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Investigation of magnetic fluids exhibiting field induced absorption peaks in the susceptibility spectra

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Measurements of the frequency and field dependent of the complex magnetic susceptibility, $\chi(\omega, H)= \chi'(\omega,H) - i\chi''(\omega, H)$, of magnetic fluids over the frequency range, 200Hz to 1 MHz, by means of the toroidal technique, are presented. The magnetic polarizing field, H, is increased from 0 to 13.6 kA/m in fifteen steps and the resulting susceptibility spectra is found to exhibit a field induced , double loss-peak over the frequency range concerned. The existence of a double absorption peak is quite unusual and represents a double or bi-modal distribution of particles sizes in the ferrofluid sample. The equations of Debye are used to determine corresponding aggregate radii whilst a measure of a distribution of relaxation times is obtained in terms of the Cole-Cole distribution parameter, α.

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1. Introduction

Ferrofluids are colloidal suspensions of single-domain magnetic particles, electrostatically or sterically stabilised in a liquid carrier and stabilised by means of a suitable organic surfactant. The particles have radii ranging from approximately 2-10 nm and when they are in suspension their magnetic properties can be described by the paramagnetism theory of Langevin, suitably modified to cater for a distribution of particle sizes. The particles are considered to be in a state of uniform magnetisation with a magnetic moment, m, given by: $m_p = M_S$ v, where M_s denotes saturation magnetisation and v is the magnetic volume of the particle.

There are two distinct mechanisms by which the magnetisation of ferrofluids may relax after an applied field has been removed: either rotational Brownian motion of the particle within the carrier liquid, with its magnetic moment, m_p, locked in an axis of easy magnetisation, or by rotation of the magnetic moment within the particle, called Néel relaxation. The time associated with the rotational diffusion is the Brownian relaxation time (τ_B) [1] where V is the hydrodynamic volume of the particle and η is the dynamic viscosity of the carrier liquid.

$$\tau_B = 3\, V\eta / kT \tag{1}$$

In the case of the Néel relaxation time (τ_N) [2], the magnetic moment may reverse direction within the particle by overcoming an energy barrier, which for uniaxial anisotropy, is given by Kv, where K is the anisotropy constant of the particle. τ_N is given by the approximate expression

$$\tau_N = \tau_0 \exp(\sigma) \tag{2}$$

where σ is the ratio of anisotropy energy to thermal energy (Kv/kT) and τ_0 has an often quoted as having an approximate value of 10^{-8} to 10^{-10} s.

Associated with a distribution of particle sizes is a distribution of relaxation times, where the effective relaxation time τ_{eff}, is given by,

$$\tau_{eff} = \tau_N \tau_B / (\tau_N + \tau_B) \tag{3}$$

the mechanism with the shortest relaxation time being dominant. As an approximate ‘rule of thumb’ [3], in the case of particles with radii greater than 6 nm the dominant mechanism is Brownian, whilst for particles with radii less that 6 nm the dominant mechanism is Néel one.

2. Complex susceptibility

Susceptibility measurements are an ideal way of investigating the dynamic properties of magnetic fluids. The frequency dependent complex susceptibility, $\chi(\omega)$, may be written in terms of its real and imaginary components, where

$$\chi(\omega) = \chi'(\omega) - i\chi''(\omega) \tag{4}$$

According to Debye's theory [4], $\chi(\omega)$ has a frequency dependence given by the equation,

$$\chi(\omega) - \chi_\infty = (\chi_o - \chi_\infty)/(1 + i\,\omega\,\tau_{eff}) \tag{5}$$

where the static susceptibility or low frequency susceptibility, χ_O, is defined as

$$\chi_o = nm^2/3kT\mu_o \qquad (6)$$

n is the number density and χ_∞ indicates the susceptibility value at very high frequencies and

$$\tau_{\varepsilon\phi\phi} = 1/\,\omega_{max} = 1/2\pi f_{max}. \qquad (7)$$

Thus by determining f_{max} and utilizing equations (1) or (2) the average particle size can be readily determined.

In a typically well dispersed ferrofluid one does not generally find a loss-peak in the susceptibility profile covering a frequency range 100Hz to 1MHz, however a loss-peak is found in the case of aggregated fluids

The relation between $\chi'(\omega)$ and $\chi''(\omega)$ and their dependence on frequency, $\omega/2\pi$, can be displayed by means of the magnetic analogue of the Cole-Cole plot [5] where the data fits a depressed circular arc. In the Cole-Cole case the circular arc cuts the $\chi'(\omega)$ axis at an angle of $\alpha\pi/2$; α is referred to as the Cole-Cole parameter and is a measure of the distribution of relaxation times.

The magnetic analogue of the Cole-Cole circular arc is described by the equation

$$\chi(\omega) = \chi_\infty + (\chi_O - \chi_\infty)/[(1 + (i\,\omega\,\tau_O)^{1-\alpha}),\;\; 0 < \alpha < 1 \qquad (8)$$

which for $\alpha = 0$, reduces to that of equation (5). Furthermore, in the rare event of finding two loss-peaks the spectra can be described by the approximate expression [6],

$$\chi(\omega) - \chi_\infty = (\chi(\omega) - \chi_\infty)\,(A/(1+i\omega\tau_A) + B/(1+i\omega\tau_B)) \qquad (9)$$

τ_A and τ_B are the average relaxation times of the individual sections of the overall distribution function. Fig. 1(A) shows a typical theoretical plot of such a spectra with loss-peaks at 100 Hz and 100 kHz whilst Fig. 1(B) shows the corresponding Cole-Cole plot of Fig. 1(A). However since ferrofluids contain a distribution of particle sizes it is necessary in fitting to the actual data, to modify equation (9) to take account of this fact, giving,

$$\chi(\omega) - \chi_\infty = (\chi(\omega) - \chi_\infty)(A/(1+i(\omega\tau_A)^{(1-\alpha_1)}) + (B/(1+i(\omega\tau_B))^{(1-\alpha_2)}) \qquad (10)$$

Plots 1(a) and 1(b) of Fig. 2 show the unpolarised susceptibility spectra which does not display a loss-peak. The remaining plots of $\chi'(\omega)$ against f (Hz) indicate how χ' decreases with increasing H. This is as predicted by theory [7].

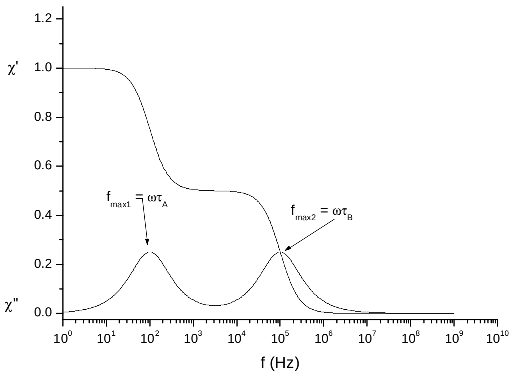

Fig. 1(a). Theoretical plot of susceptibility spectra displaying two loss- peaks.

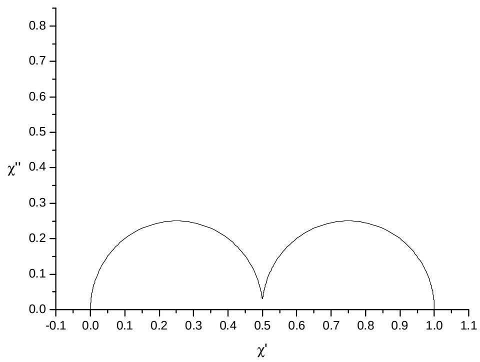

Fig. 1(b). Cole-Cole plot of susceptibility data of Fig 1(a).

3. Measurements

The investigated sample consisted of a colloidal suspension of magnetite particles of mean particle radius 5 nm, in water, with oleic acid as surfactant; the saturation magnetization was 363 Gauss. Complex susceptibility measurements were performed over the frequency range 200 Hz to 1 MHz, and polarising field over the range 0-13.6 kA/m field.

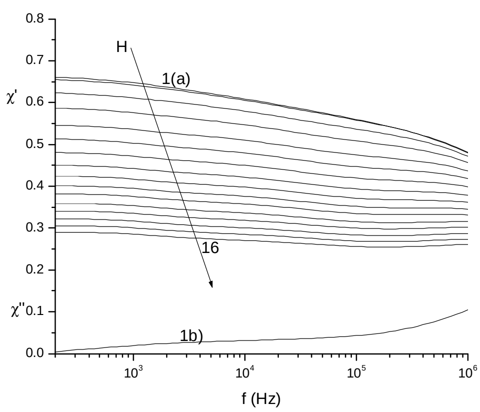

Fig. 2. Plots 1(a) and 1(b) show the unpolarised susceptibility spectra which does not display a loss-peak whilst the remaining plots of against $\chi'(\omega)$ against f (Hz) indicate how χ' decreases with increasing H.

From Fig. 3 it can be seen how the corresponding χ'' component varies with increasing H. Initially there is no loss-peak but one starts to emerge as the field is increased to 4.5 kA/m, with a peak frequency of f_{max3} = 39 kHz. With a further increase in H, two separate loss-peaks emerge until at H=13.6 kA/m the peaks occur at f_{max1}=2.6 kHz and f_{max2}=30 kHz. From equations (1) and (7) the resulting relaxation times and average particle radii are as shown in Table 1.

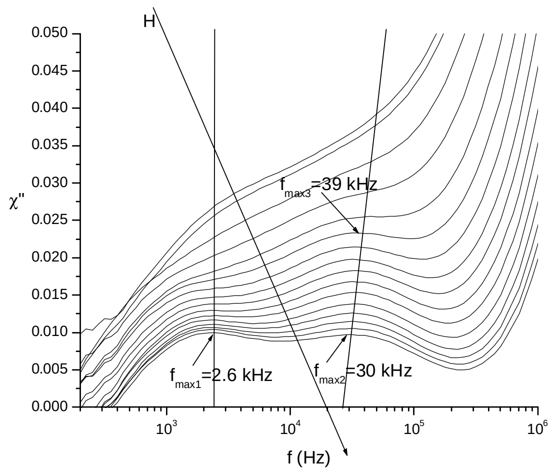

Fig. 3. Plot of variation in $\chi'(\omega)$ against f(Hz), with polarizing field H, exhibiting bi-model peaks at 2.6 kHz and 30 kHz.

Table 1.

f_{max} (kHz)	τ_B(μ s)	Radius r (nm)
1. 2.6	61.2	27.2
2. 30	5.3	12.0
3. 39	4.1	11.0

An estimate of the distribution of relaxation times, in terms of the Cole-Cole parameter α, was obtained by fitting the data to equation (10). Fig. 4 shows the Cole-Cole fit to the data at the point of emergence of the double loss-peak at H= 5.45 kA/m with two values of α, α_1=0.0025 and α_2= 0.485. An increase in H results in α_1 increasing and α_2 decreasing. Fig. 5 shows the Cole-Cole plot for H= 8.18 kA/m with two distinct Cole-Cole plots where α_1= 0.047 and α_2 = 0.391. Fig. 6 shows the plot for the final value of H=13.6 kA/m with α_1 = 0.072 and α_2 =0.017. Thus over the range 5.45kA/m to 13.6 kA/m, α_1 increased from 0.0025 to 0.072 whilst α_2 decreased from 0.485 to 0.017 indicating that the distribution of relaxation times centered on the loss-peak, $f_{max\,1}$, increased whilst the opposite happened to the distribution of relaxation times centered on the loss-peak, $f_{max\,2}$.

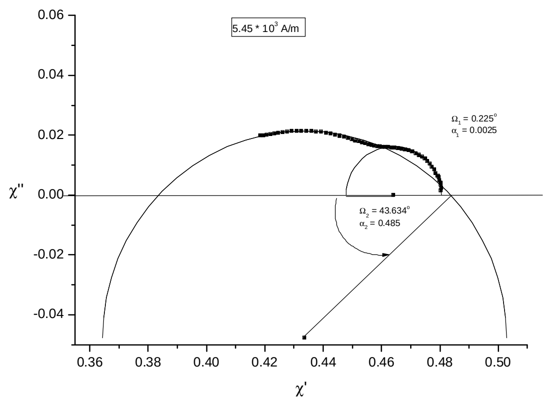

Fig. 4. Cole-Cole plot for H= 5.54 kA/m.

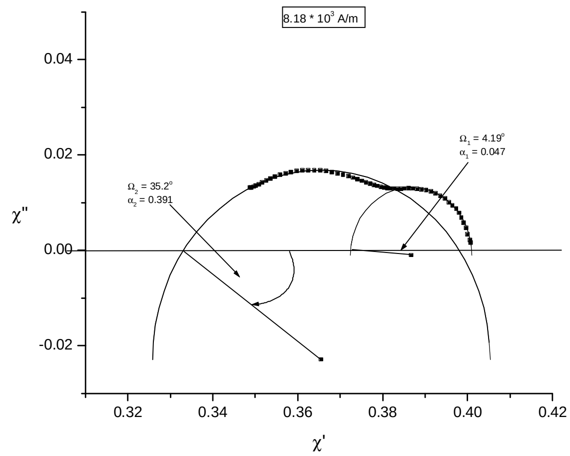

Fig. 5. Cole-Cole plot for H=8.18 kA/m.

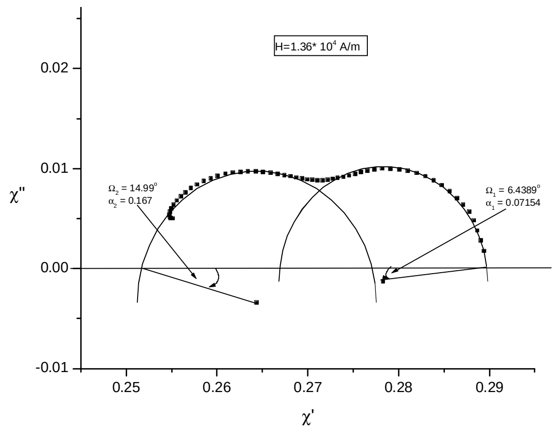

Fig. 6. Cole-Cole plot for H= 13.6 kA/m.

4. Conclusions

Measurements of the frequency and field dependent complex magnetic susceptibility of a water based magnetic fluid over the frequency range, 200 Hz to 1 MHz are presented. It has been shown how the application of a magnetic polarizing field, H, resulted initially in a single loss-peak occurring at a frequency of, f_{max3} = 39 kHz, in the $\chi''(\omega)$ against f(Hz) profile; this peak was transformed into two double loss-peak with loss-peak frequencies of f_{max1} = 2.6 kHz and f_{max2} = 30 kHz. By means of the Debye equations the corresponding particle size were shown to be 1) 27.2nm, 2) 12 nm and 3) 11nm. Application of the Cole-Cole equations enabled a measure of the Cole-Cole distribution parameter, α, as a function of H, to be obtained for the distribution of relaxation times centered on f_{max1} and f_{max2}.

References

[1] W. F.Brown, J. Appl. Phys. **34**, 1319 (1963).
[2] L. Néel, Ann. Géophys. **5**, 99 (1949).
[3] P. C. Fannin, J. Magn. Magn. Mater **136**, 49 (1994).
[4] P. Debye, Polar Molecules, The Chemical Catalog Company, New York, (1929).
[5] K. S. Cole, R. H. Cole, J. Chem. Phys. **9**, 341 (1941).
[6] P. C. Fannin, Adv. Chem. Phys.," Wideband Measurement and Analysis Techniques for the Determination of the Frequency dependent Complex Susceptibility of Magnetic Fluids, John Wiley, New York.Editors, I Prigogine and Stuart A. Rice **104**, 181 (1998).
[7] P. C. Fannin, A. T. Giannitsis, J. Mol. Liq. **114**, 89 (2004).

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