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Investigation of ferromagnetic resonance in magnetic fluids by means of the short-circuited coaxial line technique

P C Fannin†, T Relihan† and S W Charles‡

† Department of Microelectronics & Electrical Engineering, Trinity College, Dublin 2, Ireland
‡ Department of Chemistry, University College of North Wales, Bangor LL57 2UW, UK

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Abstract. The short-circuit coaxial transmission line technique has been used to investigate ferromagnetic resonance in magnetic fluids over the frequency range 0.1–6 GHz. Measurements of the complex susceptibility, $\chi(\omega) = \chi'(\omega) - i\chi''(\omega)$, of magnetic fluid samples of magnetite and cobalt particles, of approximate median diameters 10 and 5.8 nm are presented. The presence of loss peaks in the $\chi''(\omega)$ components at approximate frequencies of 1.2 and 2.5 GHz, respectively, coupled with the transition of the $\chi'(\omega)$ components from positive to negative values at slightly higher frequencies, is indicative of ferromagnetic resonance. Appropriate equations for the calculation of the complex susceptibility are presented.

1. Introduction

The determination of the complex impedance, $Z = R + iX$, of a material specimen by means of the short-circuited coaxial transmission line technique is a well-established method originating from the work of Roberts and von Hippel [1, 2]. The short-circuit produces a maximum magnetic field and a minimum electric field at the sample, thus making the technique particularly suited to the measurement of the magnetic properties of test samples. Fluids have a particular advantage over solids in that they readily fill the coaxial test cell which has almost radial electric field and concentric magnetic field. In the case of an inductive load, from the measurement of the resistive, R_L, and the reactive X_L component, the complex permeability, $\mu(\omega) = \mu'(\omega) - i\mu''(\omega)$, and hence the complex susceptibility, $\chi(\omega) = \chi'(\omega) - i\chi''(\omega)$, are readily determined by means of the equations developed in the theory section of this paper. From the plots of $\chi'(\omega)$ and $\chi''(\omega)$ against frequency in hertz, one can identify the onset of the phenomena of ferromagnetic resonance either by the presence of a peak in the $\chi''(\omega)$ component or by the $\chi'(\omega)$ component going negative.

Ferromagnetic resonance [3] in a uniaxial particle is characterized by the precession of the magnetic moment, m_p, about its axis of easy magnetization with an angular frequency ω_0, where [4]

$$\omega_0 = \gamma H_A \mu_0 \qquad (1)$$

and a relaxation time

$$\tau_0 = (\alpha\omega_0)^{-1} = M_S/(2\mu_0\gamma\alpha K). \qquad (2)$$

τ_0 has an often quoted approximate value of 10^{-8}–10^{-12} s [5, 6], α is a damping constant, M_S is the saturation magnetization per unit volume, γ is the magnetogyric ratio, K is the anisotropy constant and H_A is the internal field given by

$$H_A = 2K/M_S. \qquad (3)$$

From equations (1) and (3) ω_0 may be written as

$$\omega_0 = 2K\mu_0\gamma/M_S. \qquad (4)$$

It has been shown [4] that, in the case in which a radiofrequency field is applied perpendicular to H_A, the motion of the magnetic moment has a typical resonant character with the real and imaginary components of the AC susceptibility having a Lorentz form. Ferrofluids consist of a distribution of particle sizes, whose magnetic moment may (in the frequency range considered here), reverse direction within the particle by overcoming an energy barrier, which for uniaxial anisotropy is given by Kv, where v is the particle volume. The probability of such a transition is approximately equal to $\exp\sigma$, where σ is the ratio of anisotropy energy to thermal energy, $Kv/(kT)$. This reversal, or switching time, is referred to as the Néel relaxation time, τ [7], where Néel estimated the relaxation time τ to be

$$\tau = \tau_0 \exp\sigma. \qquad (5)$$

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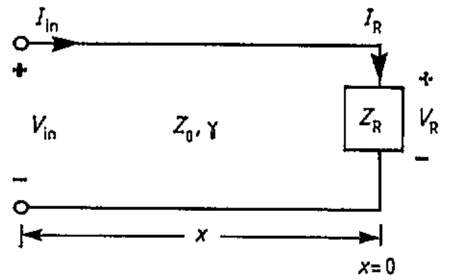

Figure 1. The equivalent circuit of the transmission line terminated in a load impedance Z_R.

Brown [8] developed Néel's work and arrived at his expressions for high and low barrier heights, described as

$$\tau_N = \tau_0 \sigma^{-1/2} \exp(\sigma) \qquad \sigma \geq 2 \qquad (6)$$
$$= \tau_0 \sigma \qquad \sigma \ll 1.$$

2. Theory

The input impedance of a transmission line at a distance x from a terminating load, Z_R, as shown in figure 1, is given by [9]

$$Z_{in} = \frac{V_{in}}{I_{in}} = \frac{V_R \cosh(\gamma x) + Z_0 I_R \sinh(\gamma x)}{I_R \cosh(\gamma x) + (V_R/Z_0) \sinh(\gamma x)} \qquad (7)$$

where Z_0 is the characteristic impedance and γ is the propagation constant which may be written as

$$\gamma = \alpha + \mathrm{i}\beta \qquad (8)$$

where α is the attenuation coefficient, $\beta = 2\pi/\lambda$ is the phase change coefficient and λ is the operating wavelength in the line. In the case in which the line has a very low loss it is assumed that $\alpha \approx 0$ and $\gamma = \mathrm{i}\beta$ and equation (7) becomes

$$Z_{in} = Z_0 \frac{Z_R + \mathrm{i} Z_0 \tan(\beta x)}{Z_0 + \mathrm{i} Z_R \tan(\beta x)}. \qquad (9)$$

When the load is a short circuit, namely $Z_R = 0$ and $V_R = 0$, then the input impedance is

$$Z_{in} = \mathrm{i} Z_0 \tan(\beta x). \qquad (10)$$

Consider the case of a short-circuited, air-filled coaxial transmission line, terminated with a toroidal sample of material of thickness or depth d, as shown in figure 2 with Z_0 and γ being the characteristic impedance and propagation constant of the air-filled line, respectively.

The shorted load section of figure 2 can be modelled as that of figure 1 with Z_R being the input impedance, as illustrated in figure 3 and with Z_l and γ_l the characteristic impedance and propagation constant of the sample-filled line. In this case it cannot be assumed that the line is lossless so the general equation (equation (7)) has to be used. Since the line is shorted, the input impedance, Z_R, may be written as

$$Z_R = Z_l \tanh(\gamma_l d). \qquad (11)$$

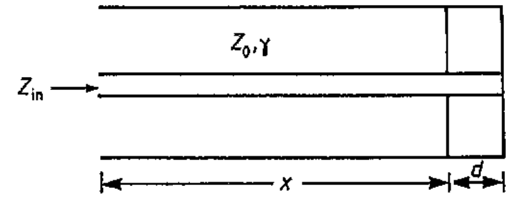

Figure 2. The model of a coaxial transmission line terminated in a toroidal sample with a short circuit.

The intrinsic impedance Z of a medium with absolute values of complex permeability μ, is given by

$$Z = \mathrm{i}\omega\mu/\gamma \qquad (12)$$

whereas the characteristic impedance, Z_l, of a coaxial line containing such a medium as the dielectric is [10]

$$Z_l = \frac{1}{2\pi} Z \ln\left(\frac{b}{a}\right) \qquad (13)$$

where b is the radius of the outer conductor and a is the radius of the inner conductor of the coaxial line. From equations (12) and (13), equation (11) then becomes

$$Z_R = A \frac{\mathrm{i}\omega\mu_l}{\gamma_l} \tanh(\gamma_l d) \qquad (14)$$

where $A = [1/(2\pi)] \ln(b/a)$. A significant simplification can be now made if it is assumed that $\gamma_l d \ll 1$, namely that the sample depth is much less than the wavelength of electromagnetic radiaton in the sample medium.

Under this assumption, $\tanh(\gamma_l d) \approx \gamma_l d$ and equation (14) becomes

$$Z_R = \mathrm{i} A \omega \mu_l d. \qquad (15)$$

Substituting in equation (9) for Z_R, one obtains

$$Z_{in} = Z_0 \frac{\mathrm{i} A \omega \mu_l d + \mathrm{i} Z_0 \tan(\beta x)}{Z_0 - A\omega\mu_l d \tan(\beta x)} \qquad (16)$$

which in terms of the permeability of the sample, μ_l, reduces to

$$\mu_l = Z_0 \frac{Z_{in} - \mathrm{i} Z_0 \tan(\beta x)}{\mathrm{i} A\omega d Z_0 + Z_{in} A \omega d \tan(\beta x)}. \qquad (17)$$

On elimination of the constant factor A, which incorporates the dimensions of the coaxial line, one obtains

$$\frac{\mu_l}{\mu_0} = \frac{\lambda}{2\pi d} \frac{(Z_{in}/Z_0) - \mathrm{i}\tan(\beta x)}{(Z_{in}/Z_0)\tan(\beta x) + \mathrm{i}}. \qquad (18)$$

Noting that $\mu_l = \mu_0 \mu_r$ and separating into real and imaginary components, $\mu_r = \mu_r' - \mathrm{i}\mu_r''$, with $Z_{in} = R_{in} + \mathrm{i}X_{in}$ and $Z_0 = R_0$, one obtains

$$\mu_r' = \frac{\lambda}{2\pi d}(R_{in}^2 \tan(\beta x) + [X_{in} - R_0 \tan(\beta x)]$$
$$\times [X_{in} \tan(\beta x) + R_0])/([R_{in} \tan(\beta x)]^2$$
$$+ [X_{in} \tan(\beta x) + R_0]^2) \qquad (19)$$

$$\mu_r' = \frac{\lambda}{2\pi d}(R_{in}[X_{in} \tan(\beta x) + R_0] - [X_{in} - R_0 \tan(\beta x)]$$
$$\times [R_{in} \tan(\beta x)])/([(R_{in} \tan(\beta x)]^2$$
$$+ [X_{in} \tan(\beta x) + R_0]^2). \qquad (20)$$

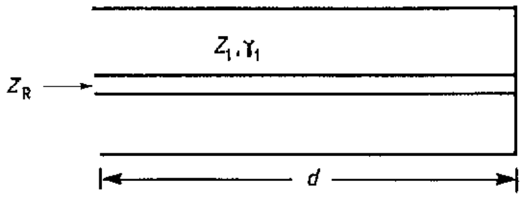

Figure 3. The model of a short-circuited toroidal sample.

Now, since

$$\mu'(\omega) = 1 + \chi'(\omega) \quad (21)$$

$$\mu''(\omega) = \chi''(\omega) \quad (22)$$

the relative, complex susceptibility components are readily determined from impedance measurements of the short-circuited line.

3. Measurement and results

The measurements reported here on a coaxial cell of 3 mm inner diameter and 7 mm outer diameter, over the frequency range 0.1–6 GHz, were made under computer control by the HP 8753C Network Analyser. This instrument automatically measures the reflection and transmission characteristics of devices by the use of the S, or scattering parameters, which are a measure of the ratio of the power reflected from a device to the power incident on a device. When the instrument is operated in a one-port mode it measures the S_{11} parameter. Now

$$S_{11} = (Z_R - Z_0)/(Z_R + Z_0) \quad (23)$$

where Z_R is the load impedance and Z_0 is the characteristic impedance of the instrument. The instrument has the capability of converting the S_{11} measurements to the real and imaginary components of Z_R by computing the equation

$$Z_R = Z_0(1 + S_{11})/(1 - S_{11}). \quad (24)$$

Thus in the case of an inductive load it automatically measures the resistive component, R_L, and the reactive component, X_L, respectively.

Using the technique presented, the complex susceptibility of three ferrofluid samples of (i) magnetite in isopar M, (ii) cobalt in toluene and (iii) cobalt ferrite in isoparm, with average particle diameters 10, 5.8 and 9.2 nm, respectively, was investigated over the frequency range 0.1–6 GHz. In order to satisfy the requirement that $\gamma_1 d \ll 1$, namely that the sample depth was much less than the wavelength of electromagnetic radiation in the sample medium, measurements over the frequency range 100–400 MHz were obtained using a sample depth, d, of 10 mm and a sample depth of 1.5 mm was used thereafter for the rest of the frequency range.

Figure 4 shows the results obtained for samples (i) and (ii); these normalized plots of $\chi'(\omega)$ and $\chi''(\omega)$ against f (Hz) show that, for both samples, the $\chi''(\omega)$ component is constant up to a frequency of approximately 400 MHz whilst at the same time the corresponding $\chi'(\omega)$ component decreases. These effects are a manifestation of the contribution of the relaxational Néel components to the

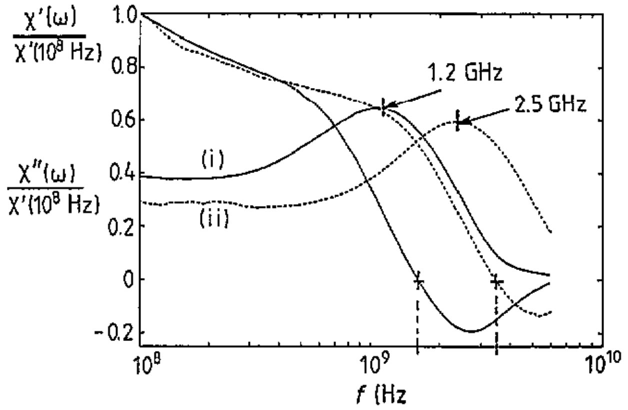

Figure 4. A normalized plot of $\chi'(\omega)$ and $\chi''(\omega)$ against f (Hz) for samples (i) and (ii).

susceptibility. Beyond 400 MHz a relaxation to resonance transition occurs with the $\chi'(\omega)$ components going negative at approximately 1.7 and 3.6 GHz respectively; for sample (iii), the change in $\chi'(\omega)$ to a negative value was found to be outside the frequency range of the measuring instrument and, apart from noting that this was to be expected, no further comment can be made.

The values of K for each of the samples used in the following calculations are typical of those obtained from low-temperature decay of remanence measurements and from torque magnetometry. For magnetite (inverse spinel structure) particles, shape anisotropy dominantes the magnetocrystalline anisotropy, resulting in a uniaxial anisotropy with K typically in the range $(2\text{–}5)\times 10^4$ J m^{-3} [11, 12]. The corresponding saturation magnetization is approximately 0.4 T. The cobalt ferrite particles studied here were produced by the thermolysis of cobalt octacarbonyl and have a poorly crystalline structure and a uniaxial anisotropy with K typically 2×10^5 J m^{-3}, and a saturation magnetization of 1 T [12, 13]. Finally, for cobalt ferrite particles the magnetocrystalline anisotropy dominates any contribution to shape and thus the particles exhibit cubic anisotropy with an approximate K value of 1.5×10^6 J m^{-3} [14]. Substituting these values into equation (4) results in theoretical resonant frequencies of 3.5, 14 and 350 GHz, respectively. It should be noted that both resonance and relaxation of the magnetic moment can take place within particles and that resonance becomes significant for these particles with $\sigma > 0.7$ [4]. For the three systems studied here the values of σ for particles of samples (i), (ii) and (iii) are 2, 5 and 200, respectively; that is, significantly greater than 0.7, indicating that a substantial amount (> 50 vol%) of the particles contribute to the observed resonance.

4. Conclusion

Having developed and presented the appropriate equations, the short-circuit coaxial transmission line technique has been used to determine the complex susceptibility, $\chi(\omega) = \chi'(\omega) - i\chi''(\omega)$, of magnetic fluid samples of magnetite, cobalt and cobalt ferrite particles, with approximate median diameters of 10, 5.8 and 9.2 nm respectively, over the frequency range 0.1–6 GHz. These measurements have revealed the presence of a resonance behaviour characterized by the precession of the magnetic moment about the easy axis of magnetization in a particle. The transition, from negative to positive values, of the $\chi'(\omega)$ components of samples (i) and (ii), at approximate frequencies of 1.7 and 3.6 GHz, respectively, is indicative of the ferromagnetic resonance effect. In the case of sample (iii), the change in $\chi'(\omega)$ to a negative value was found to be outside the frequency range of the measuring instrument.

These measured resonant frequencies differ from the approximate theoretical values of 3.5 and 14 GHz, as determined by equation (4), with the greatest discrepancy occurring in the case of the cobalt particles. However, it should also be borne in mind that equation (4) is applicable for the case of a single particle and not a distribution of particles with a corresponding distribution of K, as exists for the case of magnetic fluids.

It is of interest to note that the technique presented is suitable for measurements at higher frequencies; the upper frequency of 6 GHz here being due to the limitation of the network analyser. The authors have also used the technique to measure the magnetic properties of non-liquid samples, such as magnetic tapes.

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