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# Transient and steady linear response of dielectric particles in a high bias field subject to a weak AC probe field

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**Abstract**
A dilute suspension of spherical particles with permanent electric dipole moment and additional polarizability is subject to a strong DC external electric field. In addition, a weak AC probe field is suddenly switched on at time $t = 0$. The linear response of the polarization is described by solving the first-order Smoluchowski equation for the orientational distribution function. Both the transient and the steady response are obtained. The transient is given explicitly as a sum of exponentially decaying functions of time characterized by a set of decay rates and associated amplitudes. A fast numerical method is used to calculate the spectrum of decay rates and the amplitudes of the decaying modes from the Laplace transform of the perturbed distribution function. The steady response is characterized by a complex susceptibility which is given explicitly in terms of the decay rates and amplitudes that characterize the transient. The transient solution, its mean decay time and the susceptibility are all given as explicit functions of the frequency of the probe field. The results for simple permanent dipoles are compared with the much richer results for dipoles with polarizability as well.

## 1. Introduction

The dynamics of suspensions of mesoscale particles on intermediate and long timescales is described by a generalized diffusion equation incorporating both translational and rotational diffusion of the suspended particles [1, 2]. One of the oldest problems involving this kind of dynamics is the study of the electric polarization of a fluid of dipolar particles undergoing rotational diffusion [3–7]. For a dilute fluid, subject to a weak external electric field, the analysis of Debye gave simple analytical expressions for the polarization relaxation function when the external field is suddenly turned off and for the linear response of the polarization to a weak external sinusoidal field [3, 4]. In the Debye theory the underlying dynamics is given by the Smoluchowski equation describing rotational diffusion of the dipoles. Because it neglects inertial rotational effects, such a theory is only partially successful in describing molecular fluids [4]. However, for dilute suspensions of colloid-sized particles which have dielectric or magnetic properties (as in a ferrofluid [8, 9]), the Smoluchowski dynamics should be an excellent description.

The original Debye problem has interesting extensions both to relaxation phenomena occurring when a strong external field suddenly changes its magnitude or direction rather than being switched off, and to the differential susceptibility of a suspension subject to a strong DC background field [11–15]. The rotational dynamics also can be used to discuss the Kerr effect for dielectric particles or the Néel effect in magnetic particles [8, 10, 14, 16, 17]. In recent years a method of treating these strong-field non-linear relaxation problems by a continued-fraction approach has been worked out which gives exact closed-form expressions for the frequency dependence of the relaxation functions [7, 11, 14]. However, there is a more direct approach which leads quickly to a representation of the relaxation functions as a superposition of modes which decay exponentially in time [15–17]. The decay rates and associated amplitudes of these modes are easily determined from the poles and residues of the Laplace transform of the expansion coefficients of the orientational distribution function.

In the present work we study the linear response of a dilute suspension of spherical dielectric particles in a strong DC bias field to a weak AC perturbing field which is suddenly switched on at time $t = 0$. In earlier work [11], the steady-state linear susceptibility was obtained indirectly from the continued-fraction solution for the after-effect function [4] associated with the relaxation problem. The steady-state non-linear susceptibility has more recently been studied numerically in the context of a matrix continued-fraction formalism [18, 19]. We show that our analysis of the relaxation problem [15, 17] carries over easily to the linear response calculation in a strong bias field, giving both the transient and the steady response to the AC perturbing field. We show that the transient contribution, like the relaxation functions studied earlier, is a superposition of exponentially decaying modes and, more surprisingly, find that the frequency-dependent susceptibility can be expressed in terms of the transient solution. This representation displays the frequency dependence of the susceptibility explicitly as a sum of poles allowing easy numerical evaluation of the differential susceptibility in strong background fields at all frequencies.

Although the theory we present is equally applicable to electric or magnetic dipolar particles, for comparability with our earlier calculations we shall use the electrical picture throughout. For experimental study, magnetic suspensions [8, 9] may be preferable for exploring a wider range of the parameters introduced below. In section 2 we introduce the rotational Smoluchowski equation and its equilibrium solution in a strong external DC field. In section 3 we derive the equations giving the linear response to a weak longitudinal AC probe field, in section 4 we solve these equations by Laplace transformation and in section 5 we simplify the solution by removing the steady-state pole in the Laplace transform solution. In section 6 we give simple explicit expressions for the transient solution, for its mean decay time and for the complex susceptibility describing the steady-state response. In section 7 we give the corresponding results for a transverse probe field, in section 8 we present numerical studies and in section 9 we present conclusions.

## 2. Polar and polarizable particles

We consider a dilute colloidal suspension of anisotropic spherical particles which carry a permanent electric dipole moment $\boldsymbol{\mu} = m\boldsymbol{u}$ where the unit vector $\boldsymbol{u}$ specifies the direction of the dipole, and which, in addition, are polarizable with low-frequency electric polarizabilities

$\alpha_1$, $\alpha_2$, respectively parallel to and perpendicular to the particle symmetry axis specified by $\boldsymbol{u}$. For a dilute suspension of such mesoscale particles with slow rotational diffusion, we may neglect both inertial effects and dipole–dipole interaction effects. To describe the macroscopic polarization of the suspension it then suffices to study the single-particle orientational distribution function, $W(\boldsymbol{u}, t)$, which satisfies the rotational Smoluchowski equation

$$\frac{\partial W}{\partial t} = \mathcal{D}W = D_R \frac{\partial}{\partial \boldsymbol{u}} \cdot \left( \frac{\partial W}{\partial \boldsymbol{u}} + \beta \frac{\partial V}{\partial \boldsymbol{u}} W \right), \tag{1}$$

where $D_R$ is the rotational diffusion coefficient, $\beta = 1/k_B T$ and $V$ is the potential energy of a single particle in a uniform applied field $\boldsymbol{E}$:

$$V(\boldsymbol{u}) = -m\boldsymbol{u} \cdot \boldsymbol{E} - \tfrac{1}{2}(\alpha_1 - \alpha_2)(\boldsymbol{u} \cdot \boldsymbol{E})^2. \tag{2}$$

The Smoluchowski operator, $\mathcal{D}$, is expressed in terms of the gradient operator on the unit sphere, $\partial/\partial \boldsymbol{u}$, which, in spherical polar coordinates, has the form

$$\frac{\partial}{\partial \boldsymbol{u}} = \boldsymbol{e}_\theta \frac{\partial}{\partial \theta} + \frac{\boldsymbol{e}_\phi}{\sin\theta} \frac{\partial}{\partial \phi}, \tag{3}$$

with the usual spherical unit vectors $\boldsymbol{e}_\theta$, $\boldsymbol{e}_\phi$. For a suspending fluid of viscosity $\eta$, and for mesoscale particles of radius $a$, the rotational diffusion coefficient is given as $D_R = k_B T/8\pi\eta a^3$. A relaxation time $\tau_R$ is defined by $\tau_R = 1/D_R$ in terms of which the Debye relaxation time is $\tau_D = \tau_R/2$.

We assume that up to time $t = 0$ the suspension is in equilibrium in a constant uniform field $\boldsymbol{E}_0$ which is directed along the $z$-axis, $\boldsymbol{E}_0 = E_0 \boldsymbol{e}_z$. The equilibrium distribution function is given by the Boltzmann expression

$$W_0(\boldsymbol{u}) = \exp[-\beta V_0(\boldsymbol{u})]/Z(\xi_0, \sigma_0), \tag{4}$$

with the potential energy $V_0$, in field $\boldsymbol{E}_0$, given as

$$-\beta V_0(\boldsymbol{u}) = \xi_0 \cos\theta + \sigma_0 \cos^2\theta, \tag{5}$$

where $\theta$ is the spherical polar angle between the $z$-axis and the dipole direction $\boldsymbol{u}$. The parameters $\xi_0 = \beta m E_0$ and $\sigma_0 = \frac{1}{2}\beta(\alpha_1 - \alpha_2)E_0^2$ have the property that $\sigma_0/\xi_0^2$ is independent of the magnitude $E_0$ of the background field since the polarizabilities $\alpha_1$, $\alpha_2$ are assumed to be field-independent constants. The partition function $Z(\xi_0, \sigma_0)$ is given as

$$Z(\xi_0, \sigma_0) = \int \exp[-\beta V_0(\boldsymbol{u})]\, \mathrm{d}\boldsymbol{u} = 2\pi \int_{-1}^{1} \exp[\xi_0 x + \sigma_0 x^2]\, \mathrm{d}x. \tag{6}$$

For a dilute suspension of particles with number density $n$, the equilibrium polarization in field $\boldsymbol{E}_0$ at time $t = 0$ is given by

$$\boldsymbol{P}_0 = n\langle \boldsymbol{\mu} \rangle_0 = nm \int \boldsymbol{u} W_0(\boldsymbol{u})\, \mathrm{d}\boldsymbol{u} = nm\boldsymbol{e}_z \int \cos\theta\, W_0(\boldsymbol{u})\, \mathrm{d}\boldsymbol{u} = nm\langle P_1(\cos\theta)\rangle_0 \boldsymbol{e}_z, \tag{7}$$

where $P_1(\cos\theta)$ is the Legendre polynomial of order 1. For later use it is convenient to introduce the function $M_\ell(\xi_0, \sigma_0)$ defined as the average of the Legendre polynomial $P_\ell(\cos\theta)$ with respect to $W_0(\boldsymbol{u})$:

$$M_\ell(\xi_0, \sigma_0) = \langle P_\ell(\cos\theta)\rangle_0 = \int P_\ell(\cos\theta) W_0(\boldsymbol{u})\, \mathrm{d}\boldsymbol{u}. \tag{8}$$

To evaluate the $M_\ell$ we introduce the expansion

$$\exp[\xi_0 x + \sigma_0 x^2] = \sum_{\ell=0}^{\infty} \frac{(2\ell + 1)}{2} N_\ell(\xi_0, \sigma_0) P_\ell(x), \tag{9}$$

where the $P_\ell(x)$ are Legendre polynomials and the coefficient functions $N_\ell(\xi_0, \sigma_0)$ have a representation derived elsewhere [16, 17] in terms of Hermite polynomials $H_n$:

$$N_\ell(\xi_0, \sigma_0) = (2\mathrm{i}\sqrt{\sigma_0})^\ell \sum_{p=0}^{\infty} \frac{2(\ell+p)!}{p!(2\ell+2p+1)!}(-\sigma_0)^p H_{\ell+2p}\left(\frac{-\mathrm{i}\xi_0}{2\sqrt{\sigma_0}}\right). \tag{10}$$

It follows from these definitions that we can express the partition function and the $M_\ell$ as

$$Z(\xi_0, \sigma_0) = 2\pi N_0(\xi_0, \sigma_0), \tag{11}$$

$$M_\ell(\xi_0, \sigma_0) = N_\ell(\xi_0, \sigma_0)/N_0(\xi_0, \sigma_0). \tag{12}$$

## 3. Linear response to a longitudinal probe field

At time $t = 0$, with the system in equilibrium under the influence of the field $\boldsymbol{E}_0$, an additional sinusoidal probe field of the form $\boldsymbol{E}_1 \mathrm{e}^{\mathrm{i}\omega t}$ is suddenly switched on, where the constant, uniform field $\boldsymbol{E}_1$ is weak compared with the background field, $|\boldsymbol{E}_1| \ll |\boldsymbol{E}_0|$. We ask for the linear response of the polarization to this perturbing field. To obtain the response it is necessary to solve the Smoluchowski equation (1) to first order in the perturbation $\boldsymbol{E}_1$. Thus we expand to first order the potential,

$$V(\boldsymbol{u}) = V_0(\boldsymbol{u}) + V_1(\boldsymbol{u}) + \cdots, \tag{13}$$

the distribution function,

$$W(\boldsymbol{u}, t) = W_0(\boldsymbol{u}) + W_1(\boldsymbol{u}, t) + \cdots, \tag{14}$$

and the Smoluchowski equation. Since $W_0(\boldsymbol{u})$ is a solution of the zeroth-order Smoluchowski equation, we find that we must solve the inhomogeneous equation

$$\frac{\partial W_1(\boldsymbol{u}, t)}{\partial t} = \mathcal{D}_0 W_1(\boldsymbol{u}, t) + D_R \frac{\partial}{\partial \boldsymbol{u}} \cdot \left(\beta \frac{\partial V_1}{\partial \boldsymbol{u}} W_0(\boldsymbol{u})\right), \tag{15}$$

subject to the initial condition $W_1(\boldsymbol{u}, 0) = 0$. The normalization condition on the distribution function implies that

$$\int W_1(\boldsymbol{u}, t)\, \mathrm{d}\boldsymbol{u} = 0. \tag{16}$$

The operator $\mathcal{D}_0$ is the Smoluchowski operator for the unperturbed system with potential $V_0$.

The perturbing field $\boldsymbol{E}_1$ may be oriented in any direction relative to $\boldsymbol{E}_0$ but, in linear response, we can decompose it into longitudinal (parallel to $\boldsymbol{E}_0$) and transverse (normal to $\boldsymbol{E}_0$) components and consider each separately. For simplicity, we first suppose that $\boldsymbol{E}_1$ is purely longitudinal, $\boldsymbol{E}_1 = \boldsymbol{E}_\parallel = E_\parallel \boldsymbol{e}_z$, which gives for the perturbing potential $V_1$ the expression

$$\beta V_1(\boldsymbol{u}, t) = -[\beta m + \beta(\alpha_1 - \alpha_2)(\boldsymbol{u} \cdot \boldsymbol{E}_0)](\boldsymbol{u} \cdot \boldsymbol{E}_\parallel)\mathrm{e}^{\mathrm{i}\omega t} = -\left[1 + \frac{2\sigma_0}{\xi_0}\cos\theta\right]\cos\theta\,\xi_\parallel \mathrm{e}^{\mathrm{i}\omega t}, \tag{17}$$

where $\xi_\parallel = \beta m E_\parallel$ and $\xi_\parallel \ll \xi_0$. Using this expression for $\beta V_1$ and the gradient operator as given in equation (3), we obtain the inhomogeneous term driving equation (15) as

$$D_R \frac{\partial}{\partial \boldsymbol{u}} \cdot \left(\beta \frac{\partial V_1}{\partial \boldsymbol{u}} W_0(\boldsymbol{u})\right) = D_R W_0(\boldsymbol{u}) g_\parallel(\boldsymbol{u}) \xi_\parallel \mathrm{e}^{\mathrm{i}\omega t}, \tag{18}$$

where $g_\parallel(\boldsymbol{u})$ may be expressed in terms of Legendre polynomials as

$$g_\parallel(\boldsymbol{u}) = \sum_{\ell=0}^{\infty} g_{\ell\parallel} P_\ell(\cos\theta), \tag{19}$$

with non-zero expansion coefficients

$$g_{0\|} = -\frac{2}{3}\xi_0 - \frac{16}{15}\frac{\sigma_0^2}{\xi_0} \qquad g_{1\|} = 2 - \frac{12}{5}\sigma_0,$$
$$g_{2\|} = \frac{2}{3}\xi_0 + 8\frac{\sigma_0}{\xi_0} - \frac{16}{21}\frac{\sigma_0^2}{\xi_0}, \tag{20}$$
$$g_{3\|} = \frac{12}{5}\sigma_0, \qquad g_{4\|} = \frac{64}{35}\frac{\sigma_0^2}{\xi_0},$$

and $g_{\ell\|} = 0$ for $\ell > 4$.

Since the inhomogeneous term is proportional to $W_0(\boldsymbol{u})$, it is advantageous here, as also in the earlier study of relaxation functions [15–17], to factor out the equilibrium distribution, writing

$$W_1(\boldsymbol{u}, t) = W_0(\boldsymbol{u})\xi_\| f_\|(\boldsymbol{u}, t). \tag{21}$$

The inhomogeneous Smoluchowski equation (15) then leads to an equation for $f_\|(\boldsymbol{u}, t)$:

$$\frac{\partial f_\|(\boldsymbol{u}, t)}{\partial t} = \mathcal{L}_0 f_\|(\boldsymbol{u}, t) + D_R g_\|(\boldsymbol{u})\mathrm{e}^{\mathrm{i}\omega t}, \tag{22}$$

where we introduce the adjoint Smoluchowski operator $\mathcal{L}_0$ given in spherical polar coordinates by

$$\mathcal{L}_0 = D_R\left(\frac{\partial}{\partial \boldsymbol{u}} - \beta\frac{\partial V_0}{\partial \boldsymbol{u}}\right)\cdot\frac{\partial}{\partial \boldsymbol{u}}$$
$$= D_R\left[\frac{1}{\sin\theta}\frac{\partial}{\partial\theta}\left(\sin\theta\frac{\partial}{\partial\theta}\right) + \frac{1}{\sin^2\theta}\frac{\partial^2}{\partial\phi^2} - (\xi_0\sin\theta + 2\sigma_0\cos\theta\sin\theta)\frac{\partial}{\partial\theta}\right]. \tag{23}$$

Because the perturbing longitudinal field preserves the axial symmetry of the equilibrium solution $W_0$, we can solve the equation for $f_\|(\boldsymbol{u}, t) = f_\|(\theta, t)$ by expansion in Legendre polynomials:

$$f_\|(\theta, t) = \sum_{\ell=0}^{\infty} B_{\ell 0}(t)P_\ell(\cos\theta). \tag{24}$$

Inserting this expansion in equation (22) gives a set of first-order coupled equations for the coefficients $B_{\ell 0}(t)$:

$$\frac{\mathrm{d}B_{\ell 0}}{\mathrm{d}t} = -D_R\left[\ell(\ell+1)B_{\ell 0} + \xi_0\frac{(\ell-1)\ell}{(2\ell-1)}B_{\ell-10} - \xi_0\frac{(\ell+1)(\ell+2)}{(2\ell+3)}B_{\ell+10}\right.$$
$$+ 2\sigma_0\frac{(\ell-2)(\ell-1)\ell}{(2\ell-3)(2\ell-1)}B_{\ell-20} - 2\sigma_0\frac{\ell(\ell+1)}{(2\ell-1)(2\ell+3)}B_{\ell 0}$$
$$\left. - 2\sigma_0\frac{(\ell+1)(\ell+2)(\ell+3)}{(2\ell+3)(2\ell+5)}B_{\ell+20}\right] + D_R g_{\ell\|}\mathrm{e}^{\mathrm{i}\omega t}, \tag{25}$$

where the $g_{\ell\|}$ are given in equation (20). Note that $B_{00}(t)$ obeys the equation

$$\frac{\mathrm{d}B_{00}}{\mathrm{d}t} = D_R\left(\frac{2}{3}\xi_0 B_{10}(t) + \frac{4}{5}\sigma_0 B_{20}(t)\right) + D_R g_{0\|}\mathrm{e}^{\mathrm{i}\omega t}, \tag{26}$$

but that the equations for $B_{\ell 0}(t)$, for $\ell \geqslant 1$, are independent of $B_{00}(t)$. Thus we need solve the coupled system (25) for $\ell \geqslant 1$ only, and then $B_{00}(t)$ is obtained from (26).

For times $t > 0$, the normalization condition imposes a sum rule on the $B_{\ell 0}(t)$. Using (16), (21), (24) and recalling (8) we find the result

$$\sum_{\ell=0}^{\infty} B_{\ell 0}(t)M_\ell(\xi_0, \sigma_0) = 0. \tag{27}$$

This sum rule provides a useful test of the accuracy of the solution for the $B_{\ell 0}(t)$.

The polarization can now easily be expressed to first order in terms of the $B_{\ell 0}(t)$. We introduce a dimensionless polarization $\boldsymbol{F}(t)$ via

$$\boldsymbol{P}(t) = n\langle\boldsymbol{\mu}\rangle_t = nm\int \boldsymbol{u}W(\boldsymbol{u},t)\,\mathrm{d}\boldsymbol{u} = nm\boldsymbol{F}(t). \tag{28}$$

By axial symmetry, $\boldsymbol{F}(t)$ has only a $z$-component which is calculated from the first-order distribution function (14) as

$$F_z(t) = M_1(\xi_0,\sigma_0) + \xi_\parallel \sum_{\ell=0}^{\infty} B_{\ell 0}(t)\frac{1}{2\ell+1}(\ell M_{\ell-1}(\xi_0,\sigma_0) + (\ell+1)M_{\ell+1}(\xi_0,\sigma_0)). \tag{29}$$

## 4. Laplace transform solution

To solve the time-dependent coupled equations (25) we first carry out a Laplace transform to convert the differential equations into linear algebraic equations. Thus we define

$$\hat{B}_{\ell 0}(s) = D_R\int_0^\infty \exp[-st/\tau_R]B_{\ell 0}(t)\,\mathrm{d}t, \tag{30}$$

and, using the initial condition $B_{\ell 0}(0) = 0$, obtain the algebraic system

$$\begin{aligned}\left[-s-\ell(\ell+1)+2\sigma_0\frac{\ell(\ell+1)}{(2\ell-1)(2\ell+3)}\right]\hat{B}_{\ell 0} - \xi_0\frac{(\ell-1)\ell}{(2\ell-1)}\hat{B}_{\ell-10} + \xi_0\frac{(\ell+1)(\ell+2)}{(2\ell+3)}\hat{B}_{\ell+10}\\ -2\sigma_0\frac{(\ell-2)(\ell-1)\ell}{(2\ell-3)(2\ell-1)}\hat{B}_{\ell-20} + 2\sigma_0\frac{(\ell+1)(\ell+2)(\ell+3)}{(2\ell+3)(2\ell+5)}\hat{B}_{\ell+20}\\ = -\frac{g_{\ell\parallel}}{s-\mathrm{i}\omega\tau_R}.\end{aligned} \tag{31}$$

After solving this system for $\ell \geqslant 1$, $\hat{B}_{00}(s)$ may be obtained from

$$-s\hat{B}_{00}(s) + \frac{2}{3}\xi_0\hat{B}_{10}(s) + \frac{4}{5}\sigma_0\hat{B}_{20}(s) = -\frac{g_{0\parallel}}{s-\mathrm{i}\omega\tau_R}. \tag{32}$$

The general structure of the algebraic equations (31), (32) implies that the $\hat{B}_{\ell 0}(s)$ are meromorphic functions of $s$ with poles at positions $s_j = -\lambda_{0j}$, $j = 1, 2, 3, \ldots$, on the negative real $s$-axis, and, in addition, a pole at $s = \mathrm{i}\omega\tau_R$ corresponding to the steady solution driven by the perturbing field. There is no pole in $\hat{B}_{00}(s)$ at $s = 0$ in spite of the form of (32). If one looks explicitly at the system (31) at $s = 0$, one finds that the solution at this value of $s$ is $\hat{B}_{10}(0) = -1/\mathrm{i}\omega\tau_R$, $\hat{B}_{20}(0) = -(4\sigma_0/3\xi_0)(1/\mathrm{i}\omega\tau_R)$, $\hat{B}_{\ell 0}(0) = 0$ for $\ell > 2$, thus cancelling the apparent pole in $\hat{B}_{00}(s)$.

Denoting the residue of $\hat{B}_{\ell 0}(s)$ at $s = -\lambda_{0j}$ by $p_{\ell 0j}$ and the residue at $s = \mathrm{i}\omega\tau_R$ by $q_{\ell 0}$, we can use the inverse Laplace transform to write the solution for $B_{\ell 0}(t)$, when $\ell \geqslant 1$, as

$$B_{\ell 0}(t) = \sum_{j=1}^{\infty} p_{\ell 0j}\exp[-\lambda_{0j}t/\tau_R] + q_{\ell 0}\exp[\mathrm{i}\omega t]. \tag{33}$$

From (32) we find for $B_{00}(t)$

$$\begin{aligned}B_{00}(t) = -\sum_{j=1}^{\infty}\left(\frac{2\xi_0}{3}p_{10j} + \frac{4\sigma_0}{5}p_{20j}\right)\frac{1}{\lambda_{0j}}\exp[-\lambda_{0j}t/\tau_R]\\ + \frac{1}{\mathrm{i}\omega\tau_R}\left(\frac{2\xi_0}{3}q_{10} + \frac{4\sigma_0}{5}q_{20} + g_{0\parallel}\right)\exp[\mathrm{i}\omega t].\end{aligned} \tag{34}$$

Note that $B_{00}(t)$ is well behaved at $\omega = 0$ for the same reason that $\hat{B}_{00}(s)$ does not have a pole at $s = 0$ as explained above. The solution obtained here for the $B_{\ell 0}(t)$ consists of a superposition of a transient term made up of a sum of exponentially decaying modes and a steady term proportional to $\exp[\mathrm{i}\omega t]$. The transient component has the same form as the relaxation functions defined in our earlier work [15] but in the present case, all the residues $p_{\ell 0j}$, $q_{\ell 0}$ are dependent on the frequency $\omega$ of the perturbing field.

From the $B_{\ell 0}(t)$ we obtain the complex dimensionless polarization $F_z(t)$ as

$$F_z(t) = M_1(\xi_0, \sigma_0) + \xi_\| M_1(\xi_0, \sigma_0) B_{00}(t) + \xi_\| \sum_{j=1}^{\infty} P_{0j}(\omega) \exp[-\lambda_{0j} t/\tau_R] + \xi_\| Q_0(\omega) \exp[\mathrm{i}\omega t], \tag{35}$$

where

$$\begin{aligned} P_{0j}(\omega) &= \sum_{\ell=1}^{\infty} p_{\ell 0j}(\omega) \frac{1}{2\ell+1} (\ell M_{\ell-1}(\xi_0, \sigma_0) + (\ell+1) M_{\ell+1}(\xi_0, \sigma_0)), \\ Q_0(\omega) &= \sum_{\ell=1}^{\infty} q_{\ell 0}(\omega) \frac{1}{2\ell+1} (\ell M_{\ell-1}(\xi_0, \sigma_0) + (\ell+1) M_{\ell+1}(\xi_0, \sigma_0)). \end{aligned} \tag{36}$$

The physical polarization is given by the real part of $F_z(t)$.

## 5. Removing the steady-state pole

In the solution given above, all amplitudes $p_{\ell 0j}(\omega)$, $q_{\ell 0}(\omega)$ have a frequency dependence which can only be obtained by numerical evaluation of these residues at a range of $\omega$-values. However, it is possible to make the frequency dependence more explicit if we factor out the pole at $s = \mathrm{i}\omega\tau_R$ from the $\hat{B}_{\ell 0}(s)$. To do this, define functions $\hat{C}_{\ell 0}(s)$ by setting

$$\hat{B}_{\ell 0} = \frac{\hat{C}_{\ell 0}(s)}{s - \mathrm{i}\omega\tau_R}. \tag{37}$$

After making this substitution into the system (31) the factor $1/(s - \mathrm{i}\omega\tau_R)$ disappears from the equations and the inhomogeneous term is simply given by $-g_{\ell\|}$ which depends only on $\xi_0$, $\sigma_0$ but not on $\omega$. The solution $\hat{C}_{\ell 0}(s)$ again has poles at the same values of $s$ on the negative real axis, $s_j = -\lambda_{0j}$, but no pole at $s = \mathrm{i}\omega\tau_R$. Moreover, if we denote the residue of $\hat{C}_{\ell 0}(s)$ at $s_j = -\lambda_{0j}$ by $d_{\ell 0j}$, then $d_{\ell 0j}(\xi_0, \sigma_0)$ depends on $\xi_0$, $\sigma_0$ but not on $\omega$. The residues $q_{\ell 0}$ of $\hat{B}_{\ell 0}(s)$ at $s = \mathrm{i}\omega\tau_R$ are now simply given as

$$q_{\ell 0} = \hat{C}_{\ell 0}(\mathrm{i}\omega\tau_R). \tag{38}$$

The frequency dependence of the amplitudes $p_{\ell 0j}$, $P_{0j}$ in the transient terms now becomes explicit. We find

$$p_{\ell 0j} = -\frac{d_{\ell 0j}(\xi_0, \sigma_0)}{\lambda_{0j} + \mathrm{i}\omega\tau_R}, \qquad P_{0j} = -\frac{D_{0j}(\xi_0, \sigma_0)}{\lambda_{0j} + \mathrm{i}\omega\tau_R}, \tag{39}$$

with

$$D_{0j}(\xi_0, \sigma_0) = \sum_{\ell=1}^{\infty} d_{\ell 0j}(\xi_0, \sigma_0) \frac{1}{2\ell+1} (\ell M_{\ell-1}(\xi_0, \sigma_0) + (\ell+1) M_{\ell+1}(\xi_0, \sigma_0)). \tag{40}$$

The solution for $B_{00}(t)$ now becomes

$$B_{00}(t) = \frac{2}{15} \sum_{j=1}^{\infty} \frac{5\xi_0 d_{10j} + 6\sigma_0 d_{20j}}{\lambda_{0j}(\lambda_{0j} + \mathrm{i}\omega\tau_R)} \exp[-\lambda_{0j} t/\tau_R] + \frac{1}{\mathrm{i}\omega\tau_R} \left[ \frac{2\xi_0}{3} (\hat{C}_{10}(\mathrm{i}\omega\tau_R) - 1) + \frac{4\sigma_0}{5} \left( \hat{C}_{20}(\mathrm{i}\omega\tau_R) - \frac{4\sigma_0}{3\xi_0} \right) \right] \exp[\mathrm{i}\omega t]. \tag{41}$$

The frequency dependence in $q_{\ell 0}$ and $Q_0$ is not yet explicit, but to find it involves only solving the equations for $\hat{C}_{\ell 0}(s)$ at $s = \mathrm{i}\omega\tau_R$ rather than solving the equations for $\hat{B}_{\ell 0}(s)$ and then extracting the residue at $s = \mathrm{i}\omega\tau_R$.

## 6. Transients and susceptibility

The dimensionless polarization $F_z(t)$ as given in (35) is a superposition of a constant background field value, a transient and a steady term. By subtracting off the constant background contribution we can define differential transient and steady components of the linear response, denoted as $T_z(t)$ and $S_z(t)$ respectively:

$$\Delta F_z(t) = F_z(t) - M_1(\xi_0, \sigma_0) = \xi_{\|}(T_z(t) + S_z(t)), \tag{42}$$

with the transient given as

$$T_z(t) = \frac{2M_1(\xi_0, \sigma_0)}{15} \sum_{j=1}^{\infty} \frac{5\xi_0 d_{10j} + 6\sigma_0 d_{20j}}{\lambda_{0j}(\lambda_{0j} + \mathrm{i}\omega\tau_R)} \exp[-\lambda_{0j} t/\tau_R] - \sum_{j=1}^{\infty} \frac{D_{0j}}{\lambda_{0j} + \mathrm{i}\omega\tau_R} \exp[-\lambda_{0j} t/\tau_R]. \tag{43}$$

From the steady term we define a dimensionless complex susceptibility $\chi_{\|}(\omega)$ by

$$S_z(t) = \tfrac{1}{3}\chi_{\|}(\omega) \exp[\mathrm{i}\omega t]. \tag{44}$$

From the solution for $F_z(t)$ we find for the susceptibility

$$\chi_{\|}(\omega) = \frac{3}{\mathrm{i}\omega\tau_R} M_1(\xi_0, \sigma_0)\left[\frac{2\xi_0}{3}(\hat{C}_{10}(\mathrm{i}\omega\tau_R) - 1) + \frac{4\sigma_0}{5}\left(\hat{C}_{20}(\mathrm{i}\omega\tau_R) - \frac{4\sigma_0}{3\xi_0}\right)\right] + 3\sum_{\ell=1}^{\infty} \hat{C}_{\ell 0}(\mathrm{i}\omega\tau_R)\frac{1}{2\ell+1}(\ell M_{\ell-1}(\xi_0, \sigma_0) + (\ell+1)M_{\ell+1}(\xi_0, \sigma_0)). \tag{45}$$

The normalization for $\chi_{\|}(\omega)$ is chosen such that for polar particles ($m \neq 0, \alpha_1 = 0, \alpha_2 = 0$) in zero background field ($\boldsymbol{E}_0 = 0$) it reduces to the Debye function [3, 4]

$$\chi_{\|}^{D}(\omega) = \frac{1}{1 + \mathrm{i}\omega\tau_D}. \tag{46}$$

The frequency dependence of $\chi_{\|}(\omega)$ as expressed in (45) is still implicit, involving the $\hat{C}_{\ell 0}(s)$ evaluated at $s = \mathrm{i}\omega\tau_R$. However, using the initial condition, $\Delta F_z(0) = \xi_{\|}(T_z(0) + S_z(0)) = 0$, we get an alternative expression for $\chi_{\|}$ in terms of the transient solution, $\chi_{\|}(\omega) = -3T_z(0)$. In this way of expressing $\chi_{\|}$ the dependence on $\omega$ is completely explicit:

$$\chi_{\|}(\omega) = -\frac{2M_1(\xi_0, \sigma_0)}{5} \sum_{j=1}^{\infty} \frac{5\xi_0 d_{10j} + 6\sigma_0 d_{20j}}{\lambda_{0j}(\lambda_{0j} + \mathrm{i}\omega\tau_R)} + 3\sum_{j=1}^{\infty} \frac{D_{0j}}{\lambda_{0j} + \mathrm{i}\omega\tau_R}. \tag{47}$$

Finally, we note that from the real part of the transient term, $T_z'(t) = \operatorname{Re} T_z(t)$, we can define a normalized relaxation function

$$\Gamma_z(t) = T_z'(t)/T_z'(0), \tag{48}$$

and its associated mean relaxation time

$$\tau_{zM}(\omega) = \int_0^{\infty} \Gamma_z(t)\, \mathrm{d}t. \tag{49}$$

By use of (43) we get the result

$$\tau_{zM}(\omega)/\tau_R = \left[\frac{2M_1(\xi_0,\sigma_0)}{15}\sum_{j=1}^{\infty}\frac{5\xi_0 d_{10j}+6\sigma_0 d_{20j}}{\lambda_{0j}(\lambda_{0j}^2+\omega^2\tau_R^2)} - \sum_{j=1}^{\infty}\frac{D_{0j}}{\lambda_{0j}^2+\omega^2\tau_R^2}\right] \times \left[\frac{2M_1(\xi_0,\sigma_0)}{15}\sum_{j=1}^{\infty}\frac{5\xi_0 d_{10j}+6\sigma_0 d_{20j}}{\lambda_{0j}^2+\omega^2\tau_R^2} - \sum_{j=1}^{\infty}\frac{\lambda_{0j}D_{0j}}{\lambda_{0j}^2+\omega^2\tau_R^2}\right]^{-1}. \tag{50}$$

We see from the results in this section that the transient response, the mean relaxation time of the transient and the susceptibility can all be given in terms of the decay rates $\lambda_{0j}(\xi_0,\sigma_0)$ and the associated residues $d_{\ell 0j}(\xi_0,\sigma_0)$ of the $\hat{C}_{\ell 0}(s)$. In these exact expressions the dependence on the frequency $\omega$ of the perturbing field is completely explicit.

## 7. Response to a transverse probe field

If we consider a transverse probe field instead of a longitudinal one we can obtain similar results to those in section 6 with only slightly more effort. For simplicity we assume that the weak probe field lies now along the $x$-axis, $\boldsymbol{E}_1 = \boldsymbol{E}_\perp = E_\perp \boldsymbol{e}_x$. Writing $\boldsymbol{u}$ in terms of spherical polar coordinates as $\boldsymbol{u} = (\sin\theta\cos\phi, \sin\theta\sin\phi, \cos\theta)$ we obtain the perturbing potential for transverse fields as

$$\beta V_1(\boldsymbol{u},t) = -[\beta m + \beta(\alpha_1-\alpha_2)(\boldsymbol{u}\cdot\boldsymbol{E}_0)](\boldsymbol{u}\cdot\boldsymbol{E}_\perp)\mathrm{e}^{\mathrm{i}\omega t} = -\left[1+\frac{2\sigma_0}{\xi_0}\cos\theta\right]\sin\theta\cos\phi\,\xi_\perp \mathrm{e}^{\mathrm{i}\omega t}, \tag{51}$$

with $\xi_\perp = \beta m E_\perp$. The inhomogeneous driving term analogous to (18) is now

$$D_R\frac{\partial}{\partial\boldsymbol{u}}\cdot\left(\beta\frac{\partial V_1}{\partial\boldsymbol{u}}W_0(\boldsymbol{u})\right) = D_R W_0(\boldsymbol{u})g_\perp(\boldsymbol{u})\xi_\perp \mathrm{e}^{\mathrm{i}\omega t}, \tag{52}$$

where now $g_\perp(\boldsymbol{u})$ can be expressed in terms of associated Legendre functions [20] $P_\ell^1(\cos\theta)$ as

$$g_\perp(\boldsymbol{u}) = \sum_{\ell=1}^{\infty} g_{\ell\perp}P_\ell^1(\cos\theta)\cos\phi, \tag{53}$$

with non-zero expansion coefficients

$$g_{1\perp} = 2-\frac{4}{5}\sigma_0, \qquad g_{2\perp} = \frac{1}{3}\xi_0 + 4\frac{\sigma_0}{\xi_0} - \frac{4}{21}\frac{\sigma_0^2}{\xi_0}, \qquad g_{3\perp} = \frac{4}{5}\sigma_0, \qquad g_{4\perp} = \frac{16}{35}\frac{\sigma_0^2}{\xi_0}, \tag{54}$$

and $g_{\ell\perp} = 0$ for $\ell > 4$.

Factoring out the equilibrium distribution as in (21), $W_1(\boldsymbol{u},t) = W_0(\boldsymbol{u})\xi_\perp f_\perp(\boldsymbol{u},t)$, we find the inhomogeneous equation for $f_\perp(\boldsymbol{u},t)$:

$$\frac{\partial f_\perp(\boldsymbol{u},t)}{\partial t} = \mathcal{L}_0 f_\perp(\boldsymbol{u},t) + D_R g_\perp(\boldsymbol{u})\mathrm{e}^{\mathrm{i}\omega t}, \tag{55}$$

with initial value $f_\perp(\boldsymbol{u},0) = 0$. Since $g_\perp(\boldsymbol{u})$ no longer has axial symmetry, we can solve the equation for $f_\perp(\theta,\phi,t)$ with the expansion

$$f_\perp(\theta,\phi,t) = \sum_{\ell=1}^{\infty}(B_{\ell 1}(t)\mathrm{e}^{\mathrm{i}\phi}P_\ell^1(\cos\theta) + B_{\ell -1}(t)\mathrm{e}^{-\mathrm{i}\phi}P_\ell^{-1}(\cos\theta)). \tag{56}$$

Again we obtain a set of coupled first-order differential equations for the coefficient functions $B_{\ell 1}(t)$, $B_{\ell -1}(t)$. For the $B_{\ell 1}(t)$ these have the form

$$\frac{dB_{\ell 1}}{dt} = -D_R\left[\ell(\ell+1)B_{\ell 1} + \xi_0\frac{(\ell-1)^2}{(2\ell-1)}B_{\ell-11} - \xi_0\frac{(\ell+2)^2}{(2\ell+3)}B_{\ell+11}\right.$$
$$+ 2\sigma_0\frac{(\ell-2)^2(\ell-1)}{(2\ell-3)(2\ell-1)}B_{\ell-21} - 2\sigma_0\frac{[\ell(\ell+1)-3]}{(2\ell-1)(2\ell+3)}B_{\ell 1}$$
$$\left. - 2\sigma_0\frac{(\ell+2)(\ell+3)^2}{(2\ell+3)(2\ell+5)}B_{\ell+21}\right] + \frac{1}{2}D_R g_{\ell\perp}e^{i\omega t}. \quad (57)$$

The equations for $B_{\ell -1}$ are slightly different in form but one can show that the solutions for the $B_{\ell -1}(t)$ are simply expressed in terms of the $B_{\ell 1}(t)$ by

$$B_{\ell -1}(t) = -\ell(\ell+1)B_{\ell 1}(t). \quad (58)$$

The dimensionless polarization as defined in (28) now has a constant component in the $z$-direction and a time-dependent component in the $x$-direction:

$$\boldsymbol{F}_{Trans}(t) = \boldsymbol{e}_z M_1(\xi_0, \sigma_0) + \boldsymbol{e}_x\frac{\xi_\perp}{2}\sum_{\ell=1}^{\infty}\left(B_{\ell 1}(t) - \frac{1}{\ell(\ell+1)}B_{\ell -1}(t)\right)\frac{\ell(\ell+1)}{2\ell+1}$$
$$\times\, (M_{\ell-1}(\xi_0,\sigma_0) - M_{\ell+1}(\xi_0,\sigma_0)). \quad (59)$$

Using (58) and subtracting off the constant background gives for the transverse linear response

$$\Delta F_x(t) = \xi_\perp\sum_{\ell=1}^{\infty}B_{\ell 1}(t)\frac{\ell(\ell+1)}{2\ell+1}(M_{\ell-1}(\xi_0,\sigma_0) - M_{\ell+1}(\xi_0,\sigma_0)). \quad (60)$$

The method of solution now follows the pattern of the longitudinal case. Laplace transformation converts the differential equations (57) to linear algebraic equations:

$$\left[-s - \ell(\ell+1) + 2\sigma_0\frac{[\ell(\ell+1)-3]}{(2\ell-1)(2\ell+3)}\right]\hat{B}_{\ell 1} - \xi_0\frac{(\ell-1)^2}{(2\ell-1)}\hat{B}_{\ell-11} + \xi_0\frac{(\ell+2)^2}{(2\ell+3)}\hat{B}_{\ell+11}$$
$$- 2\sigma_0\frac{(\ell-2)^2(\ell-1)}{(2\ell-3)(2\ell-1)}\hat{B}_{\ell-21} + 2\sigma_0\frac{(\ell+2)(\ell+3)^2}{(2\ell+3)(2\ell+5)}\hat{B}_{\ell+21} = -\frac{1}{2}\frac{g_{\ell\perp}}{s - i\omega\tau_R}. \quad (61)$$

Unlike the longitudinal case, there is no term like $B_{00}(t)$ which can be eliminated in terms of other coefficients. The system (61) has the same mathematical structure as the system (31). The $\hat{B}_{\ell 1}(s)$ have poles on the negative real $s$-axis at points $s_j = -\lambda_{1j}$, $j = 1, 2, 3, \ldots$, with residues $p_{\ell 1j}$ and a pole at $s = i\omega\tau_R$ with residue $q_{\ell 1}$. The analogues of (35) and (36) are

$$\Delta F_x(t) = \xi_\perp\sum_{j=1}^{\infty}P_{1j}(\omega)\exp[-\lambda_{1j}t/\tau_R] + \xi_\perp Q_1(\omega)\exp[i\omega t], \quad (62)$$

and

$$P_{1j}(\omega) = \sum_{\ell=1}^{\infty}p_{\ell 1j}(\omega)\frac{\ell(\ell+1)}{2\ell+1}(M_{\ell-1}(\xi_0,\sigma_0) - M_{\ell+1}(\xi_0,\sigma_0)),$$
$$Q_1(\omega) = \sum_{\ell=1}^{\infty}q_{\ell 1}(\omega)\frac{\ell(\ell+1)}{2\ell+1}(M_{\ell-1}(\xi_0,\sigma_0) - M_{\ell+1}(\xi_0,\sigma_0)). \quad (63)$$

Just as in section 5 we can remove the steady-state pole by introducing functions $\hat{C}_{\ell 1}(s)$ via the definition

$$\hat{B}_{\ell 1} = \frac{\hat{C}_{\ell 1}(s)}{s - i\omega\tau_R}. \quad (64)$$

The function $\hat{C}_{\ell 1}(s)$ has poles at $s_j = -\lambda_{1j}$ with associated residues $d_{\ell 1j}(\xi_0, \sigma_0)$ which are independent of $\omega$. We find

$$p_{\ell 1j} = -\frac{d_{\ell 1j}(\xi_0, \sigma_0)}{\lambda_{1j} + \mathrm{i}\omega\tau_R}, \qquad P_{1j} = -\frac{D_{1j}(\xi_0, \sigma_0)}{\lambda_{1j} + \mathrm{i}\omega\tau_R}, \tag{65}$$

with

$$D_{1j}(\xi_0, \sigma_0) = \sum_{\ell=1}^{\infty} d_{\ell 1j}(\xi_0, \sigma_0) \frac{\ell(\ell+1)}{2\ell+1} (M_{\ell-1}(\xi_0, \sigma_0) - M_{\ell+1}(\xi_0, \sigma_0)). \tag{66}$$

Finally, we introduce transient and steady components of the transverse response:

$$\Delta F_x(t) = \xi_\perp (T_x(t) + S_x(t)), \tag{67}$$

with

$$T_x(t) = -\sum_{j=1}^{\infty} \frac{D_{1j}}{\lambda_{1j} + \mathrm{i}\omega\tau_R} \exp[-\lambda_{1j} t/\tau_R]. \tag{68}$$

A transverse susceptibility $\chi_\perp(\omega)$ is defined by

$$S_x(t) = \tfrac{1}{3}\chi_\perp(\omega) \exp[\mathrm{i}\omega t], \tag{69}$$

which can be expressed as

$$\chi_\perp(\omega) = 3 \sum_{\ell=1}^{\infty} \hat{C}_{\ell 1}(\mathrm{i}\omega\tau_R) \frac{\ell(\ell+1)}{2\ell+1} (M_{\ell-1}(\xi_0, \sigma_0) - M_{\ell+1}(\xi_0, \sigma_0)). \tag{70}$$

The initial condition, $\Delta F_x(0) = 0$, gives the alternative expression

$$\chi_\perp(\omega) = 3 \sum_{j=1}^{\infty} \frac{D_{1j}}{\lambda_{1j} + \mathrm{i}\omega\tau_R}. \tag{71}$$

From the real part of the transient term, $T_x' = \mathrm{Re}\, T_x(t)$, we define a transverse relaxation function

$$\Gamma_x(t) = T_x'(t)/T_x'(0), \tag{72}$$

and its mean relaxation time

$$\tau_{xM}(\omega) = \int_0^\infty \Gamma_x(t)\, \mathrm{d}t. \tag{73}$$

From (68) we can express this as

$$\tau_{xM}(\omega)/\tau_R = \left[\sum_{j=1}^{\infty} \frac{D_{1j}}{\lambda_{1j}^2 + \omega^2\tau_R^2}\right] \Bigg/ \left[\sum_{j=1}^{\infty} \frac{\lambda_{1j} D_{1j}}{\lambda_{1j}^2 + \omega^2\tau_R^2}\right]. \tag{74}$$

Once again, as in the longitudinal case, the frequency dependence of $T_x(t)$, $\tau_{xM}(\omega)$ and $\chi_\perp(\omega)$ has been made explicit.

## 8. Numerical results

From the results of sections 6 and 7 it is clear that to study the transient response, the mean relaxation time of the transient and the steady-state susceptibility we must calculate the relaxation rates $\lambda_{mj}$ and the amplitudes $d_{\ell mj}$ $(m = 0, 1)$ from the algebraic equations for the $\hat{C}_{\ell m}(s)$. These equations are identical in form with those for the $\hat{B}_{\ell m}(s)$ in equations (31) and (61) except that the factor $1/(s - \mathrm{i}\omega\tau_R)$ on the right-hand side is omitted. As explained in earlier work [15–17] the method of solution is truncation of the algebraic equations at

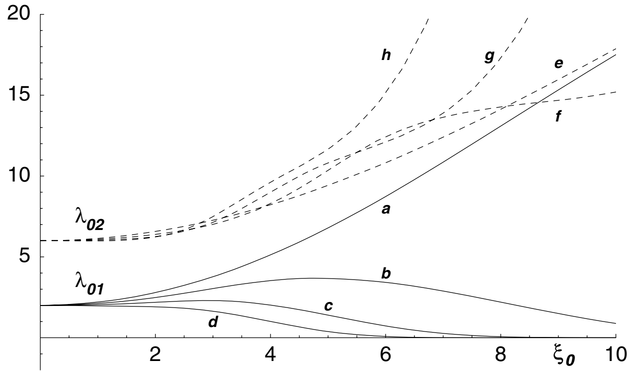


**Figure 1.** Plots of the two lowest relaxation rates $\lambda_{01}$ (solid) and $\lambda_{02}$ (dashed) for the longitudinal case. Curves $a$ and $e$ correspond to pure dipoles ($r_0 = 0$) while curves $b$, $c$, $d$ and $f$, $g$, $h$ correspond respectively to $r_0 = 0.1$, 0.2, 0.3.

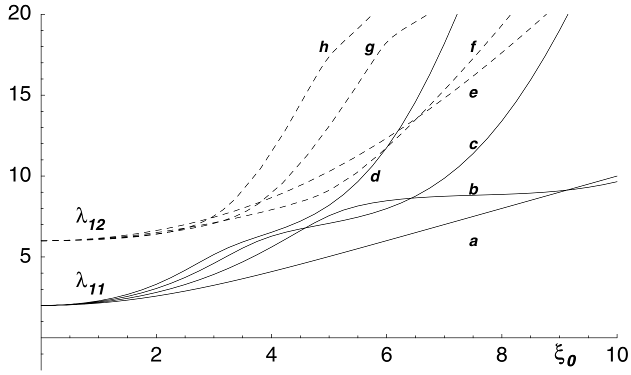


**Figure 2.** Plots of the two lowest relaxation rates $\lambda_{11}$ (solid) and $\lambda_{12}$ (dashed) for the transverse case. Curves $a$ and $e$ correspond to pure dipoles ($r_0 = 0$) while curves $b$, $c$, $d$ and $f$, $g$, $h$ correspond respectively to $r_0 = 0.1$, 0.2, 0.3.

sufficiently high order, $\ell_{Max} = N$. The relaxation rates are computed then as the roots $s_j = -\lambda_{mjN}$ of the characteristic polynomial of the truncated equation set using the function 'Characteristic Polynomial' of Mathematica 4.0. The amplitudes $d_{\ell mj}$ are approximated by the residues of $\hat{C}_{\ell m}(s)$ at the poles $\{\lambda_{mjN}\}$ and are found by numerical contour integration of $\hat{C}_{\ell m}(s)$ around each of the poles in the complex $s$-plane. Convergence of the approximation is most rapid for pure dipoles and becomes less rapid as $\sigma_0$ increases relative to $\xi_0$. Convergence is easily checked by varying the truncation order $N$. For three-figure accuracy of all numbers reported here, we found $N \leqslant 35$ was sufficient.

In figures 1 and 2 we show the dependence on the external field $\xi_0$ of the two lowest decay rates $\lambda_{m1}$, $\lambda_{m2}$ for both the longitudinal ($m = 0$) and transverse ($m = 1$) cases. For permanent dipoles ($\sigma_0 = 0$, curves $a$ and $e$), these decay rates increase monotonically as $\xi_0$ increases. In earlier relaxation calculations [15–17] we have shown this for the lowest six modes $\lambda_{mj}$, $j = 1, \ldots, 6$. For polarizable particles there is a significant change. As noted earlier in section 2, for dielectric particles the ratio $r_0 = \sigma_0/\xi_0^2$ is independent of the strength of the background field and is characteristic of the particle polarizability relative to its intrinsic dipole moment. In figures 1 and 2 we show also the roots $\lambda_{mj}$ for the three cases

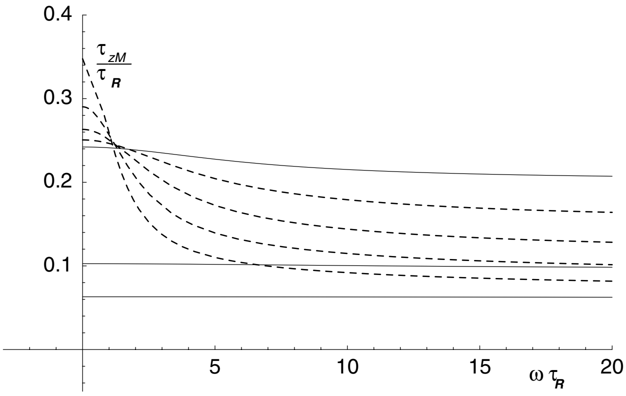


**Figure 3.** Plots of the longitudinal transient relaxation time as a function of frequency. The solid curves from the top down correspond respectively to pure dipoles with $\xi_0 = 3, 6, 9$. The dashed curves describe polarizable particles with $\xi_0 = 3$ and polarizability ratios $r_0 = 0.1, 0.2, 0.3, 0.4$ corresponding respectively to the dashed curves ordered from the top down at the high-frequency end.

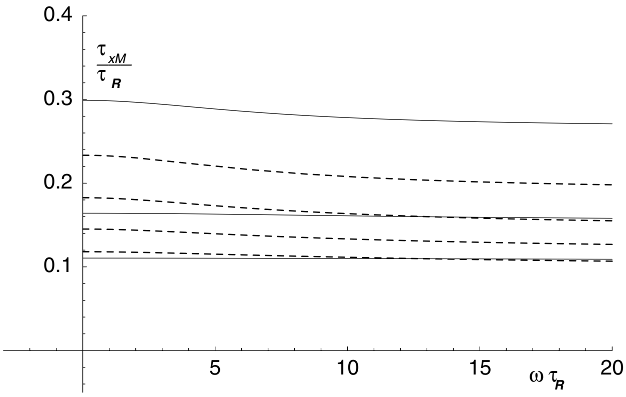


**Figure 4.** Plots of the transverse transient relaxation time as a function of frequency. The solid curves from the top down correspond respectively to pure dipoles with $\xi_0 = 3, 6, 9$. The dashed curves describe polarizable particles with $\xi_0 = 3$ and polarizability ratios $r_0 = 0.1, 0.2, 0.3, 0.4$ corresponding respectively to the dashed curves ordered from the top down.

$r_0 = 0.1, 0.2, 0.3$ (curves $b$, $c$, $d$ and $f$, $g$, $h$) corresponding to increasing polarizability. For $\lambda_{0j}$ with $j \geqslant 2$ and for all $\lambda_{1j}$ the relaxation rates again increase as $\xi_0$ increases but with weak oscillations appearing. However, in the longitudinal case the rate $\lambda_{01}$ can decrease and approach zero at large values of $\xi_0$ with the ratio $r_0$ fixed but non-zero. A similar behaviour was seen in our earlier relaxation calculations [16].

Such a decreasing value of $\lambda_{01}(\xi_0, \sigma_0)$ for polarizable particles can produce a qualitative change in the susceptibility and the transient relaxation time $\tau_{zM}$ at low frequencies $\omega$ since $\lambda_{01}$ occurs in denominators of equations (47) and (50) in the form $\lambda_{01} + \mathrm{i}\omega\tau_R$ or $\lambda_{01}^2 + \omega^2\tau_R^2$ which can each become small at low frequencies. The small denominators are relevant of course only if the numerators, given in terms of the amplitudes $d_{\ell mj}$ and $D_{mj}$, remain appreciable in magnitude. In tables 1 and 2 we list the amplitudes $d_{\ell m1}$ associated with the lowest root $\lambda_{m1}$ for $\ell = 1, \ldots, 6$ for pure dipoles ($r_0 = 0$) and for polarizable particles ($r_0 = 0.1, 0.2, 0.3, 0.4$) in a background field corresponding to $\xi_0 = 3$. In tables 3 and 4 we give the $D_{mj}$ for the same

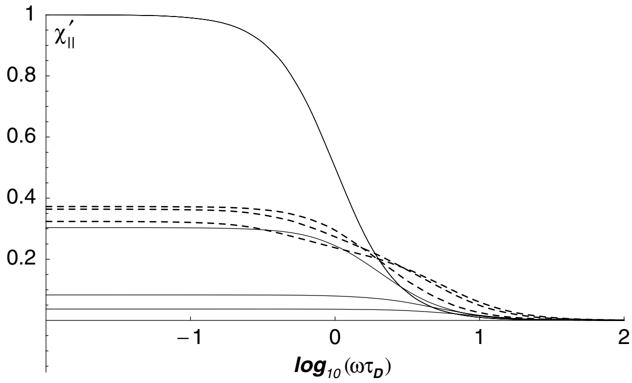


**Figure 5.** Plots of the real part of the longitudinal susceptibility $\chi'_{\parallel}$. The solid curves from the top down correspond to pure dipoles with $\xi_0 = 0$ (the Debye susceptibility), $\xi_0 = 3, 6, 9$ respectively. The dashed curves represent polarizable particles with $\xi_0 = 3$ and polarizability ratios $r_0 = 0.1, 0.3, 0.4$ corresponding respectively to the dashed curves ordered from the top down at the vertical intercept end of the graph.

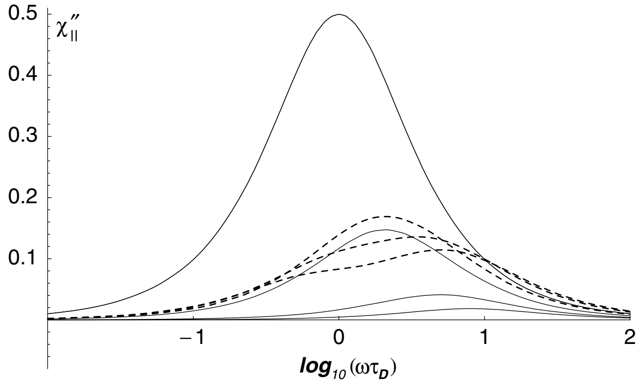


**Figure 6.** Plots of the dissipative part of the longitudinal susceptibility $\chi''_{\parallel}$. The solid curves from the top down correspond to pure dipoles with $\xi_0 = 0$ (the Debye susceptibility), $\xi_0 = 3, 6, 9$ respectively. The dashed curves represent polarizable particles with $\xi_0 = 3$ and polarizability ratios $r_0 = 0.1, 0.3, 0.4$ corresponding respectively to the dashed curves ordered from the top down at the centre of the graph ($\omega\tau_D = 1$).

parameter values and for $j = 1, \ldots, 6$. We note that for all values of $r_0$ the amplitudes $d_{\ell m1}$ decrease in magnitude as $\ell$ increases but there can be appreciable weight associated with more than one $\ell$-value. For the weights $D_{mj}$ we see that for pure dipoles ($r_0 = 0$) the dominant weight is in the slowest mode, $j = 1$, but that as $r_0$ increases, the weights spread appreciably across several modes and, for $r_0 = 0.3, 0.4$, the weights $D_{m2}$ associated with the next slowest mode, $j = 2$, become dominant. In spite of this, the weights $d_{101}$, $d_{201}$, $D_{01}$ are sufficiently large that the anomalous rate $\lambda_{01}$ has a significant effect easily seen in the mean decay time $\tau_{zM}$ shown in figure 3. For pure dipoles we see there that $\tau_{zM}(\omega)$ is only weakly dependent on frequency and decreases monotonically as $\xi_0$ increases. However, for polarizable particles, as $r_0$ increases at fixed $\xi_0 = 3$, there is a more and more marked enhancement of $\tau_{zM}(\omega)$ at low frequencies. This is in contrast to the behaviour of $\tau_{xM}(\omega)$ for transverse perturbations shown in figure 4 where both pure dipoles and polarizable particles behave similarly with only a weak

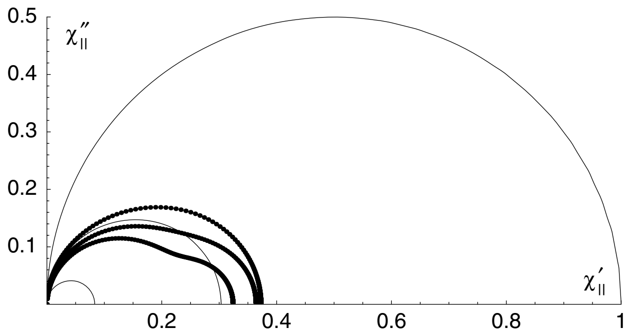


**Figure 7.** Cole–Cole plots of $\chi''_{\|}$ against $\chi'_{\|}$. The solid curves (from the outermost to innermost) correspond to pure dipoles with $\xi_0 = 0$ (the Debye susceptibility), $\xi_0 = 3, 6$ respectively. The heavy dotted curves represent polarizable particles with $\xi_0 = 3$ and polarizability ratios $r_0 = 0.1$ (outermost curve) $r_0 = 0.3$ (middle curve) and $r_0 = 0.4$ (innermost curve).

**Table 1.** Amplitudes $d_{\ell 01}$ for $\ell = 1, \ldots, 6$ and varying polarizability $\sigma_0 = r_0\xi_0^2$ with $\xi_0 = 3$.

| $r_0$ | $d_{101}$ | $d_{201}$ | $d_{301}$ | $d_{401}$ | $d_{501}$ | $d_{601}$ |
|---|---|---|---|---|---|---|
| 0.0 | 5.957 | −2.938 | 1.046 | −0.296 | 0.0700 | −0.0142 |
| 0.1 | 5.827 | −3.117 | 0.793 | −0.032 | −0.0395 | 0.0102 |
| 0.2 | 4.472 | −2.402 | 0.357 | 0.198 | −0.0932 | −0.0004 |
| 0.3 | 3.058 | −1.578 | 0.046 | 0.280 | −0.0812 | −0.0251 |
| 0.4 | 1.964 | −0.948 | −0.102 | 0.258 | −0.0452 | −0.0420 |

**Table 2.** Amplitudes $d_{\ell 11}$ for $\ell = 1, \ldots, 6$ and varying polarizability $\sigma_0 = r_0\xi_0^2$ with $\xi_0 = 3$.

| $r_0$ | $d_{111}$ | $d_{211}$ | $d_{311}$ | $d_{411}$ | $d_{511}$ | $d_{611}$ |
|---|---|---|---|---|---|---|
| 0.0 | 2.166 | −0.501 | 0.115 | −0.024 | 0.0044 | −0.0007 |
| 0.1 | 3.790 | −1.205 | 0.239 | −0.015 | −0.0058 | 0.0016 |
| 0.2 | 6.012 | −2.559 | 0.504 | 0.052 | −0.0501 | 0.0059 |
| 0.3 | 6.956 | −3.657 | 0.721 | 0.201 | −0.1308 | 0.0042 |
| 0.4 | 1.964 | −0.948 | −0.102 | 0.258 | −0.0452 | −0.0420 |

**Table 3.** Amplitudes $D_{0j}$ for $j = 1, \ldots, 6$ and varying polarizability $\sigma_0 = r_0\xi_0^2$ with $\xi_0 = 3$.

| $r_0$ | $D_{01}$ | $D_{02}$ | $D_{03}$ | $D_{04}$ | $D_{05}$ | $D_{06}$ |
|---|---|---|---|---|---|---|
| 0.0 | 2.435 | −0.709 | 0.067 | −0.001 | 0.0000 | 0.0000 |
| 0.1 | 2.516 | −0.128 | −0.113 | 0.033 | 0.0009 | 0.0000 |
| 0.2 | 2.029 | 2.091 | −0.958 | 0.071 | 0.0270 | −0.0008 |
| 0.3 | 1.451 | 5.516 | −2.212 | −0.205 | 0.1542 | 0.0008 |
| 0.4 | 0.969 | 9.613 | −2.963 | −1.462 | 0.4961 | 0.0301 |

enhancement near $\omega = 0$ and with a decrease of $\tau_{xM}$ as either the field $\xi_0$ or the polarizability $r_0$ increases.

The complex susceptibility is best discussed in terms of real and negative imaginary components which, for longitudinal perturbations, we write as

$$\chi_{\|} = \chi'_{\|} - \mathrm{i}\chi''_{\|}, \tag{75}$$

and correspondingly for $\chi_{\perp}$ in the transverse case. The negative imaginary part $\chi''_{\|}$ as defined

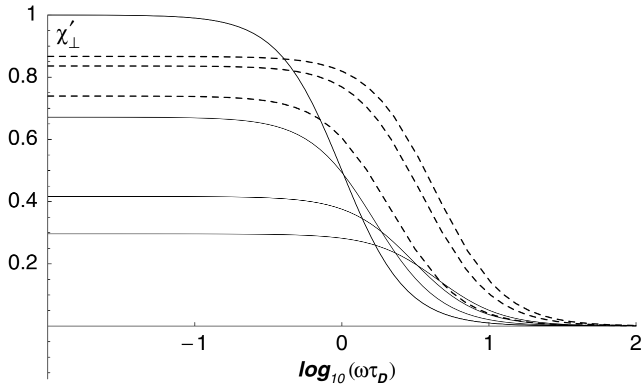


**Figure 8.** Plots of the real part of the transverse susceptibility $\chi'_\perp$. The solid curves ordered from the top down at the vertical intercept end correspond to pure dipoles with $\xi_0 = 0$ (the Debye susceptibility), $\xi_0 = 3, 6, 9$ respectively. The dashed curves represent polarizable particles with $\xi_0 = 3$ and polarizability ratios $r_0 = 0.1, 0.3, 0.4$ corresponding respectively to the dashed curves ordered from the bottom up.

**Table 4.** Amplitudes $D_{1j}$ for $j = 1, \ldots, 6$ and varying polarizability $\sigma_0 = r_0\xi_0^2$ with $\xi_0 = 3$.

| $r_0$ | $D_{11}$ | $D_{12}$ | $D_{13}$ | $D_{14}$ | $D_{15}$ | $D_{16}$ |
|---|---|---|---|---|---|---|
| 0.0 | 0.692 | 0.078 | 0.006 | 0.0002 | 0.0000 | 0.0000 |
| 0.1 | 0.794 | 0.282 | 0.053 | 0.0059 | 0.0004 | 0.0000 |
| 0.2 | 0.715 | 0.687 | 0.144 | 0.0261 | 0.0032 | 0.0003 |
| 0.3 | 0.409 | 1.324 | 0.264 | 0.0643 | 0.0107 | 0.0013 |
| 0.4 | 0.969 | 9.613 | −2.963 | −1.4621 | 0.4961 | 0.0301 |

in (75) is the dissipative component which is always $\geqslant 0$. For pure dipoles we can compare our numerical results for $\chi_\parallel$ with those calculated from the after-effect function in [11]. In figures 5 and 6 we plot $\chi'_\parallel$, $\chi''_\parallel$ against $\log_{10}(\omega\tau_D)$ to facilitate comparison with this earlier work. The results for pure dipoles are shown as solid curves corresponding first to the Debye limit as given in (46) ($\xi_0 = 0$), and then to external field values $\xi_0 = 3, 6, 9$. There is complete numerical agreement with the earlier calculations [11] for $\xi_0 = 3, 6$. In addition we show as dashed curves on the same plot the susceptibility for polarizable particles with $\xi_0 = 3$ and $\sigma_0 = r_0\xi_0^2$ for $r_0 = 0.1, 0.3, 0.4$. For increasing polarizability ($r_0 = 0.3, 0.4$) there is a qualitative change of behaviour from the results for pure dipoles. This is best seen in a Cole–Cole plot [4, 21] of $\chi''_\parallel$ against $\chi'_\parallel$ as shown in figure 7. On such a plot the Debye susceptibility (46) is a perfect semicircle of radius 0.5. If the susceptibility can be well approximated by the Debye form but with a single relaxation time $\tau_M$ replacing $\tau_D$ and an altered amplitude, then we should again get an almost semicircular plot of reduced radius. For pure dipoles, shown as solid lines for $\xi_0 = 0$ (Debye), $\xi_0 = 3, 6$, the plots are closely semicircular, as is the plot (heavy dots) for a polarizable particle with $\xi_0 = 3$ and $\sigma_0 = 0.1\xi_0^2$. For greater polarizability, corresponding to $r_0 = 0.3, 0.4$, the plots are significantly distorted from semicircles, consistent with the behaviour of $\tau_{zM}(\omega)$ shown in figure 3 which is greatly enhanced at low frequencies.

The transverse susceptibility $\chi_\perp$ is shown in figures 8–10. Once again the pure dipole results for $\chi'_\perp$, $\chi''_\perp$ agree with the earlier after-effect function calculation [11]. However, the effect of polarizability is much less dramatic in a qualitative sense than in the longitudinal case but more important quantitatively. We observe from these plots that the magnitude of $\chi_\perp$ is

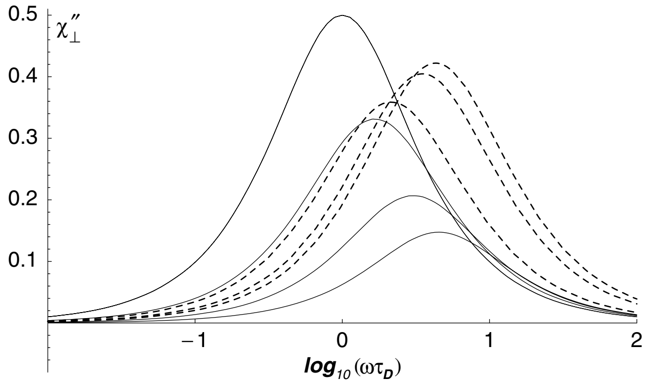


**Figure 9.** Plots of the dissipative part of the transverse susceptibility $\chi''_\perp$. The solid curves ordered from the top down (at the maximum point) correspond to pure dipoles with $\xi_0 = 0$ (the Debye susceptibility), $\xi_0 = 3, 6, 9$ respectively. The dashed curves represent polarizable particles with $\xi_0 = 3$ and polarizability ratios $r_0 = 0.1, 0.3, 0.4$ corresponding respectively to the dashed curves ordered from the bottom up (at the maximum point).

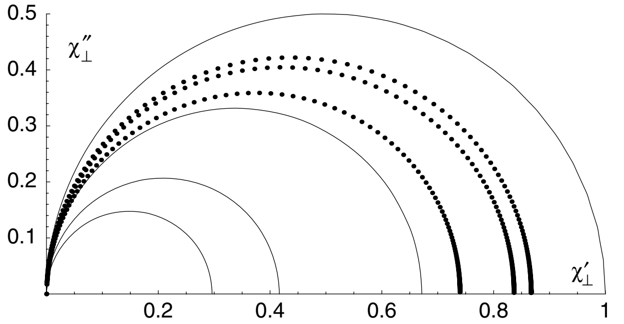


**Figure 10.** Cole–Cole plots of $\chi''_\perp$ against $\chi'_\perp$. The solid curves (from the outermost to innermost) correspond to pure dipoles with $\xi_0 = 0$ (the Debye susceptibility), $\xi_0 = 3, 6, 9$ respectively. The heavy dotted curves represent polarizable particles with $\xi_0 = 3$ and polarizability ratios $r_0 = 0.1$ (innermost curve) $r_0 = 0.3$ (middle curve) and $r_0 = 0.4$ (outermost curve).

greater than the magnitude of $\chi_\parallel$ for the same values of background field $\xi_0$ and polarizability $\sigma_0$. Moreover, we see in figures 8–10 that the effect of increasing polarizability is to enhance $\chi_\perp$ in magnitude rather than to qualitatively change its frequency dependence as happens with $\chi_\parallel$. The Cole–Cole plot in figure 10 shows too that a single-relaxation-time approximation will be reasonable even for polarizable particles with $r_0 = 0.4$ unlike in the longitudinal case. Again this conclusion is consistent with the behaviour of $\tau_{xM}(\omega)$ seen in figure 4 where there is little frequency dependence.

## 9. Discussion and conclusions

We have shown that the linear response of the polarization of a dilute suspension of dielectric particles to a weak AC probe in the presence of a strong DC background field can be directly calculated in the same manner as the relaxation functions for the system were calculated earlier [15–17]. The transient response and its mean relaxation time are given in the same way as the earlier relaxation functions in terms of a set of exponentially decaying modes, completely characterized by a set of decay rates and associated amplitudes. The steady-state response is described by a susceptibility which can also be expressed in terms of the decay rates and amplitudes of the transient. We have obtained simple explicit expressions for the transient, its mean relaxation time and the susceptibility.

For simple permanent dipoles we get the same results as earlier calculations based on the after-effect formalism [11]. However, we have also examined the effect of additional polarizability which has a marked but different impact upon the longitudinal and transverse response. The most striking feature in the longitudinal case arises from the slowest decay rate $\lambda_{01}$ which can get smaller as the polarizability $\sigma_0$ increases relative to the background field $\xi_0$. This leads to a departure of the susceptibility $\chi_{\parallel}$ from the simple single-relaxation-time Debye description. For pure dipoles both susceptibilities get smaller as the background field $\xi_0$ increases with the longitudinal susceptibility decreasing more rapidly than the transverse susceptibility. However, in the transverse case, the presence of polarizability enhances the susceptibility $\chi_{\perp}$ relative to the pure dipole case. If experimental measurements of both the transient and steady response are possible, our results provide a simple way to describe both sets of data with a common set of rates $\lambda_{mj}$ and amplitudes $d_{\ell mj}$.

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