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Broad-band measurement of the complex susceptibility of magnetic fluids

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Abstract. We report on the measurement, over the frequency range 10 Hz to 3 GHz, of the complex susceptibility, $\chi(\omega) = \chi'(\omega) - i\chi''(\omega)$, of a magnetic fluid with manganese ferrite particles of median diameter 9.4 nm in a hydrocarbon carrier. This upper frequency was realized by use of a HP Network Analyser which operates over the frequency range 300 kHz to 3 GHz and has enabled a more complete susceptibility–frequency profile to be determined. The presence of a loss-peak at 30 MHz in the $\chi''(\omega)$ component is a demonstration of the phenomenon of superparamagnetism in small ferromagnetic particles whilst the transition of $\chi'(\omega)$ to a negative value at 65 MHz is indicative of ferroresonance.

1. Introduction

A ferrofluid is a colloidal suspension of a distribution of single-domain ferromagnetic particles dispersed in a carrier liquid and stabilized by a suitable organic surfactant. Being single-domain, the particles are considered to be in a state of uniform magnetization with magnetic moment, m, given by

$$m = M_s v \tag{1}$$

where M_s (Wb m^{-2}) denotes saturation magnetization and v is the volume of the particle. The magnetic moments are fixed in orientation relative to the particles themselves because of magnetic anisotropy. The direction of the magnetic moment is referred to as the axis of easy magnetization and equilibrium of the moment may be attained by either of two relaxation mechanisms.

In the first case the magnetic moment rotates together with the particle and this is referred to as Brownian rotation [1] with relaxation time τ_B, where

$$\tau_B = 3V\eta/kT \tag{2}$$

and V is the hydrodynamic volume of the particle and η is the dynamic viscosity of the carrier liquid. In the case of the second relaxation mechanism, the magnetic moment may reverse direction within the particle by overcoming an energy barrier, Kv, where K is the effective magnetic anisotropy constant of the particle. This reversal, or switching time, is referred to as the Néel relaxation time, τ_N [2] and in terms of Brown's [3] expressions for high and low barrier heights is described approximately as

$$\tau_N = \begin{cases} \tau_0 \sigma^{-1/2} \exp(\sigma) & \sigma \gg 1 \\ \tau_0 \sigma & \sigma \ll 1 \end{cases} \tag{3}$$

with τ_0 having an approximate value of 10^{-9} s.

One should note that the formulae of equation (3) are simple approximate formula and currently there is considerable debate regarding the value of τ_0 [4] and indeed the equation itself [5].

τ_0 can be determined from the expression

$$\tau_0 = M_s/2\mu_0\gamma\alpha K \tag{4}$$

where γ is the gyromagnetic ratio and α is a damping constant. α is not accurately known but is generally approximated to 0.1 or 0.01 [6, 7] and K is a function of both particle size and shape. Thus it is quite apparent why τ_0 has only an approximate value and also why it plays a significant role in the determination of τ_N of (3).

A distribution of particle sizes implies the existence of a distribution of relaxation times, and in general both relaxation mechanisms contribute to magnetization with an effective relaxation time τ_{eff} [8], where

$$\tau_{eff} = \tau_N\tau_B/(\tau_N + \tau_B). \tag{5}$$

The dominant mechanism of a particle will be that with the shortest relaxation time. Thus if $\tau_N \gg \tau_B$, then from (5) $\tau_{eff} = \tau_B$, whilst if $\tau_N \ll \tau$, $\tau_{eff} = \tau_N$.

The magnetic moment of single-domain particles exceeds the moment of an individual atom by four to five orders of magnitude and their behaviour in a magnetic field is referred to as being 'superparamagnetic'. Now the phenomenon of superparamagnetism in a distribution of small ferromagnetic particles manifests itself as a maximum in the imaginary component of the magnetic susceptibility at a frequency f_{max}, which is related to the effective relaxation time τ_{eff} by the expression

$$f_{max} = 1/(2\pi\tau_{eff}). \tag{6}$$

Furthermore, for a particular value of σ, transition

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from relaxation to resonance [9] can occur with f_{max} being given by

$$f_{max} = \omega_r \alpha/(\sigma 2\pi) \tag{7}$$

where ω_r is the gyromagnetic frequency and α the damping parameter of equation (4).

However, it is difficult to predict in advance where f_{max} should occur for any particular ferrofluid sample and thus one has to rely on a technique that measures f_{max} directly. One effective and proven technique is the toroidal technique of Fannin *et al* [10] whereby, as a result of swept measurements taken over a wide frequency range, the real and imaginary components of the complex AC magnetic susceptibility of a ferrofluid, $\chi'(\omega)$ and $\chi''(\omega)$ respectively, are determined. Here we report on such measurements up to a frequency of 3 GHz.

2. Complex susceptibility

The theory developed by Debye [11] to account for anomalous dielectric dispersion in dipolar fluids has been used [12, 13] to account for the analogous case of magnetic fluids. Debye's theory holds for spherical particles when the magnetic dipole–dipole interaction energy, U, is small relative to the thermal energy kT.

The complex frequency-dependent magnetic susceptibility, $\chi(\omega)$, may be written in terms of its real and imaginary components, where

$$\chi(\omega) = \chi'(\omega) - i\chi''(\omega). \tag{8}$$

According to Debye's theory the complex susceptibility, $\chi(\omega)$, has a frequency-dependence given by the approximate equation

$$\chi(\omega) = \chi_0/(1 + i\omega\tau_{eff}) \tag{9}$$

$$= \chi_0/(1 + \omega^2\tau_{eff}^2) - i\omega\tau_{eff}\chi_0/(1 + \omega^2\tau_{eff}^2) \tag{10}$$

with

$$\chi_0 = nm^2/3kT\mu_0 \tag{11}$$

where n is the particle number density.

Equation (9) demonstrates how $\chi'(\omega)$ falls monotonically whilst the $\chi''(\omega)$ component has a maximum at $\omega_{max}\tau_{eff} = 1$ and it is this significant occurrence that enables one to determine the effective relaxation time, τ_{eff}, and the corresponding particle size for the ferrofluid sample in a relatively simple manner.

3. Measurement and results

The measurements reported here, over the frequency range 300 kHz to 3 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,

$$S_{11} = (Z_L - Z_0)/(Z_L + Z_0) \tag{12}$$

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

$$Z_L = Z_0(1 + S_{11})/(1 - S_{11}). \tag{13}$$

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

The instrument has a number of options for reducing noise and enhancing signal level. Averaging is one such option, which enables one to choose the number of times a measurement sweep is to be repeated, with data from each sweep being automatically averaged into the previous average. Also, an electrical delay facility exists whereby one can compensate for the electrical effects of any connection between the instrument and the device to be measured. Both these facilities lead to more accurate measurements.

A number of ferrofluid samples were tested and typical of the results obtained is that shown in figure 1 for manganese ferrite ($Mn_{0.7}Fe_{0.1}Fe_2O_4$) particles in Isopar-M. This is a normalized plot of $\chi'(\omega)$ and $\chi''(\omega)$ against $\log[f(\text{Hz})]$ up to a frequency of 3 GHz for a sample with an average particle size of 9.4 nm, a standard deviation of 0.46, a σ of 1.2 and a M_s of 0.03 T. The graph is in fact a composite of low-frequency measurements from 10 Hz to 300 kHz obtained by use of a HP LF bridge [13] and those obtained by use of the Network Analyser. The latter results were obtained using an averaging factor of 20, which means that this section of the graph represents the final outcome of 20 swept measurements. The maximum of the loss-peak of $\chi''(\omega)$ occurs at a frequency of 30 MHz whilst the frequency at which $\chi'(\omega)$ goes negative is approximately 65 MHz.

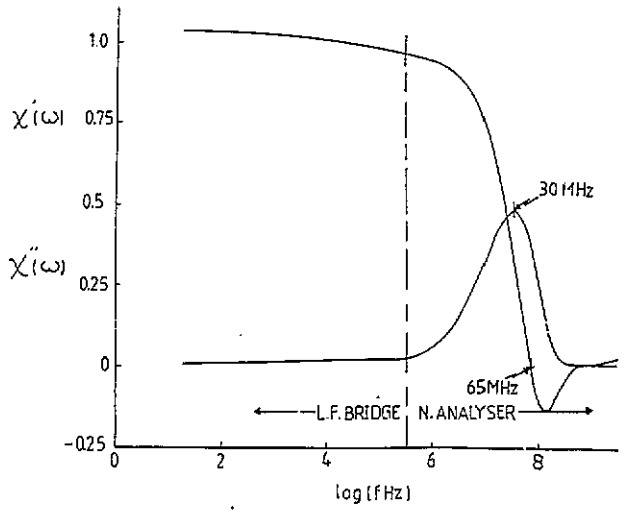

Figure 1. Normalized plot of $\chi'(\omega)$ and $\chi''(\omega)$ against $\log[f(\text{Hz})]$ over the frequency range 10 Hz to 3 GHz.

This latter occurrence is indicative of a ferroresonance [9, 14], which is qualitatively similar to that predicted by Shliomis *et al* [15].

From equation (7)

$$\omega_{max} = \omega_r \alpha / \sigma \tag{14}$$

where ω_r is the gyromagnetic frequency and α the damping parameter. For the parameters given and assuming a gyromagnetic frequency of 1 GHz, this results in a value of $\alpha = 0.036$, which is within the approximate range of 0.01 to 0.1 given in [6]. However, whilst this latter value is that which is generally quoted for α, one is reminded that this value was determined for a colloidal suspension of nickel [16]. With some ferrofluid samples it is possible to have two loss-peaks in the susceptibility profile [17], indicating the existence of two particle size distributions in the suspension, or indeed if the second loss-peak occurs in the region of approximately 1 kHz, the presence of large aggregates. The results obtained here over the frequency range 10 Hz to 300 kHz show no evidence that such a situation prevails and that the ferrofluid sample is apparently one that is well dispersed.

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