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RAPID COMMUNICATION

Measurement of the Néel relaxation of magnetic particles in the frequency range 1 kHz to 160 MHz

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Abstract. Measurements of complex susceptibility have been made on three magnetic fluids containing very small magnetic particles of median radii 2.6, 3.05 and 3.6 nm with narrow particle-size distribution. The data presented are the most detailed yet reported in the frequency range 1 kHz to 160 MHz and the first to show virtually complete loss peaks (imaginary component of the complex susceptibility) for particles relaxing by the Néel mechanism.

Up to the present time, measurements of the AC complex susceptibility, $\chi(\omega)$, of magnetic fluids [1, 2], where $\chi(\omega) = \chi'(\omega) - i\chi''(\omega)$, have in general been restricted to the frequency region below 13 MHz. Unfortunately, for studies of colloidal suspensions of magnetic particles (magnetic fluids) the frequency region of particular interest, i.e. where the Néel relaxation of the magnetic moments of the particles occurs, is to be found at and above this limiting frequency. As a result, the loss peak in $\chi''(\omega)$, the imaginary component of the susceptibility, has not always been observed in its entirety. In the cases where the maximum in $\chi''(\omega)$ has been observed at or just below 13 MHz a lack of confidence in the measurements has existed because of the fact that this frequency lies at the extreme edge of the operational frequency range of the experimental measuring system [3].

For magnetic fluids, two relaxation mechanisms can occur, one by rotational Browning diffusion characterized by a relaxation time τ_B where

$$\tau_B = 3V^1\eta/kT \qquad (1)$$

and the other by Néel relaxation with a relaxation time τ_N[4], where

$$\tau_N = f_0^{-1}(KV/kT)^{-1/2}\exp(KV/kT) \qquad (2)$$

where V^1 and V are the hydrodynamic and magnetic volumes respectively, η is the viscosity, K is the magnetic anisotropy constant and f_0^{-1} has a value of approximately 10^{-9} s.

For values of the median radius of the lognormal volume fraction of particles between 2.5 and 5 nm, the frequency f_{max} at which the maximum in the loss peak ($\chi''(\omega_{max})$) occurs, assuming Néel relaxation only, is given by $\omega\tau_N = 1$ which for the values quoted occurs approximately between 100 MHz and 1 MHz respectively, depending on the value of K. For fluids containing small particles, say 2.5 nm, and using typical values of M_s (saturation magnetization per unit volume) and K for magnetite, the assumption that only Néel relaxation need be considered is valid. This is not necessarily so for large particles (>5 nm). For larger particle size distributions, the larger particles will undoubtedly relax by the Brownian mechanism but the frequencies at which this would occur are generally well below the frequencies expected for Néel relaxation.

In the systems studied here, namely 3 colloidal suspensions of magnetite in a hydrocarbon, values of M_s and K are known approximately. The former can be obtained from a knowledge of the saturation moment of the fluid and the latter by measurement of the thermal decay of remanence. A typical value of M_s for particles of magnetite is approximately 0.44 T and typical values of K are in the region 10^4 to 4×10^4 J m^3 [5, 6].

Here we report on the measurement of the complex susceptibility of three magnetic fluids containing particles having a lognormal volume distribution with median radii 2.6, 3.05 and 3.6 nm and standard deviation $\sigma = 0.2$, in the frequency range 1 kHz to 160 MHz, using a measuring technique similar to that described previously [3]. These measurements have provided much more reliable information about the

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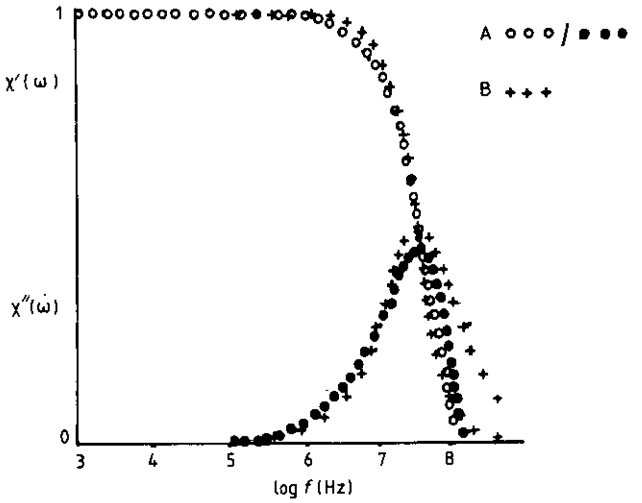

Figure 1. (A) Normalized plot of $\chi'(\omega)$ and $\chi''(\omega)$ against log f Hz for a 3.05 nm sample; (B) Debye profile.

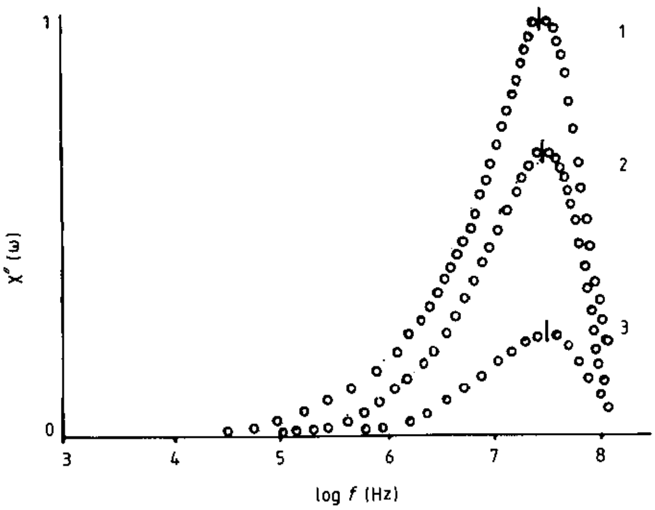

Figure 2. Normalized plots of $\chi''(\omega)$ against log f Hz for three samples: (1) 3.6 nm; (2) 3.05 nm; and (3) 2.6 nm.

shape and frequency maximum of the loss peak ($\chi''(\omega)$) than has been obtained hitherto.

Figure 1 shows a plot of $\chi'(\omega)$ and $\chi''(\omega)$ against $\log f$ for the 3.05 nm sample, whilst figure 2 shows plots of $\chi''(\omega)$ against $\log f$ for the three samples. The loss peaks occur at 34, 31 and 29 MHz for increasing particle size.

Some very limited measurements performed to high frequencies (10^8 Hz) have been reported previously [7]. However, the data presented here are to our knowledge the most detailed measurements made at high frequencies and the first that show virtually complete loss peaks for particles relaxing by the Néel mechanism.

The theory developed by Debye to account for the anomalous dielectric dispersion in dipolar fluids has been used [1, 3] to account for the analogous magnetic case of magnetic fluids. Debye's theory holds for spherical particles when the magnetic dipole–dipole interaction energy U_m is small compared with the thermal energy kT. This condition is satisfied for the three fluids studied here where $U_m/kT = 0.06$ to 0.2.

According to Debye's theory,

$$\chi(\omega) = \chi_0/(1 + i\omega\tau_N) \tag{3}$$

$$= [\chi_0/(1 + \omega^2\tau_N^2)] - i[\omega\tau_N\chi_0/(1 + \omega^2\tau_N^2)] \tag{4}$$

where χ_0 is the static susceptibility.

This expression gives the theoretical maximum of $\chi''(\omega_{max})/\chi_0$ to be 0.5. From figure 1 it is seen that the experimental value is 0.46, very close to the ideal Debye model value. In fact, the solid line shown in figure 1 is the actual Debye profile assuming a single particle size distribution for a value of $K = 1.1 \times 10^4\ \mathrm{J\,m^3}$.

Such measurements also open up the possibility of verifying the expression for f_0 derived by Aharoni (8) shown in equation (5), where

$$f_0 = \gamma_0 K M_s^{-1} \tag{5}$$

where γ_0 is the magnetogyric ratio and f_0^{-1} is usually quoted to be a constant equal to $10^9\ \mathrm{s^{-1}}$ [4].

In order to obtain accurate measurements of f_0, it is necessary to know the value of KV accurately. This is rather difficult because not only do all magnetic fluids have a distribution of particle size but they also have a distribution of K. The latter distribution arises because the shape of the particles varies considerably from particle to particle giving rise to a distribution of shape anisotropies that contribute to the values of K.

Recent measurements on frozen magnetic fluids by El-Hilo *et al* [6] have shown that it is possible to use a blocking temperature model, originally developed by Dormann [9], to obtain a mean blocking temperature and standard deviation, thereby providing a measure of the distribution of energy barriers (KV) for the system. Typical blocking temperatures are usually less than 100 K. Thus, although values of KV would be appropriate for AC measurements at these low temperatures, their appropriateness for room temperature measurements will depend on whether the shape contribution to K is dominant.

In order to pursue the verification of Aharoni's expression a programme of work is currently being planned to undertake susceptibility measurements at frequencies greater than those reported here and at low temperatures (>4 K).

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