

Dynamic susceptibilities of an assembly of dipolar particles in an elastic environment

Yu. L. Raikher,^{1,*} V. V. Rusakov,¹ W. T. Coffey,² and Yu. P. Kalmykov^{3,4}

¹Laboratory of Complex Fluids, Institute of Continuous Media Mechanics of the Urals Branch of the Russian Academy of Sciences, 614013, Perm, Russia

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

³Centre d'Etudes Fondamentales, Université de Perpignan, 52 Avenue de Villeneuve, 66860, Perpignan Cedex, France

⁴Institute of Radio Engineering and Electronics of the Russian Academy of Sciences, 141120, Fryazino, Moscow Region, Russia

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Theoretical model to describe magnetodynamics of a ferrogel, i.e., an assembly of ferromagnetic nanoparticles embedded in a gel, is proposed. The reorientations of the particles are determined by the influence of the elastic matrix and the rotational Brownian motion. Due to the interplay between these two factors, the main parameter characterizing the static magnetic susceptibility of the system is the ratio of the elastic modulus of the matrix times particle volume to the thermal energy. It is shown that the main components of the dynamic magnetic-susceptibility tensor are determined by the combinations of the reference rates of several processes inherent to the system, namely, the elastic restoration of the particle orientation, Brownian rotary diffusion, and viscous relaxation of the particle angular momentum. In the framework of the model, absorption of the energy of an alternating external field by a ferrogel is studied. With allowance for the ever present interaction of elastic and Brownian forces, the effective relaxation times for the longitudinal and transverse components of the ferrogel magnetization are evaluated.

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I. INTRODUCTION

The properties of composites obtained by embedding magnetic nanoparticles in easily deformable (soft) structures (sometimes called *complex magnetic fluids*) are rather fascinating. On the one hand, having been incorporated into supramolecular structures, such particles can produce new magnetically controlled (smart) materials, e.g., ferroliquid crystals [1–3] or ferrogels [4,5]. On the other hand, when distributed over the bulk of the matrix in small amounts, the same particles can be used as magnetically driven mechanical microprobes thus allowing one to obtain information on the rheology of the carrier media on the scale comparable with the particle size, that is ≥ 10 nm [6]. Indeed, single-domain particles possess a high specific dipolar moment so that the response of the system to a probing ac field may be easily recorded even with relatively unsophisticated equipment. However, one needs an appropriate theoretical framework to analyze the data, i.e., it is necessary to understand what physical mechanisms contribute essentially to the magnetodynamic spectra of soft magnetic systems.

When a disperse system is sufficiently dilute—which is definitely so if the objective is to investigate the properties of the matrix itself—the main contribution to the spectra comes from the behavior of *individual* particles. Unlike the situation in more coarse suspensions, in nanosystems the orientational dynamics is always a result of joint action of the deterministic (applied field, elasticity, and viscosity of the matrix) and fluctuational (Brownian diffusion) factors. This circumstance is of vital importance [6,7] for the understanding of the dynamical spectra of complex magnetic fluids.

In what follows we consider the orientational dynamics of an assembly of noninteracting Brownian dipolar particles residing in an elastic matrix. We remark, that unlike the studies [6,7] of fluid matrices with a nonequilibrium elasticity (the Maxwell fluid), our objective here is a magnetic spectrum of a ferrogel. Therefore, we consider a matrix that possesses a true (equilibrium) elasticity. The particles are assumed to be magnetically hard and the coupling of an individual particle with the matrix is modeled by an orientational potential that is a quadratic function with respect to the angular deviation of the particle magnetic moment from its equilibrium direction. The latter is imposed by the matrix, and depending on the history of the sample, two limiting cases are conceivable. (a) If the gel forms in the presence of a strong magnetic field, this direction would be the same for all the particles. Then the system is macroscopically anisotropic (uniaxial) and is characterized by a symmetrical susceptibility tensor with two main components—the longitudinal (χ_{\parallel}) and transverse (χ_{\perp}) ones—which are found by summation of the corresponding single-particle contributions. (b) If the gel was created in the absence of a field, the particle axes distribution is random, and the equilibrium direction of a particle magnetic moment exists only as a mesoscopic parameter. Then, the system as a whole is isotropic, and its macroscopic susceptibility is constructed according to the well-known rule, see Ref. [8], for example, as a weighted average $\bar{\chi} = \frac{1}{3}(\chi_{\parallel} + 2\chi_{\perp})$.

As a dissipative part of the particle interaction with the matrix, we take the viscous friction in the Stokes approximation. To simplify the theory, the particle is modeled by a plane rotator, so that its only degree of freedom is the angle ϑ describing the deviation of the particle dipolar moment $\boldsymbol{\mu}$ from its equilibrium direction. Such a scheme is in common use, see Ref. [9], for example, in the theory of dielectric response of polar fluids.

*Corresponding author. FAX: +7 3422 336957. Email address: raikher@icmm.ru

II. REFERENCE TIMES

The equation of the orientational motion for a particle with a moment of inertia I writes

$$I\ddot{\vartheta} = -\frac{\partial U}{\partial \vartheta} + Q(t), \quad (1)$$

where the first term on the right-hand side is the regular torque produced by the elastic environment of the particle and the external field H . Assuming a harmonic (quadratic) potential, we have

$$U = \frac{1}{2}K\vartheta^2 - \mu(H_{\parallel}\cos\vartheta + H_{\perp}\sin\vartheta), \quad (2)$$

where the indices of the field components correspond to the directions $\vartheta=0$ and $\vartheta=\pi/2$.

In the Stokes approximation, the torque Q in Eq. (1) incorporates the usual friction torque and the white noise [9]:

$$Q(t) = -\zeta\dot{\vartheta} + y(t), \quad \langle y(t)y(t') \rangle = 2\zeta T\delta(t-t'), \quad (3)$$

where the term $y(t)$ accounts for the thermal motion of a particle at temperature T (the Boltzmann constant is set hereafter to unity). We assume that by the order of magnitude $\zeta = 6\eta v$, i.e., ζ is the drag coefficient of a spherical particle of volume v in a liquid with the viscosity η .

With the elastic potential (2), the dissipation mechanism (3), and in the absence of an external field, Eq. (1) has the form of the Langevin equation for a torsional oscillator

$$I\ddot{\vartheta} + \zeta\dot{\vartheta} + K\vartheta = y(t). \quad (4)$$

The ratio of the inertial torque to the viscous one in Eq. (4) defines the first of the reference times of the problem

$$\tau_I = I/\zeta. \quad (5)$$

During this interval the particle angular velocity thermalizes, i.e., the velocity distribution function assumes the Maxwell form [10].

In an elastic carrier medium, one may introduce the natural frequency of the torsional oscillation of the particle

$$\omega_K = \sqrt{K/I} = 1/\sqrt{\tau_K\tau_I}. \quad (6)$$

In combination with τ_I it defines the reference time of elastic restoration, namely,

$$\tau_K = \zeta/K = (\omega_K^2\tau_I)^{-1}. \quad (7)$$

Since the Brownian particle is in equilibrium with its environment (heat bath), one can introduce one more reference time scale defining the thermal frequency

$$\omega_T = \sqrt{T/I} = 1/\sqrt{\tau_D\tau_I}. \quad (8)$$

In turn, combination of ω_T with τ_I renders the well-known Debye time of the thermal orientational relaxation

$$\tau_D = \zeta/T. \quad (9)$$

Thus, the model system possesses three independent parameters with the dimensions of time. Moreover, as is shown below, in a typical nanosuspension (magnetic fluid, for example) these times differ from one another by several orders of magnitude. Under such circumstances, it should be expected that the response of the system to an external field $H(t)$ essentially depends on the interrelation between the period of the excitation and the reference times of the problem. However, before commencing the analysis of the possible response modes, let us check the range of applicability for the Stokes approximation. As is well known [11], this approach resumes a small Reynolds number. For a particle of a size a moving with the velocity $u = \omega a$, where ω is the oscillation frequency, this condition is

$$\text{Re} \sim \rho_f a u / \eta \sim \rho_p a^5 \omega / \zeta \sim \omega \tau_I \ll 1, \quad (10)$$

where we set that the density of the liquid matrix ρ_f and that of the particle ρ_p have the same order of magnitude. Substituting the reference frequencies—elastic ω_K from Eq. (6) and thermal ω_T from Eq. (8)—it appears that the Stokes approximation (10) is valid as long as

$$\tau_I \ll \tau_K, \tau_D, \quad (11)$$

i.e., the inertial time should be the smallest of all the reference times.

It is convenient to introduce the dimensionless times of the Debye (thermal) and elastic relaxation as

$$t_D = \tau_D/\tau_I = \zeta^2/TI, \quad t_K = \tau_K/\tau_I = \zeta^2/KI, \quad (12)$$

so that condition (11) corresponds to $t_D, t_K \gg 1$. The parameter t_D for suspensions has often been estimated, and the condition $t_D \gg 1$ may be written as the inequality $T/a\eta^2 \ll 1$, which for $T \lesssim 10^3$ K and $\eta \gtrsim 10^{-2}$ P holds as soon as $a \gtrsim 10^{-9}$ cm, i.e., under all the conceivable conditions.

To estimate the time t_K , we first remark that the elasticity modulus of a polymeric gel is of the high elasticity (entropy) origin, so that in order of magnitude it is $E \sim \nu T$, where ν is the number of links in a unit volume. From dimensional considerations it follows $K \sim E\nu \sim \nu T\nu$, that is the elastic constant is the temperature times the number of links in the volume of the matrix that is equal to that of the particle. We assume that the particle ‘‘senses’’ the presence of an elastic matrix if, having been embedded there, it dislodges at least one link, that is when $\nu v \sim 1$. Then $K \gtrsim T$, so that t_K has the same order of magnitude as t_D , for which case the inequality (11) is already proven. For the particles of the size $a \sim 10$ nm, the condition $\nu v \sim 1$ renders $E \sim 0.4$ atm, so that the value obtained, namely, $\nu \sim 10^{18}$ cm $^{-3}$ is close to that of real gel-formation thresholds [12] in polymers. However, this estimation needs to be used carefully since, in a certain sense, it yields the upper boundary of the tested range. Indeed, it assumes that the particle interacts with the matrix only in a steric (excluded-volume) way. Therefore, this mechanism becomes effective only for the networks that are so dense that on the average the interchain cell is too small for a particle. On the other hand, had the particles been ad-

sorbed on the junctions or chain segments of the network, the value of νv may become much lower than unity.

III. EQUILIBRIUM FLUCTUATIONS OF THE ORIENTATION ANGLE AND ANGULAR VELOCITY

In order to obtain the equilibrium correlation function for the orientation angle, we use the Langevin equation (4). The general solution of the homogeneous equation is sought in the form $\vartheta(t) \propto \exp(\lambda t)$ yielding the characteristic equation

$$\lambda^2 + \frac{1}{\tau_I} \lambda + \omega_K^2 = 0, \quad (13)$$

whose roots, determining the rates of the relaxation processes in the system, are

$$\lambda_{1,2} = -\frac{1}{2\tau_I} [1 \pm \sqrt{1 - 4(\omega_K \tau_I)^2}]. \quad (14)$$

Using definition (12) for the dimensionless elastic time and the fact that in the physically relevant range $t_K \gg 1$, we may simplify Eq. (14), by setting

$$\sqrt{1 - 4(\omega_K \tau_I)^2} \approx \sqrt{1 - 4/t_K} \approx 1 - 2/t_K - 2/t_K^2. \quad (15)$$

This transforms formulas (14) into

$$\lambda_1 = -\frac{1}{\tau_I} \left(1 - \frac{1}{t_K}\right), \quad \lambda_2 = -\frac{1}{\tau_K} \left(1 + \frac{1}{t_K}\right), \quad (16)$$

which clearly demonstrates a marked difference in the magnitude of the roots.

The general solution of Eq. (4) with allowance for the initial conditions $\vartheta(0) = \vartheta$ and $\dot{\vartheta}(0) = \Omega$ is

$$\begin{aligned} \vartheta(t) = & \frac{\vartheta}{\Delta\lambda} (\lambda_1 e^{\lambda_2 t} - \lambda_2 e^{\lambda_1 t}) + \frac{\Omega}{\Delta\lambda} (e^{\lambda_1 t} - e^{\lambda_2 t}) \\ & + \frac{1}{I\Delta\lambda} \int_0^t (\exp \lambda_1(t-u) - \exp \lambda_2(t-u)) y(u) du, \end{aligned} \quad (17)$$

where $\Delta\lambda = \lambda_1 - \lambda_2$. To find the correlation function, we multiply Eq. (17) by the initial value ϑ of the orientational angle and take the average over the equilibrium ensemble. In equilibrium one has $\langle \vartheta^2 \rangle = T/K$, $\langle \vartheta \Omega \rangle = 0$, and $\langle \vartheta y(u) \rangle = 0$ for all $u > 0$. The nonzero contribution comes only from the first term so that

$$\begin{aligned} \langle \vartheta(t) \vartheta(0) \rangle = & \frac{T}{K\Delta\lambda} (\lambda_1 e^{\lambda_2 t} - \lambda_2 e^{\lambda_1 t}) \\ = & \frac{T}{K} \begin{cases} 1 - (K/2I)t^2 & \text{for } t \ll \tau_I \\ \exp(-t/\tau_K) & \text{for } t \gg \tau_I. \end{cases} \end{aligned} \quad (18)$$

Another way to obtain the correlation function that is more cumbersome but very useful methodically, is as follows. Since we construct a description for a stationary state, expression (17) should hold in the long-time range as well.

In this limit $\lambda_{1,2} t \gg 1$, so that in the general formula (17) one needs to retain only the solution of the inhomogeneous problem

$$\begin{aligned} \vartheta(t) = & \frac{1}{I\Delta\lambda} \int_{-\infty}^t (e^{\lambda_1(t-u)} - e^{\lambda_2(t-u)}) y(u) du \\ \equiv & \int_{-\infty}^t f(t-u) y(u) du. \end{aligned} \quad (19)$$

Then the correlation function takes the form of a double integral:

$$\langle \vartheta(t) \vartheta(0) \rangle = \int_{-\infty}^t du \int_{-\infty}^0 du' f(t-u) f(-u') \langle y(u) y(u') \rangle,$$

which is easily taken with the aid of Eq. (3). The result of integration once again leads to formula (18) as it must do.

Differentiating Eq. (17) with respect to time, one finds the time dependence of the angular velocity. The expression for its correlator

$$\langle \Omega(t) \Omega(0) \rangle = \frac{\langle \Omega^2 \rangle}{\Delta\lambda} (\lambda_1 e^{\lambda_1 t} - \lambda_2 e^{\lambda_2 t}) \approx \frac{2T}{I} \exp(-t/\tau_I) \quad (20)$$

is obtained just as Eq. (18) and confirms the definition of the relaxation time τ_I given above.

Comparing formulas (18) and (20) one finds that in the physically relevant range of material parameters ($\tau_K \gg \tau_I$) the relaxation goes in two stages. First, in a rather fast manner (during a time $\sim \tau_I$) the equilibrium with respect to the particle angular velocities settles. Then, in a much slower fashion (on the time scale $\sim \tau_K$) the equilibrium with respect to the orientation angle is achieved. Note that we refer only to monotonic relaxation. An oscillatory regime, although formally not forbidden, requires the condition $\omega_K \tau_I \gtrsim 1$, which, see Sec. II, cannot be realized within a physically meaningful domain of material parameters. For example, in order to get $\omega_K \tau_I \gtrsim 1$ in a gel, the number of junctions should have been increased by four orders of magnitude while the viscosity should have been kept constant at a value close to that of water.

To end this section, we find the asymptotic form of the correlation function for small times. To do that, we expand $\vartheta(t)$ in a Taylor series, multiply the expression obtained by the initial value ϑ , and take the average over the statistical ensemble. The result is

$$\langle \vartheta(t) \vartheta \rangle = \langle \vartheta^2 \rangle + \langle \dot{\vartheta} \vartheta \rangle t + \frac{1}{2} \langle \ddot{\vartheta} \vartheta \rangle t^2. \quad (21)$$

Due to ergodicity, ensemble averaging is equivalent to that over time, so that $\langle \vartheta \dot{\vartheta} \rangle = 0$ and $\langle \vartheta \ddot{\vartheta} \rangle = -\langle \dot{\vartheta}^2 \rangle$. Therefore,

$$\langle \vartheta(t) \vartheta \rangle = \langle \vartheta^2 \rangle - \frac{1}{2} \langle \Omega^2 \rangle t^2, \quad t \ll \tau_I. \quad (22)$$

This expression is usually called the dynamic limit of the angular correlation function. It could be derived also from the general equation (18) in the small-time limit. We remark that since expression (22) follows from the ergodicity hy-

pothesis and the assumption that the observation time is short, it should hold for more complicated models as well.

IV. STATIC SUSCEPTIBILITIES

The equilibrium distribution function for the particles is, as usual, given by the Gibbs law. For the potential (2) and with the standard expression $\frac{1}{2}I\Omega^2$ for the kinetic energy, one has

$$W_0(\vartheta, \Omega) = Z^{-1} \exp \left[-\frac{I\Omega^2}{2T} - \frac{K\vartheta^2}{2T} + \frac{\mu}{T}(H_{\parallel} \cos \vartheta + H_{\perp} \sin \vartheta) \right], \quad (23)$$

where Z is the partition function determined by the normalizing condition $\int d\vartheta \int d\Omega W_0 = 1$.

First, we consider the longitudinal static susceptibility, i.e., the one along the axis imposed by the elastic potential. For simplicity, we assume that this direction is the same for all the particles of the assembly. In particular, this means that even in the absence of the probing field the system has a nonzero magnetization

$$M = \mu n \langle \cos \vartheta \rangle,$$

where n is the particle number density. Switching on a magnetic field H_{\parallel} , imparts an additional orientation to the dipole moments, so that the magnetization acquires the increment

$$\delta M_{\parallel}(H) = \mu n [\langle \cos \vartheta \rangle_H - \langle \cos \vartheta \rangle]. \quad (24)$$

Here the angular brackets denote the statistical averaging and the subscript indicates the value of the magnetic field, at which it is taken. The absence of a subscript means the case of zero field. In the approximation, linear in H and with allowance for the normalizing condition, the equilibrium distribution function (23) transforms into

$$W_0 = \sqrt{\frac{K}{2\pi T}} \exp \left(-\frac{K\vartheta^2}{2T} \right) \times \left[1 + \frac{\mu H_{\parallel}}{T} (\cos \vartheta - \langle \cos \vartheta \rangle) + \frac{\mu H_{\perp}}{T} \sin \vartheta \right], \quad (25)$$

and the longitudinal susceptibility writes

$$\chi_{\parallel}(0) = \lim_{H_{\parallel} \rightarrow 0} \frac{\delta M_{\parallel}}{H_{\parallel}} = \frac{n\mu^2}{T} [\langle \cos^2 \vartheta \rangle - \langle \cos \vartheta \rangle^2]. \quad (26)$$

The equilibrium averages are evaluated easily with the aid of the formula

$$\begin{aligned} \langle \cos N\vartheta \rangle &= \sqrt{\frac{K}{2\pi T}} \int_{-\infty}^{\infty} d\vartheta \exp \left(-\frac{K\vartheta^2}{2T} + iN\vartheta \right) \\ &= \exp \left(-\frac{N^2 T}{2K} \right). \end{aligned} \quad (27)$$

Applying this to Eq. (26), one gets

$$\chi_{\parallel}(0) = \frac{n\mu^2}{T} e^{-T/K} \left[\cosh \left(\frac{T}{K} \right) - 1 \right]. \quad (28)$$

In a similar way, for $H = H_{\perp}$ we get the transverse static susceptibility

$$\chi_{\perp}(0) = \frac{n\mu^2}{T} \langle \sin^2 \vartheta \rangle = \frac{n\mu^2}{T} e^{-T/K} \sinh \left(\frac{T}{K} \right). \quad (29)$$

For vanishing elasticity ($K \ll T$), expressions (28) and (29) tend to the same limit

$$\chi_{\parallel}(0) = \chi_{\perp}(0) = n\mu^2/2T,$$

which, as it must do, coincides with the already known result for a system of free rotators [6,10].

In the opposite case of high rigidity ($K \gg T$) it follows from formula (29) that the transverse susceptibility tends to the finite athermal limit

$$\chi_{\perp}(0) = n\mu^2/K, \quad K \gg T. \quad (30)$$

For the longitudinal susceptibility the rigorous athermal limit is zero. According to Eq. (28), it is approached as

$$\chi_{\parallel}(0) = n\mu^2 T/2K^2, \quad K \gg T. \quad (31)$$

Indeed, in a rigid gel, the particle is almost perfectly oriented along the macroscopic anisotropy axis on account of its coupling to the matrix only. Therefore, application of a magnetic field along the same direction can do practically nothing for the further enhancement of orientation. From Eq. (1), in the static fluctuation-free ($Q=0$) limit one arrives at the standard equilibrium condition

$$\partial U / \partial \vartheta = 0. \quad (32)$$

For the longitudinal response ($H = H_{\parallel}$) Eq. (32) yields

$$K\vartheta + \mu H_{\parallel} \sin \vartheta = 0.$$

This equation does not have a solution that is linear with respect to the field strength implying that the longitudinal susceptibility becomes nonzero only when thermal fluctuations are taken into account, see expression (31). For the transverse susceptibility the finite athermal limit can be, of course, be obtained directly from Eq. (32) as well. At $H = H_{\perp}$ one has

$$K\vartheta = \mu H_{\perp} \cos \vartheta \approx \mu H_{\perp}.$$

Expressing ϑ from here and substituting it into $\chi_{\perp} = n\mu\vartheta/H_{\perp}$, formula (30) is recovered.

V. DYNAMIC SUSCEPTIBILITY

The linear response of any system to a weak probing field $H(t)$ can be represented [8] as an integral

$$M(t) = \int_0^\infty d\tau \mathcal{K}(\tau) H(t-\tau). \quad (33)$$

This relationship explicitly takes into account the causality principle, according to which the memory function $\mathcal{K}(\tau)$ must vanish in the long-time limit. $\mathcal{K}(\infty)=0$. For a harmonic signal $H(t)=he^{-i\omega t}$ relation (33) allows to define a generalized linear susceptibility as

$$M(t) = he^{-i\omega t} \int_0^\infty d\tau e^{i\omega\tau} \mathcal{K}(\tau) = he^{-i\omega t} \chi(\omega). \quad (34)$$

The memory function \mathcal{K} is associated with particular physical characteristics of a system using the correspondence principle. Setting the above-found static susceptibility—see Eqs. (28) and (29)—equal to the limit of Eq. (34) at $\omega=0$, one finds

$$\chi_\alpha(0) = \frac{n\mu^2}{T} \langle x_\alpha^2 \rangle = \int_0^\infty d\tau \mathcal{K}_\alpha \equiv \int_0^\infty d\tau \left(-\frac{dG_\alpha}{d\tau} \right) = G_\alpha(0), \quad (35)$$

where, according to Eqs. (28) and (29) we set

$$x_\alpha = \begin{cases} \cos \vartheta - \langle \cos \vartheta \rangle & \text{for } \alpha = \parallel \\ \sin \vartheta & \text{for } \alpha = \perp. \end{cases} \quad (36)$$

Thus, as the fluctuation-dissipation theorem predicts (see, Ref. [10], for example), one finds that the linear susceptibility of the system is determined by the equilibrium dipolar correlation function. The final expression for χ in the normalized form is

$$\chi_\alpha(\omega)/\chi_\alpha(0) = 1 + i\omega \int_0^\infty d\tau e^{i\omega\tau} G_\alpha(\tau) \quad (37)$$

with

$$G_\parallel = \frac{\langle \cos \vartheta_t \cos \vartheta \rangle - \langle \cos \vartheta \rangle^2}{\langle \cos^2 \vartheta \rangle - \langle \cos \vartheta \rangle^2}, \quad G_\perp = \frac{\langle \sin \vartheta_t \sin \vartheta \rangle^2}{\langle \sin^2 \vartheta \rangle^2}. \quad (38)$$

As it should be, in an isotropic system that has no rigidity ($K=0$), one gets $\langle \cos \vartheta \rangle=0$ and thus $G_\parallel = G_\perp$.

In the model under discussion, both components of the dipolar correlation function could be calculated exactly [10]. An essential simplification arises because the Langevin equation (4) is linear in the phase variable ϑ . Since the white noise $y(t)$ is a Gaussian random process, the variable $\vartheta(t)$ has the same statistics on account of the linearity of the Langevin equation. Taking the Gaussian average, one finds

$$\langle e^{ix} \rangle = \langle \cos x \rangle = \exp\left[-\frac{1}{2} \langle x^2 \rangle\right]. \quad (39)$$

With the aid of formula (39) and standard trigonometric relationships, we have for the dipolar correlation functions

$$G_\parallel(t) = \frac{\cosh \langle \vartheta_t \vartheta \rangle - 1}{\cosh \langle \vartheta^2 \rangle - 1}, \quad G_\perp(t) = \frac{\sinh \langle \vartheta_t \vartheta \rangle}{\sinh \langle \vartheta^2 \rangle}. \quad (40)$$

The general expression (18) for the angular correlation function has been derived in Sec. III. Its substitution into Eq. (40) enables one to write down the dependence of the susceptibility on the material parameters of the system as an integral (37). The latter, however, cannot be evaluated in closed form. Thus one cannot derive a simple analytical expression for χ valid for an arbitrary set of material parameters. However, a number of important simplifications are possible. To arrive at them, we represent Eq. (18) as

$$\begin{aligned} \langle \vartheta_t \vartheta \rangle &= \langle \vartheta^2 \rangle \left\{ \frac{1+b}{2b} \exp[-\gamma(1-b)t] \right. \\ &\quad \left. - \frac{1-b}{2b} \exp[-\gamma(1+b)t] \right\} \\ &= \langle \vartheta^2 \rangle \exp(-\gamma t) \frac{\sinh(\beta t + \psi)}{\sinh \psi}, \end{aligned} \quad (41)$$

where we denote $\gamma \equiv 1/(2\tau_l)$, $b \equiv \sqrt{1-4/t_K} \equiv \tanh \psi$, and $\beta \equiv \gamma b$.

To be specific, we take the transverse correlation function from Eq. (40). Expanding it in the Taylor series with respect to the angular correlation function and using the binomial expansion we have

$$\begin{aligned} \sinh \langle \vartheta_t \vartheta \rangle &= \sum_{n=0}^{\infty} \frac{\langle \vartheta^2 \rangle^{2n+1}}{(2n+1)!} \frac{\exp[-\gamma(2n+1)t]}{(2 \sinh \psi)^{2n+1}} \\ &\quad \times \sum_{k=0}^{2n+1} C_{2n+1}^k (-1)^k \exp[(2n+1-2k) \\ &\quad \times (\beta t + \psi)]. \end{aligned} \quad (42)$$

Taking into account the relation between the parameters ψ and b , Eq. (42) may be rewritten as

$$\begin{aligned} \sinh \langle \vartheta_t \vartheta \rangle &= \sum_{n=0}^{\infty} \left[\frac{(1+b) \langle \vartheta^2 \rangle}{2b} \right]^{2n+1} \frac{1}{(2n+1)!} \\ &\quad \times \sum_{k=0}^{2n+1} \left(\frac{b-1}{b+1} \right)^k C_{2n+1}^k \exp(-\gamma_{n,k}^\perp t) \end{aligned} \quad (43)$$

with

$$\langle \vartheta^2 \rangle = T/K, \quad \gamma_{n,k}^\perp = \gamma[(2n+1)(1-b) + 2kb].$$

Substituting Eq. (43) in Eq. (37) and integrating with respect to time, we obtain

$$\begin{aligned} \frac{\chi_\perp(\omega)}{\chi_\perp(0)} &= 1 + \frac{i\omega}{\sinh \langle \vartheta^2 \rangle} \sum_{n=0}^{\infty} \left[\frac{(1+b) \langle \vartheta^2 \rangle}{2b} \right]^{2n+1} \frac{1}{(2n+1)!} \\ &\quad \times \sum_{k=0}^{2n+1} \left(\frac{b-1}{b+1} \right)^k \frac{C_{2n+1}^k}{\gamma_{n,k}^\perp - i\omega}. \end{aligned} \quad (44)$$

In like manner, the longitudinal susceptibility may be transformed (see also Ref. [10]) and takes the form

$$\frac{\chi_{\parallel}(\omega)}{\chi_{\parallel}(0)} = 1 + \frac{i\omega}{\cosh\langle\vartheta^2\rangle - 1} \sum_{n=1}^{\infty} \left[\frac{(1+b)\langle\vartheta^2\rangle}{2b} \right]^{2n} \frac{1}{(2n)!} \times \sum_{k=0}^{2n} \left(\frac{b-1}{b+1} \right)^k \frac{C_{2n}^k}{\gamma_{n,k}^{\parallel} - i\omega}, \quad (45)$$

where

$$\gamma_{n,k}^{\parallel} = \gamma[2n(1-b) + 2kb].$$

VI. HIGH-RIGIDITY/LOW-TEMPERATURE LIMIT

Formulas (44) and (45) are very convenient in order to analyze the case $T < K$, which is natural to call *the high-rigidity limit*. As the above estimation shows, for a polymeric gel the onset of this inequality is roughly equivalent to the condition that the particle-matrix steric interaction is established.

Apparently, in the high-rigidity limit the macroscopic anisotropy is most pronounced, and the two susceptibility components differ considerably. To obtain the approximate formula for χ_{\perp} , we retain in expansion (44) only the term linear in the parameter T/K and make use of the approximate relation $b = \sqrt{1 - 4/t_K} \approx 1 - 2/t_K$. The result is

$$\begin{aligned} \frac{\chi_{\perp}(\omega)}{\chi_{\perp}(0)} &= 1 + i\omega \left[\left(1 + \frac{1}{t_K} \right) \frac{1}{\gamma_{0,0}^{\perp} - i\omega} - \frac{1}{t_K(\gamma_{0,1}^{\perp} - i\omega)} \right] \\ &= \frac{1}{1 - i\omega\tau_K} + \frac{i\omega\tau_K}{(t_K - i\omega\tau_K)(1 - i\omega\tau_K)} \\ &\approx \frac{1}{1 - i\omega\tau_K - \omega^2\tau_I\tau_K} \quad (\tau_K \gg \tau_I). \end{aligned} \quad (46)$$

Therefore, in the lowest order the relation describing the transverse susceptibility of a suspension in a rigid ($T \ll K$) gel has the structure of the Rocard formula. The latter type of frequency dependence is well known in molecular spectroscopy, see Ref. [10], for example. We would like to remind, however, that in the physically relevant range of material parameters ($\tau_K \gg \tau_I$) Eq. (46), despite the fact that it is quadratic in ω , does not display any resonance and its behavior is close to the Debye susceptibility. The main function of the additional term in the denominator is to prevent the divergence of the integral absorption (“ultraviolet catastrophe”) by ensuring the correct behavior at $\omega \rightarrow \infty$. Its subsidiary effect is that at some high frequency the real part of χ_{\perp} , already being very small, becomes negative, see insets in Figs. 1 and 2. We remark also that formula (46) can be derived directly from the Langevin equation (4) if we neglect fluctuations.

In order to evaluate the temperature correction to the transverse susceptibility, we sum Eq. (44) up to $n=1$ allowing the smallness of the parameter τ_I/τ_K . This yields

$$\frac{\chi_{\perp}(\omega)}{\chi_{\perp}(0)} = \frac{1}{1 - i\omega\tau_K - \omega^2\tau_I\tau_K} \left[1 - \frac{i\omega\tau_K(T/K)^2}{3(3 - i\omega\tau_K)} \right]. \quad (47)$$

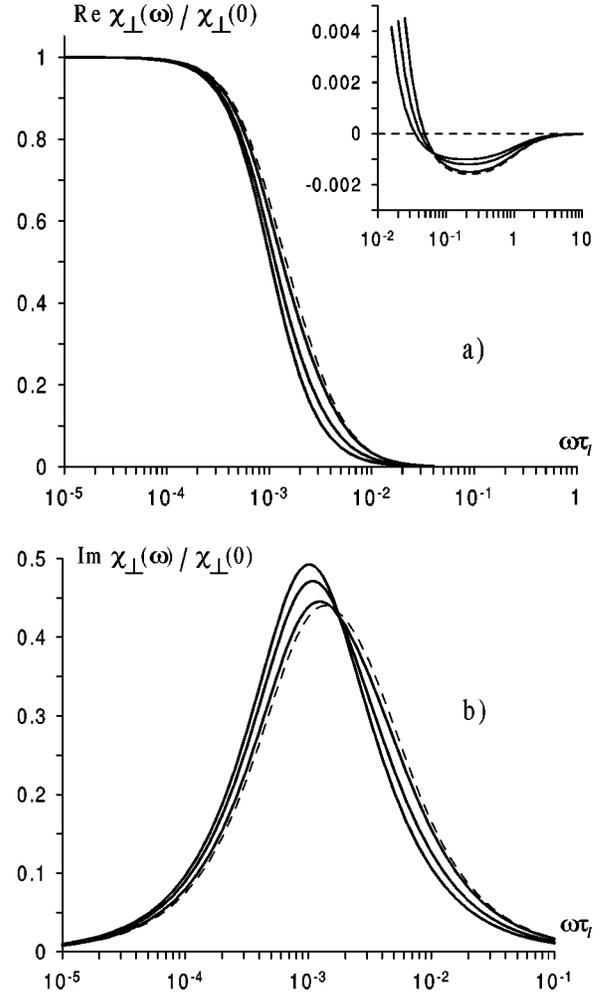


FIG. 1. (a) Transverse susceptibility for $T/K < 2$; solid curves (left to right) show the real part by the exact formula (44) at, respectively, $T/K = 0.5, 1.0$, and 1.5 ; dashed curve shows the result obtained with the approximate formula (47) for $T/K = 1.5$; for lower values, exact and approximate curves do not resolve within the scale of the figure. Inset shows the details of the curve behavior at the high-frequency end. (b) Imaginary part of the transverse susceptibility for the same values of T/K .

Calculation (with the same accuracy) of the longitudinal susceptibility with the aid of Eq. (45) yields

$$\frac{\chi_{\parallel}(\omega)}{\chi_{\parallel}(0)} = \frac{1}{1 - \frac{1}{2}i\omega\tau_K - \frac{1}{2}\omega^2\tau_I\tau_K} \left[1 - \frac{i\omega\tau_K(T/K)^2}{12(4 - i\omega\tau_K)} \right]. \quad (48)$$

Comparison of formulas (47) and (48) shows, as one might have expected, that in the dynamic limit (see Ref. [13], for example) the longitudinal magnetization relaxes twice as fast as the transverse one $\tau_{\parallel} = \tau_{\perp}/2 = \tau_K/2$. The corrections $\propto T^2$ to the Rocard-like dispersion factors in Eqs. (47) and (48) are new. For both orientations, they considerably improve the quality of the approximation. In Fig. 1 we show the results of numerically exact evaluation of the transverse susceptibility by Eq. (44) against the analytic dependence (47). We remark that although the approximation is formally justified only for

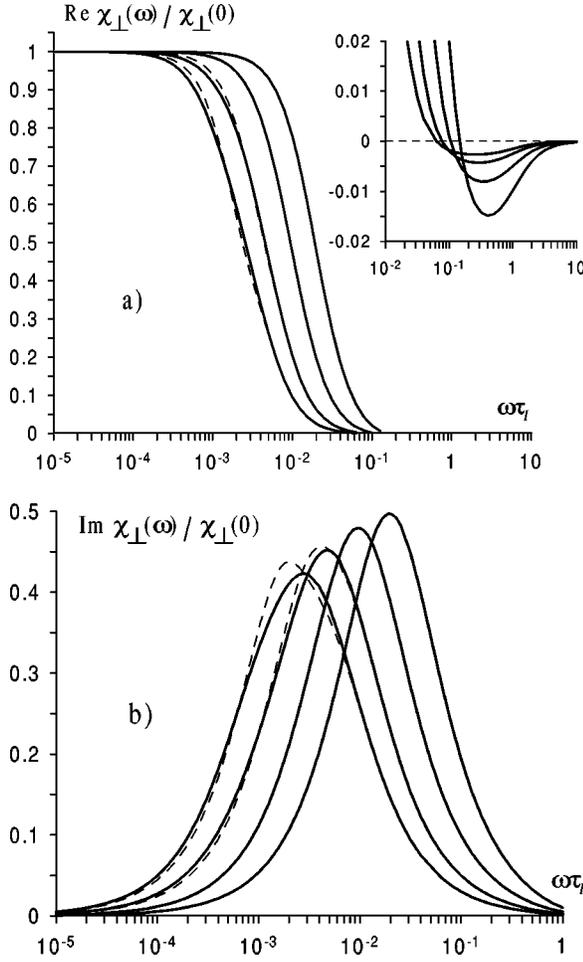


FIG. 2. (a) Transverse susceptibility for $T/K \geq 3$; solid curves (left to right) show the real part by the exact formula (44) at, respectively, $T/K = 3, 5, 10, 20$; dashed curves show the result obtained with the approximate formula (52) for $T/K = 3$ and 5; for higher values, the exact and approximate curves do not resolve within the scale of the figure. Inset shows the details of the curve behavior at the high-frequency end. (b) Imaginary part of the transverse susceptibility for the same values of T/K .

$T < K$, the curves by Eq. (47) resemble the exact one rather closely at $T/K \geq 1$ as well in the whole frequency range.

It should be noted that formulas (47) and (48) are easily derived from the approximate dipolar correlation functions. The pertinent relationships follow from Eq. (40) when formula (18) and the smallness of the parameters T/K and τ_I/τ_K are taken into account:

$$\begin{aligned}
 G_{\perp}(t) &= e^{-t/\tau_K} + \frac{\tau_I}{\tau_K} (e^{-t/\tau_K} - e^{-t/\tau_I}) \\
 &\quad + \frac{T^2}{6K^2} (e^{-3t/\tau_K} - e^{-t/\tau_K}), \\
 G_{\parallel}(t) &= e^{-2t/\tau_K} + 2\frac{\tau_I}{\tau_K} (e^{-2t/\tau_K} - e^{-t/\tau_I}) \\
 &\quad + \frac{T^2}{12K^2} (e^{-4t/\tau_K} - e^{-2t/\tau_K}).
 \end{aligned}
 \tag{49}$$

Substituting Eq. (49) in Eq. (37), after integration and some rearrangements one recovers Eqs. (47) and (48).

VII. LOW-RIGIDITY/HIGH-TEMPERATURE LIMIT

The opposite relation between elasticity and temperature, i.e., $T \gg K$, corresponds to weak elastic effects so that the particles are almost free. In this limit the difference between the correlation functions (40) tends to zero exponentially:

$$G_{\parallel}(t) = G_{\perp}(t) = \exp[\langle \vartheta_i \vartheta \rangle - \langle \vartheta^2 \rangle], \tag{50}$$

indicating that the transverse and longitudinal susceptibilities should approach a common limiting form.

The angle correlation function entering Eq. (50) is given by a general formula (18). We evaluate it with the quadratic accuracy in the small parameter τ_I/τ_K and consider the intermediate time asymptotics $t \ll \tau_K \rightarrow \infty$. The result is

$$\begin{aligned}
 \langle \vartheta_i \vartheta \rangle - \langle \vartheta^2 \rangle &= -\frac{t}{\tau_D} + \frac{\tau_I}{\tau_D} (1 - e^{-t/\tau_I}) \\
 &\quad + \frac{\tau_I^2}{\tau_D \tau_K} [3 - 2t/\tau_I + \frac{1}{2}(t/\tau_I)^2 \\
 &\quad - (t/\tau_I + 3)e^{-t/\tau_I}].
 \end{aligned}
 \tag{51}$$

The first two terms of this expansion constitute the well-known expression for the angular correlation function of a free particle [10]. The effect of rigidity is rendered by the last term, note that in the limit considered it contains an additional power of the small parameter τ_I/τ_K .

Substituting Eq. (51) in Eq. (50) and integrating the dipolar correlation function so obtained, one finds on the basis of the linear-response theory (37) the approximate expression

$$\frac{\chi(\omega)}{\chi(0)} = \frac{1}{1 - i\omega\tau_D - \omega^2\tau_I\tau_D} \left[1 + \frac{K}{T} \frac{i\omega\tau_D}{(1 - i\omega\tau_D)^2} \right], \quad T \gg K.
 \tag{52}$$

In deriving this equation, we have taken into account the smallness of τ_I/τ_D . Note that in the limit under consideration the subscript of χ is not relevant.

As formula (52) shows, at high temperatures the main term of the magnetic susceptibility of a ferrogel once again—compare Eqs. (47) and (48)—assumes the Rocard form. However, the parameter in this new equation is τ_D and not τ_K , so that now Eq. (52) is akin to the susceptibility of a dipolar suspension with a simple viscous carrier [10].

The exact dependencies $\chi_{\perp}(\omega)$ by Eq. (44) for $T/K \geq 3$ are presented in Fig. 2 in comparison with the approximate expression (52). Here the temperature-dependent correction to the Rocard form is $\propto 1/T$. As the graphs show, the approximation obtained works fairly well down to $T/K \geq 5$ in the whole frequency range.

According to Fig. 2(a), the real part of χ in a weakly elastic medium does not differ qualitatively from that rendered by the standard Rocard factor. However, for the imaginary part, see Fig. 2(b), one easily notices the difference. Namely, at any finite temperature, the maximum of the ab-

sorption line is lowered in comparison with the Rocard (or equivalent Debye) level that in our units is 0.5 and is temperature independent.

This deformation of the absorption peak is also frequency dependent. Separating the imaginary part of Eq. (52) and neglecting its difference from the Debye one, i.e., setting $\omega^2 \tau_I \tau_D \ll 1$, we have

$$\frac{\chi''(\omega)}{\chi(0)} = \frac{X}{1+X^2} \left[1 + \frac{K}{T} \frac{1-3X^2}{(1+X^2)^2} \right], \quad (53)$$

where $X = \omega \tau_D$. From Eq. (53) it follows that the ‘‘elastic’’ correction is positive at low frequencies, passes through zero at $X = 1/\sqrt{3}$ and becomes negative. Inside the negative region at $X = \sqrt{5/3}$ it assumes minimum of $-9K/16T$.

Therefore, in the high-temperature limit the anisotropy of the susceptibility is exponentially small in the reduced elasticity τ_I/τ_K . However, the contribution from the elastic mechanism to the isotropic part of the susceptibility is linear in this parameter. Analysis of the limiting temperature cases shows that in each of them the susceptibility may be written in the Rocard form. As one expects, the effective relaxation time in the high-temperature case coincides with the Debye time of orientational diffusion τ_D , while in the low-temperature limit it tends to the elastic relaxation time τ_K .

The overall behavior of the transverse susceptibility in a wide temperature range is presented in Fig. 3. As is apparent, from Fig. 3(b), the maximum of the absorption line goes down and at $T/K \approx 3$ falls as low as 0.41, that is about 80% of the pure Debye value, which in the units used here is 0.5.

In Fig. 3(c) the curves $\chi_{\perp}(\omega)$ are plotted in double-logarithmic coordinates. This reveals the presence of two asymptotics in the absorption line: the low-frequency wing that depends linearly on the frequency, and the high-frequency end where the dependence is cubic. In what follows we derive the corresponding approximate expressions.

VIII. HIGH-FREQUENCY ASYMPTOTICS

In the high-frequency range, the behavior of the correlation functions at short times is essential. Hence, it is convenient to expand the angular correlator in Taylor series with respect to time and to cut off all the terms higher than t^3 . This yields

$$\langle \vartheta_t \vartheta \rangle = \langle \vartheta^2 \rangle - \frac{\omega_T^2 t^2}{2} + \frac{\omega_T^2 t^3}{6 \tau_I}, \quad t \rightarrow 0. \quad (54)$$

With this expansion, using formula (40), we obtain the transverse dipolar correlation function as

$$G_{\perp}(t) = 1 - \left(1 - \frac{t}{3 \tau_I} \right) \frac{\omega_T^2 t^2}{2} \coth \langle \vartheta^2 \rangle. \quad (55)$$

Substituting this in Eq. (37) and performing some simple rearrangements, the desired asymptotic expression for the susceptibility is

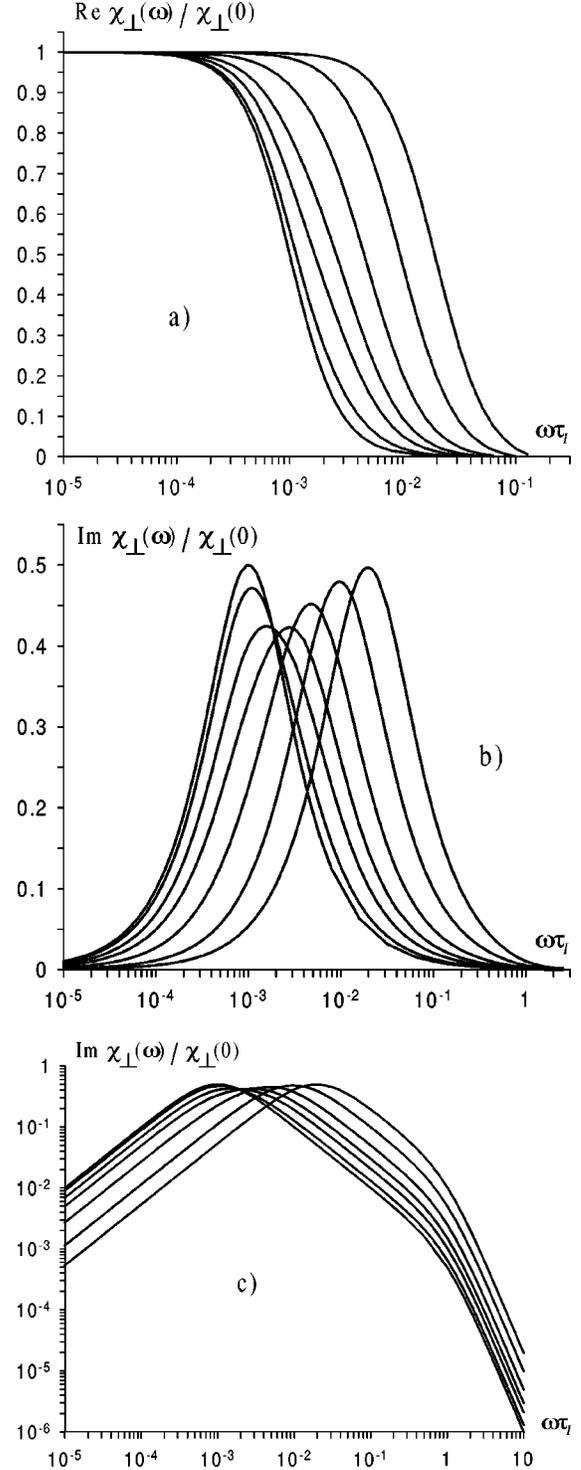


FIG. 3. (a) Real parts of the transverse susceptibility curves (left to right) at, respectively, $T/K = 0.1, 1, 2, 3, 5, 10$ and 20 . (b) Imaginary part of the transverse susceptibility for the same values of T/K . (c) Imaginary part of the transverse susceptibility for the same values of T/K double-logarithmic plot.

$$\frac{\chi_{\perp}(\omega)}{\chi_{\perp}(0)} = \left(-1 + \frac{i}{\omega \tau_I} \right) \frac{\omega_T^2}{\omega^2} \coth \left(\frac{T}{K} \right), \quad \omega \rightarrow \infty. \quad (56)$$

The longitudinal susceptibility is found in like manner as

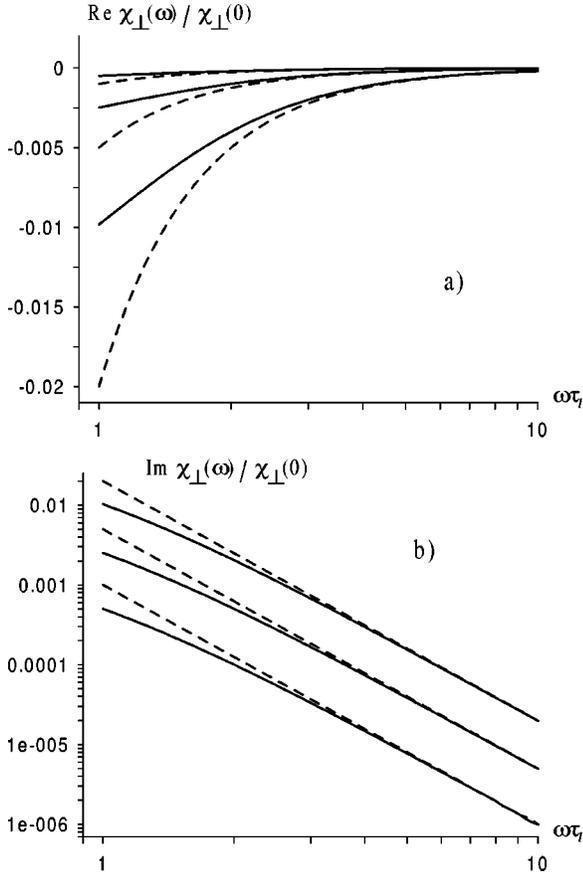


FIG. 4. (a) High-frequency asymptotics of the transverse susceptibility. Solid lines are obtained by the numerically exact solution of Eq. (44), dashed ones by the approximate formula (56); the values of T/K are 0.5, 5 and 20 ranging from top to bottom in (a) and reversely in (b).

$$\frac{\chi_{\parallel}(\omega)}{\chi_{\parallel}(0)} = \left(-1 + \frac{i}{\omega\tau_I} \right) \frac{\omega_T^2}{\omega^2} \coth\left(\frac{T}{2K}\right), \quad \omega \rightarrow \infty. \quad (57)$$

Formulas (56) and (57) confirm that the high-frequency asymptotics are $1/\omega^2$ for the real part of the susceptibility and $1/\omega^3$ for the imaginary one. In the high-temperature limit ($T \gg K$) the coth function in Eq. (56) tends to unity, so that the high-frequency asymptotics of the absorption line is proportional to the thermal frequency $\omega_T^2 = T/I$. In the low-temperature limit, that is $T \ll K$, the function $\coth(T/K) \rightarrow K/T$, so that the asymptotic value of absorption is proportional to $K\omega_T^2/T = \omega_K^2 = K/I$, i.e., eigenfrequency of the elasticity-driven oscillations. These conclusions are illustrated in Figs. 4(a), 4(b), where these asymptotics are compared to the exact results obtained with the aid of Eq. (44).

IX. LOW-FREQUENCY ASYMPTOTICS. EFFECTIVE RELAXATION TIMES

From the basic formula (37) of linear-response theory it follows that the low-frequency wing of the absorption line may be described by the expression

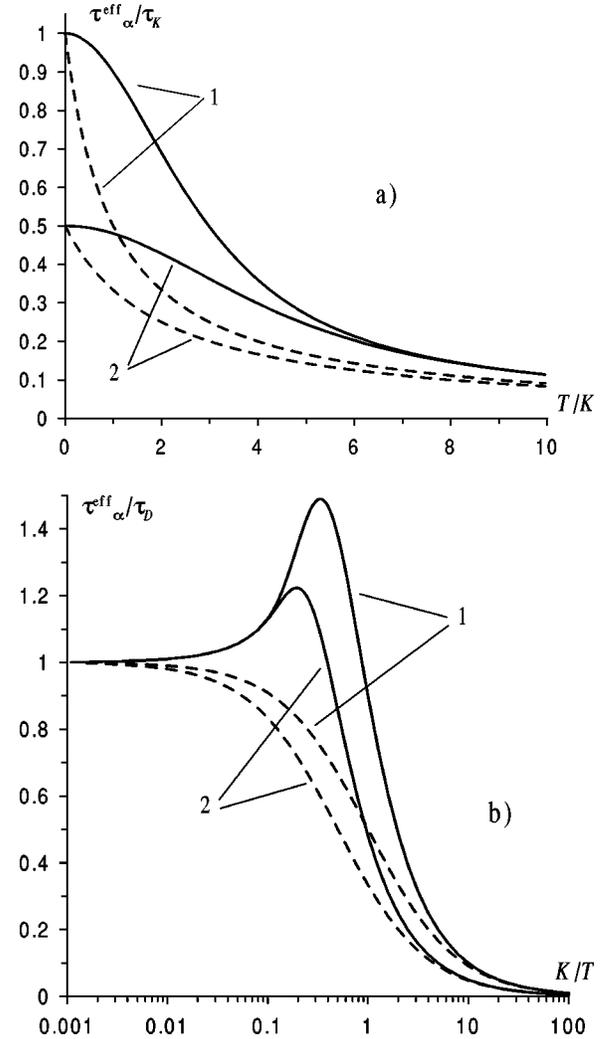


FIG. 5. (a) Effective relaxation times: transversal (1) and longitudinal (2), scaled with τ_K , as functions of the reduced temperature T/K (solid lines); the corresponding intuitive expressions (sums of partial relaxation rates) are shown by dashed lines. (b) The same relaxation times, scaled with τ_D , as functions of the inverse temperature K/T (solid lines); the intuitive expressions are shown by dashed lines.

$$\frac{\chi_{\alpha}(\omega)}{\chi_{\alpha}(0)} = 1 + i\omega\tau_{\alpha}^{\text{eff}}, \quad \omega \rightarrow 0, \quad (58)$$

where

$$\tau_{\alpha}^{\text{eff}} \equiv \int_0^{\infty} G_{\alpha}(t) dt \quad (59)$$

is the effective time characterizing the final stage of the equilibrium settling. Formula (58) explicitly shows that the low-frequency asymptotics are $\chi'_{\perp} \propto \text{const}(\omega)$ and $\chi''_{\perp} \propto \omega$.

Comparing expression (58) with the general formula (44) at $\omega \rightarrow 0$ we have

$$\tau_{\perp}^{\text{eff}} = \frac{1}{\sinh(T/K)} \sum_{n=0}^{\infty} \left(\frac{(1+b)\langle \vartheta^2 \rangle}{2b} \right)^{2n+1} \times \sum_{k=0}^{2n+1} \left(\frac{b-1}{b+1} \right)^k \frac{1}{k!(2n+1-k)! \gamma_{n,k}^{\perp}}, \quad (60)$$

where

$$b = \sqrt{1 - 4/t_K}, \quad \gamma_{n,k}^{\perp} = [(2n+1)(1-b) + 2kb]/2\tau_{\perp}.$$

We reiterate that Eq. (60) is valid for arbitrary values of material parameters. For the case of interest $\tau_K \gg \tau_{\perp}$, formula (60) simplifies considerably and takes the form

$$\begin{aligned} \frac{\tau_{\perp}^{\text{eff}}}{\tau_K} &\approx \frac{1}{\sinh(T/K)} \sum_{n=0}^{\infty} \frac{(T/K)^{2n+1}}{(2n+1)(2n+1)!} \\ &= \frac{1}{\sinh(T/K)} \int_0^{T/K} \frac{\sinh u}{u} du = \frac{\text{Shi}(T/K)}{\sinh(T/K)}, \end{aligned} \quad (61)$$

thus reducing it to one hyperbolic integral function. In like manner for the effective time of longitudinal relaxation, one has

$$\begin{aligned} \frac{\tau_{\parallel}^{\text{eff}}}{\tau_K} &\approx \frac{1}{\cosh(T/K) - 1} \sum_{n=1}^{\infty} \frac{(T/K)^{2n}}{2n2n!} \\ &= \frac{1}{\cosh(T/K) - 1} \int_0^{T/K} \frac{\cosh(u) - 1}{u} du. \end{aligned} \quad (62)$$

The temperature behavior of the effective relaxation times is presented in Figs. 5(a) and 5(b). For comparison, in the dashed lines we show the results of the intuitive approach, where the average rate $(\tilde{\tau}^{\text{eff}})^{-1}$ of a process that undergoes in several stages is assumed to be the sum of the partial rates, i.e., inverse relaxation times. For our case

$$(\tilde{\tau}_{\perp}^{\text{eff}})^{-1} = \tau_K^{-1} + \tau_D^{-1}, \quad (\tilde{\tau}_{\parallel}^{\text{eff}})^{-1} = 2\tau_K^{-1} + \tau_D^{-1}. \quad (63)$$

In Fig. 5(a) the effective times are scaled with the elastic time τ_K that dominates in the low-temperature case. Accord-

ingly, at $T \rightarrow 0$ (dynamic limit, no fluctuations) both effective times are finite, the transverse time being exactly twice as large as the longitudinal one. As the temperature grows and/or the rigidity decreases, the relaxation by orientational diffusion becomes more efficient, and the effective time goes down tending to the Debye expression $\propto 1/T$.

In Fig. 5(b) the same effective times are presented scaled with the Debye time τ_D , which controls the high-temperature case. Here the abscissa axis is inverted with respect to that of Fig. 5(a) so that the left end of the plot corresponds now to a high-temperature case. There the macroscopic anisotropy of the gel is very weak, and both effective times approach each other exponentially, see Eq. (50). In the low-temperature range $K/T \gg 1$, the dynamic limit, where $\tau_{\perp} = 2\tau_{\parallel}$, comes in effect, compare Fig. 5(a). However, in none of the cases does the intuitive approach (63) seem appropriate to describe the real effective times $\tau_{\alpha}^{\text{eff}}$ except maybe in trivial limits $T/K \rightarrow 0$ and $K/T \rightarrow 0$.

Finally, we present the asymptotic relations for the effective times obtained from Eqs. (61) and (62):

$$\begin{aligned} \tau_{\perp}^{\text{eff}} &= \begin{cases} \tau_K \left[1 - \frac{T^2}{9K^2} \right] & \text{for } T \ll K \\ \tau_D \left[1 + \frac{K}{T} \right] & \text{for } T \gg K, \end{cases} \\ \tau_{\parallel}^{\text{eff}} &= \begin{cases} \frac{1}{2} \tau_K \left[1 - \frac{T^2}{24K^2} \right] & \text{for } T \ll K \\ \tau_D \left[1 + \frac{K}{T} \right] & \text{for } T \gg K. \end{cases} \end{aligned} \quad (64)$$

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