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ORIGINAL PAPER

Nonlinear normal and anomalous response of non-interacting electric and magnetic dipoles subjected to strong AC and DC bias fields

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Abstract The perturbation theory approach via the Smoluchowski equation to the nonlinear dielectric relaxation of noninteracting permanent electric dipoles (Coffey and Paranjape, Proc R Ir Acad A 78:17, 1978) and the analogous Brownian magnetic relaxation of ferrofluids where Néel relaxation is ignored is revisited for the particular case of a strong dc bias field superimposed on a *strong* ac field. Unlike weak ac and strong bias dc fields, a frequency-dependent dc term now appears in the response as well as additional nonlinear terms at the fundamental and second harmonic frequencies. These may be experimentally observable particularly in the ferrofluid application. The corresponding results for the dc term for anomalous relaxation based on the fractional Smoluchowski equation are also given.

Keywords Nonlinear relaxation · Debye theory · Ferrofluids · Brownian motion · Anomalous relaxation

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1 Introduction

The Debye [1] theory of dielectric relaxation of non-interacting rigid electric dipoles under the *combined* influence of their rotational Brownian motion and a time-varying applied field predicting dispersion and absorption of microwave (GHz) radiation by polar fluids has been extended by Coffey and Paranjape [2] to include terms cubic in the applied field using perturbation theory. The small perturbation parameter is as usual the ratio of the interaction energy of a dipole with the applied field to the thermal energy kT (k is the Boltzmann constant and T is the absolute temperature). In particular, they considered the response to a strong alternating (ac) field alone and a weak field superimposed on a strong dc one. In the second of these cases, the ac field was supposed so weak that terms in its square and higher are negligible. The response exhibits typical nonlinear behavior in so far as it always depends on the *precise form* of the driving fields unlike the linear response.

Subsequently, the perturbation calculation was verified numerically for the strong ac field situation by Déjardin and Kalmykov [3] who also considered the *strong* ac and dc field case [4]. They achieved this by solving the differential-recurrence relation generated by the rotational Smoluchowski equation [5] governing the relaxation process using matrix continued fraction methods in the frequency domain. All the results are summarized in section 7.6 of [5]. Following the work of Coffey and Paranjape [2], Déjardin et al. [6,7] extended the perturbation calculation to include the *nonlinear* ac terms in the constant plus ac field case showing that the *combined* effect of the two strong fields is to give rise to additional dispersion and absorption phenomena which do not appear at all when only the linear term in the ac field is considered. These comprise a *time-independent* but *frequency-dependent* dc term in the response as well as a second harmonic contribution and one at the fundamental frequency which is cubic in the ac field. This term also does not appear if the nonlinear response due to a strong ac field alone is calculated. Despite these novel features in the combined field nonlinear response, experimental investigations of the nonlinear dielectric response seem to have been largely confined to that due to the strong ac field alone. For example, the results of Coffey and Paranjape [2] for the strong ac field have been favourably compared with nonlinear response measurements by De Smet et al. [8] and Jadżyn et al. [9,10].

Now, for *electric* dipoles which typically have a small dipole moment, it is often difficult to realize experimentally the strong nonlinear response conditions because of the consequent small value of the interaction energy between a dipole and the electric field. However, in a ferrofluid consisting of blocked single-domain ferromagnetic particles in a colloidal suspension, it is much easier to create the strong nonlinear regime because of the large magnetic moment, $10^4-10^5\ \mu_{\hat{B}}$, of such particles. This feature of a typical ferrofluid particle was recognized by Fannin et al. [11,12] who were able to detect nonlinear relaxation effects due to strong ac fields in the magnetic susceptibility of a ferrofluid. The terminology "blocked" refers to the fact that the solid state-like or Néel [5,13–15] magnetization relaxation mechanism over the internal anisotropy-Zeeman energy barriers inside the single-domain particle due to the shuttling action of the Brownian motion [13–15] is assumed to be inoperative. Finally, we should recall that the Debye theory is based on the extension of Einstein's theory [5] of the translational Brownian motion to orientational relaxation. Now that theory pertains to a relatively very large particle of size visible in a microscope (e.g., a pollen grain) immersed in a "sea" of very small particles. Therefore, one would expect that the ferrofluid situation, where the relaxation effects begins to appear at low MHz frequencies because of the great size of the particles, provides a much more suitable vehicle for the verification of the Debye theory than minute electric dipoles.

It is the purpose of this paper to revisit the perturbation calculation of the combined field situation with a view toward encouraging the experimental detection of the frequency-dependent dc term as well as the nonlinear effects due to the interaction of the two fields at the fundamental and second harmonic frequencies as well as the term with the fundamental frequency which also appears in the cubic response. Thus, we shall briefly re-derive for purposes of clarity the combined field response. In particular, we shall highlight the frequency dependence of the dc term and we shall also show how the calculation may be extended to anomalous relaxation governed by a fractional Fokker-Planck equation [5].

2 Differential-recurrence relation for the statistical moments

The basis of the Debye theory [1] of orientational relaxation of polar fluids is the rotational diffusion Smoluchowski equation for the evolution of the probability distribution function $W(\theta, t)$ in the configuration space of polar angles of an ensemble of rigid noninteracting electric dipoles of moment $\boldsymbol{\mu}$, undergoing rotational Brownian motion at absolute temperature T under the influence of an external time-varying electric field $\mathbf{E}(t)$ which reads

$$2\tau \frac{\partial W}{\partial t} = \frac{1}{\sin\theta} \frac{\partial}{\partial\theta} \left[\sin\theta \left(\frac{\partial W}{\partial\theta} - \frac{1}{kT} WM \right) \right], \qquad (1)$$

In Eq. (1), $W(\theta, t)\sin\theta \mathrm{d}\theta$ is the probability that at time t a dipole has an orientation lying between colatitudes θ and $\theta + \mathrm{d}\theta$ relative to the direction of $\mathbf{E}(t)$, $M(\theta, t)$ is the torque acting on the dipole due to $\mathbf{E}(t)$, $\tau = \zeta/(2kT)$ is the Debye relaxation time where $\zeta = 8\pi\eta a^3$ is the viscous drag coefficient of a dipole which is treated as a rigid sphere of radius a rotating in a fluid of viscosity η representing all the microscopic degrees of freedom of the surroundings, and $W(\theta, t)$ is the surface density of orientations of dipoles on the unit sphere. Here we consider a strong unidirectional field $\mathbf{E}_0$ superimposed on a *strong* alternating field $\mathbf{E}\cos\omega_0 t$, so that

$$\begin{aligned} M(\theta, t) &= -\mu E(t)\sin\theta = -\mu(E_0 + E\cos\omega t)\sin\theta \\ &= -\mu E_0 (1 + \lambda\cos\omega t)\sin\theta, \end{aligned} \qquad (2)$$

where $\lambda = E/E_0$. Furthermore, the torque M is axially symmetric so that $W(\theta, t)$ is independent of the azimuthal angle φ.

The general solution of Eq. (1) is of the form of the Fourier series

$$W(\theta, t) = \sum_{n=0}^{\infty} (n + 1/2)\, f_n(t)\, P_n(\cos\theta), \tag{3}$$

where $f_n(t) = \langle P_n(\cos\theta)\rangle(t)$ are the expectation values of the Legendre polynomials of order n (statistical moments), viz.,

$$f_n(t) = \langle P_n(\cos\theta)\rangle(t) = \frac{\int_{-1}^{1} W(\theta, t)\, P_n(\cos\theta)\, \mathrm{d}\cos\theta}{\int_{-1}^{1} W(\theta, t)\, \mathrm{d}\cos\theta}. \tag{4}$$

On substituting Eq. (3) into Eq. (1) and making use of the recurrence and orthogonality relations for the Legendre polynomials $P_n(z)$, we easily find that the $f_n(t)$ satisfy the differential-recurrence relation [5]

$$\frac{\mathrm{d}}{\mathrm{d}t} f_n(t) = -\frac{n(n+1)}{2\tau}\left\{ f_n(t) - \frac{\mu E(t)}{kT(2n+1)} \left[f_{n-1}(t) - f_{n+1}(t)\right]\right\} \tag{5}$$

which has stationary solution (i.e., pertaining to the forced response) for arbitrary $\mathbf{E}(t)$

$$f_n(t) = \frac{n(n+1)\xi_0}{2\tau(2n+1)} \int_{-\infty}^{t} \mathrm{e}^{-\frac{n(n+1)}{2\tau}(t-u)} e(u) \times \left[f_{n-1}(u) - f_{n+1}(u)\right] \mathrm{d}u, \tag{6}$$

where for $M(\theta, t)$ given by Eq. (2) we have $e(t) = 1 + \lambda\cos\omega t$ and $\xi_0 = \mu E_0/(kT)$. Equation (6) indicates that if we can calculate the Fourier coefficients $f_n(t)$ for given $\mathbf{E}(t)$ we have the time evolution of the observables. In particular, we shall be interested in averages of the Legendre polynomial of order 1 $f_1(t) = \langle P_1(\cos\theta)\rangle(t)$ pertaining to the dielectric response.

3 Successive approximation solution for $f_1(t)$

The rotational diffusion equation (1) and its alternative representation as a differential-recurrence relation equation (5) (which may also be derived from the appropriate Langevin equation [5]), although in itself a linear equation implicitly contains all the nonlinear behavior. The perturbation procedure to determine corrections to the linear response may be implemented as follows. We write out the integral equation (6) for the first few $f_n(t)$ and solve the resulting hierarchy of integral equations by the method of successive approximations assuming that $\xi_0 < 1$ in order to maintain convergence. The formal solutions for $\langle P_1(\cos\theta)\rangle(t)$ will be then rendered by the perturbation method.

We have in general for the dielectric response

$$\langle P_1(\cos\theta)\rangle(t) = \frac{\xi_0}{3\tau}\left\{ \int_{-\infty}^{t} \mathrm{e}^{-\frac{t-u}{\tau}} e(u)\, \mathrm{d}u - \frac{\xi_0^2}{5\tau^2} \int_{-\infty<u_2\le u_1\le u\le t} \mathrm{e}^{-\frac{t-u}{\tau}} \mathrm{e}^{-\frac{3(u-u_1)}{\tau}} \times \mathrm{e}^{-\frac{u_1-u_2}{\tau}} e(u_2)\, e(u_1)\, e(u)\, \mathrm{d}u_2 \mathrm{d}u_1 \mathrm{d}u + \cdots \right\}. \tag{7}$$

Notice that the leading term in Eq. (7) is simply the linear dielectric response. So far the procedure is entirely general. Next for the particular time variation given in Eq. (2), the solutions are best obtained using two-sided Fourier transforms. Consequently, we find after elementary but tedious manipulations that for the nonlinear dielectric response

$$\begin{aligned}
\langle P_1(\cos\theta)\rangle(t) &= \frac{\xi_0}{3}\left\{ 1 - \frac{\xi_0^2}{15} - \frac{\lambda^2\xi_0^2}{5\left(1+\omega^2\tau^2\right)}\left(\frac{1}{6} + \frac{3+\omega^2\tau^2}{9+\omega^2\tau^2}\right)\right. \\
&+ \frac{\lambda}{1+\omega^2\tau^2}\left[1 - \frac{\xi_0^2}{15}\frac{27+\omega^2\tau^2-2\omega^4\tau^4}{\left(1+\omega^2\tau^2\right)\left(9+\omega^2\tau^2\right)}\right]\cos\omega t \\
&+ \frac{\lambda\omega\tau}{1+\omega^2\tau^2}\left[1 - \frac{\xi_0^2}{15}\frac{42+19\omega^2\tau^2+\omega^4\tau^4}{\left(1+\omega^2\tau^2\right)\left(9+\omega^2\tau^2\right)}\right]\sin\omega t \\
&- \frac{\lambda^2\xi_0^2}{90\left(1+\omega^2\tau^2\right)} \\
&\times \left[\frac{\left(81-153\omega^2\tau^2-62\omega^4\tau^4-8\omega^6\tau^6\right)\cos 2\omega t}{\left(1+4\omega^2\tau^2\right)\left(9+\omega^2\tau^2\right)\left(1+(4/9)\omega^2\tau^2\right)}\right. \\
&+ \left.\frac{\omega\tau\left(252+88\omega^2\tau^2+16\omega^4\tau^4\right)\sin 2\omega t}{\left(1+4\omega^2\tau^2\right)\left(9+\omega^2\tau^2\right)\left(1+(4/9)\omega^2\tau^2\right)}\right] \\
&- \frac{\lambda^3\xi_0^2}{30\left(1+\omega^2\tau^2\right)}\left[\frac{\left(27-13\omega^2\tau^2\right)\cos\omega t}{18\left(1+\omega^2\tau^2\right)\left(1+(4/9)\omega^2\tau^2\right)}\right. \\
&+ \frac{\omega\tau\left(21+\omega^2\tau^2\right)\sin\omega t}{9\left(1+\omega^2\tau^2\right)\left(1+(4/9)\omega^2\tau^2\right)} \\
&+ \frac{\left(3-17\omega^2\tau^2\right)\cos 3\omega t}{6\left(1+9\omega^2\tau^2\right)\left(1+(4/9)\omega^2\tau^2\right)}
\end{aligned}$$

$$+\frac{\omega\tau\left(7-3\omega^2\tau^2\right)\sin 3\omega t}{3\left(1+9\omega^2\tau^2\right)\left(1+(4/9)\omega^2\tau^2\right)}\Bigg]\Bigg\}$$
$$+O\left(\xi_0^5\right). \tag{8}$$

This pertains to normal nonlinear dielectric relaxation of noninteracting rigid dipoles under the combined influence of strong constant and ac fields and with appropriate changes of notation also pertains to magnetic relaxation of a blocked ferrofluid. Its striking features over and above the single ac field case are the appearance of a frequency-dependent dc term $O(\lambda^2)$ accompanied by terms in the second harmonic of the applied field $O(\lambda^2)$ and a correction $O(\lambda^3)$ at the fundamental ac frequency, to the third harmonic term. In the weak ac and strong dc field case, all that appears is the correction $\lambda\xi_0^2$ at the fundamental frequency, to the linear response as well as the frequency-independent $\xi_0^2/15$ term due to the action of the strong dc field alone. The frequency-dependent but time-independent term [the first line of Eq. (8)] is also the result previously obtained by Déjardin et al. [7] confirming the present perturbation calculation. The appearance of the frequency-dependent dc and the other harmonic terms in Eq. (8), alluded to above suggests that experiments like those described in [11,12,16] should be made on ferrofluid systems with the objective of detecting these terms. The methods we have described may be extended to a mean-field potential whereupon the integral equation (6) above becomes vector-valued, details are available in [17–19].

Notice that in applying Eq. (8) to the magnetization of an assembly of noninteracting magnetic dipoles in superimposed ac and dc fields as in a ferrofluid it is customary [19] to write the applied field as $\mathbf{H}_0 + \mathbf{H}\cos\omega t$ and the resulting magnetization as

$$M_H(t) = \chi_0(\xi,\xi_0,\omega) + \sum_{k=1}^{\infty} \xi^k Re\left[\chi_k(\xi,\xi_0,\omega)e^{ik\omega t}\right] \tag{9}$$

where $\xi_0 = \mu H_0/(kT)$, $\xi = \mu H/(kT)$, the dc term is given by

$$\chi_0(\xi,\xi_0,\omega) = \chi_S H_0\left[1 - \frac{1}{15}\xi_0^2 - \frac{1}{60}\left(\frac{5}{1+\omega^2\tau_D^2} + \frac{1}{1+\omega^2\tau_D^2/9}\right)\xi^2 + \cdots\right], \tag{10}$$

$\chi_S = N_0\mu^2/(3kT)$ is the static susceptibility, $\boldsymbol{\mu}$ is the magnetic dipole moment of a ferrofluid particle, and N_0 is the number of particles per unit volume. Here, the Debye relaxation time is now denoted by τ_D in order to distinguish it from the exponentially long over barrier or Néel relaxation time. Equation (10) by inspection of Eq. (8) with suitable replacements is entirely equivalent to the dc term in that equation.

4 Generalization to anomalous relaxation

Now one of the most noteworthy features of the dielectric relaxation of disordered materials such as glass forming liquids and amorphous polymers is the failure of the Debye theory [1] of normal dielectric relaxation to adequately describe the low frequency spectrum of the linear susceptibility. The relaxation process in such disordered systems is characterized by the temporally nonlocal behavior arising from the energetic disorder which produces obstacles or traps simultaneously delaying the motion of the particle and producing memory effects. It appears that a significant amount of experimental data on anomalous relaxation of disordered systems and complex liquids supports the following empirical expressions for the complex dielectric susceptibility spectra, namely, the Cole–Cole equation [20]

$$\chi_{CC}(\omega) = \frac{\chi_S}{1+(i\omega\tau)^\sigma}, \tag{11}$$

the Cole–Davidson equation [21]

$$\chi_{CD}(\omega) = \frac{\chi_S}{(1+i\omega\tau)^\nu}, \tag{12}$$

and the Havriliak–Negami equation [22]

$$\chi_{HN}(\omega) = \frac{\chi_S}{[1+(i\omega\tau)^\sigma]^\nu}, \tag{13}$$

which is a combination of the Cole–Cole and Cole–Davidson equations. Here τ is a characteristic relaxation time usually known as the Debye relaxation time, χ_0 is the static susceptibility, and $\sigma(0 < \sigma \leq 1)$ and $\nu(0 < \nu \leq 1)$ are parameters with values usually obtained by fitting experimental data. In the context of the linear susceptibility, Eqs. (11)–(13), the Cole–Cole parameter σ is a *broadening* parameter because the dielectric loss spectrum broadens as σ is reduced, while the Cole–Davidson parameter ν in Eqs. (12) and (13) is a *skewing* parameter. Detailed discussions of anomalous relaxation behavior in complex disordered systems and various underlying microscopic models a reader can find, e.g., in [5,23–33]. Equations (11)–(13) are phenomenological generalizations of the Debye equation for the complex susceptibility, viz.,

$$\chi_{\mathrm{D}}(\omega) = \frac{\chi_S}{1 + i\omega\tau} \tag{14}$$

which may be derived using a variety of microscopic models of the relaxation process. For example, Debye [1] extended Einstein's treatment of the translational Brownian motion to the rotational Brownian motion of noninteracting permanent dipoles subjected to an external time-varying field. It might also happen that the motion which prevails is different for different kinds of dipoles. Moreover, both large and small jump transitions may exist simultaneously. The above observations lead us to the second microscopic (relaxator) model considered by Debye [1] (and much extended by Fröhlich [34]), which is a Poisson-like process, where relaxation occurs due to the crossing by large jumps of rare members of an assembly of dipoles over a potential barrier due to the shuttling action of thermal agitation. This model also produces a relaxation spectrum of the form of Eq. (14); however, the over barrier relaxation time has Arrhenius-like behavior as it depends exponentially on the height of the potential barrier.

The fractional kinetic equations incorporating the Cole–Cole, Cole–Davidson, and Havriliak–Negami relaxation processes can be written by extending a hypothesis of Nigmatullin and Ryabov [23]. They noted that for a system characterized by the single exponential relaxation function $f(t) = \mathrm{e}^{-t/\tau}$ and, hence, the Debye equation for the complex susceptibility, Eq. (14), the conventional kinetic equation describing the ac stationary response to a forcing function $F(t) = F\mathrm{e}^{\mathrm{i}\omega t} \sim E\mathrm{e}^{\mathrm{i}\omega t}$, namely,

$$\left(\tau \frac{\mathrm{d}}{\mathrm{d}t} + 1\right) f(t) = F(t) \tag{15}$$

may be generalized to a fractional kinetic equation of fractional order ν so describing a system with Cole–Davidson anomalous relaxation behavior as

$$\left(\tau \, {}_{-\infty}D_t^1 + 1\right)^\nu f(t) = F(t). \tag{16}$$

From now on operator equations of the type $(\tau \, {}_{-\infty}D_t^1 + 1)^\nu$ are to be understood as series of fractional operators via the binomial expansion

$$(a+b)^\nu = \sum_{n=0}^{\infty} \frac{(-1)^n(-\nu)_n}{n!} a^{\nu-n} b^n, \tag{17}$$

where $(a)_n = \Gamma(n+a)/\Gamma(a)$ is a Pochhammer symbol, the fractional derivative ${}_{-\infty}D_t^\alpha$ is given by the Riemann-Liouville definition [35–37]

$${}_{-\infty}D_t^\alpha [f(t)] = \frac{1}{\Gamma(1-\alpha)} \frac{\mathrm{d}}{\mathrm{d}t} \int_{-\infty}^{t} \frac{f(t')\mathrm{d}t'}{(t-t')^\alpha}, \tag{18}$$

and $\Gamma(z)$ is the gamma function and $0 < \alpha < 1$. Assuming adiabatic switching on of the ac field $F(t) = F\mathrm{e}^{\mathrm{i}\omega t}$, the solution of Eq. (16) yields the Cole–Davidson Eq. (12) [23]. According to Nigmatullin and Ryabov [23], the fractional exponent ν reflects the discontinuous character of the anomalous relaxation process and represents the fractal dimension of the set over which the relaxation times are statistically distributed. The Fröhlich relaxator model [34] as modified to the fractional diffusion by Nigmatullin and Ryabov may serve as an example of such a process. For the Cole–Cole relaxation, the underlying kinetic equation is given by [25]

$$\left(\tau^\sigma \, {}_{-\infty}D_t^\sigma + 1\right) f(t) = F(t). \tag{19}$$

The physical meaning of the parameter σ is the *fractal dimension* of the set of waiting times which is the scaling of the waiting time segments in the random walk with magnification. The fractional exponent σ measures the statistical self-similarity (or how the whole looks similar to its parts) of the waiting time segments [25]. In like manner, combining the fractional diffusion Eq. (19) describing Cole–Cole relaxation and Eq. (16) describing Cole–Davidson relaxation, one may also introduce the fractional kinetic equation [25]

$$\left(\tau^\sigma \, {}_{-\infty}D_t^\sigma + 1\right)^\nu f(t) = F(t). \tag{20}$$

Equation (20) represents a fractional generalization of the normal kinetic Eq. (15) to incorporate the Havriliak–Negami anomalous relaxation. For the two particular cases $\nu = 1, 0 < \sigma < 1$ and $\sigma = 1, 0 < \nu < 1$, Eq. (20) reduces to Eqs. (19) and (16), respectively. The fractional derivatives in Eqs. (16), (19), and (20) are memory functions with a slowly decaying power law kernel in the time. Such behavior arises from random torques with an anomalous waiting time distribution.

The nonlinear dielectric relaxation treated in Sect. 3 via the rotational diffusion model may be extended to anomalous relaxation by using the above fractional kinetic equation approach. Here we consider as a definite example only the Cole–Cole relaxation mechanism characterizing by the anomalous exponent σ (other relaxation mechanisms can be treated in like manner; see, e.g., [31]). The generalization of the theory based on a fractional version of the Smoluchowski Eq. (1), namely, [5,27,29]

$$2\tau^{\sigma}\ {}_{-\infty}D_t^{\sigma} W = \frac{1}{\sin\theta}\frac{\partial}{\partial\theta}\left[\sin\theta\left(\frac{\partial W}{\partial\theta} - \frac{1}{kT}WM\right)\right], \tag{21}$$

has been fully explained in [5,27,29]. Here the general solution of Eq. (21) is also of the form of the Fourier series, Eq. (3). Now, just as for the normal diffusion, we can obtain from Eq. (21) the fractional analogue of the recurrence Eq. (5) for the response functions $f_n(t) = \langle P_n(\cos\vartheta)\rangle(t)$ [31]

$$\left(\tau_n^{\sigma}\ {}_{-\infty}D_t^{\sigma} + 1\right) f_n(t) = \frac{\xi_0 + \xi\cos\omega t}{2n+1}\left[f_{n-1}(t) - f_{n+1}(t)\right], \tag{22}$$

where $\tau_n^{\sigma} = 2\tau^{\sigma}/n(n+1)$. Under linear response conditions, $\xi << 1$, and $\xi_0 = 0$, Eq. (22) yields the linear susceptibility from Eq. (11). Moreover, just as for the normal diffusion, Eq. (22) also allows one to evaluate the nonlinear ac stationary responses (see for details [31]). In particular, we have the generalization of Eq. (10), viz.,

$$\chi_0(\xi,\xi_0,\omega) = \chi_S H_0\left[1 - \frac{1}{15}\xi_0^2 - \frac{1}{60}Re\left(\frac{5}{1+(i\omega\tau_D)^{\sigma}} + \frac{1}{1+(i\omega\tau_D)^{\sigma}/3}\right)\xi^2 + \cdots\right]. \tag{23}$$

Such a generalization is likely to be important as the Cole–Cole relaxation behavior has proved useful in the analysis of magnetic and dielectric relaxation data.

5 Conclusion

In this paper, we have emphasized the rectifying effect of a strong bias field superimposed on a strong ac field on the electric polarization (or magnetization) of an assembly of noninteracting dipolar particles. Furthermore, we have suggested that experiments should be designed so as to detect the frequency-dependent dc nonlinear response introduced by the bias field. In this context, the appearance of individual nonlinear fundamental and third harmonic frequency components in Eq. (8) is also important because the latter frequency components may on occasion be easier to detect than the frequency-dependent dc one. Moreover, because they constitute part of the relaxation process they will also serve as experimental evidence of a frequency-dependent dc response. We have demonstrated how the anomalous nonlinear dielectric and magnetic relaxation can be treated by using fractional kinetic equations. The results obtained can explain the anomalous nonlinear relaxation of complex dipolar systems, where the relaxation process is characterized by a broad distribution of relaxation times. The advantage of having kinetic equations incorporating the anomalous relaxation then becomes apparent as it is now possible to study the effect of the nonlinear anomalous behavior on fundamental parameters associated with the fractional diffusion. We finally remark that the perturbation method of the calculation of nonlinear ac responses is quite general. For example, the method can also be applied to the calculation of the dynamic Kerr effect ac response of *polar* and *anisotropically polarizable* molecules as well as to nonlinear dielectric and Kerr effect relaxation of molecules under the influence of a mean-field potential.

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