

On the influence of cyclic polarising fields on the properties of magnetic fluids

P. C. FANNIN*, C. MAC OIREACHTAIGH

Department of Electronic and Electrical Engineering, Trinity College, Dublin 2, Ireland

Measurement of the frequency dependent, complex susceptibility, $\chi(\omega) = \chi'(\omega) - i\chi''(\omega)$, of magnetic fluids, by means of the toroidal technique, is a well established technique for investigating the dynamic properties of magnetic fluids, under the influence of an external polarising field, H . Here we show how data obtained from such polarised measurements, in conjunction with the Weierstrass hypothesis, can be used to determine the isotropic properties of a magnetic fluid sample. The Weierstrass hypothesis dictates that, for the fluid to be isotropic, a plot of $\chi(\omega, H)$ vs H should be symmetrical about the vertical axis. Here we demonstrate this to be so for a colloidal suspension of magnetite in water.

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1. Introduction

The study of magnetic nanoparticles has become of increasing interest in recent years due to their fundamental scientific interest and also to their suitability for use in a diverse range of applications, including magnetic data storage, magnetic seals, actuators, loudspeakers and drug targeting. In many applications the fluid is subjected to a polarizing magnetic field. Thus it is important to be able to accurately determine whether this process changes the magnetic fluids such that they do not revert to their original state, upon removal of the polarising field.

Hence there exists a need to determine if the properties of the unmagnetised fluid remain isotropic after being subjected to a cycle of magnetization. The investigations are realized through the measurement of the frequency and field -dependent, complex susceptibility, $\chi(\omega, H) = \chi'(\omega, H) - i\chi''(\omega, H)$, of the magnetic fluids, by means of the toroidal technique[1]. This technique uses a polarizing winding which enables the direction of the polarizing field to be reversed by simply reversing the polarity of the dc voltage applied to it. Furthermore from measurement of $\chi(\omega)$ at frequencies below 1 MHz, (by means of this technique) relaxation mechanisms both Brownian[2] and Néel [3], can be investigated and readily identified in any magnetic fluid sample.

Magnetic fluids consist of colloidal suspensions of nanoparticles of ferromagnetic or ferrimagnetic materials dispersed in a carrier liquid and stabilised by a suitable organic surfactant. The surfactant coating creates an entropic repulsion between particles [4], such that thermal agitation alone is sufficient to maintain them in a stable dispersion. The particles are single-domain and are considered to be in a state of uniform magnetisation with magnetic dipole moment (Wb m),

$$m = M_s v \quad (1)$$

where M_s is the saturation magnetisation (Wb/m²) of the of the material and v is the magnetic volume of the particle. When in suspension their magnetic properties can be described by the Langevin function ($L(\xi)$), suitably modified to take account of a distribution of particle sizes. The magnetisation M is described by the Langevin expression,

$$M = M_s [\coth \xi - 1/\xi]. \quad (2)$$

$\xi = mH/kT$, where k is Boltzmann's constant and H the magnetizing field.

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

$$\chi(\omega) = \chi'(\omega) - i\chi''(\omega), \quad (3)$$

is a proven [5, 6] method for determining the dynamic properties of magnetic fluids. It has been shown that the theory of Debye[7] developed to account for the anomalous dielectric dispersion in dipolar fluids may be used[8, 9, 10] to account for the analogous case of magnetic fluids. According to Debye's theory, $\chi(\omega)$ has a frequency dependence given by the equation,

$$\chi(\omega) = (\chi_0 - \chi_\infty)/(1 + i\omega\tau_{\text{eff}}) + \chi_\infty \quad (4)$$

where the static susceptibility

$$\chi_0 = (nm^2/3kT\mu_0) \quad (5)$$

n is the particle number density, τ_{eff} is the effective relaxation time, T is the temperature, μ_0 is the permeability

of free space and χ_∞ is the high frequency susceptibility at a frequency below that of resonance.

The magnetic moment of the particles may relax through either rotational Brownian motion of the particle within the carrier liquid, with relaxation time τ_B or through the Néel mechanism with relaxation time τ_N .

The Brownian relaxation time τ_B is given by

$$\tau_B = 4\pi r^3 \eta / kT$$

where r is the hydrodynamic radius of the particle, η is the dynamic viscosity of the carrier liquid and k is Boltzmann's constant.

In the case of the Néel relaxation mechanism, 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. This reversal time may be described approximately in terms of Brown's [11] expressions for high and low barrier heights, as,

$$\begin{aligned} \tau_N &= \tau_0 \sigma^{-1/2} \exp(\sigma), \quad \sigma \geq 2 \\ &= \tau_0 \sigma, \quad \sigma \ll 1. \end{aligned} \quad (7)$$

τ_0 is a damping time having an often-quoted approximate value of 10^{-9} s [12] and $\sigma = Kv/kT$, where K is the anisotropy.

Magnetic fluids have a distribution of particle sizes and have an effective relaxation time

τ_{eff} [13], which in terms of τ_B and τ_N , is given by,

$$\tau_{\text{eff}} = \tau_N \tau_B / (\tau_N + \tau_B), \quad (8)$$

the mechanism with the shortest relaxation time being dominant. τ_{eff} is related to the frequency f_{max} of the maximum of the $\chi''(\omega)$ component, by the expression,

$$\tau_{\text{eff}} = 1/2\pi f_{\text{max}}. \quad (9)$$

In the case where $\tau_N \gg \tau_B$, from (8) and (6),

$$\tau_{\text{eff}} = 1/2\pi f_{\text{max}} = \tau_B = 4\pi r^3 \eta / kT \quad (10)$$

As the work presented involves the application of a polarising field to a sample, it is appropriate to briefly mention that, assuming the Langevin function for the magnetization of the fluid, an expression for the field dependence of the A.C susceptibility, $\chi(\omega, H)$, can be written as follows[14],

$$\chi(\omega, H) = \frac{\chi_0 (1 + f(H)) - \chi_\infty}{1 + i\omega\tau_{\text{eff}}} + \chi_\infty \quad (11)$$

with,

$$(1 + f(H)) = 3 \left[1 + \left(\frac{kT}{mH} \right)^2 - \coth^2 \left(\frac{mH}{kT} \right) \right] \quad (12)$$

and where $\xi = mH/kT$.

For increasing values of polarising field, equations (11) and (12) predict, respectively,

- a reduction in both $\chi'(\omega)$ and $\chi''(\omega)$ with increasing biasing field and,
- a corresponding shift in f_{max} to higher frequencies.

These points are clearly demonstrated by theoretical plots of the real and imaginary components of equation (11) against f (Hz) as shown in Fig. 1. Here it is shown how, in response to increasing H , both $\chi'(\omega)$ and $\chi''(\omega)$ decrease in magnitude whilst there is a corresponding increase in f_{max} .

This polarizing technique has previously been successfully used to investigate the loss- tangent, heat loss[15] and also hysteresis effects in magnetic fluids[16].

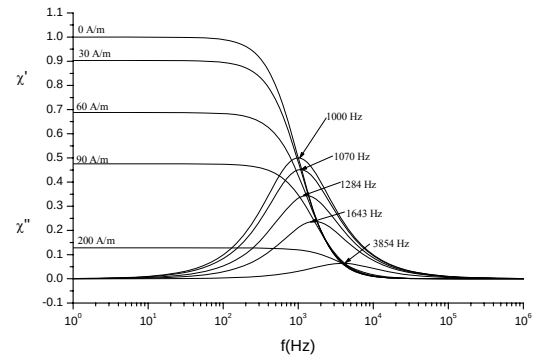


Fig 1. Theoretical plots of the real, $\chi'(\omega, H)$, and imaginary, $\chi''(\omega, H)$, components of eq (11).

The objective of this work is to investigate the isotropic properties of a magnetic fluid and this is achieved by measuring the fluids complex susceptibility, $\chi(\omega, H)$, for a cyclic variation of H from 0 up to +13.6 kA/m back down to 0, down to -13.6 kA/m and back up to 0 kA/m, over the frequency range, 100 Hz to 1 MHz.

2. Theory

If the susceptibility, χ , depends upon ω and on polarising field, H , then according to the Weierstrass hypothesis [17] we may write,

$$\chi(\omega, H) = a_0(\omega) + a_1(\omega)H + a_2(\omega)H^2 + a_3(\omega)H^3 + a_4(\omega)H^4 + \dots \quad (13)$$

and for $\omega=0$,

$$\chi(0, H) = a_0(0) + a_1(0)H + a_2(0)H^2 + a_3(0)H^3 + a_4(0)H^4 \dots \quad (14)$$

If the fluid is isotropic, then $\chi(\omega, H) = \chi(\omega, -H)$. Consequently, under such conditions,

$$a_0(\omega) + a_1(\omega)H + a_2(\omega)H^2 + a_3(\omega)H^3 + a_4(\omega)H^4 = a_0(\omega) - a_1(\omega)H + a_2(\omega)H^2 - a_3(\omega)H^3 + a_4(\omega)H^4 \dots, \quad (15)$$

which is only possible if,

$$a_1(\omega) = a_3(\omega) = a_5(\omega) = 0. \quad (16)$$

Hence we expect to be able to expand $\chi(\omega, H)$ in a series of even powers of H , i.e.

$$\chi(\omega, H) = a_0(\omega) + a_2(\omega)H^2 + a_4(\omega)H^4 + \dots + a_{2n}(\omega)H^{2n}, \quad (17)$$

which means that the graph of $\chi(\omega, H)$ vs H is symmetrical about the vertical axis. If χ is complex then we can say, $\chi(\omega, H) = \chi'(\omega, H) - i\chi''(\omega, H)$, where χ' and χ'' are the real and imaginary components of susceptibility respectively and it can readily be shown that in the case of the fluid being isotropic, a plot of the individual complex components vs H , are also symmetrical about the vertical axis.

Symmetry can be confirmed by fitting the data to a polynomial and observing whether the magnitude of the odd coefficients are zero, or close to zero as required by (16).

3. Measurements and results

Complex magnetic susceptibility measurements, over the frequency range 100 Hz to 1 MHz, were made by means of the toroidal technique [17] in conjunction with a Hewlett Packard RF Bridge 4291A. A high permeability toroid wound with twenty excitation turns was used. A second coil comprising of 3 turns was also wound on the toroid and connected to a stabilized D.C supply to provide biasing magnetic fields, H , with H being varied from 0 to ± 13.6 kA/m. Measurements were performed on a number of samples and here we present the results obtained for a 110 G colloidal suspension of magnetite in water, of mean radius 5.5nm (as obtained from electron micrograph); the surfactant was oleic acid.

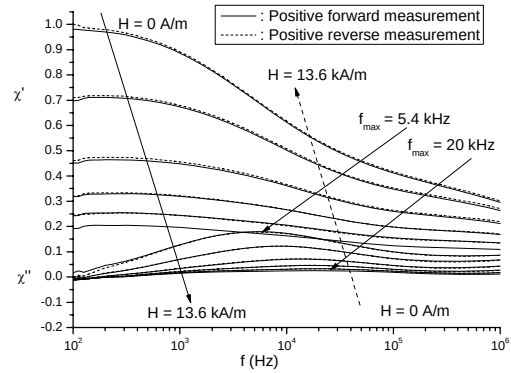


Fig 2. Plot of $\chi'(\omega, H)$ and $\chi''(\omega, H)$ (Positive forward and reverse) against f (Hz) for the sample.

Fig 2. shows six plots of $\chi'(\omega, H)$ and $\chi''(\omega, H)$ against $\omega/2\pi = f$ (Hz), obtained for the sample over the polarising field range; the polarizing field, H , being varied from 0 up to +13.6 kA/m back down to 0, down to -13.6 kA/m and back up to 0 kA/m; the approximate values of H used being, 0, 2.7kA/m, 5.5 kA/m, 8.2 kA/m, 10.9 kA/m and 13.6 kA/m. This behaviour of $\chi'(\omega, H)$ and $\chi''(\omega, H)$ as a function of frequency, is as predicted by equations (11) and (12). From Fig 2 it is apparent that there is very little difference between the profiles of $\chi'(\omega, H)$ and also those of $\chi''(\omega, H)$ at each value of H , indicating that the cyclic variation of the polarising field has not introduced any hysteresis into the sample.

Fig 3 shows the corresponding results obtained for the polarizing field, H , being varied from 0 down to -13.6 kA/m and back up to 0 kA/m; again it is apparent that there is little difference between the susceptibility profiles. Thus it is clear, that over the complete cycle of polarisation, the fluid apparently remains unaltered, i.e. it remains isotropic.

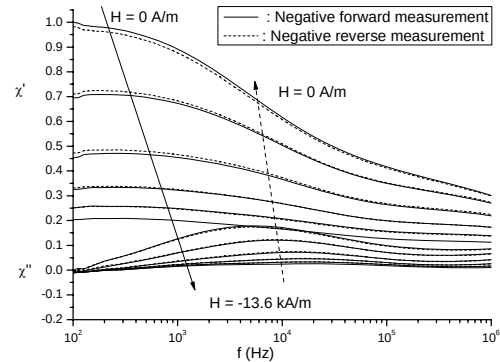


Fig 3. Plot of $\chi'(\omega, H)$ and $\chi''(\omega, H)$ (Negative forward and reverse) against f (Hz) for the sample.

From Figs. 2 and 3, it can also be seen that at low frequencies, say 100 Hz, χ'' is negligible thereby enabling the approximation $\chi'(100 \text{ Hz}) \sim \chi_0$ to be used. Thus Fig. 4 shows a plot of χ_0 against H and it is clear that the data points for each value of H are coincident and that the plots are symmetrical about the zero axis thereby confirming that the isotropic properties of the sample have been unaffected.

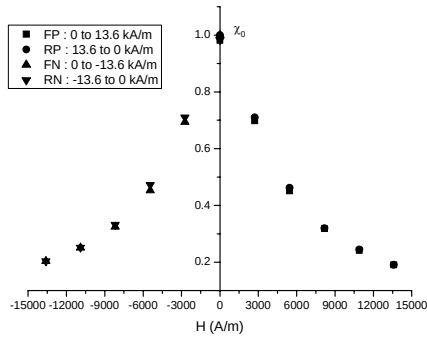


Fig. 4. Plots of χ_0 against H over polarising field range, 0 up to +13.6 kA/m (FP) back down to 0 (RP), down to -13.6 kA/m (FN) and back up to 0 (RM).

In terms of the proposed model, we recall that, for the magnetic fluid to be isotropic, $\chi'(\omega, H) = \chi'(\omega, -H)$ and $\chi''(\omega, H) = \chi''(\omega, -H)$; this fact is demonstrated by the following examples in,

i) Figs. 5(a) and 5(b) which show the χ' and χ'' data as a function of H at a frequency of 500 Hz,

ii) Figs. 6(a) and 6(b) show the χ' and χ'' data as a function of H at a frequency of 1 kHz, iii) Figs. 7(a) and 7(b) show the χ' and χ'' data as a function of H at a frequency of 10 kHz.

In all of the above cases, these plots are shown fitted to a 9th order polynomial and from inspection of the corresponding resulting odd coefficients one can determine how good an approximation is made to equation (16).

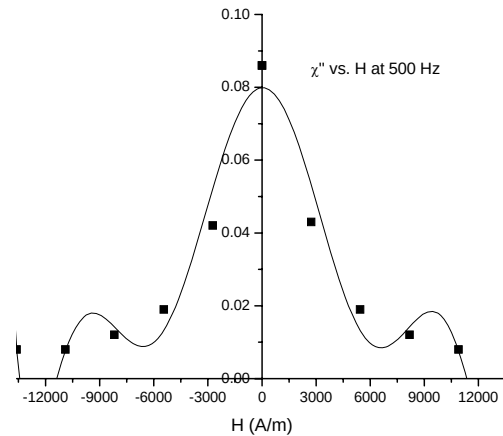


Fig. 5(b). Fit to χ'' against H data at 500 Hz.

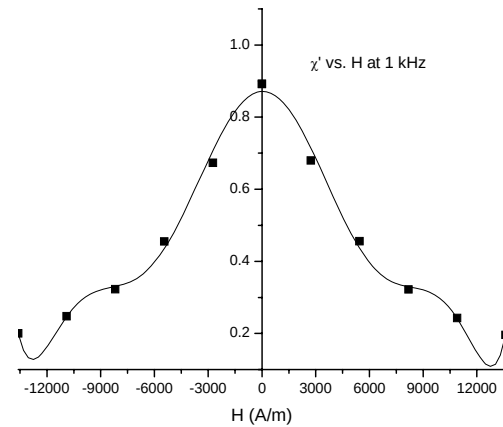


Fig. 6(a). Fit to χ' against H data at 1000 Hz.

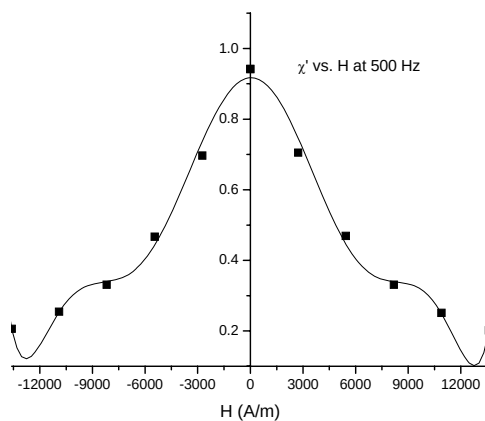


Fig. 5(a). Fit to χ' against H data at 500 Hz.

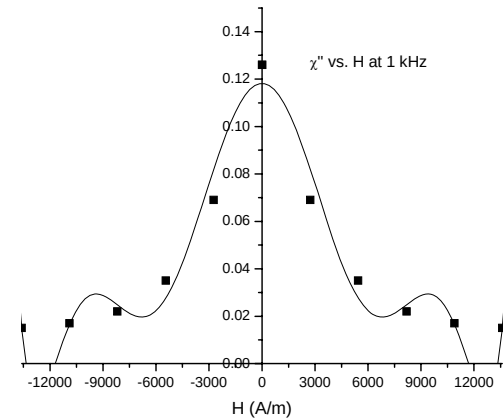


Fig. 6(b) Fit to χ'' against H data at 1000 Hz.

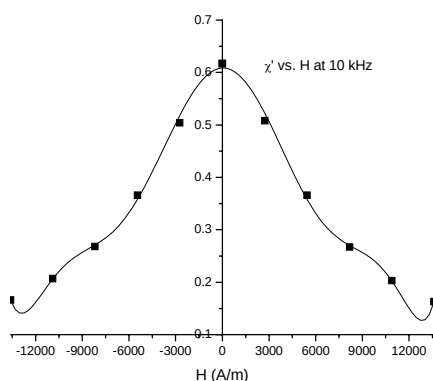


Fig 7(a). Fit to χ' against H data at 10 kHz.

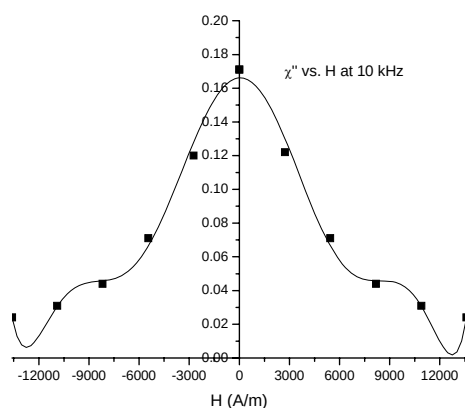


Fig 7(b) Fit to χ'' against H data at 10 kHz.

Finally, Fig 8 shows plots of χ' data as a function of H at spot frequencies of 100 Hz, 500 Hz, 1 kHz, 10 kHz, 50 kHz and 100 kHz.

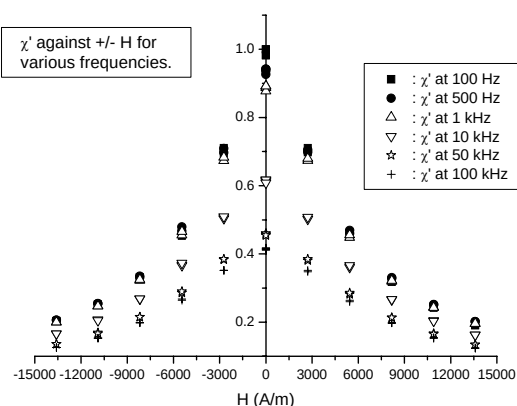


Fig. 8. Plots of χ' against H data at 100 Hz, 500 Hz, 1 kHz, 10 kHz, 50 kHz and 100 kHz.

Thus it has been clearly demonstrated that, in all the examples given, the plots are symmetrical about the vertical axis through the origin, thereby confirming that the isotropic properties of the sample have been unaffected

4. Conclusion

The object of this work was to study the possible influence which the application of a cyclic polarizing field to a magnetic fluid would have on its isotropic properties, upon removal of the field.

To this end, measurements of the frequency and field dependent complex magnetic susceptibility, $\chi(\omega, H) = \chi'(\omega, H) - i\chi''(\omega, H)$, of a water based magnetic fluid over the frequency range, 100 Hz to 1 MHz and polarizing field range, 0 to ± 13.6 kA/m were undertaken.

Plots of $\chi'(\omega, H)$, vs H and $\chi''(\omega, H)$ vs H , were shown to be symmetrical about the vertical axis, thereby satisfying the Weierstrass hypothesis, (which states that for the fluid to be isotropic, such plots must be symmetrical about the vertical axis), and confirming that the cyclic variation in H had negligible effect the isotropic properties of the fluid sample.

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*Corresponding author: pfannin@tcd.ie