J. Phys. D: Appl. Phys. **21** (1988) 533–534. Printed in the UK

LETTER TO THE EDITOR

The field dependence of the complex frequency-dependent susceptibility of magnetic fluids

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Received 21 December 1987

Abstract. A technique for the measurement of the field dependence of the complex frequency-dependent susceptibility of magnetic fluids together with some results for a colloidal dispersion of magnetite are reported.

Measurements of the real $\chi'(\omega)$ and imaginary $\chi''(\omega)$ components of the AC susceptibility of magnetic fluids (ferrofluids) have been reported [1–3] using a novel technique described by Fannin and co-workers [1].

Here a development of this technique is described, which enables the AC susceptibility to be measured in the presence of a constant polarising magnetic field, and some preliminary results are presented.

The test cell consists of a slot cut in a toroid of very high permeability, around which is wound a coil for exciting the alternating field. Measurements of the impedance of the coil, with the slot empty and filled with a magnetic fluid, enables values of $\chi'(\omega)$ and $\chi''(\omega)$ to be obtained [1]. On the toroid is also wound a second coil, comprising two turns, connected to a stabilised DC supply, to provide the biasing magnetic field, H_0. Between the biasing coil and the power supply is a network which ensures that the AC susceptibility measurements are unaffected. Fields of up to 9 kA m^{-1} can be achieved in the slot using a current of 1.5 A.

Several magnetic fluids have been studied using this technique but for the sake of brevity the result of only one sample, namely a colloidal suspension of magnetite in water, with a saturation magnetisation of 8×10^{-3} T, is presented here. The variations of $\chi'(\omega)$ and $\chi''(\omega)$ with frequency ($\omega/2\pi$) and biasing field are presented in figures 1 and 2.

It has been shown [1–4] that the theory of Debye [5] for the complex permittivity of polar dielectrics can be applied to ferrofluids, and that the following relationships hold:

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

$$\chi(\omega) = \chi'(\omega) - \mathrm{i}\chi''(\omega) \qquad (2)$$

$$\chi_0 = nm_{\mathrm{p}}^2/3kT\mu_0 \qquad (3)$$

and

$$\tau_{\mathrm{eff}} = 4\pi\eta r^3/kT \qquad (4)$$

where n is the number density of particles, m_{p} is the particle magnetic moment, η is the dynamic viscosity of the carrier liquid, r is the hydrodynamic radius of the particle, and τ_{eff} is the effective relaxation time.

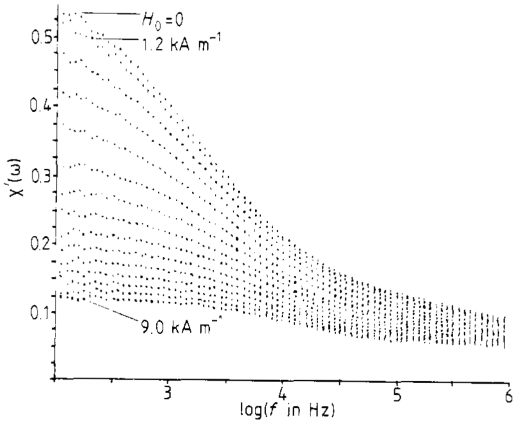

Figure 1. A plot of $\chi'(\omega)$ versus log(frequency) for polarising fields of $H_0 = 0$ and from $H_0 = 1.2$–9 kA m^{-1} in steps of 0.6 kA m^{-1}.

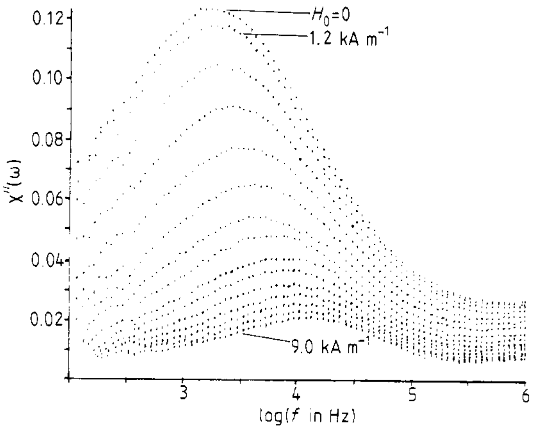

Figure 2. A plot of $\chi''(\omega)$ versus log(frequency) for polarising fields of $H_0 = 0$ and from $H_0 = 1.2$–$9\,\mathrm{kA\,m^{-1}}$ in steps of $0.6\,\mathrm{kA\,m^{-1}}$.

Assuming a Langevin function for the magnetisation of the fluid, an expression for the field dependence of the AC susceptibility $\chi(\omega, H_0)$ can be written as follows

$$\begin{aligned}\chi(\omega, H_0) &= \chi_0(1 + f(H_0))/(1 + \mathrm{i}\omega\tau_{\mathrm{eff}})\\ &= [\chi_0(1 + f(H_0))/(1 + \omega^2\tau_{\mathrm{eff}}^2)]\\ &\qquad - \mathrm{i}[\omega\tau_{\mathrm{eff}}\chi_0(1 + f(H_0))/(1 + \omega^2\tau_{\mathrm{eff}}^2)] \qquad (5)\end{aligned}$$

with

$$(1 + f(H_0)) = 3\left[1 + \left(\frac{kT}{m_{\mathrm{p}}H_0}\right)^2 - \coth^2\left(\frac{m_{\mathrm{p}}H_0}{kT}\right)\right] \qquad (6)$$

where k is the Boltzmann constant and T is the absolute temperature.

Equation (6) predicts a reduction in both $\chi'(\omega)$ and $\chi''(\omega)$ with increasing biasing field as shown in figures 1 and 2.

Plots of $\chi'(\omega)$ as a function of H_0 for a range of frequencies (0.03 Hz to 1 MHz) are given in figure 3.

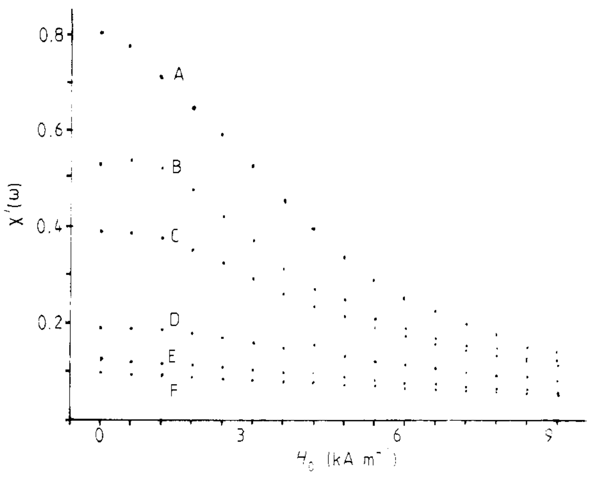

Figure 3. A plot of $\chi'(\omega)$ versus polarising field, H_0, for the frequency range 0.03 Hz to 1 MHz. Curves: A, 0.03 Hz; B, 100 Hz; C, 1.0 kHz; D, 10 kHz; E, 0.1 MHz; F, 1.0 MHz.

These curves have the same form as that predicted by equation (6). A detailed analysis of these curves which includes the particle-size distribution in the colloid will be the subject of a much more detailed publication.

Included in figure 3 is the curve for the zero frequency (actually 0.03 Hz) susceptibility $\chi'(0, H_0)$ obtained from measurement of the DC magnetisation curve for the fluid.

Examination of figure 2 shows that the frequency, $f_{\max} = \omega_{\max}/2\pi$, corresponding to the maximum value of $\chi''(\omega)$, increases with increase in biasing magnetic field. From equation (4)

$$\omega_{\max} = \tau_{\mathrm{eff}}^{-1} = kT/4\pi\eta r^3 \qquad (7)$$

so that the observed increase in $f_{\max}$ is indicative of a decrease in average particle radius from 40 to 20 nm. The large values of these radii are consistent with the presence of aggregates of particles which are typical of water-based fluid. Independent measurements of the aggregate size using photocorrelation spectroscopy [6], pulsed birefringence measurements [7] and measurements of sedimentation rates [8] are consistent with the observations made in this Letter and elsewhere.

The decrease in the average particle radius with increase in biasing field can be explained as follows. The effect of the steady magnetic field produces a greater alignment of the moments of the larger particles than the smaller ones.

Thus with increasing bias field, the larger particles are effectively prevented from contributing to $\chi'(\omega)$ and $\chi''(\omega)$, hence explaining the apparent decrease in particle size.

Measurements on other magnetic fluids produce similar results. For fluids in which little aggregation is present, the value of $f_{\max}$ is shifted to much higher frequencies ($<10^6$ Hz), consistent with a very much smaller effective particle size, nearer to the actual size of an individual particle.

A detailed analysis of all these measurements and of the curves given in figures 1, 2 and 3 is currently being undertaken.

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