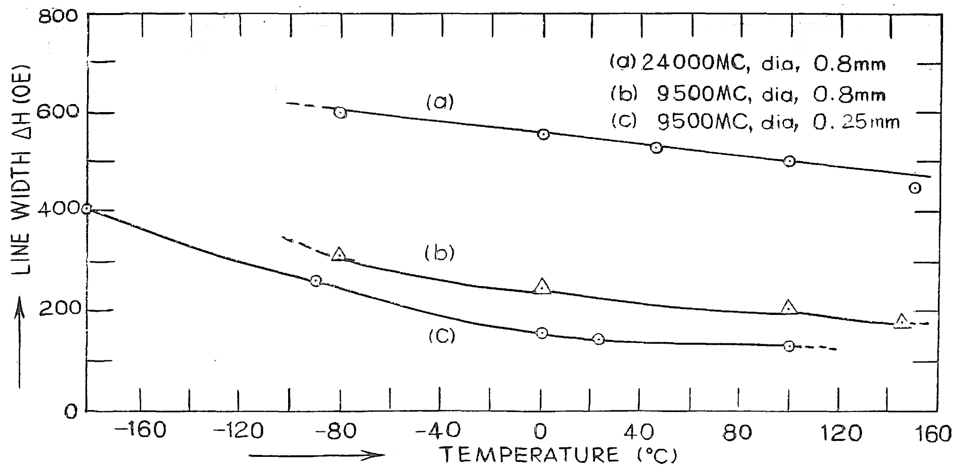

Fig. 3 Temperature dependedce of the line width ΔH of $Ni_{0.7}Cd_{0.3}Fe_2O_4$ at 9500 MC and 24000 MC.

The line width increases at lower temperatures in a similar manner to Healy's result(14) on nickel ferrite and to Tannenwald's result(15) on manganese ferrite.

This tendency seems to arise from the low conductivity(1) of the sample and the increase of the anisotropy field at low temperatures. The line broadening owing to the electron migration between Fe^{++} and Fe^{+++} is considered to be small due to the low conductivity of the sample.

According to the curve (c) in Fig. 3, the value of ΔH does not change remarkably in the temperature range from 0°C to 100°C and shows a tendency to increase markedly with decreasing temperature below 0°C.

The graphs also indicate that both the size effect and the frequency dependence are remarkable in the line width. The curves (a) and (b) in Fig. 3 were obtained by the absorption measurement using a conventional resonance apparatus described elsewhere.(2)(12)

As the obtained absorption curves are closely Lorentzian over the measured region, the line width ΔH was assumed to be equal to $\sqrt{3}\Delta H_m$, where ΔH_m means the line width taken between the points of maximum slope of the absorption curve.

The g-value of the sample varies from 2.13 at -180°C to 2.03 at 100°C and the internal field H_i takes values from 300 oersted to 200 oersted in the same temerature range.

It was also found that the internal field of the used ferrite varies with temperature in a similar fashon to the saturation magnetization. The g-value did practically not chage in the temperature region from 0°C to 100°C, but it gradually increased with decreasing temperature below 0°C.

The internal field correction[10] is particularly important in determining the g-values of polycrystalline specimens.

In polycrystalline ferrite, indeterminate internal field H_i seems to be a resultant of both the anisotropy field and the demagnetizing field which arises from the porosity and a special grain structure of ferrite material. The temperature dependence of various quantities observed in the ferromagnetic resonance experiment on Ni-Cd ferrite $Ni_{0.7}Cd_{0.3}Fe_2O_4$ is shown in the table, where H_z and H_{eff} are respectively the dc magnetic field and the effective field in the sample at resonance and g_{-app} means the apparent g-factor and g_{eff} the effective g-factor.

TABLE. Temperature dependence of various quantities of $Ni_{0.7}Cd_{0.3}Fe_2O_4$ at 9500MC.

TEMP.(°C)	H_z (OE)	H_{eff} (OE)	g_{app}	g_{eff}	ΔH_m (OE)	ΔH (OE)
−180	2880	3180	2.36	2.13	226	400
− 90	3010	3260	2.25	2.08	153	262
0	3080	3320	2.20	2.04	100	173
25	3110	3330	2.18	2.03	90	156
100	3130	3330	2.16	2.03	80	138

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