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LETTER TO THE EDITOR

On the low-field, low-frequency susceptibility of magnetic fluids

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Abstract. A formula is derived for the complex, frequency-dependent, low-field relative susceptibility of a magnetic fluid which takes account of magnetic fluctuations of the intra-particle Néel-type and those induced by the rotational Brownian motion of the particles.

Magnetic fluids consist of colloidal dispersions of ferromagnetic particles. The particles being small, with radii of the order of 10 nm or less, consist of single magnetic domains (Shliomis and Raikher 1980). If I_s (Wb m^{-2}) denotes the saturation magnetisation, the magnetic moment, $\boldsymbol{m}$ (Wb m), of a spherical particle, of volume v, is given by

$$\boldsymbol{m} = I_s v \boldsymbol{s} = I_s(4\pi a^3/3)\boldsymbol{s} \tag{1}$$

where a is the radius and $\boldsymbol{s}$ is a unit vector parallel to $\boldsymbol{m}$.

The magnitude of $\boldsymbol{m}$ is constant, but its direction will fluctuate in a random manner for two reasons. Firstly, within the particle $\boldsymbol{m}$ will change from one easy direction of magnetisation to another by surmounting an energy barrier Kv (Néel 1949). Secondly, the particle being surrounded by a viscous fluid will perform rotational Brownian motion (Debye 1929) which will impose further changes in the direction of $\boldsymbol{m}$.

In a magnetic fluid in which the particle number density, N/V, is small enough to permit the neglect of magnetic dipole interaction between particles, the low-field, zero-frequency susceptibility, $\chi'(0)$, has the value

$$\chi'(0) = Nm^2/3kTV\mu_0 = \chi_s \tag{2}$$

if all particles have the same size; k and T are, respectively, the Boltzmann constant and the absolute temperature and μ_0 is the absolute permeability of free space.

For a uniaxial material, $\boldsymbol{m}$ will have two, anti-parallel equilibrium, or easy directions relative to axes fixed in the particle. If we assume that the time taken for a reversal of magnetisation is short compared with the time $\boldsymbol{m}$ spends in a particular direction, relative to the particle, we may treat these changes of sign of the magnetisation as a Poisson process. Therefore, we may characterise the magnetisation of a particle with respect to axes fixed in the particle by the function $m\,r(t)\boldsymbol{b}$, in which $\boldsymbol{b}$ is a unit vector along one of the easy directions and $r(t)$ is a stochastic function that changes suddenly, between $+1$

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and −1. As Kenrick (1929) has shown, the time average

$$\begin{aligned}\overline{r(t')r(t'+t)} &= \exp(-2\gamma|t|)\\ &= \exp(-|t|/\tau_N)\end{aligned} \tag{3}$$

in which γ is the mean residence time of the magnetisation along an easy direction and τ_N is the Néel relaxation time.

Provided inertia effects may be neglected we may treat the internal fluctuations in the sense of $\boldsymbol{m}$ and the orientational fluctuations as statistically independent. Under these circumstances the component $m_z(t') = \boldsymbol{m}\cdot\boldsymbol{k}$ of $\boldsymbol{m}$ along the space z axis at time t' may be written

$$\begin{aligned}m_z(t') &= m\,r(t')\boldsymbol{b}\cdot\boldsymbol{k}\\ &= m\,r(t')\cos\theta(t')\end{aligned} \tag{4}$$

in which $\theta(t')$ is the angle between the unit vector $\boldsymbol{b}$, fixed in the particle, and the z direction.

The autocorrelation function

$$\begin{aligned}c(t) &= \overline{m_z(t')m_z(t'+t)}\\ &= m^2\overline{r(t')r(t'+t)\cos\theta(t')\cos\theta(t'+t)}\end{aligned} \tag{5}$$

simplifies to

$$c(t) = m^2\overline{[r(t')r(t'+t)]}\,\overline{[\cos\theta(t')\cos\theta(t'+t)]}. \tag{6}$$

Debye (1929) has shown that

$$\overline{[\cos\theta(t')\cos\theta(t'+t)]} = (1/3)\exp(-|t|/\tau_D). \tag{7}$$

The Debye relaxation time, τ_D, due to Brownian motion, has the value

$$\tau_D = (4\pi\eta a^3)/(kT) \tag{8}$$

where η is the kinematic viscosity of the surrounding fluid. From (3), (6) and (7) we find that

$$c(t) = (m^2/3)\exp\{-|t|[(\tau_N + \tau_D)/\tau_N\tau_D]\} \tag{9}$$

in agreement with the conjecture of Shiliomis (1974a, b).

Using the fact that the complex, frequency ($=\omega/2\pi$)-dependent, relative susceptibility $\chi(\omega) = \chi'(\omega) - i\chi''(\omega)$ satisfies the equation (Scaife 1971)

$$\chi(\omega) = -(N/kTV\mu_0)\int_0^\infty \frac{dc(t)}{dt}\exp(-i\omega t)\,dt \tag{10}$$

we find from (9) that

$$\chi(\omega) = (Nm^2/3kTV\mu_0)(1 + i\omega\tau_{eff})^{-1} \tag{11}$$

where the effective relaxation time

$$\tau_{eff} = \tau_N\tau_D/(\tau_N + \tau_D). \tag{12}$$

Brown (1963a, b) has determined the dependence on v of the Néel relaxation time τ_N to be

$$\tau_N = \begin{cases} \tau_1 v^{-1/2} \exp(Kv/kT) & Kv > 2kT \\ \tau_2 v & Kv \ll kT \end{cases} \tag{13}$$

τ_1, τ_2 being constants independent of v. Shliomis (1974a, b) has pointed out that a critical value for the particle volume, $v_c = (4\pi a_c^3/3)$, occurs when $\tau_D = \tau_N$. So we have two possibilities:

$$\tau_{eff} \simeq \tau_D \qquad a > a_c \qquad \tau_N \gg \tau_D \tag{14a}$$

and

$$\tau_{eff} \simeq \tau_N \qquad a < a_c \qquad \tau_N \ll \tau_D. \tag{14b}$$

In the first, (14*a*), case we speak of the magnetic moment being *blocked* since $\boldsymbol{m}$ will maintain its direction, relative to axes fixed in the particle, for a long time compared with the Debye rotational relaxation time, τ_D. In the second case, (14*b*), we may ignore the effect of rotational Brownian motion on the magnetic susceptibility since the directional fluctuations of $\boldsymbol{m}$ are fast compared with the random changes in particle orientation induced by the thermal agitation of the surrounding fluid molecules.

In actual magnetic fluids there will be a range of values of a. If the inter-particle magnetic energy is small compared with kT, we may replace (11) by

$$\chi(\omega) = (I_s^2/3kT\mu_0) \int_0^\infty \frac{v^2 f(v)\, \mathrm{d}v}{[1 + \mathrm{i}\omega\tau_{eff}(a)]}. \tag{15}$$

$f(v)\,\mathrm{d}v$ is the number density of particles with volumes between v and $v + \mathrm{d}v$. Equations (11) and (15) are restricted to cases where $\chi_s \lesssim 1$, the external field h is such that $(mh/kT) < 1$, and to values of ω well below the Larmor precession frequency. The case of a uniaxial material was chosen for simplicity of exposition; the general form of (11) and (15) will apply to other classes of ferromagnetic particles.

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