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Origin of line broadening in Co-substituted NiZnCu ferrites

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A systematic variation in linewidth with Co concentration at the *X* band (9.38 GHz) was observed in polycrystalline Co-substituted NiZnCu ferrites. Also, the temperature dependence of the linewidth and complex permeability were measured for two different Co concentrations. The linewidth shows a minimum with Co concentration. The contribution of porosity, magnetic anisotropy, and eddy current to line broadening was calculated. The line broadening due to eddy current was negligible and the line broadening due to porosity and magnetic anisotropy explain well the variation of linewidth with Co concentration. The temperature at which the linewidth increases rapidly increases with Co concentration. This temperature is consistent with the temperature of the second maximum peak in the temperature dependence of permeability. Therefore, the rapid increase in linewidth with temperature is attributed to the rapid increase in magnetocrystalline anisotropy of divalent cobalt ions. [S0021-8979(00)28608-8]

I. INTRODUCTION

NiZnCu ferrites are used as low-temperature sintered materials for chip inductors at high frequencies up to a few GHz. Substitution of a small concentration of cobalt to host ferrites improves the high-frequency properties, such as magnetic losses and the quality factor, at the sacrifice of initial permeability, which is attributed to the local induced anisotropy by divalent cobalt ions.[1] Because of the very strong positive magnetocrystalline anisotropy of cobalt ions, weak negative anisotropy of host ferrites can be compensated to nearly zero with a small concentration of cobalt.[2] Both phenomena are due to a degenerate orbital ground state of a divalent cobalt ion when this ion is in a crystalline field of trigonal symmetry.[3]

A small concentration of cobalt ions causes line broadening by the same reason. Haas and Callen[4] suggested that line broadening is determined by a magnon–magnon scattering mechanism that is caused by the ion-to-ion variation in spin-orbit coupling. However, Teale[5] found that this magnon scattering by cobalt did not make a significant contribution to the linewidth. In 1970, Vrehen[6] suggested that nonuniformities in the anisotropy field lead to a coupling of uniform precession to spin waves, thus to a decrease in the relaxation time of uniform precession and to a line broadening in polycrystalline nickel–cobalt ferrites.

In our previous report,[7] the decrease of initial permeability with cobalt concentration in $(Ni_{0.2}Cu_{0.2}Zn_{0.6})_{1.02-x}Co_xFe_{1.98}O_4$ was attributed to local induced anisotropy by cobalt ions. In this study, the origin of line broadening in polycrystalline cobalt-substituted NiZnCu ferrites is discussed.

II. EXPERIMENT

Cobalt-substituted nickel–zinc–copper ferrites of the composition $(Ni_{0.2}Cu_{0.2}Zn_{0.6})_{1.02-x}Co_xFe_{1.98}O_4 (0 \leqslant x \leqslant 0.05)$ were prepared by the conventional mixed oxide method. Samples were sintered at 900 °C for 2 h in an air atmosphere. Ferromagnetic resonance (FMR) was recorded using an *X*-band (9.38 GHz) spectrometer (Bruker, model ER 301). Spherical samples with a diameter of 500 μm were prepared by air propelling the specimens around the walls of an abrasive cup. The surface of these spherical samples was polished with an abrasive paper down to 10 μm. Complex permeabilities were measured using an impedance analyzer (Hewlett Packard, model HP 4194A) on toroidal specimens. Electrical resistivity was measured at room temperature using the dc four-point-probe method on bar specimens. Porosity was measured by the Archimedes' method.

III. RESULTS

Linewidths and *g* factors at room temperature increase with cobalt concentration as shown in Fig. 1. The linewidth has a local minimum at $x=0.030$, but the linewidth and *g*

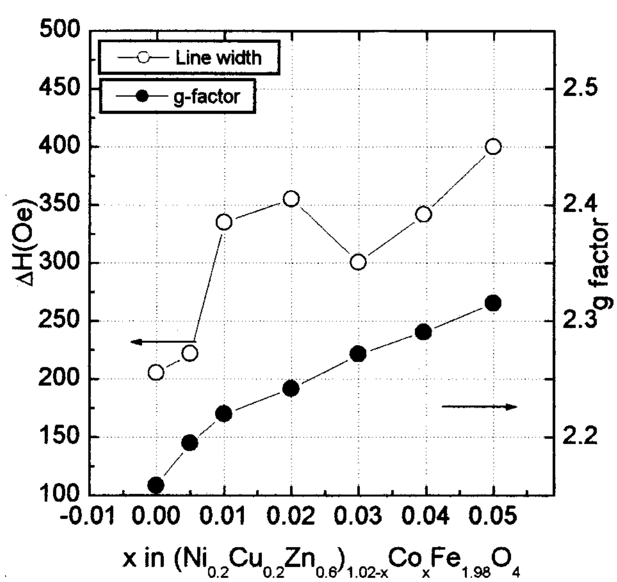

FIG. 1. Variation of linewidths with Co concentrations.

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TABLE I. Co concentration [x in $(Ni_{0.2}Cu_{0.2}Zn_{0.6})_{1.02-x}Co_xFe_{1.98}O_4$] dependence of porosity, saturation magnetization measured at 10 kOe, electrical resistivity at room temperature, and calculated magnetocrystalline anisotropy K_1 by the one-ion model.

Composition (x)	Porosity (%)	Saturation magnetization (G)	Electrical resistivity ($10^{10}\ \Omega$ cm)	K_1 (10^4 erg/cm^3)
0.000	4.7	3650	3.3	−2.7
0.005	4.7	3760	1.4	−0.8
0.010	6.3	3730	1.4	1.1
0.020	6.1	3830	5.0	4.9
0.030	4.7	4000	2.5	8.7
0.040	4.7	3950	2.4	12.5
0.050	5.6	4050	1.4	16.3

factor increase with cobalt concentration. Sirvitz and Saunders[8] and Vrehen[6] attributed a minimum in linewidths with cobalt concentration in polycrystalline nickel–cobalt ferrites to the compensation of magnetocrystalline anisotropy.

Magnetocrystalline anisotropies with cobalt concentration were calculated by the one-ion model.[3] The detail calculation procedure is shown in our previous work.[7] Calculation results at room temperature are summarized in Table I, but a minimum in anisotropy occurs at $x=0.005$. Saturation magnetization at 10 kOe, porosity, and electrical resistivity at room temperature are also summarized in Table I.

The temperature dependence of linewidths is plotted for the compositions of $x=0.000$, 0.005, and $x=0.030$ in Fig. 2. The linewidth of the sample of $x=0.000$, 0.005, and 0.030 is nearly constant between 150 and 250 K, between 210 and 330 K, and between 290 and 370 K, respectively. It is noteworthy that below these temperatures linewidths increase rapidly only for the cobalt-substituted compositions, $x=0.005$ and 0.030. Linewidths near the Curie temperature decrease because the specimens gradually lose their ferrimagnetic property.

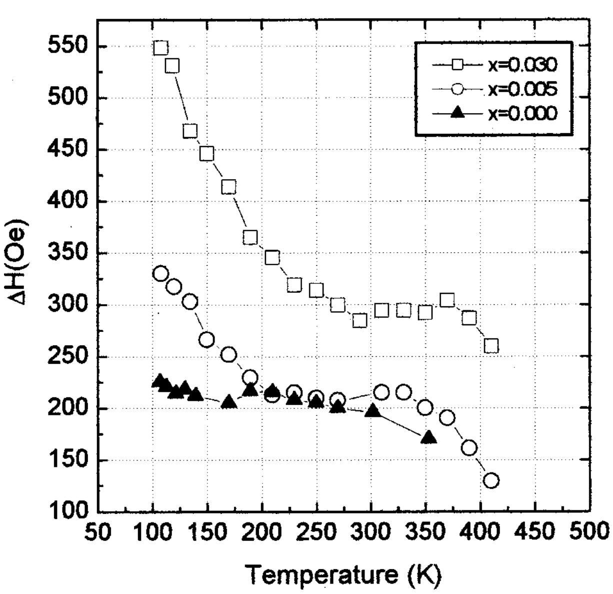

FIG. 2. Temperature dependence of linewidths for $(Ni_{0.2}Cu_{0.2}Zn_{0.6})_{1.02-x}Co_xFe_{1.98}O_4$.

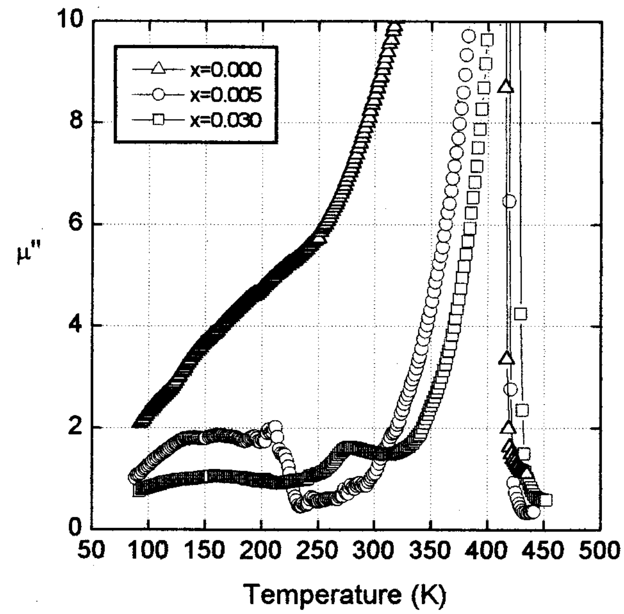

FIG. 3. Temperature dependence of real parts of complex permeability for $(Ni_{0.2}Cu_{0.2}Zn_{0.6})_{1.02-x}Co_xFe_{1.98}O_4$.

The temperature dependence of the real parts of complex permeability, that is, magnetic losses, is shown in Fig. 3. The second maximum losses are at 210 K for the composition of $x=0.005$, and at 290 K for $x=0.030$. There is only a slight bump near 150 K and no clear maximum for the composition of $x=0.000$. The second maximum in the imaginary parts of the complex permeability, the initial permeability that is not shown in Fig. 3, also occurs at the same temperatures. These second maximum peaks (SMP) are known to occur when magnetocrystalline anisotropy crosses zero with decreasing temperature.[2] In this case, the compensation of negative anisotropy of host ferrites is due to the positive anisotropy of cobalt ions. The temperature of SMP increases from 210 to 290 K as cobalt concentration (x) increases from 0.005 to 0.030.

IV. DISCUSSION

A. Variation of linewidth and *g* factor with cobalt concentration

In polycrystalline specimens, the total linewidth is a sum of various contributions of anisotropy, porosity, eddy current loss, and others. Therefore, the linewidth can be expressed as follows:[9]

$$\Delta H=\Delta H_{\mathrm{porosity}}+\Delta H_{\mathrm{anisotropy}}+\Delta H_{\mathrm{eddy}}+\Delta H_{\mathrm{others}}. \tag{1}$$

The linewidth depends on the size and surface roughness of the spherical specimens.[10] But variation of the linewidth with cobalt concentration in Fig. 1 is not due to these factors because the specimens are of the same size and have the same surface roughness. Line broadening by eddy current loss is negligible because the electrical resistivities of these compositions are of the order of magnitude $10^{10}\ \Omega$ cm. In this case, line broadening by eddy current loss is about 1×10^{-8} Oe.

Line broadening by porosity and by random orientation of the anisotropy axes in polycrystal can be expressed as[9]

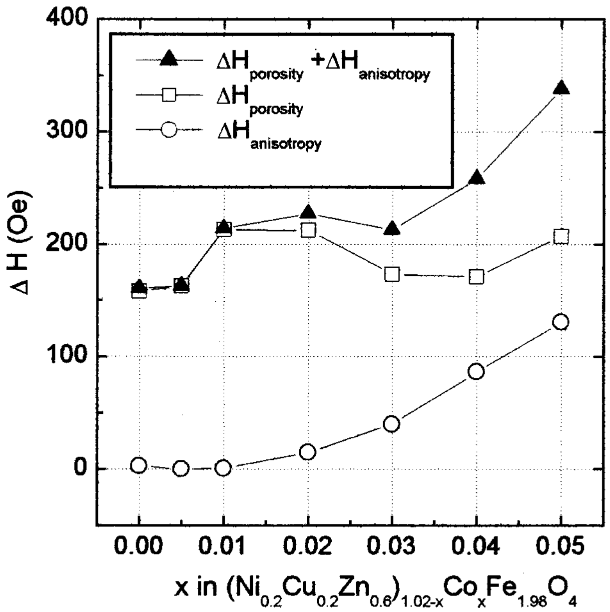

FIG. 4. Calculated linewidths due to anisotropy broadening and porosity broadening.

$$\Delta H_{\text{porosity}} = \frac{8\pi}{45} 4\pi M_s \frac{1}{\cos\theta_u} p(1-p), \tag{2}$$

$$\Delta H_{\text{anisotropy}} = \frac{H_a^2}{4\pi M_s}\left(\frac{8\pi\sqrt{3}}{21}\right)\left(\frac{\Omega^2 - 1/3\Omega + 19/360}{\sqrt{(\Omega-1/3)^3(\Omega-2/3)}}\right), \tag{3}$$

with $\Omega = \dfrac{\omega}{\gamma 4\pi M_s}$, $\quad H_a = \dfrac{2K_1}{M_s}$,

for $K_1 > 0$, $\quad H_a = \dfrac{4|K_1|}{3M_s}$,

for $K_1 < 0$,

where ω is the resonance frequency, $4\pi M_s$ is the saturation magnetization, p is the porosity, γ is the gyromagnetic ratio, H_a is the anisotropy field, K_1 is the first-order anisotropy constant, and θ_u is the polar angle of the magnon having the same energy as the uniform precession, in the limit as wave vector k approaches zero. For a spherical sample, $\cos\theta_u$ is approximately 1/3.

From Eqs. (2) and (3), $\Delta H_{\text{porosity}}$ and $\Delta H_{\text{anisotropy}}$ were calculated and are shown in Fig. 4. The maximum difference in porosity in the specimens is 1.6% and this corresponds to the variation in line broadening of 60 Oe. Line broadening by anisotropy is almost zero at $x=0.005$ because the magnetocrystalline anisotropy crosses zero at this composition.

The increase of the g factor with cobalt concentration indicates that the interaction of spin-orbital coupling increases with cobalt concentration due to the degenerate orbital ground state of cobalt ions.[11]

Though the sum of the calculated $\Delta H_{\text{porosity}}$ and $\Delta H_{\text{anisotropy}}$ is smaller than the measured value by about 50 Oe, the calculated variation of the linewidth with cobalt concentration is consistent with the measured values. This indicates that line broadening in polycrystalline ferrites can be explained well by porosity and anisotropy. The remaining 50 Oe may be attributed to ΔH_{others}, which includes the linewidth contributed by various relaxation mechanisms, surface roughness, and the size of the specimens.

B. Temperature dependence of linewidth

The valence exchange, slowly relaxing ions, or rapidly relaxing ions, can increase the linewidth with decreasing temperature.[9] The valence exchange relaxation is important in high-conductive ferrites, but in very low-conductive ferrites, like the samples in this work, the rapid increase in linewidth with decreasing temperature cannot be explained by this mechanism. A divalent cobalt ion is a candidate for rapidly relaxing ions due to its degenerate orbital ground state.[12] Teale[5,13] reported that rapid relaxation by cobalt ions occurs below 10 K but rapidly relaxing ion assumptions are not justified above 100 K, though the rapid increase in linewidth occurs below 200 K in cobalt-substituted ferrites.

The temperature at which the linewidth increases rapidly (Fig. 2) is consistent with the temperature of SMP (Fig. 3). Also, these temperatures increase with cobalt concentration. It is evident that the rapid increase in the linewidth is due to cobalt substitution. It should be noted that the magnetocrystalline anisotropy of divalent cobalt ions has a strong temperature dependence. Therefore, the contribution to line broadening by the anisotropy of divalent cobalt ions is dominant below the temperature of SMP, so the linewidth increases rapidly with decreasing temperature.

V. CONCLUSION

Line broadening in polycrystalline cobalt-substituted NiZnCu ferrites is well explained by porosity and anisotropy broadening theory. The rapid increase in the linewidth with decreasing temperature is attributed to the rapid increase in magnetocrystalline anisotropy due to divalent cobalt ions.

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