SIZE DEPENDENT FMR LINE-SHIFT AND LINE-WIDTH STUDIES IN POLYCRYSTALLINE FERRITES AND GARNETS

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Abstract. — In resonance and relaxation studies of polycrystalline ferrites and garnets, size dependent contributions arise from effects due to eddy current, inhomogeneous demagnetization and surface pits while size-independent contributions arise due to anisotropy and porosity. The experimentally observed size dependent contributions to line-shift and line-width have been satisfactorily explained. The existing theory for porosity contribution to ΔH in several cases gives results much higher than what is experimentally observed. On assuming that pores have a distribution of ellipsoidal shapes in the sample values of ΔH in agreement with experiments have been obtained. The theory for size dependent contribution to line-shift has been shown to apply also to Zn$_x$Fe$_{2.5}$O$_4$ system which has relatively high conductivity compared to other ferrites investigated so far.

1. Introduction. — In FMR studies of polycrystalline magnetic insulators contributions from a large number of sources to the internal field have to be taken into account. If the sample is located at the base of the cavity the resonance frequency $\omega$, and the line-width $\Delta \omega$ are given by

$$\omega = \gamma (H_{\text{ap}} + H_a + H_p + H_s + H_d)$$

$$\Delta \omega = \Delta H_1 + \Delta H_2 + \Delta H_3 + \Delta H_4 + \Delta H_{\text{id}} + \Delta H_{\text{pit}}$$

where the subscripts stand for the following : exp, experimental ; a, anisotropy ; p, porosity ; e, eddy current within the cavity wall ; id, inhomogeneous demagnetization ; i, intrinsic. We consider only the spherical shape which is often used in experiments.

The size dependent contributions arising from eddy current, inhomogeneous demagnetization and pit have been experimentally investigated by us in detail and shown to agree satisfactorily with our analysis [1, 2]. An estimate of $\Delta H_1$ obtained by subtracting ($\Delta H_e + \Delta H_p$) from the experimentally observed diameter independent contribution, $\Delta H_0$, yields unrealistic values in several cases. This has been shown [2] to arise owing to overestimation of the porosity contribution obtained using the analysis of Schlömann [3], Geschwind-Clogston [4] and Sparks [5]. Among these theories the contributions from the polycrystalline pores is considered on the spherical-cavity model. Assuming a distribution of ellipsoidal shapes for the pores, reasonable values of $\Delta H_p$ have been obtained in agreement with experiment.

If the sample has finite conductivity a contribution from it to the line-shift and line-width has to be added in eqs. (1) and (2). The contribution to line-shift is small even in case of metals [6, 7] and can be ignored. An approximate calculation of the line-width arising from the loss due to eddy current in the bulk of the sample has been made by Sparks for cylindrical sample geometry. A similar calculation for the spherical shape carried out by us yields

$$\Delta H_e = \frac{16}{15} \frac{\pi^2}{c^2} \sigma R^2 \omega^2$$

where $R$ is the radius of the sample and $\sigma$ is the conductivity. Our resonance measurements on Zn$_x$Fe$_{2.5}$O$_4$ are shown to be in broad agreement with these results.

2. Line broadening due to porosity. — We assume that N pores of different ellipsoidal shapes are present in a spherical sample. We also assume that one of their principal axes coincides with the magnetization of the sample. The $i$-th pore can be considered as a hole with magnetic moment, $-\mathbf{M}_i$, placed at the centre of the
cavity which interacts with the field produced by the poles on the surface of the pore. Hence the total free energy neglecting anisotropy, magnetostriction and exchange is

\[ F = -\mathbf{M} \cdot \mathbf{H}_0 - \frac{1}{2} \mathbf{M} \cdot \mathbf{N} \cdot \mathbf{M} - \frac{1}{2} \sum_{i=1}^{N} \frac{v_i}{V} \mathbf{M} \cdot kN_i \mathbf{M}_z \]

(4)

where \( \mathbf{N} \) is the demagnetization dyadic for the shape of the sample, \( N_i^z \) is the z-component of the demagnetization dyadic for the shape of the \( i \)-th pore and \( V \) is the volume of the sample. The magnetization in the direction of \( z \) has been assumed to be much larger than the components along the \( x \)- and \( y \)-directions. The resonance frequency is then given by the average over the \( N \)-pores, so the line-shift due to porosity considering equal volume for each pore is given by

\[ H_p' = \frac{v}{V} \left< \frac{N_z}{4\pi} \right> 4\pi M \]

(5)

where \( v/V \) is the porosity of the sample.

The line-width contribution is calculated assuming that the tangential component of the magnetic field is continuous at the ellipsoidal surface. So the uniform internal demagnetizing field is continuous at the surface of the ellipsoid parallel to it. The field within the medium is then considered to vary as the field of a dipole placed at the centre of an equivalent spherical cavity but having magnetic moment of

\[ \frac{v}{V} \left< \frac{N_z}{4\pi} \right> \mathbf{M} \]

The remaining part of the calculation follows that of Sparks and we obtain

\[ \Delta H_p' = 9 \left< \frac{N_z}{4\pi} \right> \Delta H_p \]

(6)

where \( \Delta H_p \) is the expression obtained by Sparks [5];

\[ \Delta H_p = \frac{\pi}{2} 4\pi M \frac{v}{V} \frac{0.109(3 \cos^2 \theta_u + 1.4)}{\cos \theta_u} \frac{\omega_i}{\omega_i}. \]

(7)

The values of

\[ \left< \frac{N_z}{4\pi} \right> \text{ and } \left< \left( \frac{N_z}{4\pi} \right)^2 \right> \]

obtained for \( c/a \) varying continuous from 1/6 to 6 are 0.166 and 0.06 respectively. With these values the results obtained for \( H_p' \), \( \Delta H_p' \) are given for NiZn, MgMn and YIG in table 1. The values of the intrinsic line-width calculated with the present \( \Delta H_p' \) values are in better agreement with the single crystal data.

**Table I**

<table>
<thead>
<tr>
<th>Ferrite composition</th>
<th>( 4\pi M ) (gauss)</th>
<th>Porosity ( v/V )</th>
<th>( H(0) ) (Oe)</th>
<th>( \Delta H_0 = \Delta H_i + \Delta H_a + \Delta H_p ) (Oe)</th>
<th>( \Delta H_a ) (Oe)</th>
<th>( \Delta H_p ) (Oe)</th>
<th>( \Delta H_p ) (Oe)</th>
</tr>
</thead>
<tbody>
<tr>
<td>YIG</td>
<td>1750</td>
<td>0.01</td>
<td>3584</td>
<td>30</td>
<td>10</td>
<td>17</td>
<td>+3</td>
</tr>
<tr>
<td>NiFe$_2$O$_4$</td>
<td>3270</td>
<td>0.03</td>
<td>3014</td>
<td>500</td>
<td>189</td>
<td>82</td>
<td>+229</td>
</tr>
<tr>
<td>Ni$<em>{0.75}$Zn$</em>{0.25}$Fe$_2$O$_4$</td>
<td>5100</td>
<td>0.07</td>
<td>3126</td>
<td>110</td>
<td>66</td>
<td>203</td>
<td>-159</td>
</tr>
<tr>
<td>Ni$<em>{0.5}$Zn$</em>{0.5}$Fe$_2$O$_4$</td>
<td>4800</td>
<td>0.07</td>
<td>3278</td>
<td>95</td>
<td>30</td>
<td>195</td>
<td>-130</td>
</tr>
<tr>
<td>Ni$<em>{0.25}$Zn$</em>{0.75}$Fe$_2$O$_4$</td>
<td>2300</td>
<td>0.02</td>
<td>3500</td>
<td>120</td>
<td>1</td>
<td>44</td>
<td>+75</td>
</tr>
<tr>
<td>MgFe$_2$O$_4$</td>
<td>1600</td>
<td>0.02</td>
<td>3350</td>
<td>600</td>
<td>340</td>
<td>31</td>
<td>+229</td>
</tr>
<tr>
<td>Mg$<em>{0.75}$Mn$</em>{0.25}$Fe$_2$O$_4$</td>
<td>2920</td>
<td>0.03</td>
<td>3340</td>
<td>300</td>
<td>69</td>
<td>91</td>
<td>+140</td>
</tr>
<tr>
<td>Mg$<em>{0.5}$Mn$</em>{0.5}$Fe$_2$O$_4$</td>
<td>3285</td>
<td>0.07</td>
<td>3245</td>
<td>100</td>
<td>55</td>
<td>198</td>
<td>-153</td>
</tr>
<tr>
<td>Mg$<em>{0.25}$Mn$</em>{0.75}$Fe$_2$O$_4$</td>
<td>4010</td>
<td>0.04</td>
<td>3340</td>
<td>170</td>
<td>33</td>
<td>123</td>
<td>+14</td>
</tr>
<tr>
<td>MnFe$_2$O$_4$</td>
<td>4850</td>
<td>0.09</td>
<td>3000</td>
<td>470</td>
<td>20</td>
<td>248</td>
<td>+202</td>
</tr>
</tbody>
</table>

* \( H(0) \) = Extrapolated value of the resonance field for zero diameter.

**Table II**

| Effective g Factors of Ferrous-Zinc Ferrites |

<table>
<thead>
<tr>
<th>Ferrite composition</th>
<th>( 4\pi M ) (gauss)</th>
<th>Porosity ( v/V )</th>
<th>( H(0) ) (Oe)</th>
<th>( \Delta H_{obs} ) (Oe)</th>
<th>( H_p ) (Oe)</th>
<th>( H_a ) (Oe)</th>
<th>( g_{eff} ) (Oe)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fe$_3$O$_4$</td>
<td>5500</td>
<td>0.079</td>
<td>2150</td>
<td>2500</td>
<td>73</td>
<td>160</td>
<td>2.98</td>
</tr>
<tr>
<td>Zn$<em>{0.2}$Fe$</em>{2.8}$O$_4$</td>
<td>6650</td>
<td>0.064</td>
<td>2450</td>
<td>2500</td>
<td>71</td>
<td>75</td>
<td>2.73</td>
</tr>
<tr>
<td>Zn$<em>{0.4}$Fe$</em>{2.6}$O$_4$</td>
<td>6160</td>
<td>0.068</td>
<td>2600</td>
<td>2400</td>
<td>70</td>
<td>25</td>
<td>2.63</td>
</tr>
<tr>
<td>Zn$<em>{0.6}$Fe$</em>{2.4}$O$_4$</td>
<td>4460</td>
<td>0.054</td>
<td>3000</td>
<td>1000</td>
<td>40</td>
<td>15</td>
<td>2.32</td>
</tr>
</tbody>
</table>

* \( H(0) \) = Extrapolated value of the resonance field for zero diameter.
This is true particularly for the YIG sample. This analysis is also in agreement with the measurements made by Dionne [8] on manganese doped YIG which, as shown by him, are not in agreement with the Geschwind-Clogston-Sparks analysis.

3. Line-width and line-shift in ferrous-zinc ferrites. —

The resonance data for most members of the Fe$_3$O$_4$ ferrite series is not available in literature. The method of preparation and equilibrium magnetic properties of the samples made by us are described elsewhere [9]. Our measurements of the line-shift and line-width for $x = 0, 0.2, 0.4, 0.6$ and for different diameters have yielded the data given in Table II and III. The resonance curve for Zn$_{0.4}$Fe$_{2.6}$O$_4$ is given in figure 1. The curves for other compositions are similar. On account of large line-widths the error in resonance field is of the order of +50 Oe and in line-width is of the order of 10 %.

The value (2.99) of $g_{\text{eff}}$ for Fe$_3$O$_4$ is not unreasonable. From EPR studies for octahedral coordination, $g_{\text{eff}}$ for divalent iron has been reported [10] to be 3.428 and 3.30. In magnetically ordered systems this value is expected to be lower on account of larger overlap and quenching of the orbital contribution.

As seen from Table III the values of observed $\Delta H$ are in fair agreement with the theoretical values. In view of the approximation used in the calculations of the line-width contribution from six different sources, the agreement is satisfactory.

4. Conclusion. — We have obtained an expression for the porosity contribution to line-shift and line-width on a more realistic model regarding the shape of the pores. It has been assumed that the pores are ellipsoidal in shape having the same volume but varying values of $c/a$. The results obtained are in better agreement than the existing theories. The analysis of size dependent factors in FMR earlier obtained for insulators have been shown to be applicable also for systems like ferrous zinc ferrite where the contribution from conductivity cannot be ignored.

References