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## Ferromagnetic Resonance in Various Ferrites at Low and High Temperatures

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#### Synopsis

Ferromagnetic resonances in manganese, copper, and magnesium ferrites and nickelzinc binary ferrites were tried at low and high temperatures at the frequency of 9310 MC; in the temperature range of magnetic transition for manganese ferrite, double peaks were observed; with lowering of the temperature, a small peak at low field overlapped the ordinary large peak showing an asymmetrical resonance curve which tended to be symmetrical at the lowest temperature. g-factors and the half line widths were determined up to the Curie temperatures. In general, the line width of simple ferrites decreases monotonously to the Curie temperature, but those of binary ferrites containing zinc ferrite first decreases rapidly with rising temperature from the liquid nitrogen temperature and then increases near the Curie temperature; especially, it is most interesting to notice that the curve of line width versus temperature for high zinc ferrite in Zinc-Nickel ferrites shows minimum and maximum, the behaviour seems to correspond to a type predicted by Néel, which appears in the diagram for saturation magnetization versus temperature.

Moreover, we could determined g-factor of various Ni-Zn binary ferrites just at Curie point, by using paramagnetic resonance condition; the value of g-factor monotonously decreases with increasing content of zinc ferrite. The fact is quite in contradistinction to the change of g-value with varying zinc content in Zn-Ni ferrites at room and low temperatures.

The size effect reported by Beljers and Polder could not be observed for nickel ferrite and Ni-Zn binary ferrite at the frequency of 9310 Mc/sec., but recently it was observed at 23,500 Mc/sec. at room and low temperature on several ferrites.

#### I. Introduction

There have been many studies on the physical properties of spinel type oxide from the magnetic and the electric measurements, furthermore, by means of the ferromagnetic resonance experiment.

From these measurements, the magnetic transition of magnetite at a temperature of  $-160^{\circ}$ C has been studied, (1),(2),(3) and especially, from the experiment on X-ray and the changes of electrical conductivity accompanying the transition, Verwey<sup>(4)</sup> stressed that the anomaly of magnetite could be attributable to a rearrangement of ferric ion Fe<sup>+++</sup> and of ferrous ion Fe<sup>++</sup> in 16C site.

Recently, one of the present writers discovered the magnetic transition of nickel ferrite which occurred at  $-120^{\circ}$ C and seems to have been finished at a lower

<sup>(1)</sup> L. R. Rickford, Phys. Rev. Phys. Rev., 78 (1950), 449.

<sup>(2)</sup> C. A. Domenicalli, Phys. Rev. 78 (1950), 458.

<sup>(3)</sup> T. Okamura and Y. Torizuka, Rci. Rep. RITU, A-Vol. 3, No. 2 (1951), 215,

<sup>(4)</sup> E. J. W. Verwey and P. W. Haaymen, Physica, 8 (1941), 970.

temperature than  $-195^{\circ}$ C, for which the ferromagnetic resonance experiment<sup>(5)</sup> was carried out, and furthermore, the transition was investigated by measuring the magnetic property and electrical conductivity at a low temperature,<sup>(6)</sup> and the problem of applying Verwey model to the transition was discussed.

We found recently also magnetic anomaly of the same type of Mn-ferrite at a low temperature  $^{(7)}$  in the range of  $+20^{\circ}$ C to  $-150^{\circ}$ C, for which the resonance experiments were made at the wave length of 3.22 cm, and the results have been presented.

The microwave resonance experiment in ferrites at high temperature was first undertaken by the present writers<sup>(8)</sup> for cobalt ferrite and also in Co-Zn binary ferrite at a low temperature,<sup>(9)</sup> and many interesting results were obtained. We performed the same experiment on manganese ferrite, copper ferrite, magnesium ferrite and nickel-zinc ferrites, up to the Curie point, and the resonance field, g-factor and the half line widths were determined at various high and low temperatures.

#### II. Preparation of specimen

#### MnOFe<sub>2</sub>O<sub>3</sub>:

Manganese carbonate and Fe<sub>2</sub>O<sub>3</sub>, the latter being prepared by heating ferroussulphate at 800°C in the air, were mixed in the desired mol ratio and the mixed powder was heated at 1,200°C for three hours. The sintered block thus obtained was crushed to the size below 200 mesh.

The powder obtained was pressed at  $2\sim5$  ton/cm<sup>2</sup> in the cylindrical form, whose diameter was 1 cm and  $3\sim4$  mm in thickness. This compact was heated again in the air at 1,300°C for 3 hours and then quenched in the air. MgOFe<sub>2</sub>O<sub>3</sub>:

MgO powder was prepared by firing chemical pure magnesium powder and was mixed with  $Fe_2O_3$  and pressed at 2 ton/cm<sup>2</sup>. The compact obtained was sintered at 1,200°C for three hours and then quenched in the air.  $CuOFe_2O_3$ 

CuO powder was prepared by heating Cu  $(NO_3)_2$  at 800°C for 5 hours and was mixed with Fe<sub>2</sub>O<sub>3</sub> and pressed at 3 ton/cm<sup>2</sup>. The compact obtained was sintered at 1,050° for 3 hours and then quenched in the air.

#### Ni-Zn binary ferrite:

Nickel carbonate and ZnO powder prepared by firing pure zinc powder, were mixed with Fe<sub>2</sub>O<sub>3</sub> thoroughly in the desired mol ratio, and the mixed powder was pressed at 2 ton/cm<sup>2</sup> in the form of rectanguler plate; the compact was first

<sup>(5)</sup> T. Okamura, Nature, No 711, July 28 (1951), 162.

T. Okamura and Y. Torizuka, Sci. Rep. RITU, A-Vol. 3, No. 2 (1951), 219,

<sup>(6)</sup> T. Okamura and J. Simoisaka Sci. Rep. RITU, A-Vol. 3, No. 2 (1951), 223.

<sup>(7)</sup> T. Okamura and J. Simoizaka, Phys. Rev. 83 (1951), 664.

<sup>(8)</sup> T. Okamura, Y. Torizuka and Y. Kojima, Phys. Rev. to be published. Details to be published by Y. Torizuka, in Sci. Rep. RITU.

<sup>(9)</sup> Y. Torizuka, Sci. Rep. RITU, to be published.

sintered at 1,000°C for 3 hours. The sintered block was crushed to the size with the sieve of 200 mesh, and the powder obtained was again pressed at the pressure of 2 ton/cm² in a thin cylindrical form. Next, the cylindrical compact was sintered at 1,200°C for three hours and then quenched in the air.

From the sintered block thus obtained, thin disk specimens  $2 \text{ mm} \sim 3.5 \text{ mm}$  in diameter,  $0.2 \text{ mm} \sim 0.4 \text{ mm}$  in thickness were polished out, whose density always showed above 80%, in accordance with the calcuated value.

The dimensions of specimens used for the present experiment are given in Table 1.

				<b>É</b> z €
Mat	erial	Form	Thickness (mm)	Diameter (mm)
MnOFe <sub>2</sub> CuOFe <sub>2</sub> C MgOFe <sub>2</sub> C NiOFe <sub>2</sub> O Ni-Zn fe	${ m QFe_2O_3} { m disk} { m disk} { m disk}$		0.30 0.23 0.25	2.80 2.70 3.40 1.5, 2.0, 2.5. 1.5, 2.0, 2.5.
NiOFe <sub>2</sub> O <sub>3</sub>	ZnOFe <sub>2</sub> O <sub>3</sub> %			
100 80 60 40 20	0 20 40 60 80	disk disk disk disk disk	0.15 0.15 0.20 0.15 0.20	1.70 2.10 1.90 2.40 2.25

Table 1.

#### III. Resonance experiment on Mn-ferrite at low temperature\*

Experimental apparatus and the method were the same as for the magnetic resonance experiment on the antiferromagnetic materials; (10) the rectangular resonant cavity at low temperature made of transparent silica was used, at the bottom of which the specimen was fixed by means of drop of vacuum compound; the cavity containing the specimen was first cooled down at the liquid nitrogen temperature, and the relative absorptions with varying static magnetic field were observed, while warming it at the rate of 0.5°C per minute from the lowest temperature to higher temperatures.

Figs. 1, 2 and 3 show the curves of relative absorption with varying applied magnetic field from room temperature down to  $-190^{\circ}$ .

In the temperature range of  $+25^{\circ}$  to  $-70^{\circ}$ , double peaks were observed; with falling temperature, a small peak overlapped the ordinary large peak, which showed an asymmetrical resonance curve tending to be symmetrical as shown in Figs. 2 and 3.

From the result of magnetic measurement, it had previously been determined that the transition of the manganese ferrite would begin at  $+25^{\circ}$  and finish at  $-150^{\circ}$ , below which the intensity magnetization showed almost constant value. (7)

<sup>\*</sup> The result was reported in abridgment to Phys. Rev; Vol. 83, No. 4, 847~848, August 15, 1951.

<sup>(10)</sup> T. Okamura, Y. Torizuka' and Y. Kojima, Sci. Rep. RITU, A-Vol. 3, No. 2 (1951), 209.

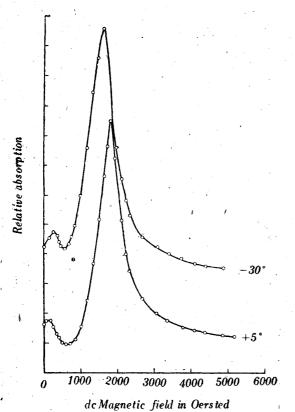


Fig. 1 Curves of absorption versus dc magnetic field for  $MnOFe_2O_3$ ; temperature range  $+5^{\circ}$  to  $-30^{\circ}$ .

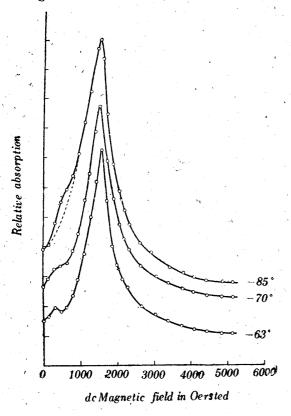


Fig. 2 Curves of absorption versus dc magnetic field for MnOFe<sub>2</sub>O<sub>8</sub>; temperature range  $-63^{\circ}$  to  $-85^{\circ}$ .

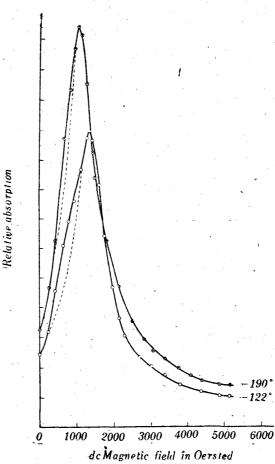


Fig. 3 Curves of absorption versus dc magnetic field for  $MnOFe_2O_3$ ; temperature range  $-122^\circ$  to  $-190^\circ$ .

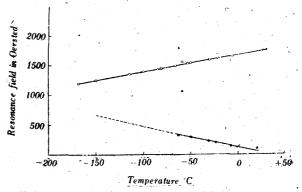


Fig. 4 Resonance fields for high and low peaks versus temperature.

To ascertain the behaviour of these double peaks more precisely, the resonance field strengths were plotted as a function of temperature as shown in Fig. 4; the open circles denote a resonance field for high field peak, the solid circles low field peak, respectively. From this figure, it can be concluded that the manner of the change of resonance fields with varying temperature is qualitatively the same in the case of nickel ferrite. Thus, the double peaks in MnOFe<sub>2</sub>O<sub>3</sub> may also be considered to be due to a mixed state of crystal that appears in the process of transition. The problem will be discussed in connection with the single crystal which has recently been prepared in our laboratory.

#### IV. Resonance experiment on Mn-ferrite at high temperature

The resonant cavity at high temperature was used, by which the resonance experiment on cobalt ferrite at high temperature was made. (11) Details for the apparatus and procedure were therefore, neglected here.

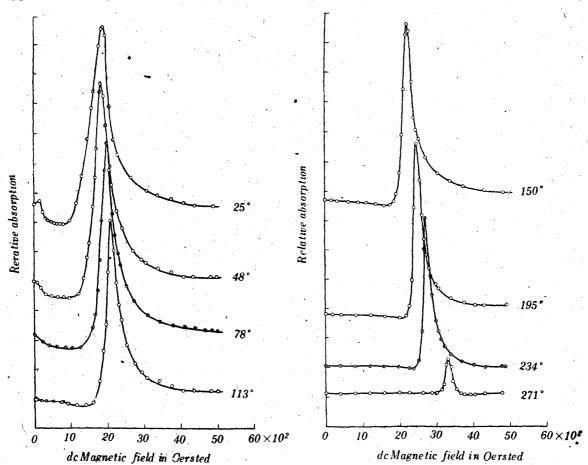


Fig. 5a Curves of absorption versus dc magnetic field for MnOFe<sub>2</sub>O<sub>2</sub>; temperature range 25° to 113°.

Fig. 5b Curves of absorption versus dc magnetic field for MnOFe<sub>2</sub>O<sub>3</sub>; temperature range 150° to 271°.

Figs. 5a, b show the curves of the relative absorption with varying dc magnetic field at various temperatures, from room temperature up to near the Curie temperature. As seen from these figures, the line shape becomes narrower, and

<sup>(11)</sup> Y. Torizuka, Sci. Rep. RITU, to be published.

the absorption amount decreases with rising temperature up to the vicinity of the Curie point, at which the resonance disappears.

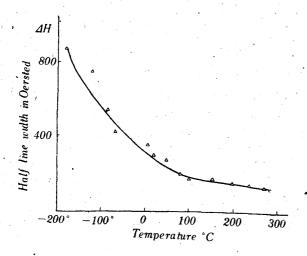


Table 2. MnOFe<sub>2</sub>O<sub>3</sub> Curie point: 303°C

Temp. °C	Line Width  ΔH (Oe)
-190	* 880
-122	760
-85	540
-70	420
-30	400
+5 $+25$ $+62$ $+100$ $+125$ $+192$	360 300 280 200 180 180
+250	160
+300	145

Fig. 6 The half width of the absorption line as a function of temperature for MnOFe<sub>2</sub>O<sub>8</sub>.

The line widths were determined at the half-maximum values of field for the resonance curves at high and low temperatures, and are shown in Fig. 6. In accompanying Table 2, the decreasing behaviour of the line width is always monotonous, but the amount different from the case of cobalt, because of the extremely large crystalline anisotropy.

Next, g-factor and the absorption amount were determined from the Fig. 5a, b at various temperatures by using the saturation magnetization obtained by

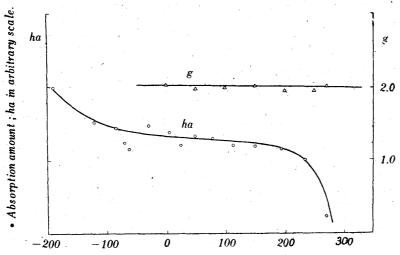


Fig. 7 The amount of the absorption at the maximum and the g-factor as functions of temperature; the open circles represent the absorption amount. the triangles represent g-factor.

Pauthonet; (12) these values are graphically shown in Fig. 7.

The g-factor of  $MnOFe_2O_3$ at room temperature was determined by many physicists with microwave resonance periments, and the magnitude of g-factor indicates that the magnetic moment of Mn-ferrite and of the whole series of Mn-Zn ferrite is due almost

entirely to the spin moments of the Fe<sup>+++</sup> and Mn<sup>++</sup> ions and that the contribution from the orbital moment is essentially zero in these materials.

<sup>(12)</sup> R. Pauthonet, Compt. Rend, 230, 1843 (1950).

In the present experiment, the magnitude of g-factor at room temperature was found to be 2.02 by assuming the density of the specimen to be 5.5. The value satisfactorily coincides with the uncorrected value 2.02 obtained by Yager and co-workers (13) at a frequency of 24,164 Mc/sec., but somewhat different from the value 2.05 as the corrected value for the size effect.

The value of g at various high temperatures are listed in Table 3; g=2.0 remains almost constant over the whole temperature range from room temperature to the Curie temperature, within the limit of error. Small discrepancy in the value of 2.0 at high temperatures seems to be due to the difficulty in determining Ms and the difference between our material and the one for which Ms was measured. Moreover, the g-factor at just about the Curie point  $303^{\circ}$ C was determined by

Table 3. MnOFe<sub>2</sub>O<sub>3</sub> Curie point: 303°C

Temp.	Ms (gauss)	g
0 50 100 156 200 250 275	430 395 350 300 240 160 90	2.02 1.97 1.99 2.01 1.96 1.94 1.93

using paramagnetic resonance condition for H=3480 and was found to be 1.97  $\pm$  0.03.

The change in absorption amount with varying temperatures shows the similar tendency to the change of saturation magnetization with temperature as shown in Fig. 7; these results are different from those in the case of cobalt ferrite. (11) The crystalline anisotropy of manganese ferrite, is very small in comparison with that of cobalt ferrite; therefore, the effect of magnetic anisotropy upon the resonance

condition is small and the amount of the intensity magnetization is only effective. The resonance fields at the absorption maximum at various temperatures were also plotted as a function of temperature and shown in Fig. 8; the resonance field increases linearly with rising temperature, and its amount es-

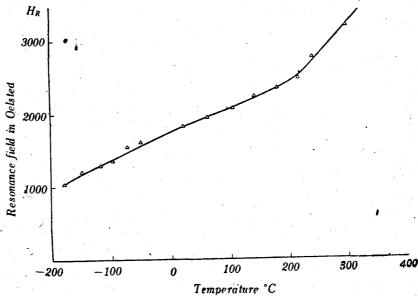


Fig. 8 The change of resonance field at the absorption maximum with varying temperature for MnOFe<sub>2</sub>O<sub>3</sub>.

pecially increases from 200°C to the Curie point. However, the amount of increase near the Curie point is much smaller than that in the case of cobalt ferrite.

<sup>(13)</sup> W. A. Yager, J. K. Galt, F. R. Merritt and F. A. Wood, Phys. Rev. 80, 744 (1950).

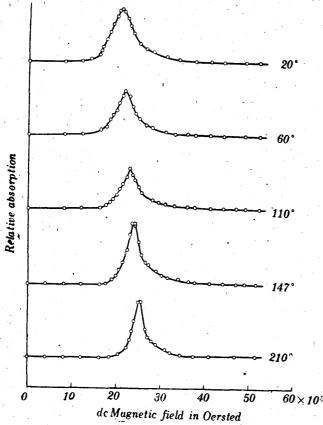


Fig. 9a Curves of absorption versus dc magnetic field for CuOFe<sub>2</sub>O<sub>3</sub>; temperature range 20° to 210°.

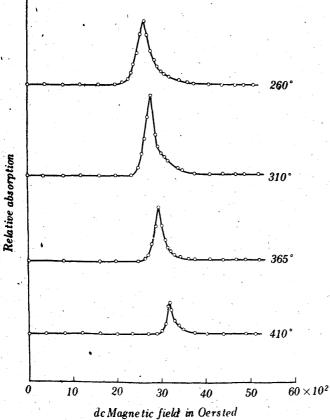


Fig. 9b Curves of absorption versus dc magnetic field for CuOFe<sub>2</sub>O<sub>3</sub>; temperature range 260° to 410°.

# V. Resonance experiment on copper and magnesium ferrites

The ferromagnetic resonances in copper and magnesium ferrites at various low and high temperatures have not yet been tried. Then, we carried out these experiments. Figs. 9 and 10 show the curve of the relative absorption with varying dc magnetic field at various temperature from room temperature to near the Curie temperature for Cu-and Mg-ferrite, respectively.

From these figures and other data at low temperatures, the half line widths were poltted as the functions of temperature as shown in Fig. 11, accompanying Tables 4 and 5 CuOFe<sub>2</sub>O<sub>3</sub> and MgOFe<sub>2</sub>O<sub>3</sub>, respectively; the line widths decreased from liquid nitrogen temperature nously with rising temperature up to near the Curie point as inthe case of Mn-ferrite described in the foregoing chapter, and the change of resonance fields with varying temperature creases slowly up to the Curie point as shown in Fig. 12. The tendency is almost the same in both materials.

Unfortunately, no values for the saturation magnetization at high temperatures have been published, except at room temperature.

Therefore, only the g-factors at room temperature can be discussed. g-facters for CuO-

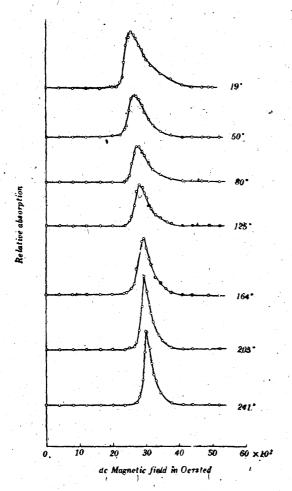


Fig. 10a Curves of absorption versus dc, magnetic field for MgOFe<sub>2</sub>O<sub>3</sub>; temperature range 19° to 241°.

Fe<sub>2</sub>O<sub>3</sub> and MgOFe<sub>2</sub>O<sub>3</sub> at room temperature were determined by us at the frequency of 9310 Mc/sec. on spherical polycrystalline specimen; the values obtained by us are 2.09 for CuOFe<sub>2</sub>O<sub>3</sub> and 2.04 for MgO-Fe<sub>2</sub>O<sub>3</sub>.<sup>(14)</sup> According to the results of Yager and co-workers at the frequency of 24,164Mc/sec., the value of MgOFe<sub>2</sub>O<sub>3</sub> is 2.03~2.06.

We determined the saturation magnetization Ms for CuOFe<sub>2</sub>O<sub>3</sub> and MgOFe<sub>2</sub>O<sub>3</sub> by

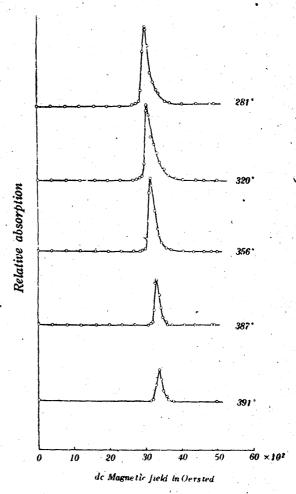


Fig. 10b Curves of absorption versus dc magnetic field for MgOFe<sub>2</sub>O<sub>3</sub>; temperature range 281° to 391°.

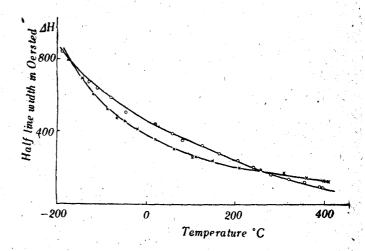


Fig. 11 The half widths of the absorption line as functions of temperature for CuOFe<sub>2</sub>O<sub>3</sub> and MgO-Fe<sub>2</sub>O<sub>3</sub>; the crosses represent CuOFe<sub>2</sub>O<sub>3</sub> and the open circles represent MgOFe<sub>2</sub>O<sub>3</sub>.

<sup>(14)</sup> T. Okamura and Y. Torizuka, Nature, Vol. 167, No. 4259 (1951), 936.

Table 4 CuOFe<sub>2</sub>O<sub>3</sub> Curie point: 430°C

emie ponit. 450 C						
, Temp. °C	Line Width  4H (Oe)					
$ \begin{array}{r} -176 \\ -145 \\ -122 \\ -99 \\ -87 \\ -65 \\ -50 \\ -20 \\ +20 \\ +62 \\ +102 \end{array} $	800 700 609 550 520 482 465 420 350 315 258					
$egin{array}{c} +110 \\ +147 \\ +210 \\ +260 \\ +310 \\ +362 \\ +400 \\ +410 \\ \hline \end{array}$	265 240 200 190 170 137 122 122					

Table 5 MgOFe<sub>2</sub>O<sub>3</sub> Curie point: 432°C

Temp.	Line Width  4H (Oe)
-190	840
-130	680
-110	640
-82	588
-50	502
+16	435
+51	388
+79	338
+124	316
+164	270
+204	240
+242	200
+281	160
+320	157
+336	116
+390	98
+398	97

comparing them with the value of NiOFe<sub>2</sub>O<sub>3</sub> at room temperature, and found them to be 260 gauss and 140 gauss, respectively. These values agree satisfactrily

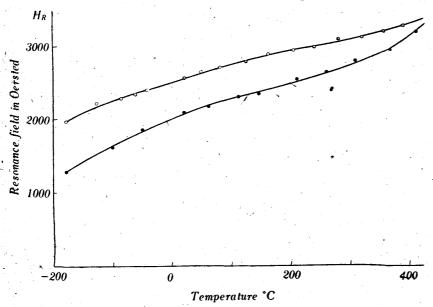


Fig. 12 Resonance fields at the absorption maximum as functions of temperature for CuOFe<sub>2</sub>O<sub>3</sub> and MgOFe<sub>2</sub>O<sub>3</sub>; the open circles represent MgOFe<sub>2</sub>O<sub>3</sub> and the solid circles represent CuOFe<sub>2</sub>O<sub>3</sub>.

with the values obtained by Gorter. (15) By using these values g-factors of CuOFe<sub>2</sub>O<sub>3</sub> and MgO- $Fe_2O_3$ could determined in the case of the present disk formed specimen and were found to be 2.08 and 2.04, respectively, as in the case of sphere formed specimen.(14) Thus, in the experiments at the frequency of 9310 Mc/sec.,

size effect seems to be out of question. In high temperature measurements, the absorption disappeared at the Curie point, taking small value of absorption amount at this temperature, because of

<sup>(15)</sup> E. W. Gorter, Nature, Vol. 165, No. 4203, 798 (1950). According to Groter's result, Ms for CuOFe<sub>2</sub>O<sub>3</sub> and MgOFe<sub>2</sub>O<sub>3</sub> is 250 gauss and 145 gauss, respectively.

the rapid decrease in magnetization, at which the resonance fields could be determined with the values of  $H=3450~\pm50~gauss$  and  $H=3330~\pm50~gauss$  for  $CuOFe_2O_3$  and  $MgOFe_2O_3$ ; g-factors were caluculated to be respectively  $1.93\pm0.03$  and  $2.00\pm0.03$ , by using paramagnetic resonance condition.

#### VI. Resonance experiment on binary ferrite: Ni-Zn ferrite

The ferromagnetic resonance experiments on the binary ferrite were made only at room temperature by Beljers and Polder in the case of Zn-Ni ferrites.

The present authors studied the various binary ferrites at various low temperatures by the resonance experiment, for which the g-factors were determined at low temperatures. But the resonance experiments for binary ferrite at high temperatures have not yet been performed. Therefore, the present writers made the resonance experiment on Ni-Zn binary ferrites in the temperature range of  $-190^{\circ}$  to the Curie temperature.

From the experimental results at room temperature, the g-factor and saturation moment of Ni-ferrite and Ni-Zn ferrites are both considerably greater than the spin only values that the orbital moment contribution is important in these ferrite.

Up to the present, g-factor of Ni-ferrite at room temperature was determined by Beljers and Polder, (15) Yager and co-workes (13) and the present authors. (14), (16) The values are given as follows: 2.31, 2.21 and 2.26, respectively; Beljers and Polder and the present author made the experiment at the wavelength of 3.2 cm and

Yager and co-workers at the frequency of 24,164 'Mc/sec.

Beljers and Polder first pointed out the effect of sphere size on the g-factor at the wavelength of 3.2 cm in the case of Ni-Zn ferrites. But we cold not observe such a effect in using three kinds of sphere specimens, namely, 1.5mm, 2 mm and  $2.5 \sim 3 \text{ mm}$ , at the same frequency for nickel ferrite and Ni-Zn binary ferrites. The such size effect rather depends on the density of sintered specimen, and in order to

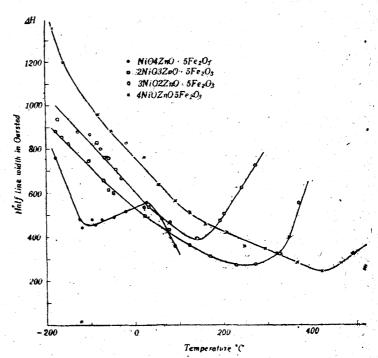


Fig. 13 The half widths of the absorption line as functions of temperature for Ni-Zn binary ferrites.

avoid the effect, we must prepare the specimen having the density above 80% for

<sup>(15)</sup> G. Beljers and D. Polder, Nature, Vol. 165, No. 4203, 800 (1950).

<sup>(16)</sup> T. Okamura, Y. Torizuka and Y. Kojima, Sci. Rep. RITU, A-Vol. 2, No. 4, 663-(1950).

the calculated value from the lattice constant. But recently we observed the size effect for g-factor on several ferrites at 23,500Mc/sec. at room and low temperature, the details will be seen in the following report.

From the experimental data, the half widths of line and the resonance fields for various Ni-Zn ferrites of NiO4ZnO5Fe<sub>2</sub>O<sub>3</sub>, 2NiO3ZnO5Fe<sub>2</sub>O<sub>3</sub>, 3NiO2ZnO5Fe<sub>2</sub>O<sub>3</sub>

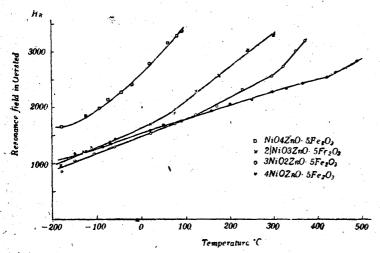


Fig. 14 Resonance fields at the absorption maximum as functions of temperature for Ni-Zn binary ferrites.

and 4NiOZnO5Fe<sub>2</sub>O<sub>3</sub> that have the Curie points at 120°C, 330°C, 395°C and 526°C, respectively, were determined in the temperature range of −190° to the Curie temperature, and are shown in Figs. 13 and 14, respectively, accomanying Tables 6 and 7.

The curve of the half line width versus temperature was diffrent from that of other simple

Table 6

1	nO5Fe <sub>2</sub> O <sub>3</sub> pint: 120°C		nO5Fe <sub>2</sub> O <sub>3</sub>	3NiO2ZnO5Fe <sub>2</sub> O <sub>3</sub> Curie point: 395°C		- II.	
Temp.	Line Width  ΔH (Oe)	Temp.	Line Width  ΔH (Oe)	Temp.	Line Width  4H (Oe)	Temp.	Line Width H (Oe)
-180 -125 -120 -97 -90 -75 -37 -20 +27 +60 +90	760 480 440 480 455 480 490 514 560 480 360	-175 -130 -102 -87 -80 -70 -62 -47 -32 +20 +30 +77 +140 +190	935 880 865 830 800 760 760 705 665 537 535 465 390 470	$\begin{array}{c c} -180 \\ -165 \\ -150 \\ -107 \\ -70 \\ -60 \\ -50 \\ +20 \\ +77 \\ +120 \\ +170 \\ +228 \\ +272 \\ +320 \\ -347 \end{array}$	880 855 820 745 655 615 600 495 435 367 312 272 273 320	-182 -160 -85 -50 +20 +50 +125 +157 +205 +247 +295 +330 +365	1360 1200 960 880 760 637 560 505 455 415 355 345 325 280
		+200 +240 +270	505 623 720	+347 +370	398 550	+420 +462 +490	240 280 320

ferrites found in the foreging chapters and from the experimental result of cobalt ferrites; that is in the case of Ni-ferrite containing low zinc ferrites, the line widths first decrease rapidly with rising temperature and then increase near 140°C, 275°C and 420°C for 2NiO3ZnO5Fe<sub>2</sub>O<sub>3</sub>, 3NiO2ZnO5Fe<sub>2</sub>O<sub>3</sub> and 4NiOZnO5Fe<sub>2</sub>O<sub>3</sub>, respectively; the amount of the increase is roughly proportional to the content of Zn-ferrite which is added to the Ni-ferrite. But the high zinc ferrite, namely, NiO4ZnO5Fe<sub>2</sub>O<sub>3</sub> ferrite, shows a different curve from the former; the curve of the

Table 7

	O5Fe <sub>2</sub> O <sub>3</sub> int: 120°C	1	nO5Fe <sub>2</sub> O <sub>3</sub> pint : 330°	3NiO2ZnO5Fe <sub>2</sub> O <sub>3</sub> Curie point: 395°C		4NiOZnO5Fe <sub>2</sub> O <sub>3</sub> Curie point: 526	
Temp. ℃	<i>H</i> <sub>R</sub> (Oe)	Temp. °C	H <sub>R</sub> (Oe)	Temp. °C	<i>H<sub>R</sub></i> (0e)	Temp. ℃	<i>H</i> <sub>R</sub> (0e)
-180 -125 -95 -85 -45 +5 +45 +60 +82 +93	1660 1860 1980 2140 2280 2420 2790 3160 3290 •	-180 -130 -112 -70 +20 +75 +123 +177 +242 +300	970 1210 1290 1380 1700 1900 2280 2560 3020 3290	-180 -150 -107 -60 +20 +76 +122 +170 +228 +295 +320 +350 +369	• 850 1050 1180 1290 1550 1700 1870 2005 2320 2560 2720 3005 3190	$\begin{array}{c} -180 \\ -150 \\ -125 \\ -85 \\ -60 \\ +20 \\ +50 \\ +90 \\ +122 \\ +158 \\ +200 \\ +245 \\ +295 \\ +328 \\ +365 \\ +420 \\ +440 \\ +488 \\ \end{array}$	850 1180 1200 1310 1430 1590 1690 1770 1870 1940 2060 2140 2280 2340 2420 2540 2640 2830

half line width with rising temperature has a minimum and maximum at ca.  $-100^{\circ}$ C and  $+20^{\circ}$ C, respectively; the width first decreases rapidly with rising temperature to ca  $-100^{\circ}$ C and then increases, passing a maximum at about  $+20^{\circ}$ C, then decreases up to the Curie point.

The behaviour of change of line width versus temperature for high zinc ferrite seems to correspond to the V type which appears in the diagram for Ms against temperature proposed by Néel's theory, (17) but from direct static measurement on Ms at high temperatures, Guillaud and Roux (18) could not find such a tendency in the Zn-Ni binary ferrite.

On the other hand, the line broadening appearing near the Curie point must be due to the decreasing behaviour of the factor  $\frac{K}{Msa-Msb}$ , where K is anisotropy constant, Msa and Msb respectively denotes the saturation magnetization of two sub-lattices; with the rise in temperature from the lowest temperature, the initial rapid decrease in the line width is essentially due to the decrease of K, and afterwards the line width increases in some temperature region, where Msa and Msb are antiparallel in approximately equal amount. Thus, the line broadening near the Curie point seems to be due to the appearance of an antiferromagnetic state. In the higher zinc ferrite, after passing through the antiferromagnetic region from  $-100^{\circ}$ C to ca  $+25^{\circ}$ C, the direction of saturation magnetization may change from Msa to Msb at  $25^{\circ}$ C, and with further rise in temperature, Msa-Msb decreases and then vanishes at the Curie point.

Generally speaking, it should be noticed that the increase of the line width with rising temperature near Curie point for Ni-Zn ferrites can not be observed

<sup>(17)</sup> L. Néel, Ann. Physique, 3 (1948), 137.

<sup>(18)</sup> C. Guillaud and M. Roux, Compt. Rend., 229 (1949), 1133.

for other simple ferrites.

The resonance fields increase with the rise of temperature and its amounts also increase rapidly near the Curie temperature.

By using the values for Ms taken from the static magnetization experiment of Guillaud and Roux, the g-factors at high temperatures are given in Table 8.

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	ZnO5Fe, point: 5		3NiO2ZnO5Fe₂O₃ Curie point: 395°C		2NiO3ZnO5Fe <sub>2</sub> O <sub>3</sub> Curie point: 330°C			NiO4ZnO5Fe <sub>2</sub> O <sub>3</sub> Curic point: 120°C			
Temp. °C	Ms (gauss)	g	Temp.	Ms (gauss)	g	Temp.	Ms (gauss)	g	Temp.	Ms (gauss)	g
20 100 150 200 250 300 350 400 450 500	420 370 340 305 275 240 205 175 125 70	2.12- 2.06 2.05 2.01 2.02 2.31 2.02 2.01 2.03 2.05	20 100 150 200 250 300 350	455 400 350 305 245 • 190 110	2.17 2.09 2.05 2.00 2.00 2.50 1.91	20 100 150 200 250	405 230 220 160 60	2.07 2.00 1.97 2.01 2.00	20 50	190 110	1.86

In general, the value of g-factor seems to be less than the value 2, when Zn-ferrite is added to Ni-ferrite. But the value of g-factor first decreases at a moderate temperature, then remains constant in the limit of error.

Lastly, in case of these ferrites, the values of g-factor are also determined just at the Curie point where the absorption disappears, by using paramagnetic resonance condition, from Table 7.

Thus, we could confirm the g-factor of Ni-Zn binary ferrites just at the Curie point, depending on the content of zinc ferrite, and the values are shown in Table 9, but the value of g-factor monotonously decreases with increasing content of zinc ferrite. The fact is quite in contradistinction to the change of g-value with varying zinc content of nickel ferrite at room and low temperatures. (13)

Table 9

Specimen	H <sub>R</sub> (Oe)	Temp. (°C)	g-value
NiO4ZnO5Fe <sub>2</sub> O <sub>3</sub>	3600	120	1.86
2NiO3ZnO5Fe <sub>2</sub> O <sub>3</sub>	3450	330	1.94
3NiO2ZnO5Fe <sub>2</sub> O <sub>3</sub>	3400	395	1.97
4NiOZnO5Fe <sub>2</sub> O <sub>3</sub>	3000	526	2.20
NiOFe <sub>2</sub> O <sub>3</sub>	2950	590	2.25

#### Summary

- 1. In the range of magnetic transition of manganese ferrite at low temperature, ferromagnetic resonance experiment was made, and the double peaks on the resonance curve was always observed in the same temperature range.
- 2. The ferromagnetic resonance in manganese ferrite at high temperatures was tried up to the Curie temperature; the line width and g-factor were determined.

The line width decreased monotonously from low temperature to the Curie point and the value of g-factor showed 2.0 in these temperature ranges in the limit of error.

3. g-factors for copper and magnesium ferrites were determined at room temperature; the value of copper and magnesium ferrites was 2.08 and 2.04, respectively.

Moreover, the temperature dependency of the line width for the both ferrites was determined; monotonous decreases of the line width against temperature were also observed.

- 4. The size effect reported by Beljers and Polder could not be observed for nickel ferrite and Ni-Zn binary ferrite at least at the frequency of 9310 Mc/sec.
- 5. For Ni-Zn binary ferrite, the changes of line width with varying temperatures were observed from liquid nitrogen temperature up to the Curie temperature; the line width first decreased with rising temperature, but increased near the Curie temperature, and such tendency could not be observed in other simple ferrites.

Especially, it is most interesting to notice that the curve of line width versus temperature for high zinc-nickel ferrites show minimum and maximum; the behaviour seems to correspond to V-type predicted by Néel.

6. g-factors for Ni-Zn binary ferrites were determined from room temperature up to the Curie temperature; especially, at Curie temperaturs, the g-factors were determined by using paramagnetic resonance condition.

Recently resonance experiments on various ferrites were studied at a shorter wavelength of 1.27 cm, details of the experiments in which the size effect, temperature and frequency dependencies of g-factor are discussed will be seen in the following report.

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According to the recent report of Dr. D. Healy, (19) the g-values of nickel ferrite on the single crystal did not change with varying temperature, but the crystal structure does not seems to be cubic at the lowest temperature by his experimental results, so the g-value at the low temperature obtained by Healy who calculated it by Kittel's formula for the cubic structure can not be compared with our results. (5)

We wish to thank Dr. Healy for sending us his interesting report.

<sup>(19)</sup> D. W. Healy, Jr; Contract N 50RI-76, Task Order No. 1, NR-078-011 Cruft Laboratory Harvard University Cambridge Massachusetts Technical Roport No. 135, August 15, 1951.