Viscoelastic ferrocolloid modelled as the Jeffreys fluid: dynamic magnetic susceptibility in the presence of a bias field

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Abstract. Orientational Brownian motion of a solid nanoparticle embedded in a viscoelastic medium modelled by the Jeffreys fluid is studied. The particle bears a magnetic moment and is subjected to a combination of a constant (bias) and a weak (probing) AC magnetic fields. First, the set of Langevin equations of the problem is presented and then on that basis a corresponding kinetic equation (KE) is derived. The solution of the KE that is linear with respect to the AC field amplitude is worked out. This solution is virtually exact since it is cast as a series that enables one to obtain the result with arbitrary accuracy. In that framework, a closed expression for the dynamic magnetic susceptibility of a statistical ensemble of magnetic particles in a viscoelastic environment is found, analyzed and compared to the previously existed approximate solution. It is shown that: (i) the dynamic elasticity of the medium has a substantial effect on the magnetic spectra; (ii) to correctly describe this effect, one has to retain a considerable number of terms in the series that presents the KE solution.

1. Introduction

For a long time, the Jeffreys model [1] works as a very helpful tool in phenomenological rheology. Comprising only three parameters, it is able to mimic a wide range of polymer media: from weak solutions to gel-like systems. That is why we chose it to investigate the essential features of magnetic nanosuspensions with viscoelastic carriers. This approach enables one to deal with the challenging problem: to advance the theory of magnetodynamic response of ferrofluids beyond the case of ordinary systems (colloids in Newtonian liquids) to a wide variety of magnetic nanosuspensions whose base behaves viscoelastically. The problem is especially important since the modern trend is to use field-driven magnetic nanoparticles as drug delivery agents [2, 3, 4, 5], heat mediators [6, 7, 8, 9], immunoassay sensors [10, 11, 12, 13] and nanosurgery tools [14, 15] in biotechnology and medicine by embedding them in, for example, analytical animal / human tissue samples, blood, lymph, cell cytosol, etc., whose rheology is far from that of simple liquids.

Another aspect of application of magnetic nanoparticles is the use in laboratory technique for active microrheology, the method where the particles are introduced in small quantities to the media which rheology is unknown or needs to be time-monitored in the course of the chemical processes undergoing in them. In this method the nanoparticles are set to motion by means of an applied magnetic field (e.g., rotary motion), and the induced dynamic magnetization signal is used for analyzing the mechanical properties of the particle environment [16, 17, 18, 19, 20].
The common feature of all those approaches is that the magnetic response of nanoparticles is strongly affected by Brownian motion which is the more intense the smaller the particles. Therefore, any correct theory ought to take this factor into account.

Given the above-presented considerations, in this paper we investigate the orientational motion of a single-domain ferromagnet particle suspended in the Jeffreys fluid. The particle with typical size about $10^{-20}$ nm is assumed to be magnetically hard that means that its magnetic moment $\vec{\mu}$ has a constant length $\mu$ and is “frozen” in the particle body. Because of that, unit vector $\vec{e} = \vec{\mu}/\mu$ in below is used as the marker of the particle orientational motion.

The particle is surrounded by the Jeffreys fluid whose mechanical properties are schematized by the Newtonian and Maxwellian rheological elements connected in parallel [1], see Fig. 1. The Newtonian element has viscosity coefficient $\eta_N$; the Maxwellian one is a chain comprising the viscous element with coefficient $\eta_M$ and a spring that models shear elasticity with modulus $G$. On a particle of hydrodynamic volume $V$, upon its motion, this fluid exerts an elastic restoring torque proportional to $G$ and two interacting viscous torques proportional to $\eta_N$ and $\eta_M$.

![Figure 1. Schematic representation of a single-domain particle in a Jeffreys fluid.](image)

2. Equations of motion and kinetic equation

On the nanoscale, the inertia effects are negligible, and, thus, the set of Langevin equations that governs the orientational motion of an embedded nanoparticle has the form [21]:

$$\frac{\partial}{\partial t} \vec{\omega} = \left( \vec{\Omega} \times \vec{e} \right),$$

$$\frac{\partial}{\partial t} \vec{\Omega} = \frac{1}{\zeta_N} \left[ -\vec{L}U + \vec{Q} + \vec{y}_N(t) \right], \quad \vec{L} = \left( \vec{e} \times \frac{\partial}{\partial \vec{e}} \right),$$

$$\left( 1 + \tau_M \frac{\partial}{\partial t} \right) \vec{\Omega} = -\zeta_M \vec{\Omega} + \vec{y}_M(t).$$

Here $\vec{e}$ is unit vector of the particle orientation, $\vec{\Omega}$ its angular velocity, $U$ the energy of particle interaction with the applied magnetic field, and $\vec{L}$ operator of infinitesimal rotation. The response coefficients of the medium are set by the conventional microrheology relations [16]:

$$\zeta_\sigma = 6\eta_\sigma V, \quad K = 6GV, \quad \sigma = N, M,$$

The random forces in equations (1) are defined via the fluctuation-dissipation theorem and delta-correlated (white noise):

$$\langle y_{i\sigma}(t)y_{j\sigma}(t + \tau) \rangle = 2T\zeta_\sigma \delta_{ij} \delta_{\sigma\sigma'} \delta(\tau) \quad \sigma = N, M;$$

in this formula and further, temperature is taken in energy units, i.e., $k_B = 1$. 

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In the set (1) phase variable $\tilde{Q}$ is the retarded mechanical response of the medium. In physical sense, vector $\tilde{Q}$ is the torque exerted on the particle on the part of the dynamic quasi-mesh the nodes of which has reference lifetime $\tau_M = \zeta_M/K = \eta_M/G$. As mentioned, in the Jeffreys fluid there are two dissipative mechanisms working in parallel. One of them is linear (Newtonian) viscous friction that accounts for the particle interaction with the low-molecular component of the medium. Another one is the retarded interaction of the Maxwell viscoelastic type that resembles the hindered motion of the particle through the polymer environment. Note that in the limit $\tau_M \to \infty$, the Jeffreys model tends asymptotically to the case of a real gel whose mesh is not physical but chemical. Indeed, upon that limiting transition, the torque $\tilde{Q}$ simplifies to: $\tilde{Q} = -K\tilde{\Omega}$ pointing out that the response of the gel mesh to mechanical perturbation has only elastic component [22]. In this particular case and for planar rotation of the particle the set (1) reduces to a 1D Kelvin model considered in [22].

The kinetic equation for the particle orientation distribution function $W(\tilde{e}, t)$ is obtained from the set (1) via a standard procedure, see [21, 23, 24], for example:

$$\frac{dW}{dt} = \left[ \frac{1}{\zeta_N} \left( \hat{L}W\hat{L} + \frac{\partial}{\partial \tilde{Q}} W \frac{\partial}{\partial \tilde{Q}} - \hat{L}W \frac{\partial}{\partial \tilde{Q}} - \frac{\partial}{\partial Q} W\hat{L} \right) + \frac{1}{\zeta_M} \frac{\partial}{\partial \tilde{Q}} W \frac{\partial}{\partial \tilde{Q}} \right] \times \left( U(\tilde{e}) + \frac{1}{2} KQ^2 + T \ln W \right);$$

(4)

here and further we use nondimensional phase variable $\tilde{Q} = Q/K$ but omit tilde for the sake of brevity. The equilibrium solution of Eq. (4) is rendered by the extended Boltzmann distribution

$$W_0 \left( \tilde{e}, \tilde{Q} \right) \propto \exp \left\{ -\frac{1}{T} \left[ U(\tilde{e}) + \frac{1}{2} KQ^2 \right] \right\}. \quad (5)$$

Note an essential feature of the system under study: in equilibrium – both in the absence and in the presence of a constant magnetic field – distribution (5) does not contain cross terms, i.e., the phase variables are statistically independent.

The orientation-dependent potential of a particle with a “frozen-in” magnetic moment is

$$U = -\mu H(\tilde{e} \cdot \tilde{h}), \quad \tilde{h} = \tilde{k} + \varepsilon(t) \tilde{p}, \quad \tilde{h}^2 = 1, \quad \tilde{p}^2 = 1. \quad (6)$$

In Eq. (6) we assume that the magnetic field comprises a constant (bias) part $H\tilde{k}$ directed along $Oz$ and an AC part $\tilde{H}_1 = H\varepsilon(t)\tilde{p}$ with $|\varepsilon(t)| \ll 1$. The constant component endows the system with uniaxial anisotropy, so that the response to a weak probing field is characterized by a susceptibility tensor

$$\chi_{ik} = n\mu \partial \langle e_i \rangle / \partial H_{1k}, \quad (7)$$

where $n$ is particle number concentration.

We evaluate susceptibility (7) by way of averaging the stochastic equations (1); with the potential $U$ this takes the form

$$\frac{\partial}{\partial t}\tilde{e} = \gamma_H (\tilde{e} \times (\tilde{h} \times \tilde{e})) + \gamma_N (\tilde{Q} \times \tilde{e}) + (\tilde{u} \times \tilde{e}), \quad \gamma_N = K/\zeta_N, \quad \gamma_H = \mu H/\zeta_N, \quad (8)$$

$$\frac{\partial}{\partial t}\tilde{Q} + \gamma \tilde{Q} = -\gamma_H (\tilde{e} \times \tilde{h}) + \tilde{u}, \quad \tilde{u} = \tilde{y}_N(t)/\zeta_N, \quad \tilde{u} = \tilde{y}_M/\zeta_M - \tilde{u}; \quad \gamma = \gamma_N + \gamma_M = \gamma_N (1 + 1/q);$$

where $q = \eta_M/\eta_N$ is the viscoelasticity parameter.

With the explicit form of potential, the equilibrium distribution function (5) takes the form

$$W_0 \left( \tilde{e}, \tilde{Q} \right) = \frac{1}{Z_\xi Z_Q} \exp \left\{ \xi \left( \tilde{e} \cdot \tilde{k} \right) - \frac{1}{2} \beta Q^2 \right\}, \quad Z_\xi = 4\pi \sinh \xi, \quad Z_Q = (2\pi/\beta)^{3/2}. \quad (9)$$
Here two nondimensional parameters are introduced: \( \xi = \mu H/T \) for the field amplitude, and \( \beta = K/T \) for inverse temperature.

The statistical moments of function (9) are expressed as

\[
c_n = \left\langle \left( M \mathbf{e} \right)^n \right\rangle = \frac{2\pi}{Z\xi} \int_{-1}^{1} dx x^n \exp(\xi x) = \frac{e^\xi + (-1)^{n+1} e^{-\xi}}{e^\xi - e^{-\xi}} - \frac{n c_{n-1}}{\xi} \quad ; \quad c_0 = 1. \tag{10}
\]

As the system at \( \varepsilon = 0 \) is uniaxially anisotropic with \( \mathbf{k} \) being the axis direction, one may introduce for it an orthogonal set of projection tensors

\[
P_{ij}^\parallel = k_i k_j, \quad P_{ij}^\perp = \delta_{ij} - k_i k_j, \quad P_{ij}^{(\alpha)} P_{jk}^{(\beta)} = P_{ik}^{(\alpha)} \delta_{\alpha\beta}, \quad \alpha = \parallel, \perp,
\]

in terms of which all the matrix operations reduce to simple scalar ones:

\[
A_{ij} = A_{\parallel} P_{ij}^\parallel + A_{\perp} P_{ij}^\perp, \quad A_{ij}^{-1} = A_{\parallel}^{-1} P_{ij}^\parallel + A_{\perp}^{-1} P_{ij}^\perp, \quad A_{ij} B_{jk} = A_{\parallel} B_{\parallel} P_{ij}^\parallel + A_{\perp} B_{\perp} P_{ij}^\perp. \tag{12}
\]

As the distribution of phase variable \( Q \) remains isotropic independently of the field presence, all its statistical averages of odd order vanish, whereas for the even-order ones we have

\[
\left\langle (Q_j)^{2n} \right\rangle_0 = \frac{1}{ZQ} \int d\tilde{Q} (\tilde{Q})^{2n} \exp(-\beta Q^2/2) = \tilde{T}^n(2n + 1)!!, \quad \left\langle (Q_j)^{2n} \right\rangle_0 = \tilde{T}^n(2n - 1)!!, \quad j = x, y, z.
\]

The probing magnetic field perturbs the equilibrium, and the distribution function \( W \) becomes time-dependent. The linear response of the system under study in the absence of bias field was studied in Ref. [21]. It turns out, however, that the functional basis developed for that case is usable as well when a constant field is on. Indeed, from the set (8) it follows that the statistical moments are linked to each other only due to the retarded friction. For instance, to solve equation for the first basis function which is of zero order with respect to \( Q \), the first-order function \( (\tilde{Q} \times \mathbf{e}) \) is required. Taking, in turn, the equation for the latter, one finds that it involves the function of second order in \( \tilde{Q} \) and so forth \textit{ad infinitum}. A thorough analysis reveals that the whole set of basis functions splits in three mutually orthogonal sub-sets (sub-bases):

\[
\tilde{M}_{2n+1} = \Psi_n^{(2)}(Q^2) \tilde{M}, \quad \tilde{K}_{2n} = \Psi_n^{(0)}(Q^2) \mathbf{e}, \quad \tilde{L}_{2n+2} = \Psi_n^{(4)}(Q^2) \left[ (\tilde{Q} \times \mathbf{e}) \tilde{Q} - (1/3) \tilde{Q}^2 \mathbf{e} \right]. \tag{14}
\]

Inside each sub-base, the functions are orthogonalized stepwise along As seen, the Gram-Schmidt procedure [25]. Requiring that the non-equilibrium distribution function is normalized, one finds the first function of the sub-basis \( K \) in the form \( \tilde{K}_0 = \delta \mathbf{e} = \mathbf{e} - \left\langle \mathbf{e} \right\rangle_0 \). Scalar factors \( \Psi_n^{(p)}(Q^2) \) which define the sub-basis functions of higher order are polynomials of argument \( Q^2 \). The power of \( \Psi_n^{(p)} \) in this argument is indicated by the subscript (i.e., \( n \)) whereas the superscript (i.e., \( p \)) is the power of the “weight” factor needed to make those polynomials mutually orthogonal:

\[
\left\langle Q^p \Psi_n^{(p)} \Psi_m^{(p)} \right\rangle_0 = \left\langle \left( \Psi_n^{(p)} \right)^2 Q^p \right\rangle_0 \delta_{nm}. \tag{15}
\]

as it follows from definitions (14) of the basis functions. The details of constructing the full basis and the technique to work with it are partially given in [21] and in detail presented in [26].

In terms of the above-described functional bases, the non-equilibrium solution of kinetic equation (4) takes the form

\[
W(\mathbf{e}, \tilde{Q}, t) = W_0(\mathbf{e}, \tilde{Q}) \left\{ 1 + \sum_{n=0,1,2,\ldots} \left[ a_n(t) \tilde{K}_{2n} + b_n(t) \tilde{M}_{2n+1} + c_n(t) \tilde{L}_{2n+2} \right] \right\}. \tag{16}
\]
Averaging the first basis function with distribution (16), one finds the explicit form of the first coefficient in that expansion:

$$
\alpha^{(0)}_i(t) = \left[ \frac{P_{ij}^\parallel}{c_2 - c_1^2} + \frac{2P_{ij}^\perp}{1 - c_2} \right] (\delta e_j). \quad (17)
$$

By progressive application of that procedure, one is able to obtain any number of expansion coefficients in Eq. (16), i.e., to evaluate the non-equilibrium distribution function with arbitrary accuracy. As soon as $W(\tilde{c}, \tilde{q}, t)$ is found, it could be used for deriving the observable magnetic moment, i.e., $\langle \tilde{c} \rangle$. A plausible way to do that is to expand phase variable $\tilde{c}$ with respect to the above-introduced set of basis functions $\tilde{K}_j$, $\tilde{M}_j$ and $\tilde{L}_j$. Then the non-equilibrium average $\langle \tilde{c} \rangle$ comes out as a series of equilibrium averages of those functions, whereas the coefficients of this series are rendered by the solution of an appropriate matrix equation of order $N$ that couples all the coefficients. This treatment is done taking equation (4) as a starting point. Skipping this very cumbersome work, in the next section we present the main results of this treatment.

3. Dynamic magnetic susceptibility

To the corresponding matrix equation a harmonic probing field $\varepsilon(t)H\tilde{p} = \varepsilon H \exp(-i\omega t)\tilde{p}$ is substituted. Linearization with respect to $\varepsilon$ yields the response at frequency $\omega$, the equations for the longitudinal and normal projections of the dynamic magnetic moment with respect to axis $\tilde{k}$ have the same general form

$$
R^{(\alpha)}_{ij}\, \chi^{(\alpha)}_j = \chi^{(\alpha)}_i, \quad \alpha = ||, \perp. \quad (18)
$$

The exciting force and response vectors in Eq. (18) are defined as

$$
\begin{align*}
(X^{(\alpha)}_j)^T &= (\langle \delta \tilde{c}, \langle \tilde{M}_1 \rangle, \langle \tilde{K}_2 \rangle, \langle \tilde{L}_3 \rangle, \langle \tilde{K}_4 \rangle, \langle \tilde{L}_5 \rangle, \ldots \rangle^{(\alpha)}, \\
(Y^{(\alpha)}_j)^T &= \varepsilon f^{(\alpha)}_i (1, -1, 0, 0, 0, \ldots); \quad f^{(1)} = \frac{1}{2} (1 - c_2), \quad f^{(\perp)} = \frac{1}{4} (1 + c_2). \quad (19)
\end{align*}
$$

where superscript $T$ denotes matrix transposition. The initial fragment of matrix $\hat{R}$ (8-th order is taken as an example) is

$$
\begin{align*}
\begin{pmatrix}
\varepsilon(\tilde{c}) & -\frac{1}{2}\beta & 0 & 0 & 0 & 0 & 0 & 0 \\
-g(\tilde{M}, \tilde{c}) & g(\tilde{M}) & \frac{1}{3}\beta & -\frac{1}{2}\beta & 0 & 0 & 0 & 0 \\
0 & -g(\tilde{M}, \tilde{K}) & g(\tilde{K}) & 0 & -\frac{1}{2}\beta & 0 & 0 & 0 \\
0 & -\frac{2}{3}g(\tilde{L}, \tilde{M}) & \frac{1}{3}\beta & \frac{1}{2}\beta & 0 & 0 & 0 & 0 \\
0 & 0 & -\frac{2}{3}g(\tilde{M}, \tilde{K}) & \gamma(\tilde{M}, \tilde{L}) & -\frac{1}{3}\beta & -\frac{1}{3}\beta & 0 & 0 \\
0 & 0 & 0 & 2\gamma(\tilde{K}, \tilde{M}) & g(\tilde{K}) & 0 & -\frac{1}{2}\beta & 0 \\
0 & 0 & 0 & 0 & -\frac{2}{3}\gamma(\tilde{L}, \tilde{M}) & g(\tilde{L}) & 0 & -\frac{1}{2}\beta \\
0 & 0 & 0 & 0 & 0 & -\frac{2}{3}\gamma(\tilde{M}, \tilde{K}) & 2\gamma(\tilde{M}, \tilde{L}) & g(\tilde{L})
\end{pmatrix}^{(20)}
\end{align*}
$$

The projection index that was omitted for brevity in (20), is recovered right below. Thus specified expressions for the diagonal elements of $\hat{R}$:

$$
g^{(\alpha)}_m(\tilde{B}) = \frac{m\beta}{2} \left( 1 + \frac{1}{q} \right) + \gamma^{(\alpha)}(\tilde{B}) - i\omega \tau D, \quad (21)
$$
where \( \tau_D = \zeta_N/2T \) is the Debye rotary relaxation time and the \( \gamma^{(a)} \) terms are

\[
\gamma^{(c)}(\vec{e}) = \frac{1 - c_2}{2(c_2 - c_1^2)}, \quad \gamma^{(M)}(\vec{M}) = \gamma^{(K)}(\vec{K}) = \frac{1 + c_2}{2(1 - c_2)}, \quad \gamma^{(L)}(\vec{L}) = \frac{3 - c_2}{2(1 + c_2)},
\]

\[
\gamma^{(M,\vec{e})} = \frac{1 - c_2}{2(c_2 - c_1^2)}, \quad \gamma^{(M,\vec{M})} = \frac{1}{\gamma^{(M,\vec{K})} - 1 + c_2}, \quad \gamma^{(M,\vec{L})} = \frac{1}{\gamma^{(M,\vec{K})} - 2(1 - c_2)}, \quad \gamma^{(M,\vec{L})} = \frac{1}{\gamma^{(M,\vec{K})} - 7 - c_2}.
\]

Those for the off-diagonal elements also depend on the strength of bias field, they are

\[
\gamma^{(M,\vec{e})} = \frac{1 - c_2}{2(c_2 - c_1^2)}, \quad \gamma^{(M,\vec{M}')} = \frac{1}{\gamma^{(M,\vec{K})} - 1 + c_2}, \quad \gamma^{(M,\vec{L})} = \frac{1}{\gamma^{(M,\vec{K})} - 2(1 - c_2)}, \quad \gamma^{(M,\vec{L})} = \frac{1}{\gamma^{(M,\vec{K})} - 7 - c_2}.
\]

In general, the field-induced anisotropy does not affect the way of linking of statistical moments, it remains the same as in the isotropic case [21]. However, the matrix elements below the main diagonal are different as they become bias-field dependent.

To solve Eq. (18) by backward sweep method (see [21, 27, 28], for example), we cast it in the form of recurrence relation

\[
C^{-2}_n X_n - B^{-1}_n X_{n-1} + A_n X_n + B^+_n X_{n-1} + C^+_n X_{n-2} = Y_n.
\]

(22)

From the above-given matrix fragment (20), it follows that the coefficients of this equation could be presented as respective components of vectors

\[
A = \left( g_0(\vec{e}), g_1(\vec{M}), g_2(\vec{K}), g_3(\vec{M}), g_4(\vec{L}), g_5(\vec{M}), g_6(\vec{K}), g_6(\vec{L}), g_6(\vec{M}, \vec{M}), \ldots \right),
\]

\[
B^- = \left( 0, -\gamma(\vec{M}, \vec{e}), \gamma(\vec{K}, \vec{M}), 0, \gamma(\vec{M}, \vec{L}), \gamma(\vec{K}, \vec{M}), 2\gamma(\vec{K}, \vec{M}), 2\gamma(\vec{K}, \vec{M}), 0, 3\gamma(\vec{M}, \vec{L}), \vec{L} \right),
\]

\[
C^- = -\frac{1}{6} \left( 0, 0, 0, 0, 0, 5\gamma(\vec{L}, \vec{M}), 10\gamma(\vec{K}, \vec{M}), 0, 17\gamma(\vec{L}, \vec{M}), 14\gamma(\vec{K}, \vec{M}), 0, 9\gamma(\vec{L}, \vec{M}), 18\gamma(\vec{K}, \vec{M}), \ldots \right),
\]

\[
B^+ = \frac{1}{2} \beta \left( -3, 2, 0, 1, 2, 0, 1, 2, 0, 1, 2, \ldots \right), \quad C^+ = -\frac{1}{2} \beta \left( 0, 1, 1, 0, 1, 0, 1, 0, 1, 0, 1, \ldots \right),
\]

\[
Y = f(1, -1, 0, 0, 0, 0, 0, 0, 0, 0, 0, \ldots).
\]

(23)

Upon substitution an expression with two sweep coefficients and a free term \(- X_n = S_n X_{n-1} + P_n X_{n-2} + a_n \) the five-term recurrence formula (22) reduces to a three-term one. After that, by comparison with (22) one arrives at the sweep equations

\[
S_n = -\left( B^- + \left( B^+ + C^+ S_{n+2} \right) P_{n+1} \right) / Z_n, \quad Z_n = A_n + S_n + 1 + \left( B^+ + C^+ S_{n+2} \right) P_n + 1, \quad P_n = C^- / Z_n, \quad a_n = \left[ Y_n - (B^+ + C^+ S_{n+2}) a_{n-1} - C^+ a_{n-2} \right] / Z_n.
\]

(24)

To derive an expression for the magnetic susceptibility, we combine the first line of (18) and the sweep relation for the second response component: \( g_0 X_1 = -\frac{1}{2} \beta X_2 = Y_1 \) and \( X_2 = S_2 X_1 + a_2 \). Sweep coefficient \( S_2 \) is obtained from recurrence relations (24) under uniform initial conditions \( S_{N+1} = P_{N+1} = a_{N+1} = 0 \), i.e., assuming that a statistical moment is the smaller the higher its order. Then varying the largest order of the moment that is taken into account gives a convenient way to control the precision of the performed calculations. In our work \( N = 10 \) ensured very good accuracy of the results.

With explicit expressions for the coefficients of Eq. equations (22), one finds, first, \( S_2 = \gamma(\vec{c}) / Z_2, \ a_2 = -Y_1 / Z_2 \) and, finally, the non-equilibrium magnetic moment \( X_1 \). Using definition (7) and restoring all the temporally omitted notations, one arrives at the susceptibility expression

\[
\chi^{(a)}(\omega, \xi) = \frac{\chi^{(a)}(0,\xi)}{\omega \tau_D}, \quad \chi^{(0,\xi)} = \frac{\pi \mu^2}{T} (c_2 - c_1^2), \quad \chi^{(0,\xi)} = \frac{\pi \mu^2}{T} (1 - c_2).
\]

(25)
here the components of equilibrium (static) susceptibility $\chi(0,0,0)$ do not depend on the relaxation mechanism and have the same form as in Refs. [29, 28].

An approximate expression for $\chi^{(a)}(\omega,\xi)$ could be obtained, as it is done in Refs. [30], in a much simpler way if to use only the first two statistical moments, i.e., to set $N = 2$. In the model under study, the viscoelastic properties of carrier medium enter the dynamic equations via the retarded friction. Because of that, the minimal set of phase variables, along with the observable quantity $\langle \varepsilon \rangle$, includes the torque $\langle \tilde{Q} \rangle$. Given that, the general formula (25) reduces to [30]:

$$
\tilde{\chi}^{(a)}(\omega,\xi) = \frac{1}{1 - i\omega\tau^{(a)}} \left( 1 + \frac{\frac{1}{2} \frac{\beta}{q} - i\omega \tau_D}{\gamma^{(a)}(\tilde{M}) + \frac{1}{2} \frac{\beta}{q} - i\omega \tau_D} \right), \quad \tau^{(a)} = \frac{\tau_D}{\gamma^{(a)}(\varepsilon)}. \quad (26)
$$

In this case the frequency behavior of the susceptibility is entirely determined by the effective elasticity $\beta$ of the medium and viscoelasticity parameter $q = \eta_M/\eta_N$. As already mentioned, at high temperatures ($\beta \ll 1$) the viscoelasticity is weak, so that the susceptibility expression reduces to a simple Debye formula with the relaxation time $\tau_D$.

For a Jeffreys matrix with substantial elasticity ($\beta \gg 1$) and under a relatively low bias field, the susceptibility formula (26) may be effectively presented as a sum of just two relaxation modes, slow ($s$) and fast ($f$):

$$
\tilde{\chi}^{(a)}(\omega,\xi) \simeq \frac{1 - 2\gamma^{(s)}(\varepsilon)/\beta}{1 - i\omega\tau_s} + \frac{2\gamma^{(s)}(\varepsilon)/\beta}{1 - i\omega\tau_f}, \quad \tau_s = \frac{\tau_D}{\gamma^{(s)}(\varepsilon)} \left[ 1 + \frac{\beta/2}{\gamma^{(s)}(\tilde{M}) + \beta/2q} \right], \quad \tau_f = \frac{2\tau_D}{\beta} = \frac{\zeta_N}{\beta}; \quad \beta > 2\gamma^{(s)}(\varepsilon), \beta \gg 1. \quad (27)
$$

We discuss the behavior of the obtained dynamic (complex) susceptibility in terms of its imaginary component $\text{Im}\tilde{\chi}^{(a)}$ that is responsible for the AC field energy absorption. In Fig. 2 along with the lines of $\text{Im}\tilde{\chi}^{(a)}$ calculated with the exact solution of KE (long series), we present the effective field (two-term) approximation from Ref. [30].

As seen, qualitatively, the approximate plots are similar to the exact ones. In particular, with the increase of matrix elasticity, the slow component of $\text{Im}\tilde{\chi}^{(a)}$ grows in magnitude whereas its peak goes to yet lower frequencies. Meanwhile, the fast component of $\text{Im}\tilde{\chi}^{(a)}$ decreases and moves to the high frequency end. Note that the shift of the low-frequency peak, although more pronounced in exact solution, is anyway very slow. From asymptotic analysis it follows that its position get indeed close to the limiting point $\omega\tau_s = \omega\tau_D(1 + q)/\gamma^{(a)}(\varepsilon)$ only at $\beta \gg 2q$, i.e., $\tau_D \gg \tau_M$. For typical viscoelastic fluids where $q \sim 10^2 - 10^3$ (see [31], for example) and the more so for gels ($q \to \infty$), this limit cannot be attained.

With the growth of bias field, the relaxation rate of the slow mode increases (its cusp moves to high-frequency end) while its height becomes smaller. Meanwhile, the position of the fast-mode peak remains fixed, and just the magnitude of the peak enhances. In a strong bias field and at weak-to-moderate elasticity $\beta < 2\gamma^{(a)}(\varepsilon)$, approximation (28) tends to a simple expression

$$
\tilde{\chi}^{(a)}(\omega,\xi) = \frac{1}{1 - i\omega\tau^{(a)}}, \quad \tau^{(a)} \approx \frac{\tau_D}{\gamma^{(a)}(\varepsilon)} \approx \left\{ \begin{array}{ll} \tau_H = \zeta_N/\mu H, & \alpha = \perp, \\ \tau_H/2, & \alpha = \parallel; \end{array} \right\}; \quad \xi \gg \beta/2. \quad (28)
$$

which reproduces the result for dynamic susceptibility of magnetically hard particles under strong bias field in a linearly viscous (Newtonian) matrix [29]. In this limit, only the fast relaxation mode that is forced by the imposed field is working whereas all the viscoelasticity effects are negligible.
Figure 2. Normalized frequency dependences of the imaginary component of dynamic magnetic susceptibility obtained by exact solution (top row, a and b) and in mean-field approximation (bottom row, c and d) for the cases of moderate $\beta = 3$ (left column, a and c) and strong elasticity $\beta = 10$ (right column, b and d); the curves in each pane correspond to dimensionless constant fields $\xi = 0.1$ (1), 1 (2), 3 (3) and 10 (4).

4. Conclusions
Appearance of the low-frequency (slow) relaxation mode besides the customary high-frequency one, is a novel effect emerging in our model, i.e., in a system where the carrier fluid is the Jeffreys viscoelastic medium. In fact, this low-frequency mode had been already found in Ref. [30] but, as it was just a low-order (effective field) approximation, its existence could not be considered as unambiguous. Now, we have demonstrated that the exact solution of the KE yields the same result. Thus, the developed high-order approximation (one may justly consider its accuracy as infinite) has shown that: (i) the slow mode really exists and (ii) the frequency dependence $\text{Im} \tilde{\chi}^\parallel(\omega)$ always has a characteristic two-peak shape.

Summing up the results, we note the following. Good general agreement between the exact and approximate plots of $\text{Im} \tilde{\chi}^\parallel$ at low values of $\beta$ (weak elasticity of the medium) is quite expectable because only by means of $\beta$ the dynamics of the particle is affected by the retarded friction. The right-hand (high-frequency) wings of the exact and approximate lines in Fig. 2 are very similar whatever the medium elasticity, see Fig. 2. To understand that, it suffices to recall that in the model under study the most rapid relaxation process is caused by the usual Stokes mechanism (viscosity coefficient $\eta_N$) that works almost independently of the retardation one. However, if the Jeffreys media has substantial elasticity, the exact and approximate susceptibility profiles $\text{Im} \tilde{\chi}^\parallel$ display notable differences. In particular, in the exact solution the leftward shift of low-frequency peak is much more pronounced than in the effective-field approximation. In other
words, in complex fluid media with substantial dynamic elasticity the contributions of higher moments (they are related to the viscoelastic relaxation) are not at all negligible.

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