Dynamics of magneto-orientational susceptibility in a viscoelastic magnetic fluid

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Abstract

Nonlinear dynamic magneto-orientational susceptibility responsible for the induced birefringence in a viscoelastic magnetic suspension is investigated. In the model, the particles are considered to be magnetically rigid and their mechanical inertia is taken into account. The rheology of the fluid matrix is of the Maxwell type. We show that the frequency spectra of the field-induced birefringence presents a number of specific lineshapes.

Measurement of a frequency-dependent response of a dilute suspension of fine magnetic particles to a weak external field seems to be a prospective method for studying micro-rheology of complex fluids. In this case the particles are well apart and, experiencing the exciting influence of the field, serve as local probes with a size ranging 10–100 nm. This spatial scale equals to that of supramolecular structures, which actually define a great many of the rheological features of a complex fluid.

To support these qualitative considerations, hereby we extend the theoretical model developed in Refs. [1–3] to describe dynamic birefringence induced by AC magnetic field in an assembly of single-domain ferroparticles in a viscoelastic fluid. Two-dimensional orientational motion of a magnetically rigid particle in a Maxwell fluid is considered. We choose the 2D representation for its simplicity. The 3D one, though far more cumbersome mathematically, should differ in results just by numerical coefficients of the order of unity [4].

In the adopted framework, a magnetic particle is a solid disk of radius \( a \) confined in an arbitrary plane passing along the direction of the applied magnetic field \( H \), which is taken for the polar axis of the coordinate system. The magnetic moment \( \mu \) of the particle has a constant absolute value and its direction inside the plane of the disk is fixed. Orientation of the vector \( \mu \) with respect to the polar axis is defined by the angle \( \vartheta \). The particle is surrounded by a viscoelastic (Maxwell) matrix whose stress relaxation time is \( \tau_M \). As it is shown in

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PII: S0304-8853(99)00035-9
Ref. [1], the Langevin equation for the rotary motion of a particle with the moment of inertia \( I \) writes

\[
\dot{\vartheta} + \frac{1}{\tau_M} \dot{\vartheta} + \frac{1}{I} \left( \frac{\zeta}{\Omega} \frac{\partial}{\partial \vartheta} + \vartheta \frac{\partial}{\partial \Omega} \right) \dot{\vartheta} = - \frac{\mu}{I \tau_M} \sin \vartheta \dot{\vartheta} + \frac{1}{I \tau_M} f(t),
\]

where \( \Omega = \dot{\vartheta} \) and \( \dot{\vartheta} = (1 + \tau_M \delta/dt) \). In Eq. (1) the noise is white:

\[
\langle f(t) f(0) \rangle = 2 \zeta T \delta(t),
\]

with \( \zeta \) being the friction coefficient of the particle.

The Fokker–Planck equation (FPE) for the distribution function \( W(\vartheta, \Omega, \dot{\vartheta}, t) \) corresponding to Eq. (1) was derived in Ref. [3]:

\[
\left( \frac{\partial}{\partial t} + \frac{\partial}{\partial \vartheta} \frac{\zeta}{\Omega} \frac{\partial}{\partial \vartheta} + \frac{\partial}{\partial \Omega} \frac{\partial}{\partial \Omega} \right) W(\vartheta, \Omega, \dot{\vartheta}, t)
= \frac{\zeta}{T^2 \tau_M^2} \frac{\partial^2}{\partial \Omega^2} W + \frac{\partial}{\partial \Omega} W
\times \{ [I \dot{\vartheta} + \zeta + \mu T M H \cos \vartheta] \Omega
+ \mu \sin \vartheta (H + \tau_M \dot{\vartheta}) \} W.
\]

It has the equilibrium solution

\[
W_{eq}(\vartheta, \Omega, \dot{\vartheta}) = Z^{-1} \exp \left[ - \frac{\Omega^2}{2 T} - \frac{\tau_M}{2 \zeta} I \vartheta^2 \right.
+ \mu H \sin \vartheta \frac{\vartheta^2}{2} + \frac{\mu H}{T} \cos \vartheta \left. \right],
\]

with the partition function

\[
Z = \frac{4 \pi^2 T}{I} \left( \frac{\zeta}{I \tau_M} \right)^{1/2} J_0(\zeta),
\]

where \( J_0(\zeta) \) is the modified Bessel function of the Langevin argument \( \zeta = \mu H / T \).

The distribution (4) defines the intrinsic scales for the angular velocity and acceleration:

\[
\langle \vartheta^2 \rangle = \Omega^2 / 2 = T / I, \quad \langle \vartheta^2 \rangle = \Omega^2 \dot{\vartheta} / 2 = T / (I \tau_M)
\]

where \( \tau_I = I / \zeta \) is the relaxation time of the particle angular momentum. Passing to the dimensionless variables

\[
v = \Omega / \Omega_T, \quad w = \dot{\vartheta} / \dot{\vartheta}_T,
\]

we seek the time-dependent solution of FPE (3) in the form

\[
W(\vartheta, \Omega, \dot{\vartheta}, t) = W_0(v, w)[1 + \Psi(\vartheta, \Omega, \dot{\vartheta}, t)],
\]

\[
W_0(v, w) = \pi^{-1} \exp(-v^2 - w^2),
\]

where \( |\Psi| \ll 1 \). Function \( \Psi \) is expanded in a power series in \( \zeta \) up to the second order. In a harmonic field the corresponding terms are

\[
\Psi^{(1)} = e^{-i \xi n} \zeta^{(0)} \sum_{q= \pm 1} A_{m,n}(q, \omega) e^{i q \vartheta} H_m(v) H_n(w),
\]

\[
\Psi^{(2)} = e^{-2 i \xi n^2} \zeta^{(0)} \sum_{q= \pm 2} B_{m,n}(q, \omega) e^{i q \vartheta} H_m(v) H_n(w),
\]

where \( H_m(x) \) are Hermite polynomials, and \( \zeta^{(0)} \) and \( \zeta^{(0)} \) are the corresponding static dipole and quadrupole susceptibilities with respect to \( \zeta \). According to the general definition of a complex magnetic susceptibility of the rank \( q \)

\[
\langle \cos q \vartheta \rangle = \zeta^q \chi_{2q}(\omega) \exp(-i q \omega t),
\]

from Eqs. (9) and (10) follow the expressions

\[
\chi(\omega)/\chi(0) = A_{0,0}(1, \omega), \quad \chi_{2q}(\omega)/\chi_{2q}(0) = B_{0,0}(2, \omega).
\]

For the case of a magnetic suspension, provided the particles are just slightly anisometric, the quadrupole susceptibility \( \chi_{2q} \) determines the dynamic field-induced birefringence, see Ref. [5], for example.

Despite a simple look of Eq. (11), evaluation of the functions \( A_{0,0} \) and \( B_{0,0} \) for a viscoelastic system is rather painstaking. Indeed, on substituting Eqs. (8) and (9) in Eq. (3) one gets an infinite set of chain-linked moment equations. Due to that, to determine the lowest coefficients, the whole set should be solved. For that we use a procedure based on the methods of matrix backward sweeping by Risken [6]. In the obtained results we conventionally distinguish three regimes of the dynamical response. Their qualitative differences are evident from Figs. 1 and 2 and are discussed below.

Weak viscoelasticity limit is defined as \( \tau_M \ll T_D \), where \( T_D = \zeta / T \) is the Debye relaxation time - the only one that determines the orientational response in a Newtonian magnetic suspension. From Eq. (9) upon using a minimally sufficient basis, analytical
For the dipolar one it is determined by condition $m + n \leq 1$ and yields:

$$\chi(\omega)/\chi(0) = \frac{1 - i\omega\tau_M}{i\omega^2\tau_D\tau_D^* - \omega^2\tau_D - i\omega(\tau_M + \tau_D) + 1}. \quad (12)$$

For the quadrupolar susceptibility, the condition of minimal sufficiency expands to $m + n \leq 2 \ [7]$. This yields for $\chi_2$ an expression of the type (12) but with a sixth-power polynomial in $\omega$ in the denominator. It happens to be too cumbersome to be presented in a short paper.

**Developed viscoelasticity** corresponds to the range $\tau_M \sim \tau_D$. There the susceptibility spectra are pronouncedly comb-like. The basic eigenfrequency $\omega_M = 1/\sqrt{\tau_D\tau_M}$ owes to the interplay of the particle inertia and the dynamic elasticity of the matrix. The comb-like shape is constructed of the multiples of $\omega_M$. To evaluate those lineshapes one has to essentially use the numerical approach, since no small parameters are available. Note that the condition of being positive applies only to $\text{Im} \chi$. The quadrupole term $\text{Im} \chi_2$ does not need to obey it.

**Strong viscoelasticity** means that $\tau_M \gg \tau_D$. Backtracing this condition to FPE, it can be shown that it allows to omit the diffusion term there. By that we come to the case known as the collisionless limit. The analytical technique for it is developed in the kinetic theory of plasma [8]. The finiteness of the obtained linewidths is due to the averaging of the particle rotational trajectories over the equilibrium thermal distribution and is known as the Landau damping. Thence the comb-like shapes smooth down into a one-cusp relaxational curve that is controlled by a single parameter that is the thermal frequency, see Eq. (6). The pertinent asymptotic formulas write ($x = \omega/\Omega_T$):

$$\chi(\omega)/\chi(0) = 1 - xe^{-x^2} \left(2 \int_0^x e^y dy - i\sqrt{\pi}\right), \quad (13)$$

$$\chi_2(\omega)/\chi_2(0) = 1 - x^2 - x \left(\frac{3}{2} - x^2\right)e^{-x^2}$$

$$\times \left(2 \int_0^x e^y dy - i\sqrt{\pi}\right), \quad (14)$$

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*Journal of Magnetism and Magnetic Materials 201 (1999) 211–214*
We remark that both expressions satisfy the corresponding Kramers–Kröenig relations.

From Figs. 1 and 2 one can see that the simple contours rendered by Eqs. (13) and (14) (they are drawn by dashes in the intermediate figures) give the positions and widths of the intricate 'wavepackets' occurring in the regime of developed viscoelasticity, accurately enough. Given that, we expect that for polydisperse systems where the fine structure will be considerably suppressed, those are the asymptotic contours which would always dominate the spectral lineshapes.

This work was partially supported by Grants 98–02–16453 from RFBR and 96–0663 from INTAS.

References