THE EFFECTIVE EIGENVALUE METHOD AND ITS APPLICATION TO STOCHASTIC PROBLEMS IN CONJUNCTION WITH THE NONLINEAR LANGEVIN EQUATION

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I. INTRODUCTION

The concept of the effective eigenvalue appears to have been originally introduced into the study of relaxation problems in statistical physics by Leontovich.[1] It was later developed and applied (sometimes implicitly) to a variety of stochastic problems in laser physics,[2-4] magnetic domains,[5, 6] polar fluids,[7] polymers,[8] nematic liquid crystals,[9, 10] etc. In the present context, namely the theory of the Brownian motion, the method constitutes a truncation procedure which allows one using simple assumptions to obtain closed-form approximations to the solution of certain infinite hierarchies of differential-difference equations in the time variables. These

equations govern the time behavior of the statistical averages characterizing the relaxation of nonlinear stochastic systems. Thus, their solution is needed to calculate observable quantities such as the relaxation times and dynamic susceptibilities of the system.

The infinite hierarchies of differential-difference equations may arise in each of three entirely equivalent ways from the nonlinear Langevin equation (it should be understood before proceeding any further that in the present review we are concerned only with the response in the long-time or noninertial limit):

1. By averaging the noninertial Langevin equation regarded as a stochastic differential-difference equation of the Stratonovich type[11]
2. By averaging the inertial Langevin equation and proceeding to the noninertial limit[11-13]
3. By constructing from the noninertial Langevin equation the Fokker-Planck equation for the transition probability $W(\{\mathbf{x}\}, t)$ in configuration space $\{\mathbf{x}\} = \{x_1, x_2, \ldots, x_n\}$ and separating the variables by assuming a solution in the form of a sum of known functions of the configuration and unknown functions of the time.[3]

The Fokker-Planck equation approach is a very powerful method for the analysis of nonlinear stochastic systems.[3] However, this approach is extremely lengthy to use in practice, especially in the case of multidimensional systems, since it involves many mathematical manipulations. Hence, an alternative approach is desirable. One of the main purposes of this review is to show how the relaxation behavior may be obtained in a simple manner directly from the nonlinear Langevin equation of the process. The work of the previous authors is also expanded upon and treated in some detail to provide an introduction to the subject.

In general, the differential-difference equations take the form of three- (or higher-order) term recurrence relations[3] between the set of statistical averages describing the dynamical behavior. Thus, the behavior of any selected average is coupled to that of all the others so forming a hierarchy of averages. The time behavior of the first-order average, for example, involves that of the second-order average, which in turn involves the third-order average, and so on.

If the recurrence relation between the averages is a three-term one, the Laplace transform of the solution for a step stimulus may be expressed as an infinite continued fraction. The characteristic equation or secular determinant of the system (the continued fraction having been expressed as a rational function of the complex frequency) will in general possess an infinite number of roots. Thus, the system will have a denumerable set of

relaxation times and corresponding relaxation modes. In an actual numerical solution of the problem successive convergents of the continued fraction are calculated until convergence is reached when a solution is deemed to have been obtained.[3] If the underlying recurrence relation is not a three-term one, numerical solutions may be obtained (albeit not in as convenient a representation as the continued fraction) by writing the set of the differential-difference equations as a first-order matrix differential equation.[14] The size of the system matrix is then successively increased until convergence is attained.

The numerical approach to the problem has the disadvantage that closed form solutions are not available. Furthermore, the qualitative behavior is not at all obvious. Several investigators (see, e.g., Refs. 6, 15, and 16) have attempted to overcome this problem by recasting the solution of the Fokker-Planck equation as a Sturm-Liouville problem and evaluating the first few eigenvalues of that equation by various methods. It is then supposed that the longest relaxation time (corresponding to the reciprocal of the lowest eigenvalue) is the only one of significance, so that the system may be described by a single relaxation mode. This always involves the assumption that the contribution of the higher-order eigenfunctions to the relaxation process is negligible.

To pose the solution as a Sturm-Liouville problem the solution of the noninertial Fokker-Planck equation which may be written down symbolically as[3]

$$\frac{\partial}{\partial t} W(\{\mathbf{x}\}, t) = L_{\mathrm{FP}}(\{\mathbf{x}\}) W(\{\mathbf{x}\}, t) \tag{1}$$

is expressed as an infinite sum of products of unknown functions $\{y_k\}$ in the configuration space and exponential time decays $\{e^{-\lambda_k t}\}$, viz.,

$$W(\{\mathbf{x}\}, t) = \sum_k c_k(t) y_k(\{\mathbf{x}\})$$

where the functions $c_k(t)$ and $y_k(\{\boldsymbol{x}\})$ obey the equations

$$\frac{\mathrm{d}}{\mathrm{d}t} c_k(t) + \lambda_k c_k(t) = 0 \tag{2}$$

and

$$L_{\mathrm{FP}}(\{\mathbf{x}\}) y_k(\{\mathbf{x}\}) = \lambda_k y_k(\{\mathbf{x}\}) \tag{3}$$

The λ_k are the eigenvalues of the Sturm-Liouville equation for which solutions $\{\lambda_k\}$ exist.

Thus, the time behavior of the average $\langle A \rangle$ of any quantity $A(\{x\})$ of interest can be evaluated by means of the formula

$$\langle A \rangle = \int A(\{\mathbf{x}\}) W(\{\mathbf{x}\}, t)\, \mathrm{d}\{\mathbf{x}\} \tag{4}$$

Because of Eq. (1), the average $\langle A \rangle$ from Eq. (4) has the form

$$\langle A \rangle = \sum_k a_k \mathrm{e}^{-\lambda_k t} \tag{5}$$

where the a_k are the weight coefficients (amplitudes) corresponding to the eigenvalues λ_k. The reformulation of the task of calculating the relaxation behavior as a Sturm-Liouville problem allows us to give the formal definition of the effective eigenvalue.

To achieve this let us now suppose that the time behavior of $\langle A \rangle$ may be approximated by

$$\langle A(t) \rangle = \langle A(0) \rangle \mathrm{e}^{-\lambda_{\mathrm{eff}} t}$$

whence according to Eq. (5)

$$\lambda_{\mathrm{eff}} = -\frac{\langle \dot{A}(0) \rangle}{\langle A(0) \rangle} = \frac{\Sigma_k a_k \lambda_k}{\Sigma_k a_k} \tag{6}$$

and τ_{eff}, the effective relaxation time, is then

$$\tau_{\mathrm{eff}} = \lambda_{\mathrm{eff}}^{-1} \tag{7}$$

It is difficult to evaluate λ_{eff} from this formula using the Sturm-Liouville equation because a knowledge of the law of formation of the eigenvalues λ_k and their corresponding amplitudes a_k is rarely available. The approach taken in this review does not attempt to calculate λ_{eff} by explicitly calculating the eigenvalue spectrum as required by Eq. (5); rather it gives λ_{eff} in terms of equilibrium averages of the distribution function.

The concept of the effective eigenvalue so far does not particularly rely on the linearity of the response. In many cases the linear response may effectively be described by a single relaxation time. This is not true for processes such as the Kerr effect relaxation of an assembly of dipolar molecules under the influence of a step stimulus. Here two effective

eigenvalues are needed[14] to describe the response because it is intrinsically nonlinear. Another example where two effective eigenvalues are needed is ferromagnetic resonance[6, 11] of assemblies of single-domain ferromagnetic particles where "entanglement" of the dipole and quadrupole moments is required to produce the resonance even in the linear response approximation. If the response is linear it is possible to considerably simplify the expression for effective relaxation times using linear response theory, as we now demonstrate.

According to linear response theory[3, 10] (where we suppose for the purpose of illustration that $\mathbf{m}(t)$ represents the instantaneous electric dipole moment of a system) the autocorrelation function of the dipole moment is given by[10, 17]

$$C_m(t) = \frac{\langle \mathbf{m}(0)\mathbf{m}(t)\rangle_0 - \langle \mathbf{m}(0)\rangle_0^2}{\langle \mathbf{m}^2(0)\rangle_0 - \langle \mathbf{m}(0)\rangle_0^2} \tag{8}$$

where the subscript zero on the angular braces denotes that the ensemble averages are to be evaluated in the absence of the external stimulus which yields the relaxation behavior.

The autocorrelation function $C_m(t)$ has a clear physical meaning. It describes relaxation of the average dipole moment $\langle \mathbf{m}\rangle$ (having switched off at $t = 0$ a small constant external electric field $\mathbf{E}$, which has been applied in the infinite past). We have

$$\langle \mathbf{m} - \mathbf{m}_0\rangle = \mathbf{E}\left(\langle \mathbf{m}^2(0)\rangle_0 - \langle \mathbf{m}(0)\rangle_0^2\right) C_m(t)$$

where $\mathbf{m}_0$ is the equilibrium value of $\langle \mathbf{m}\rangle$.
The effective eigenvalue method implies that $C_m(t)$ may be represented as a pure exponential decay. We then have with the aid of Eqs. (7) and (8)

$$\tau_{\text{eff}} = -\frac{\langle \mathbf{m}^2(0)\rangle_0 - \langle \mathbf{m}(0)\rangle_0^2}{\langle \mathbf{m}(0)\dot{\mathbf{m}}(0)\rangle} \tag{9}$$

The significance of the foregoing is that we can evaluate λ_{eff} from the underlying Langevin equation for $\mathbf{m}(t)$ without solving it since we require only the value of $\dot{\mathbf{m}}(t)$ at time $t = 0$. Thus, all the desired properties of the

system in the linear response approximation can be evaluated. For example, the complex susceptibility $\chi(\omega) = \chi'(\omega) - i\chi''(\omega)$ obeys the relation[10]

$$\chi(\omega) = \chi'(0)\left[1 - i\omega\int_0^\infty C(t)e^{-i\omega t}\right] \tag{10}$$

with $C(t)$ defined as in Eq. (8) and $\chi'(0) = \langle \mathbf{m}^2(0)\rangle_0 - \langle \mathbf{m}(0)\rangle_0^2$. If the process can be described by a single effective relaxation time then the effective eigenvalue method immediately leads to the Debye equation

$$\chi(\omega) = \frac{\chi'(0)}{1 + i\omega\tau_{\text{eff}}} \tag{11}$$

We note (for convenience we refer to Eqs. (5) and (6)) that a global characterization of the decay of the electric polarization is given by the relaxation time T defined as

$$T = \int_0^\infty C(t)\,dt = \frac{\Sigma_k a_k \lambda_k^{-1}}{\Sigma_k a_k} \tag{12}$$

namely the area under the autocorrelation function curve. This correlation time also includes contributions from all the eigenvalues but it gives no information on the different time regions of relaxation that are possible; the behavior of T and τ_{eff} is sometimes similar. If different time scales are involved, however, τ_{eff} differs from T and in view of the definition

$$\tau_{\text{eff}} = \frac{\Sigma_k a_k}{\Sigma_k a_k \lambda_k} \tag{13}$$

accurately represents the initial slope of the decay of the polarization, thereby giving precise information on the initial decay of the polarization in the noninertial limit.

We have also mentioned that it is often assumed that the longest relaxation time of the distribution function $W(\{\mathbf{x}\}, t)$ accurately represents that of the polarization decay. It is apparent that this is true only if a single eigenvalue determines the decay process (see Section II.J).

We began by supposing that the decay process in the time domain was a pure exponential, thus allowing us to derive Eq. (9) for the effective eigenvalue. Another entirely equivalent way of describing the concept was introduced by Morita[7] in the context of the hierarchy of differential-difference equations. We suppose that in the continued fraction (or failing

that, in the recurrence relation) describing the Laplace transform of the infinite hierarchy, the ratio of the Laplace transforms of the desired and higher-order averages may be replaced (in the limit of low frequencies) by their final equilibrium values. This corresponds to the final value theorem for the Laplace transform. Such a procedure serves as a well-defined method of closing the hierarchy, which simultaneously reduces the order of the characteristic equation. Furthermore, in the linear response approximation this procedure usually reduces the infinite order characteristic equation to one of the first order and closes the hierarchy at the first equation.[7, 11] (An exception to the rule being the resonance phenomena considered in Sections II.H and IV.)

In this chapter, we adopt a synergetic approach in the spirit of Haken's book,[18] and apply the concept of the effective eigenvalue in conjunction with the nonlinear Langevin equation to discuss the following stochastic problems:

1. The relaxation of the longitudinal and transverse components of the electric polarization for a nematic liquid crystal in the linear response approximation. We show in Section II and Appendix A how the hierarchy of differential-difference equations can be obtained directly from the noninertial vectorial Langevin equation for the rotational Brownian motion of a molecule with dipole moment $\boldsymbol{\mu}$ in the mean nematic field $\mathbf{E}_0(\{\boldsymbol{\mu}\})$

$$\zeta \frac{\mathrm{d}}{\mathrm{d}t}\boldsymbol{\mu}(t) = \boldsymbol{\lambda}(t) \times \boldsymbol{\mu}(t) + \left[\boldsymbol{\mu}(t) \times \mathbf{E}_0(\{\boldsymbol{\mu}(t)\})\right] \times \boldsymbol{\mu}(t) \quad (14)$$

where ζ is the friction coefficient and $\boldsymbol{\lambda}(t)$ is the white noise driving torque. Equation (14) is derived in Section II from the Euler-Langevin equation[17] for the angular velocity $\boldsymbol{\Omega}$ and the kinematic relation $\dot{\boldsymbol{\mu}} = [\boldsymbol{\Omega} \times \boldsymbol{\mu}]$. This allows us to eliminate $\boldsymbol{\Omega}$ and so to simplify the problem. First we consider the problem of dielectric relaxation in the Meier-Saupe nematic potential $U(\vartheta) = -U_0 \cos^2\vartheta$. We then use the effective eigenvalue method to obtain the complex dielectric susceptibility tensor for an arbitrary uniaxial potential of the crystalline anisotropy.

2. The Kerr effect relaxation of an assembly of noninteracting rigid dipolar molecules under the influence of a suddenly applied dc field $\mathbf{E}$. In this case two effective eigenvalues are needed to describe the response.[14] We show how the Kerr effect response can be obtained

by direct averaging of the vectorial Langevin equation for the rotational Brownian motion of a linear molecule in the dc field **E**, which is Eq. (14) with $\mathbf{E}_0 = \mathbf{E} = \text{const.}$

3. The relaxation of the magnetization and ferromagnetic resonance of an assembly of noninteracting single-domain ferromagnetic particles. This problem is distinguished from the liquid crystal problem by the fact that the underlying Langevin equation is the Landau-Lifshitz-Gilbert equation[11, 15] for the magnetization $\mathbf{M}(t)$ augmented by a random field term $\mathbf{h}(t)$, namely

$$\begin{aligned}\frac{\mathrm{d}}{\mathrm{d}t}\mathbf{M}(t) = {} & g'M_s\mathbf{M}(t) \times [\mathbf{H}_{ef}(\{\mathbf{M}(t)\}) + \mathbf{h}(t)] \\ & + h'[\mathbf{M}(t) \times [\mathbf{H}_{ef}(\{\mathbf{M}(t)\}) + \mathbf{h}(t)]] \times \mathbf{M}(t)\end{aligned} \tag{15}$$

where g' and h' are constants, $M_s = |\mathbf{M}(t)|$, $\mathbf{H}_{ef}$ is the effective field of the crystalline anisotropy. Our aim is to calculate the complex magnetic susceptibility tensor of the particle. Once again two effective eigenvalues are required to describe ferromagnetic resonance because "entanglement" of the dipole and quadrupole responses to the transversally applied ac magnetic field $\mathbf{H}(t)$ is needed to produce the resonance.[6]

Although describing diverse physical phenomena, the three problems are mathematically united by the fact that they all involve the rotational Brownian motion in three-dimensional uniaxial potentials. They are discussed in Section II. Similar problems (namely dielectric relaxation of a polar fluid in a constant electric field and magnetic relaxation of a ferrofluid in a constant magnetic field) have been already solved by this method in Refs. 5 and 24; we described these in Ref. 11. As an example of the application of the method to two-dimensional rotational Brownian motion in an N-fold cosine potential, the calculations of the longitudinal and transverse relaxation times are given in Section III.

The second major class of problem, which we consider in Section IV, is the two-dimensional Brownian motion in a tilted cosine potential:

$$U(\phi) = -(a\phi + b\cos\phi) \tag{16}$$

Applications of this model are listed in Refs. 3 and 22. Two further problems come under this heading:

4. The Josephson tunneling junction, which is made up of two superconductors separated by a thin layer of oxide.[23] We label[23] ψ_R and

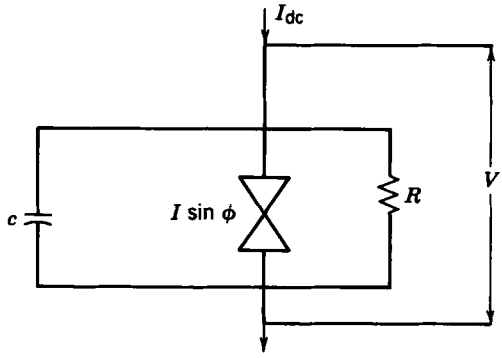


Figure 1. Equivalent circuit of Josephson junction.

ψ_L pair wave functions for the right and left superconductor, respectively. The phase difference $\phi = \phi_L - \phi_R$ between these wave functions (of the Cooper pairs) is given by the Josephson equation[22, 23]

$$\dot{\phi}(t) = \frac{2eV(t)}{\hbar} \tag{17}$$

where $V(t)$ is the potential difference across the junction, e is the charge on the electron, and $\hbar = h/2\pi$, where h is is the Planck constant. The junction is now modeled (Fig. 1)[3, 23] by a resistance R in parallel with a capacitance C, across which is connected a dc current generator I_{dc} (representing the bias current applied to the junction). At the other end of the junction (across the resistance R) is connected a phase-dependent current generator, $I \sin \phi$, representing the Josephson supercurrent due to the Cooper pairs tunneling through the junction. Since the junction operates at a temperature above absolute zero, there will be a noise current $\mathrm{i}(t)$ superimposed on the bias current. The circuit thus behaves according to the Langevin equation[3]

$$I_{dc} + \mathrm{i}(t) = C\frac{dV(t)}{dt} + \frac{1}{R}V(t) + I \sin \phi(t) \tag{18}$$

Using the Josephson equation (17), Eq. (18) may be cast into the form[3, 22]

$$\left(\frac{\hbar}{2e}\right)^2 C\ddot{\phi}(t) + \left(\frac{\hbar}{2e}\right)^2 \frac{1}{R}\dot{\phi}(t) + \frac{\hbar}{2e} I \sin \phi(t) = \frac{\hbar}{2e}[I_{dc} + \mathrm{i}(t)] \tag{19}$$

Equation (19) has the same form as that for a Brownian particle moving under the influence of both a cosine potential and a linear potential. The behavior of the system will be determined by the relative amplitudes of the constant and periodic terms. A more complete discussion of Eq. (19) is given in Refs. 3 and 23, and in the papers quoted therein. The quantities of physical interest are the mean value of the voltage $\langle V \rangle = (\hbar/2e)\langle \dot{\phi} \rangle$ and/or junction impedance to external high-frequency current. We present exact and approximate calculations of the junction impedance to an external small-signal current using Eq. (19) in the noninertial limit when we can neglect the capacitative term $C\ddot{\phi}$.

5. The use of the theory for the description of quantum noise in ring laser gyros arises because the phase difference $\phi(t)$ between the clockwise and counterclockwise running waves in such a gyro operating in the steady state is governed by the Langevin equation[19, 20]

$$\dot{\phi}(t) = a + b \sin \phi(t) + F(t) \tag{20}$$

where

$$a = -\frac{8\pi}{\lambda L}(\mathbf{A} \cdot \Omega)$$

$|\mathbf{A}|$ is the area covered by the optical path of length L along the ring cavity, Ω is the rotation rate of the gyro, λ is the wavelength of the laser, b is the backscattering coefficient, $F(t)$ is a white-noise source term. The quantity of interest in this application is the beat signal $\cos \phi(t)$, in particular, its spectrum $\alpha(\omega)$, defined as[19, 20]

$$\alpha(\omega) = \int_{-\infty}^{\infty} e^{i\omega\tau} \langle \cos \phi(0) \cos \phi(\tau) \rangle \, d\tau \tag{21}$$

The beat signal spectrum $\alpha(\omega)$ for the ring laser obtained using the numerical exact solution of Eq. (20) given in Ref. 20 is also compared with the result yielded by the effective eigenvalue method.

We note that when the tilt is equal to zero (the parameter a of Eq. (17)) the model may be applied (with appropriate changes of notation) to the theory of dielectric relaxation of an assembly of noninteracting two-dimensional dipoles, discussed briefly in Section III.

Problems 4 and 5 are of the upmost importance in the context of this review, since they can be solved exactly. A comparison of the exact solutions and those yielded by the effective eigenvalue method allows us to

comment on the accuracy of the latter. Using the foregoing problems as an example, we shall demonstrate that the effective eigenvalue method in certain circumstances yields a simple and extremely concise analytical description of the solution of complex stochastic problems. Our method of obtaining the exact solution of these problems has the merit of being considerably simpler than the previously available algorithms described in Refs. 3, 20, and 53.

Thus, the purpose of the review is twofold:

1. To demonstrate (using as examples various well-known models) how the quantities characterizing the dynamical behavior of stochastic systems in the time and/or frequency domains may be calculated by direct averaging of nonlinear Langevin equations. The problem of constructing and solving the underlying Fokker-Planck equations is thus bypassed entirely.
2. To show that the effective eigenvalue method is a valuable and powerful tool for the purpose of obtaining simple analytic solutions to nonlinear stochastic problems in certain ranges of the parameter values and for certain potentials.

II. THREE-DIMENSIONAL ROTATIONAL BROWNIAN MOTION IN AXIALLY SYMMETRIC POTENTIALS

A. The Noninertial Langevin Equation for a Nematic Liquid Crystal in the Meier-Saupe Mean Field Approximation

As our first example of Brownian motion in axially symmetric potentials we reconsider the theory of dielectric relaxation of nematic liquid crystals due to Martin et al.[16] This proceeds from the Fokker-Planck equation without explicit reference to the underlying Langevin equation. The aim is to extend the Debye theory of dielectric relaxation of assemblies of noninteracting polar molecules subjected to a weak alternating (ac) field to include the effects of a strong intermolecular potential giving rise to the nematic state.

The essence of the diffusion equation method[3] is to write down the Fokker-Planck equation (Smoluchowski equation), for the transition probability of orientations of dipoles in configuration space. This is solved[16] by the method of separation of the variables. The separation procedure yields a Sturm-Liouville equation in the space variables. The reciprocal of the lowest eigenvalue of this equation yields the longest relaxation time of the probability distribution of orientations. Furthermore, on expanding the dipole moment as a series of eigenfunctions of the Sturm-Liouville

equation and averaging over the distribution function, the orientational polarization may be expressed as an infinite set of discrete Debye type relaxation mechanisms. The relaxation times and amplitudes are then determined by the eigenvalues of the Sturm-Liouville equation. Approximate analytic solutions for the lowest eigenvalue of the distribution function may be found for high and low nematic potential barriers.[16] We term the Sturm-Liouville method method I. This is commonly used in the study of nematic liquid crystals.[16]

An alternative approach to the problem is to expand the transition probability as a series of spherical harmonics. This yields the time behavior of the transition probability as an infinite hierarchy of differential-difference equations. The lowest-order member of the hierarchy governs the time dependence of the polarization but is coupled to all the higher members by the differential-difference scheme, thus giving an infinite number of relaxation modes. In the frequency domain the hierarchy may often be written as an infinite continued fraction which facilitates its solution. The most general method of solution is effected by converting the hierarchy into the set of ordinary differential equations (see Section II.J)

$$\dot{\mathbf{X}} = \mathbf{AX} + \mathbf{B}U$$

and successively increasing the size of the system matrix **A** (Ref. 3) by means of the recurrence relations of the hierarchy until convergence is attained. The reciprocal of the lowest eigenvalue of this set yields the longest relaxation time of the system. We term this method II.

Method I, based on the Sturm-Liouville equation, has also been used in the study of the analogous relaxation process in ferromagnetic domains in conjunction with various asymptotic methods[5, 6, 15] to obtain analytic solutions for the lowest eigenvalue of the distribution function for high and low anisotropy. The contribution of higher-order eigenvalues to the polarization is neglected so that the polarization can be described by a single eigenvalue. It is then assumed that the reciprocal of the lowest eigenvalue may be identified with the relaxation time characterizing a flip of the magnetization.[6] Both methods I and II have been used in Ref. 11 in conjunction with linear response theory[10] in the study of the dispersion of the magnetic susceptibility of fine ferromagnetic particles.

The disadvantage of the diffusion equation method is that in all applications we first have to derive that equation from the Chapman-Kolmogorov equation in curvilinear coordinates and the underlying Langevin equation. Next we must either use elaborate mathematical formulae involving spherical harmonics to deduce the set of differential-

difference equations or we must study the properties of Sturm-Liouville equations, the solution of which cannot generally be given in terms of known functions. In neither method is it easy to generalize the results to an arbitrary nematic potential.

It is the purpose of this section to show how the differential-difference equations (Method II) for a nematic liquid crystal for the Meier-Saupe potential arise naturally from the noninertial Langevin equation written in the vector form and defined as a Stratonovich equation, thus bypassing the diffusion equation entirely. Having derived the differential-difference equations by averaging the Langevin equation we then show how closed form expressions of the Debye type may be obtained for the complex susceptibility. The results are given for an ac field applied parallel and perpendicular to the axis of symmetry. We then show how the results may be generalized to an arbitrary uniaxial nematic potential. The availability of these expressions rests on the assumption that the contribution to the dynamical behavior of all processes that occur on a timescale $< \frac{1}{3}\tau_D$, where τ_D is the Debye relaxation time, may be adequately approximated by their equilibrium values. This assumption may be stated more precisely, as that in the Laplace transform of the differential-difference equation describing the behavior of the mean dipole moment (here the first spherical harmonic average) the ratio of the Laplace transforms of the averages of the first- and higher-order spherical harmonics may be replaced by their final equilibrium values. This reduces the nth-order characteristic equation of the system to one of the first order. The growth and decay of the mean dipole moment is thus characterized by a single exponential with an relaxation time (the reciprocal of the effective eigenvalue λ_{eff}) which is a function of the nematic potential. A numerical analysis of our results and a comparison with the exact solution is given in Section II.J, where it is shown that the present approach yields an excellent description of the transverse relaxation and accurately describes the longitudinal relaxation for low barrier heights only.

We commence our study by considering the rotational Brownian movement of a linear molecule subject to the mean nematic field $\mathbf{E}_0$ and an external electric field $\mathbf{E}_1(t)$. The total electric field $\mathbf{E}(t)$ acting on the molecule is

$$\mathbf{E}(t) = \mathbf{E}_0 + \mathbf{E}_1(t) \tag{22}$$

The molecule contains a rigid electric dipole $\boldsymbol{\mu}$ along the axis of symmetry. The angular velocity $\boldsymbol{\omega}(t)$ of the molecule satisfies the kinematic relation[24, 32]

$$\frac{d\boldsymbol{\mu}(t)}{dt} = \boldsymbol{\omega}(t) \times \boldsymbol{\mu}(t) \tag{23}$$

We specialize Eq. (23) to the rotational Brownian motion of a molecule by supposing that $\boldsymbol{\omega}$ obeys the Euler-Langevin equation[17]:

$$I\frac{d\boldsymbol{\omega}(t)}{dt} + \zeta\boldsymbol{\omega}(t) = \boldsymbol{\lambda}(t) + \boldsymbol{\mu}(t) \times \mathbf{E}(t) \tag{24}$$

where I is the moment of inertia of the molecule about any line through the origin perpendicular to the line of symmetry, $\zeta\boldsymbol{\omega}$ is the damping torque, and $\boldsymbol{\lambda}(t)$ is the white-noise driving torque, both due to the Brownian movement. $\boldsymbol{\lambda}(t)$ satisfies

$$\overline{\lambda_i(t)} = 0 \tag{25}$$

$$\overline{\lambda_i(t)\lambda_j(t')} = 2kT\zeta\delta_{ij}\delta(t - t') \tag{26}$$

where δ_{ij} is Kronecker's delta, $i, j = 1, 2, 3$, which correspond to Cartesian axes x, y, z fixed in the molecule. This is the assumption that the random torques about different axes are statistically independent. $\delta(t)$ is the Dirac delta function. The term $\boldsymbol{\mu} \times \mathbf{E}(t)$ in Eq. (24), is the torque due to the nematic mean field $\mathbf{E}_0$ and external field $\mathbf{E}_1(t)$. The overbar indicates a statistical average.

Equation (24) includes the inertia of the molecule. The noninertial response occurs when I tends to zero or when ζ, the friction coefficient, becomes very large. In this limit the angular velocity vector is

$$\boldsymbol{\omega}(t) = \frac{\boldsymbol{\lambda}(t)}{\zeta} + \frac{\boldsymbol{\mu} \times \mathbf{E}(t)}{\zeta} \tag{27}$$

We combine this with the kinematic relation (23), yielding

$$\frac{d\boldsymbol{\mu}(t)}{dt} = \frac{\boldsymbol{\lambda}(t)}{\zeta} \times \boldsymbol{\mu}(t) + \frac{\{\boldsymbol{\mu}(t) \times \mathbf{E}(t)\} \times \boldsymbol{\mu}(t)}{\zeta} \tag{28}$$

or

$$\frac{d\boldsymbol{\mu}(t)}{dt} = \frac{\boldsymbol{\lambda}(t)}{\zeta} \times \boldsymbol{\mu}(t) + \frac{\mu^2\mathbf{E}(t)}{\zeta} - \frac{\boldsymbol{\mu}(t)\{\boldsymbol{\mu}(t) \cdot \mathbf{E}(t)\}}{\zeta} \tag{29}$$

which is the Langevin equation for the rotational motion of the dipole moment $\boldsymbol{\mu}$ of the molecule in the noninertial limit.

B. Equation of Motion for a Step Change in Field Applied Along the Polar Axis

To achieve the simplest possible presentation of the problem let us first suppose that the molecule is subject to a mean nematic field $\mathbf{E}_0 = -\text{grad}\, V$, where $V(\vartheta) = -V_0 \cos^2\vartheta$ in spherical polar coordinates, $V_0 = 3AS/2V_m^2$, where S is the order parameter, V_m is the molar volume, and A is a constant of interactions.[16] Hence,

$$\mathbf{E}_0 = \mathbf{k}\left(-\frac{\partial V}{\mu \partial \cos\vartheta}\right) = \mathbf{k}\frac{2V_0}{\mu}\cos\vartheta(t) = \mathbf{k}E_0\cos\vartheta(t) \tag{30}$$

where $E_0 = 2V_0/\mu$.

Let us suppose that at $t = 0$ a small field $\mathbf{E}_1U(t)$, where $U(t)$ is the unit step function, is applied along the z axis, so that

$$\mathbf{E}(t) = \mathbf{E}_0 + \mathbf{E}_1U(t)\mathbf{k} \tag{31}$$

$$= \frac{E_0}{\mu}\mu_z(t)\mathbf{k} + E_1U(t)\mathbf{k} \tag{32}$$

Equation (29) then becomes, with the aid of Eq. (30),

$$\dot{\mu}_x(t) = \frac{1}{\zeta}\left(\lambda_y(t)\mu_z(t) - \lambda_z(t)\mu_y(t)\right) - \frac{\mu_x(t)\mu_z(t)}{\zeta}\left[\frac{E_0\mu_z(t)}{\mu} + E_1U(t)\right] \tag{33}$$

$$\dot{\mu}_y(t) = \frac{1}{\zeta}\left(\lambda_z(t)\mu_x(t) - \lambda_x(t)\mu_z(t)\right) - \frac{\mu_y(t)\mu_z(t)}{\zeta}\left[\frac{E_0\mu_z(t)}{\mu} + E_1U(t)\right] \tag{34}$$

$$\dot{\mu}_z(t) = \frac{1}{\zeta}\left(\lambda_x(t)\mu_y(t) - \lambda_y(t)\mu_x(t)\right) + \frac{\left(\mu^2 - \mu_z^2(t)\right)}{\zeta}\left[\frac{E_0\mu_z(t)}{\mu} + E_1U(t)\right] \tag{35}$$

The desired quantity is the average behavior of the component of the dipole moment in the field direction, that is, the longitudinal component.

Equation (35) contains multiplicative noise terms: $\lambda_x(t)\mu_y(t)$ and $\lambda_y(t)\mu_x(t)$. Taking the Langevin equation for N stochastic variables $\{\xi(t)\} = \{\xi_1(t), \xi_2(t), \xi_3(t), \ldots, \xi_N(t)\}$ as

$$\frac{\mathrm{d}}{\mathrm{d}t}\xi_i(t) = h_i(\{\xi(t)\}, t) + g_{ij}(\{\xi(t)\}, t)\Gamma_j(t) \tag{36}$$

with

$$\bar{\Gamma}_j(t) = 0 \tag{37}$$

$$\overline{\Gamma_i(t)\Gamma_j(t')} = 2\delta_{ij}\delta(t - t') \tag{38}$$

and interpreting it as a Stratonovich[3] equation, Risken[3] has shown that the drift coefficient is

$$\begin{aligned} D_i(\{x\}, t) = \dot{x}_i &= \lim_{\tau \to 0} \frac{1}{\tau}\overline{(\xi_i(t + \tau) - x_i)}\Big|_{\xi_k(t) = x_k} \\ &= h_i(\{x\}, t) + g_{kj}(\{x\}, t)\frac{\partial}{\partial x_k} g_{ij}(\{x\}, t) \\ k &= 1, 2, \ldots, N \end{aligned} \tag{39}$$

The last term in Eq. (39) is called the noise-induced or spurious drift.[11] $\xi_i(t + \tau)$, $\tau > 0$, is a solution of Eq. (36), which has the sharp value $\xi_k(t) = x_k$ for $k = 1, 2, \ldots, N$. The quantities x_k in Eq. (39) are themselves random variables with the probability density function $W(\{x\}, t)$ defined such that $W\,\mathrm{d}x_k$ is the probability of finding x_k in the range x_k to $x_k + \mathrm{d}x_k$. Instead of using different symbols we have distinguished the starting values at time t from the stochastic variables by deleting the time argument as in Ref. 11.

We now use this theorem to evaluate the average of the multiplicative noise terms in Eq. (35). We have nine tensor components:

$$\begin{aligned} g_{11} &= 0 & g_{12} &= \frac{\mu_z}{\zeta} & g_{13} &= \frac{-\mu_y}{\zeta} \\ g_{21} &= \frac{-\mu_z}{\zeta} & g_{22} &= 0 & g_{23} &= \frac{\mu_x}{\zeta} \\ g_{31} &= \frac{\mu_y}{\zeta} & g_{32} &= \frac{-\mu_x}{\zeta} & g_{33} &= 0 \end{aligned} \tag{40}$$

whence with the aid of Eq. (39)

$$
\begin{aligned}
&kT\zeta g_{xj}(\{\boldsymbol{\mu}\},t)\frac{\partial}{\partial x_x}g_{3j}(\{\boldsymbol{\mu}\},t)\\
&\quad= kT\zeta\Bigg[g_{11}\frac{\partial}{\partial x_1}g_{31}+g_{12}\frac{\partial}{\partial x_1}g_{32}+g_{13}\frac{\partial}{\partial x_1}g_{33}\\
&\qquad+g_{21}\frac{\partial}{\partial x_2}g_{31}+g_{22}\frac{\partial}{\partial x_2}g_{32}+g_{23}\frac{\partial}{\partial x_2}g_{33}\\
&\qquad+g_{31}\frac{\partial}{\partial x_3}g_{31}+g_{32}\frac{\partial}{\partial x_3}g_{32}+g_{33}\frac{\partial}{\partial x_3}g_{33}\Bigg] \qquad (41)\\
&\quad= kT\zeta\Bigg[\frac{\mu_z}{\zeta}\frac{\partial}{\partial\mu_x}\left(\frac{-\mu_x}{\zeta}\right)+\left(\frac{-\mu_z}{\zeta}\right)\frac{\partial}{\partial\mu_y}\left(\frac{\mu_y}{\zeta}\right)\\
&\qquad+\frac{\mu_z}{\zeta}\frac{\partial}{\partial\mu_z}\left(\frac{\mu_y}{\zeta}\right)+\left(\frac{-\mu_x}{\zeta}\right)\frac{\partial}{\partial\mu_z}\left(\frac{-\mu_x}{\zeta}\right)\Bigg]\\
&\quad= \frac{-2kT}{\zeta}\mu_z \qquad (42)
\end{aligned}
$$

which is the noise-induced drift. We must now further average Eq. (42) over the density distribution function $W(\{\boldsymbol{\mu}\},t)$ of dipole orientations in configuration space at time t. Equation (42) then becomes $(-2kT/\zeta)\langle\mu_z\rangle$,where $\langle f\rangle$ denotes averaging a function f over the density function $W(\{\boldsymbol{\mu}\},t)$, namely $\langle f(\{\boldsymbol{\mu}\})\rangle = \int f(\{\boldsymbol{\mu}\})W(\{\boldsymbol{\mu}\},t)\,\mathrm{d}\boldsymbol{\mu}$. The averaged deterministic drift, from Eq. (35) is

$$\langle h_3\{\boldsymbol{\mu}\}\rangle = \frac{\mu^2-\langle\mu_z^2\rangle}{\zeta}E_1U(t)+\frac{\langle\mu^2\mu_z-\mu_z^3\rangle E_0}{\zeta\mu} \qquad (43)$$

Thus, the averaged equation of motion of the dipole is

$$\frac{\mathrm{d}}{\mathrm{d}t}\langle\mu_z\rangle+\frac{2kT}{\zeta}\langle\mu_z\rangle=\frac{E_1U(t)}{\zeta}\langle\mu^2-\mu_z^2\rangle+\frac{E_0}{\zeta}\langle\mu^2\mu_z-\mu_z^3\rangle \qquad (44)$$

Now

$$\mu_z=\mu\cos\vartheta=\mu P_1(\cos\vartheta) \qquad (45)$$

and so

$$\frac{d}{dt}\langle P_1(\cos\vartheta)\rangle + \frac{2kT}{\zeta}\langle P_1(\cos\vartheta)\rangle = \frac{2\mu[E_1U(t)]}{3\zeta}[1 - \langle P_2(\cos\vartheta)\rangle] + \frac{2}{5}\frac{\mu E_0}{\zeta}[\langle P_1(\cos\vartheta)\rangle - \langle P_3(\cos\vartheta)\rangle] \tag{46}$$

$$P_2(u_z) = \tfrac{1}{2}(3u_z^2 - 1) \tag{47}$$

$$P_3(u_z) = \tfrac{1}{2}(5u_z^3 - 3u_z) \tag{48}$$

are the Legendre polynomials of order 1, 2, and 3, $u_z = \cos\vartheta$.

C. Equation of Motion of the Transverse or x Component of the Dipole Moment

In the transverse case the step change in the field is applied parallel to the x axis so that we need to determine the behavior of $\langle\mu_x\rangle$. We find as before that the x component of the dipole moment satisfies

$$\frac{d}{dt}\langle\mu_x\rangle + \frac{2kT}{\zeta}\langle\mu_x\rangle = \frac{\mu^2}{\zeta}E_1U(t) - \frac{\langle\mu_x^2\rangle}{\zeta}E_1U(t) - \frac{\langle\mu_z^2\mu_x\rangle}{\mu\zeta}E_0 \tag{49}$$

Now

$$\mu_x = \mu\cos\vartheta\cos\phi = \mu X_{11}(\vartheta,\phi) \tag{50}$$

where X_{11} is the spherical harmonic of order 1 (Ref. 25). Thus, Eq. (49) becomes

$$\frac{d}{dt}\langle X_{11}\rangle + \frac{2kT}{\zeta}\langle X_{11}\rangle = -\frac{\mu E_0}{\zeta}\left(\frac{2}{15}\langle X_{31}\rangle + \frac{1}{5}\langle X_{11}\rangle\right) + \frac{\mu E_1U(t)}{3\zeta}\left[2 - \frac{1}{2}\langle X_{22}\rangle + \langle P_2\rangle\right] \tag{51}$$

$$X_{31} = \tfrac{3}{2}(5\cos^2\vartheta - 1)\sin\vartheta\cos\phi \tag{52}$$

$$X_{22} = 3\sin^2\vartheta\cos 2\phi \tag{53}$$

D. Calculation of the Effective Relaxation Time for the Parallel Component of the Dipole Moment

Equations (46) and (51) are the leading terms in the infinite hierarchy of differential-difference equations, which describe the averages $\langle X_{nm}(\vartheta, \phi)\rangle$. We show in Appendix A how the next term of the hierarchy can also be obtained by direct averaging of the Langevin equation. The calculation of the relaxation times for both parallel and perpendicular cases is accomplished by rewriting the infinite hierarchy as the set of ordinary differential equations

$$\frac{\mathrm{d}}{\mathrm{d}t}\mathbf{X} = \mathbf{A}\mathbf{X} + \mathbf{B}U(t) \tag{54}$$

and truncating at a given size of **A**, where **A** is the transition matrix and **B** is the driving force matrix. The relaxation times are the reciprocals of the roots of the characteristic equation

$$\det\{s\mathbf{I} - \mathbf{A}\} = 0 \tag{55}$$

where s denotes the complex frequency.

The disadvantage of this method is that it is, in general, impossible to get closed form expressions for the relaxation times, nor is it possible to describe the mean dipole moment by a single relaxation time except in the trivial case where the nematic potential is zero so that

$$\frac{\mu E_0}{2kT} = \sigma \ll 1 \tag{56}$$

This difficulty may be circumvented by the effective eigenvalue method, which has been used by Morita[7] in conjunction with the Fokker-Planck equation. A similar approach may be used in the context of the averaged Langevin equation, as has been demonstrated in Ref. 24 in connection with the problem of the linear response of a polar fluid in the presence of a dc bias field.

We first consider the parallel equation of motion, Eq. (46). We recall that the nematic field $\mathbf{E}_0$ has always been present and also that we are interested only in the response linear in $\mathbf{E}_1$. We therefore assume just as in Ref. 24 that

$$\langle P_n\rangle = \langle P_n\rangle_0 + \langle P_n\rangle_1 \tag{57}$$

where P_n is the Legendre polynomial of order n. The subscript 0 denotes

the average in the absence of the field $\mathbf{E}_1$ and the subscript 1 denotes the portion of the average that is linear in E_1. On substituting this equation into Eq. (46) we have

$$\begin{aligned}\frac{\mathrm{d}}{\mathrm{d}t}\{\langle P_1\rangle_0 + \langle P_1\rangle_1\} + \frac{2kT}{\zeta}\{\langle P_1\rangle_0 + \langle P_1\rangle_1\} \\ = \frac{2\mu}{3\zeta}\{E_1U(t)\}\{1 - \langle P_2\rangle_0 - \langle P_2\rangle_1\} \\ + \frac{2}{5}\frac{\mu}{\zeta}E_0\{\langle P_1\rangle_0 + \langle P_1\rangle_1 - \langle P_3\rangle_0 - \langle P_3\rangle_1\}\end{aligned} \tag{58}$$

Now $\langle P_1\rangle_0$ has reached equilibrium so that

$$\frac{2kT}{\zeta}\langle P_1\rangle_0 = \frac{2\mu}{5\zeta}E_0\{\langle P_1\rangle_0 - \langle P_3\rangle_0\} \tag{59}$$

The solution of this recurrence relation is

$$\langle P_1\rangle_0 = \langle P_3\rangle_0 = 0 \tag{60}$$

because the nematic field cannot orient the dipole. The average $\langle P_1\rangle_1$ satisfies

$$\begin{aligned}\frac{\mathrm{d}}{\mathrm{d}t}\langle P_1\rangle_1 + \frac{2kT}{\zeta}\langle P_1\rangle_1 = \frac{2\mu E_0}{5\zeta}[\langle P_1\rangle_1 - \langle P_3\rangle_1] \\ + \frac{2\mu E_1U(t)}{3\zeta}[1 - \langle P_2\rangle_0]\end{aligned} \tag{61}$$

In the last term the averages are those in the absence of E_1; because this term is multiplied by E_1 it is of the same order as the others.

Equation (61) is a three-term recurrence relation driven by a forcing function, the E_1 term. To determine eigenvalues we consider the unforced equation:

$$\frac{\mathrm{d}}{\mathrm{d}t}\langle P_1\rangle_1 + \frac{2kT}{\zeta}\langle P_1\rangle_1 = \frac{2\mu E_0}{5\zeta}[\langle P_1\rangle_1 - \langle P_3\rangle_1] \tag{62}$$

Recalling that

$$\langle P_1(\cos\vartheta(0))\rangle_1 = 0 \tag{63}$$

the Laplace transform of Eq. (62) is

$$\left(s + 2\frac{kT}{\zeta}\right)\mathscr{L}\langle P_1\rangle_1 = \frac{2\mu E_0}{5\zeta}\left[\mathscr{L}\langle P_1\rangle_1 - \mathscr{L}\langle P_3\rangle_1\right] \tag{64}$$

yielding

$$\left(s + \frac{1}{\tau_D} - \frac{2\sigma}{5\tau_D}\right)\mathscr{L}\langle P_1\rangle_1 = -\frac{2\sigma}{5\tau_D}\mathscr{L}\langle P_3\rangle_1 \tag{65}$$

where $\tau_D = \zeta/2kT$ is the Debye relaxation time. The characteristic equation of the system is

$$s + \frac{1}{\tau_D} - \frac{2\sigma}{5\tau_D}\left[1 - \frac{\mathscr{L}\langle P_3\rangle_1}{\mathscr{L}\langle P_1\rangle_1}\right] = 0 \tag{66}$$

We have stated that this will lead to a polynomial equation of high order in s yielding a discrete set of relaxation times and that one cannot in general obtain a closed form result for the lowest root. If we suppose, however, following the approach outlined in Refs. 7 and 24, that the term

$$\frac{\mathscr{L}\langle P_3\rangle_1}{\mathscr{L}\langle P_1\rangle_1}$$

may be replaced by its final (equilibrium) value (i.e., its value as t tends to infinity)

$$\frac{\lim_{t\to\infty}\langle P_3\rangle_1}{\lim_{t\to\infty}\langle P_1\rangle_1} = \frac{\lim_{s\to 0} s\mathscr{L}\langle P_3\rangle_1}{\lim_{s\to 0} s\mathscr{L}\langle P_1\rangle_1}$$

then Eq. (66) may be evaluated in terms of a single relaxation time that depends on the strength of the nematic potential. This is accomplished as follows. At equilibrium, which occurs when $t \gg \tau_D$,

$$\lim_{t\to\infty}\frac{\langle P_3\rangle_1}{\langle P_1\rangle_1} = \frac{\int_{-1}^{+1}\frac{1}{2}(5x^3 - 3x)e^{\sigma x^2 + \xi_1 x}\,dx}{\int_{-1}^{+1} x e^{\sigma x^2 + \xi_1 x}\,dx} \tag{67}$$

On expanding Eq. (67) as a Taylor series in ξ_1 we have (we retain only terms linear in ξ_1 since we are concerned with the linear response)

$$\lim_{t\to\infty} \frac{\langle P_3\rangle_1}{\langle P_1\rangle_1} = \frac{1}{2} \frac{\int_{-1}^{+1}(5x^4 - 3x^2)e^{\sigma x^2}\,dx}{\int_{-1}^{+1} x^2 e^{\sigma x^2}\,dx} \tag{68}$$

Furthermore, on defining the function F by[6]

$$F(\sigma) = \int_0^1 e^{\sigma x^2}\,dx \tag{69}$$

we have

$$\int_0^1 x^2 e^{\sigma x^2}\,dx = F'(\sigma) = \frac{1}{2\sigma}[1 - F(\sigma)] \tag{70}$$

$$\int_0^1 x^4 e^{\sigma x^2}\,dx = F''(\sigma) = \frac{1}{2\sigma}\left\{1 - \frac{3}{2\sigma}[1 - F(\sigma)]\right\} \tag{71}$$

The function $F(\sigma)$ is related to Dawson's integral $D(\sigma)$,[26]

$$D(\sigma) = \exp(-\sigma^2)\int_0^\sigma \exp(t^2)\,dt$$

by

$$D(\sqrt{\sigma}) = \sqrt{\sigma}\, e^{-\sigma} F(\sigma) \tag{72}$$

Equation (68) may now be written

$$\lim_{t\to\infty} \frac{\langle P_3\rangle_1}{\langle P_1\rangle_1} = -\frac{3}{2} + \frac{5}{2}\frac{F''(\sigma)}{F'(\sigma)} \tag{73}$$

Thus, the effective eigenvalue is

$$\lambda_{\|} = \frac{1}{\tau_D} - \frac{2\sigma}{5\tau_D}\left[1 - \lim_{t\to\infty} \frac{\langle P_3\rangle_1}{\langle P_1\rangle_1}\right] \tag{74}$$

or

$$\lambda_{\|} = \frac{1}{\tau_D}\left(1 - \sigma + \frac{\sigma F''(\sigma)}{F'(\sigma)}\right) = \frac{1}{2\tau_D}\left(\frac{F(\sigma)}{F'(\sigma)} - 1\right) \tag{75}$$

in agreement with the result of Ref. 6. There the Fokker-Planck equation combined with Leontovich's method[1] was used for the calculation of the magnetization relaxation times of single-domain ferromagnetic particles (see Section II.I). Thus,

$$\tau_{\|} = \frac{1}{\lambda_{\|}} = \frac{\tau_D}{1 - \sigma + \sigma[F''(\sigma)/F'(\sigma)]} = 2\tau_D \frac{F'(\sigma)}{F(\sigma) - F'(\sigma)} \tag{76}$$

or

$$\tau_{\|} = 2\tau_D \frac{\sqrt{\sigma} - D(\sqrt{\sigma})}{D(\sqrt{\sigma})(1 + 2\sigma) - \sqrt{\sigma}} \tag{77}$$

Since[6]

$$\frac{\sigma F''(\sigma)}{F'(\sigma)} = \sigma \frac{\frac{1}{5} + \frac{1}{7}\sigma \cdots}{\frac{1}{3} + \frac{3}{5}\sigma} \approx \frac{3}{5}\sigma \tag{78}$$

for small σ, we have

$$\tau_{\|} = \tau_D \tag{79}$$

If terms $O(\sigma^2)$ are retained

$$\tau_{\|} = \frac{\tau_D}{1 - \frac{2}{5}\sigma} = \tau_D\left(1 + \frac{2}{5}\sigma\right) \tag{80}$$

On the other hand, for very large σ, $\tau_{\|}$ becomes

$$\tau_{\|} = 2\sigma\tau_D \tag{81}$$

in sharp contrast to the result for the large dc bias field where $\tau_{\|}$ vanishes as the reciprocal of the field strength[24] and in qualitative agreement with Refs. 6 and 29, where the longitudinal relaxation time increases as the potential strength is increased.

E. Calculation of the Effective Relaxation Time for the Transverse Component of the Dipole Moment

We assume as before that

$$\langle X_{nm} \rangle = \langle X_{nm} \rangle_0 + \langle X_{nm} \rangle_1 \tag{82}$$

which leads us to the linearized equation for $\langle X_{nm} \rangle$:

$$\langle \dot{X}_{11} \rangle_1 + \frac{2kT}{\zeta} \langle X_{11} \rangle_1 = -\frac{\mu E_0}{\zeta} \left(\frac{2}{15} \langle X_{31} \rangle_1 + \frac{1}{5} \langle X_{11} \rangle_1 \right) + \frac{\mu E_1 U(t)}{3\zeta} \left[2 - \frac{1}{2} \langle X_{22} \rangle_0 + \langle P_2 \rangle_0 \right] \tag{83}$$

and as in Eq. (59)

$$\langle X_{11} \rangle_0 = 0 \tag{84}$$

Thus, the eigenvalue equation is

$$s + \frac{1}{\tau_D} + \frac{\sigma}{5\tau_D} + \frac{2\sigma}{15\tau_D} \frac{\mathscr{L}\langle X_{31} \rangle_1}{\mathscr{L}\langle X_{11} \rangle_1} = 0 \tag{85}$$

Now

$$\lim_{t \to \infty} \langle X_{11} \rangle = \langle \sin \vartheta \cos \phi \rangle = \frac{\int_0^{2\pi} \int_0^{\pi} \sin \vartheta \cos \phi \, e^{\sigma \cos^2 \vartheta + \xi_1 \sin \vartheta \cos \phi} \sin \vartheta \, d\vartheta \, d\phi}{\int_0^{2\pi} \int_0^{\pi} e^{\sigma \cos^2 \vartheta + \xi_1 \sin \vartheta \cos \phi} \sin \vartheta \, d\vartheta \, d\phi} \tag{86}$$

which in the linear approximation of ξ_1 is

$$\langle X_{11} \rangle_1 = \frac{\xi_1 \int_{-1}^{+1} (1 - x^2) e^{\sigma x^2} dx}{\int_{-1}^{+1} e^{\sigma x^2} dx} = \xi_1 \frac{F(\sigma) - F'(\sigma)}{F(\sigma)} \tag{87}$$

Likewise,

$$\langle X_{31} \rangle_1 = \frac{3}{2} \frac{\xi_1 \int_{-1}^{+1} (5x^2 - 1)(1 - x^2) e^{\sigma x^2} dx}{\int_{-1}^{+1} e^{\sigma x^2} dx} = \frac{3}{2} \xi_1 \frac{\int_{-1}^{+1} (-5x^4 + 6x^2 - 1) e^{\sigma x^2} dx}{\int_{-1}^{+1} e^{\sigma x^2} dx} = \frac{3}{2} \xi_1 \frac{(-5F'' + 6F' - F)}{F} \tag{88}$$

Thus,

$$\lim_{t\to\infty}\frac{\langle X_{31}\rangle_1}{\langle X_{11}\rangle_1}=\frac{3}{2}\frac{6F'-5F''-F}{F-F'} \tag{89}$$

whence

$$-s=\lambda_\perp=\frac{1}{\tau_D}\left[1+\frac{\sigma}{5}+\frac{2\sigma}{15}\frac{3}{2}\frac{6F'-5F''-F}{F-F'}\right] \tag{90}$$

$$=\frac{1}{\tau_D}\left[1+\sigma\frac{F'-F''}{F-F'}\right] \tag{91}$$

Now

$$\lambda_\perp=\tau_\perp^{-1} \tag{92}$$

whence

$$\tau_\perp(\sigma)=\frac{\tau_D}{1+\sigma\dfrac{F'-F''}{F-F'}}=2\tau_D\frac{F(\sigma)-F'(\sigma)}{F(\sigma)+F'(\sigma)} \tag{93}$$

or

$$\tau_\perp=2\tau_D\frac{D(\sqrt{\sigma})(2\sigma+1)-\sqrt{\sigma}}{D(\sqrt{\sigma})(2\sigma-1)+\sqrt{\sigma}} \tag{93a}$$

Let us suppose that σ is so small that only terms $O(\sigma^2)$ are retained. Then

$$\tau_\perp(\sigma)=\tau_D\left(1-\frac{\sigma}{5}\right) \tag{94}$$

while for very large σ the behavior is similar to that of a dipole in a dc bias field, viz.,

$$\tau_\perp(\sigma)=\frac{\tau_D}{\sigma} \tag{95}$$

in contrast to the result for $\tau_\parallel$ and again in agreement with the result of Refs. 6 and 29.

F. Calculation of the Frequency Dependence of the Susceptibility

Having determined the effective relaxation times for transverse and longitudinal fields, we may calculate the frequency dependence of the polarization as follows. We recall that Eq. (61) is

$$\langle \dot{P}_1 \rangle_1 + \frac{2kT}{\zeta}\langle P_1 \rangle_1 = \frac{2\mu E_0}{5\zeta}\left[\langle P_1 \rangle_1 - \langle P_3 \rangle_1\right] + \frac{2\mu E_1 U(t)}{3\zeta}\left[\langle P_0 \rangle_0 - \langle P_2 \rangle_0\right] \tag{96}$$

and that the effective eigenvalue procedure reduces this equation to

$$\langle \dot{P}_1 \rangle_1 + \frac{1}{\tau_{\|}(\sigma)}\langle P_1 \rangle_1 = \frac{2\mu E_1 U(t)}{3\zeta}\left[\langle P_0 \rangle_0 - \langle P_2 \rangle_0\right] = \frac{2\mu E_1 U(t)}{\zeta}\left[1 - \frac{F'(\sigma)}{F(\sigma)}\right] \tag{97}$$

where we have used Eqs. (69) and (70) to simplify the right side of Eq. (96). Equation (97) may be further simplified to

$$\frac{\mathrm{d}}{\mathrm{d}t}\langle P_1 \rangle_1 + \frac{1}{\tau_{\|}(\sigma)}\langle P_1 \rangle_1 = \frac{\xi_1 U(t)}{2\tau_D}\left[1 - \frac{F'(\sigma)}{F(\sigma)}\right] \tag{98}$$

by using Eq. (75). This is a first-order ordinary differential equation in $\langle P_1 \rangle_1$ with solution

$$\langle P_1 \rangle_1 = \frac{\xi_1 F'(\sigma)}{F(\sigma)} U(t)\left\{1 - \exp\left[-t/\tau_{\|}(\sigma)\right]\right\} \tag{99}$$

The rise transient of the polarization arising from $\mathbf{E}_1$, assuming N noninteracting dipoles, is then

$$N\mu\langle P_1 \rangle_1 = \frac{N\mu^2 E_1 F'(\sigma)}{kTF(\sigma)} U(t)\left\{1 - \exp\left[-t/\tau_{\|}(\sigma)\right]\right\} \tag{100}$$

or

$$P_r(t) = \chi'_{\|}(0) E_1 U(t)\left\{1 - \exp\left[-t/\tau_{\|}(\sigma)\right]\right\} \tag{101}$$

where

$$\chi'_{\parallel}(0) = \frac{N\mu^2}{kT}\frac{F'(\sigma)}{F(\sigma)}$$

is the static susceptibility. N is the number of molecules per unit volume. This is the rise transient of the polarization. The decay transient following the removal of E_1 is

$$P_{\mathrm{d}}(t) = \chi'_{\parallel}(0)E_1U(t)\mathrm{e}^{-t/\tau_{\parallel}(\sigma)} \tag{102}$$

The after-effect function $f(t)$ is thus

$$f(t) = \chi'_{\parallel}(0)^{-t/\tau_{\parallel}(\sigma)} \tag{103}$$

The frequency-dependent susceptibility arising from the imposition of an ac field $\mathbf{E}_1\mathrm{e}^{\mathrm{i}\omega t}$ may then be written (since we have limited the solution to terms linear in ξ_1) from linear response theory[10] as

$$\begin{aligned}\chi_{\parallel}(\omega) &= \chi'_{\parallel}(\omega) - \mathrm{i}\chi''_{\parallel}(\omega) = f(0) - \mathrm{i}\omega\int_0^{\infty} f(t)\mathrm{e}^{-i\omega t}\,\mathrm{d}t \\ &= \chi'_{\parallel}(0)\left[1 - \mathrm{i}\omega\int_0^{\infty}\mathrm{e}^{-t/\tau_{\parallel}(\sigma)}\,\mathrm{e}^{-\mathrm{i}\omega t}\,\mathrm{d}t\right]\end{aligned} \tag{104}$$

Thus,

$$\chi_{\parallel}(\omega) = \frac{\chi'_{\parallel}(0)}{1 + \mathrm{i}\omega\tau_{\parallel}} \tag{105}$$

This is the result for a longitudinal field and is in agreement with an earlier result of Raĭkher and Shliomis[6] for magnetic relaxation of ferromagnetic domains. From Eq. (83), the result for a transverse field is

$$\begin{aligned}\frac{\mathrm{d}}{\mathrm{d}t}\langle X_{11}\rangle_1 + \frac{2kT}{\zeta}\langle X_{11}\rangle_1 &= -\frac{\mu E_0}{\zeta}\left[\frac{2}{15}\langle X_{31}\rangle_1 + \frac{1}{5}\langle X_{11}\rangle_1\right] \\ &\quad + \frac{\mu E_1U(t)}{3\zeta}\left[2 - \frac{1}{2}\langle X_{22}\rangle_0 + \langle P_2\rangle_0\right]\end{aligned} \tag{106}$$

which with our closure procedure becomes

$$\frac{\mathrm{d}}{\mathrm{d}t}\langle X_{11}\rangle_1 + \lambda_\perp \langle X_{11}\rangle_1 = \frac{\mu E_1 U(t)}{3\zeta}\left[2 - \frac{1}{2}\langle X_{22}\rangle_0 + \langle P_2\rangle_0\right] \tag{107}$$

where

$$\langle X_{22}\rangle_0 = 0 \tag{108}$$

since

$$\int_0^{2\pi} \cos 2\phi \, \mathrm{d}\phi = 0 \tag{109}$$

This is a consequence of the axial symmetry of the nematic potential. Equation (107) then simplifies with the aid of Eq. (69) to

$$\frac{\mathrm{d}}{\mathrm{d}t}\langle X_{11}\rangle_1 + \frac{1}{\tau_\perp}\langle X_{11}\rangle_1 = \frac{\xi_1 U(t)}{2\tau_D}\left[1 + \frac{F'(\sigma)}{F(\sigma)}\right] \tag{110}$$

We then find, just as for the parallel case, that the complex susceptibility for a small transverse field $\mathbf{E}_1 e^{i\omega t}$ is

$$\chi_\perp(\omega) = \frac{\chi'_\perp(0)}{1 + i\omega\tau_\perp} \tag{111}$$

where

$$\chi'_\perp(0) = \frac{\mu^2 N}{2kT}\left[1 - \frac{F'(\sigma)}{F(\sigma)}\right]$$

and $\tau_\perp$ is given by Eq. (93).

G. The Relaxation Times for An Arbitrary Uniaxial Nematic Potential

The analysis so far has been restricted to the simplest nematic potential, namely

$$V(\vartheta) = -V_0 \cos^2 \vartheta \tag{112}$$

We now show how the analysis may be extended to yield the relaxation times for an arbitrary nematic potential. The nematic field $\mathbf{E}_0$ is the gradient of a potential $V(\{\boldsymbol{\mu}\})$ so that

$$\mathbf{E}_0(\{\boldsymbol{\mu}\}) = -\frac{\partial V}{\partial \boldsymbol{\mu}} \tag{113}$$

where

$$\frac{\partial}{\partial \boldsymbol{\mu}} = \mathbf{i}\frac{\partial}{\partial \mu_x} + \mathbf{j}\frac{\partial}{\partial \mu_y} + \mathbf{k}\frac{\partial}{\partial \mu_z} \tag{114}$$

Because $|\boldsymbol{\mu}| = \text{constant} = \mu$, V is indeterminate by an arbitrary function of μ^2, and $\mathbf{E}_0$ by an arbitrary vector along $\boldsymbol{\mu}$, which contribute nothing to $\boldsymbol{\mu} \times \mathbf{E}_0$. We confine ourselves to an uniaxial potential, where $\mathbf{E}_0(\{\boldsymbol{\mu}\})$ has only a $\mathbf{k}$ component so that

$$\mathbf{E}_0(\{\boldsymbol{\mu}\}) = -\mathbf{k}\frac{\partial}{\partial \mu_z} V(\{\boldsymbol{\mu}\}) \tag{115}$$

The total electric field acting on the molecule is

$$\mathbf{E}_T(t) = \mathbf{E}_0(\{\boldsymbol{\mu}(t)\}) + \mathbf{E}_1(t) \tag{116}$$

This consists of the axially symmetric field $\mathbf{E}_0$ and the small externally applied probe field $\mathbf{E}_1(t)$, where $\mu E_1(t) \ll kT$.

By using Eq. (116) we can rewrite the Langevin equation (24) as

$$\begin{aligned}\frac{\mathrm{d}\boldsymbol{\mu}(t)}{\mathrm{d}t} = &-\zeta^{-1}(\boldsymbol{\mu}(t) \times (\boldsymbol{\mu}(t) \times \mathbf{E}_0(\{\boldsymbol{\mu}(t)\}))) \\ &+ \mathbf{F}(\{\boldsymbol{\mu}(t)\})E_1(t) + \mathbf{L}(t)\end{aligned} \tag{117}$$

where

$$\mathbf{F}(\{\boldsymbol{\mu}(t)\}) = -\zeta^{-1}(\boldsymbol{\mu}(t) \times (\boldsymbol{\mu}(t) \times \mathbf{e})) \qquad \mathbf{e} = \frac{\mathbf{E}_1(t)}{E_1(t)} \tag{118}$$

$$\mathbf{L}(t) = \zeta^{-1}(\boldsymbol{\lambda}(t) \times \boldsymbol{\mu}(t)) \tag{119}$$

On averaging (c.f., Eqs. (44) and (49)),

$$\frac{\mathrm{d}}{\mathrm{d}t}\langle \mu_x \rangle + \frac{1}{\tau_D}\langle \mu_x \rangle = -\zeta^{-1}\langle E_0 \mu_x \mu_z \rangle + \langle F_x \rangle E_1(t) \tag{120}$$

$$\frac{\mathrm{d}}{\mathrm{d}t}\langle \mu_y \rangle + \frac{1}{\tau_D}\langle \mu_y \rangle = -\zeta^{-1}\langle E_0 \mu_y \mu_z \rangle + \langle F_y \rangle E_1(t) \tag{121}$$

$$\frac{\mathrm{d}}{\mathrm{d}t}\langle \mu_z \rangle + \frac{1}{\tau_D}\langle \mu_z \rangle = \zeta^{-1}\left\langle E_0\left(\mu^2 - \mu_z^2\right)\right\rangle + \langle F_z \rangle E_1(t) \tag{122}$$

We first consider the equation of motion $\langle \mu_z \rangle$. We suppose that a small field $\mathbf{E}_1(t) = E_1(t)\mathbf{k}$ is applied along the z axis at time $t = 0$. We require as before the linear response to $\mathbf{E}_1(t)$. We therefore assume that in Eq. (122) $\langle \mu_z \rangle$ and $\langle E_0(\mu^2 - \mu_z^2)\rangle$ can be represented as

$$\langle \mu_z \rangle = \langle \mu_z \rangle_0 + \langle \mu_z \rangle_1 \tag{123}$$

$$\left\langle E_0\left(\mu^2 - \mu_z^2\right)\right\rangle = \left\langle E_0\left(\mu^2 - \mu_z^2\right)\right\rangle_0 + \left\langle E_0\left(\mu^2 - \mu_z^2\right)\right\rangle_1 \tag{124}$$

As before, the subscript 0 denotes the equilibrium ensemble average in the absence of the field $\mathbf{E}_1(t)$ and the subscript 1 denotes the portion of the ensemble average that is linear in $\mathbf{E}_1(t)$. Thus, we have from Eqs. (122)–(124)

$$\begin{aligned} &\frac{\mathrm{d}}{\mathrm{d}t}\left(\langle \mu_z \rangle_0 + \langle \mu_z \rangle_1\right) + \frac{1}{\tau_D}\left(\langle \mu_z \rangle_0 + \langle \mu_z \rangle_1\right) \\ &\quad = \zeta^{-1}\left(\left\langle E_0\left(\mu^2 - \mu_z^2\right)\right\rangle_0 + \left\langle E_0\left(\mu^2 - \mu_z^2\right)\right\rangle_1\right) \\ &\qquad + \left(\langle F_z \rangle_0 + \langle F_z \rangle_1\right) E_1(t) \end{aligned} \tag{125}$$

Now $\langle \mu_z \rangle_0$ has reached equilibrium, so that

$$\langle \mu_z \rangle_0 = \int_0^{2\pi}\int_0^{\pi} \mu \cos\vartheta W_0(\vartheta) \sin\vartheta \, \mathrm{d}\vartheta \, \mathrm{d}\phi \tag{126}$$

where

$$W_0(\vartheta) = C \exp\left(-\frac{V(\vartheta)}{kT}\right) \tag{127}$$

is the equilibrium distribution function, ϑ and ϕ are the polar and

azimuthal angles, respectively, and C is the normalizing constant. The ensemble average $\langle \mu_z \rangle_1$ satisfies

$$\frac{\mathrm{d}}{\mathrm{d}t}\langle \mu_z \rangle_1 + \frac{1}{\tau_D}\langle \mu_z \rangle_1 = \zeta^{-1}\langle E_0(\mu^2 - \mu_z^2)\rangle_1 + \zeta^{-1}\langle \mu^2 - \mu_z^2 \rangle_0 E_1(t) \tag{128}$$

To determine $\tau_{\|}$ we consider the unforced equation:

$$\frac{\mathrm{d}}{\mathrm{d}t}\langle \mu_z \rangle_1 + \frac{1}{\tau_D}\langle \mu_z \rangle_1 = \zeta^{-1}\langle E_0(\mu^2 - \mu_z^2)\rangle_1 \tag{129}$$

The characteristic equation of the system is then

$$s + \tau_D^{-1} - \zeta^{-1}\frac{\mathscr{L}\langle E_0(\mu^2 - \mu_z^2)\rangle_1}{\mathscr{L}\langle \mu_z \rangle_1} = 0 \tag{130}$$

where the symbol $\mathscr{L}$ is the Laplace transform.

If we suppose, again following Morita,[7] that

$$\frac{\mathscr{L}\langle E_0(\mu^2 - \mu_z^2)\rangle_1}{\mathscr{L}\langle \mu_z \rangle_1} = \frac{\mathscr{L}\langle E_0(\mu^2 - \mu_z^2)\rangle - \mathscr{L}\langle E_0(\mu^2 - \mu_z^2)\rangle_0}{\mathscr{L}\langle \mu_z \rangle - \mathscr{L}\langle \mu_z \rangle_0} \tag{131}$$

may be replaced by its final (equilibrium) value (i.e., its value as t tends to infinity),

$$\lim_{t\to\infty}\frac{\langle E_0(\mu^2 - \mu_z^2)\rangle_1}{\langle \mu_z \rangle_1} = \lim_{s\to 0}\frac{s\mathscr{L}\langle E_0(\mu^2 - \mu_z^2)\rangle_1}{s\mathscr{L}\langle \mu_z \rangle_1} \tag{132}$$

then Eq. (132) may be evaluated as follows. At equilibrium ($t \to \infty$)

$$\langle \mu_z \rangle = \mu\frac{\int_0^\pi \cos\vartheta \exp[-V/kT + (\mu E_1/kT)\cos\vartheta]\sin\vartheta\,\mathrm{d}\vartheta}{\int_0^\pi \exp[-V/kT + (\mu E_1/kT)\cos\vartheta]\sin\vartheta\,\mathrm{d}\vartheta}$$

which becomes in the linear approximation of E_1

$$\begin{aligned}\langle \mu_z \rangle &\cong \mu \frac{\int_0^\pi \cos\vartheta\,[1 + (\mu E_1/kT)\cos\vartheta]\exp(-V/kT)\sin\vartheta\,d\vartheta}{\int_0^\pi (1 + (\mu E_1/kT)\cos\vartheta)\exp(-V/kT)\sin\vartheta\,d\vartheta} \\ &= \mu\langle\cos\vartheta\rangle_0 + \frac{\mu^2 E_1}{kT}\left(\langle\cos^2\vartheta\rangle_0 - \langle\cos\vartheta\rangle_0^2\right) \qquad (133) \\ &= \langle\mu_z\rangle_0 + \frac{E_1}{kT}\left(\langle\mu_z^2\rangle_0 - \langle\mu_z\rangle_0^2\right)\end{aligned}$$

Further,

$$\begin{aligned}&\langle E_0(\mu^2 - \mu_z^2)\rangle \\ &\quad = \mu^2 \frac{\int_0^\pi (1 - \cos^2\vartheta)E_0 \exp[-V/kT + (\mu E_1/kT)\cos\vartheta]\sin\vartheta\,d\vartheta}{\int_0^\pi \exp[-V/kT + (\mu E_1/kT)\cos\vartheta]\sin\vartheta\,d\vartheta} \\ &\quad \cong \mu^2 \frac{\int_0^\pi (1 - \cos^2\vartheta)E_0[1 + (\mu E_1/kT)\cos\vartheta]\exp(-V/kT)\sin\vartheta\,d\vartheta}{\int_0^\pi \exp(-V/kT)[1 + (\mu E_1/kT)\cos\vartheta]\sin\vartheta\,d\vartheta} \qquad (134) \\ &\quad = \langle E_0(\mu^2 - \mu_z^2)\rangle_0 + \frac{\mu^3 E_1}{kT}\langle E_0(\cos\vartheta - \langle\cos\vartheta\rangle_0)(1 - \cos^2\vartheta)\rangle_0\end{aligned}$$

The second term on the right side of Eq. (134) can be evaluated by noting that

$$\begin{aligned}&\langle E_0(\cos\vartheta - \langle\cos\vartheta\rangle_0)(1 - \cos^2\vartheta)\rangle_0 \\ &\quad = \frac{\int_0^\pi - [\partial V/(\mu\partial\cos\vartheta)][\cos\vartheta - \langle\cos\vartheta\rangle_0][1 - \cos^2\vartheta]\exp(-V/kT)\sin\vartheta\,d\vartheta}{\int_0^\pi \exp(-V/kT)\sin\vartheta\,d\vartheta} \\ &\quad = \frac{kT}{\mu}\frac{-[\cos\vartheta - \langle\cos\vartheta\rangle_0][1 - \cos^2\vartheta]e^{-V/kT}\Big|_0^\pi - \int_0^\pi (1 - 3\cos^2\vartheta + 2\cos\vartheta\langle\cos\vartheta\rangle_0)e^{-V/kT}\sin\vartheta\,d\vartheta}{\int_0^\pi e^{-V/kT}\sin\vartheta\,d\vartheta} \\ &\quad = -\frac{kT}{\mu}\left[1 - 3\langle\cos^2\vartheta\rangle_0 + 2\langle\cos\vartheta\rangle_0^2\right] \qquad (135)\end{aligned}$$

In the nematic phase $\langle \cos \vartheta \rangle_0$ is equal to zero. Substituting Eq. (135) into (134) we obtain

$$\left\langle E_0(\mu^2 - \mu_z^2)\right\rangle = \left\langle E_0(\mu^2 - \mu_z^2)\right\rangle_0 - E_1\left(\mu^2 - 3\langle \mu_z^2 \rangle_0\right) \tag{136}$$

The effective eigenvalue $\lambda_{\|}$ with this procedure is then

$$\begin{aligned}\lambda_{\|} &= \tau_D^{-1} - \zeta^{-1} \frac{\left\langle E_0(\mu^2 - \mu_z^2)\right\rangle - \left\langle E_0(\mu^2 - \mu_z^2)\right\rangle_0}{\langle \mu_z \rangle - \langle \mu_z \rangle_0} \\ &= \tau_D^{-1} - \zeta^{-1} \frac{\mu^2 E_1(1 - 3\langle \cos^2 \vartheta \rangle_0)}{\mu^2 E_1/kT\left(\langle \cos^2 \vartheta \rangle_0 - \langle \cos \vartheta \rangle_0^2\right)} \\ &= (2\tau_D)^{-1} \frac{1 - \langle \cos^2 \vartheta \rangle_0}{\langle \cos^2 \vartheta \rangle_0}\end{aligned} \tag{137}$$

We have used Eqs. (133) and (136) here. Thus, the longitudinal relaxation time $\tau_{\|} = \lambda_{\|}^{-1}$ may be expressed in terms of the equilibrium averages as

$$\begin{aligned}\tau_{\|} &= 2\tau_D \frac{\langle \cos^2 \vartheta \rangle_0}{1 - \langle \cos^2 \vartheta \rangle_0} \\ &= \tau_D \frac{2\langle P_2 \rangle_0 + 1}{1 - \langle P_2 \rangle_0} = \tau_D \frac{2S + 1}{1 - S}\end{aligned} \tag{138}$$

where P_2 is the Legendre polynomial of order 2 and $S = \langle P_2 \rangle_0$ is the order parameter. Equation (138) was first obtained by Meier[9, 10] from the Fokker-Planck equation.

We now calculate the transverse relaxation time $\tau_{\perp}$. We consider the same problem as above but this time the step change in the field $\mathbf{E}_1(t) = \mathbf{i}U(t)E_1(t)$ is applied parallel to the x axis so that we need to determine the behavior of $\langle \mu_x \rangle$ from Eq. (120). We find, just as before, that the eigenvalue equation is

$$s + \tau_D^{-1} - \zeta^{-1} \frac{\mathscr{L}\langle E_0 \mu_x \mu_z \rangle_1}{\mathscr{L}\langle \mu_x \rangle_1} = 0 \tag{139}$$

or

$$s + \tau_D^{-1} - \zeta^{-1} \frac{\langle E_0 \mu_x \mu_z \rangle_1}{\langle \mu_x \rangle_1} = 0 \qquad (t \to \infty) \tag{140}$$

Now

$$\langle \mu_x \rangle = \mu \frac{\int_0^\pi \int_0^{2\pi} \sin\vartheta \cos\phi \exp[-V/kT + (\mu E_1/kT)\sin\vartheta\cos\phi]\sin\vartheta \, d\vartheta \, d\phi}{\int_0^\pi \int_0^{2\pi} \exp[-V/kT + (\mu E_1/kT)\sin\vartheta\cos\phi]\sin\vartheta \, d\vartheta \, d\phi} \tag{141}$$

which in the linear approximation of E_1 is

$$\langle \mu_x \rangle \cong \frac{\mu^2 E_1}{kT} \langle \sin^2\vartheta \cos^2\phi \rangle_0 = \frac{\mu^2 E_1}{2kT}(1 - \langle \cos^2\vartheta \rangle_0) \tag{142}$$

In the same way

$$\begin{aligned} \langle E_0 \mu_x \mu_z \rangle &\cong \frac{\mu^3 E_1}{kT} \langle E_0 \cos\vartheta \sin^2\vartheta \cos^2\phi \rangle_0 \\ &= -\mu^2 E_1 (1 - 3\langle \cos^2\vartheta \rangle_0)/2 \end{aligned} \tag{143}$$

$$\langle E_0 \mu_y \rangle = 0 \tag{144}$$

We have used the same procedure as in Eq. (135) to obtain Eq. (143).

Thus, the effective eigenvalue $\lambda_\perp$ is given by

$$\begin{aligned} \lambda_\perp &= \tau_D^{-1} - \frac{\langle E_0 \mu_x \mu_z \rangle}{\langle \mu_x \rangle} = \tau_D^{-1} - \zeta^{-1} \frac{\mu^2 E_1 (1 - 3\langle \cos^2\vartheta \rangle_0)}{(\mu^2 E_1/kT)(1 - \langle \cos^2\vartheta \rangle_0)} \\ &= (2\tau_D)^{-1} \frac{1 + \langle \cos^2\vartheta \rangle_0}{1 - \langle \cos^2\vartheta \rangle_0} \end{aligned} \tag{145}$$

whence the transverse relaxation time $\tau_\perp = \lambda_\perp^{-1}$ may be expressed in terms of the equilibrium averages as

$$\begin{aligned} \tau_\perp &= 2\tau_D \frac{1 - \langle \cos^2\vartheta \rangle_0}{1 + \langle \cos^2\vartheta \rangle_0} \\ &= 2\tau_D \frac{1 - \langle P_2 \rangle_0}{2 + \langle P_2 \rangle_0} = 2\tau_D \frac{1 - S}{2 + S} \end{aligned} \tag{146}$$

Equations (138) and (146) for the relaxation times $\tau_{\parallel}$ and $\tau_{\perp}$ are valid for any axially symmetric nematic potential.

The retardation factors $g_{\parallel}$ and $g_{\perp}$ defined by

$$\tau_{\parallel} = g_{\parallel}\tau_D \qquad \tau_{\perp} = g_{\perp}\tau_D \tag{147}$$

may now be expressed entirely in terms of the order parameter S as

$$g_{\parallel} = \frac{2S+1}{1-S} \qquad g_{\perp} = \frac{2-2S}{2+S} \tag{148}$$

On eliminating the order parameter S in Eq. (148) one may deduce that

$$g_{\parallel} = \frac{2-g_{\perp}}{g_{\perp}} \tag{149}$$

$$g_{\perp} = \frac{2}{g_{\parallel}+1} \tag{150}$$

In accordance with the results of Section II.F, the longitudinal and transverse components of the complex susceptibility are given by the Debye equations

$$\chi_{\parallel}(\omega) = \frac{\chi'_{\parallel}(0)}{1+i\omega\tau_{\parallel}} \qquad \chi_{\perp}(\omega) = \frac{\chi'_{\perp}(0)}{1+i\omega\tau_{\perp}} \tag{151}$$

with relaxation times given by Eqs. (138) and (146); also

$$\chi'_{\parallel}(0) = \frac{\mu^2 N}{3kT}(2S+1) \qquad \chi'_{\perp}(0) = \frac{\mu^2 N}{3kT}(1-S) \tag{152}$$

We have shown how general formulae for $\tau_{\parallel}$ and $\tau_{\perp}$ (valid for an arbitrary uniaxial potential of the crystalline anisotropy) may be calculated directly in terms of the equilibrium order parameter S from the Langevin equation where that equation is regarded as a stochastic nonlinear equation of the Stratonovich type. This eliminates the complicated mathematical analysis arising from the Fokker-Planck equation. Our approach is based on the effective eigenvalue method which enables us in this instance to close the hierarchy of differential-difference equations by reducing the nth-order characteristic equation of the system to one of the first order. The relaxation of the polarization components is thus characterized by a

single effective eigenvalue or weighted decay rate. The effective eigenvalues $\lambda_{\parallel}$ and $\lambda_{\perp}$ give precise information on the initial decay of the polarization components. Equation (148) is in qualitative agreement with available experimental data (e.g., Ref. 28) and with previous theoretical estimates,[16, 29, 30] from which it follows that the relaxation time $\tau_{\parallel}$ increases and $\tau_{\perp}$ decreases in nematic liquid crystals as compared to the Debye relaxation time in the isotropic phase.

A comparison of the results predicted by Eq. (34) with experimental data for p, p'-heptylcyanobiphenyl[31] is given in Fig. 2. Experimental points for $g_{\perp}$ and $g_{\parallel}$ have been evaluated from Eqs. (147), where $\tau_{\perp}$ and $\tau_{\parallel}$ are regarded as experimental values and the values of the Debye relaxation times τ_D were extrapolated from those of the isotropic liquid. One may see from Fig. 2 that the theory deviates from the experimental observations. This is due to the fact that the effective eigenvalue for the longitudinal component of the polarization in a nematic liquid crystal yields a smaller relaxation time than that given by the lowest eigenvalue of the distribution function[9, 10, 16] as obtained by solution of the Sturm-Liouville equation. This is corroborated by numerical solution of the Fokker-Planck equation (transformed into a Schrödinger equation) used in laser physics for the calculation of the intensity correlation functions of the light field.

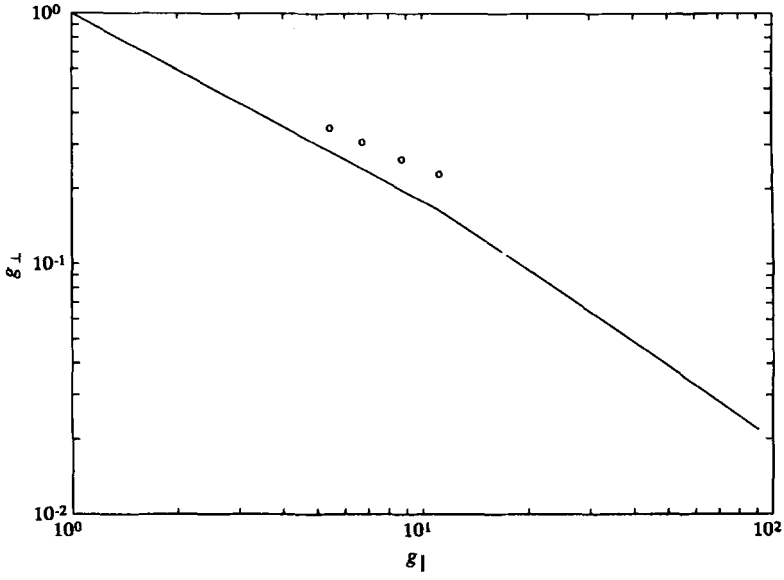


Figure 2. The retardation factor $g_{\perp}$ versus $g_{\parallel}$ for p, p'-heptylcyanobiphenyl. Experimental data (circles) from ref. [31].

Here λ_{eff} may be up to 25% greater than the lowest eigenvalue. The deviation has also been experimentally verified by Arecchi et al.[52] In Section II.J we compare the relaxation behavior yielded by the effective eigenvalue approach with the exact and asymptotic solutions of the problem.

H. Analogy with Magnetic Relaxation of Single-Domain Ferromagnetic Particles

The treatment of the relaxation behavior of nematic liquid crystals just given bears a close resemblance to the theory of magnetic relaxation of ferromagnetic domains. The theory has been reviewed by us in Volume 83 of this series. This study was initiated by W. F. Brown, Jr., who based his approach on the Fokker-Planck equation. The form of the Fokker-Planck equation obtained by Brown[15, 33] closely resembles that obtained by Debye[34] in his study of the dielectric relaxation of polar fluids. For convenience, the Fokker-Planck equation for the probability density of orientations of the magnetization $\mathbf{M}(t)$ of a single-domain ferromagnetic particle is called Brown's equation. Having obtained this equation, Brown applied the method of separation of the variables to convert the solution to a Sturm-Liouville problem.

The Sturm-Liouville method involves calculations of considerable mathematical complexity.[6, 15, 33] In view of this difficulty we have reconsidered in Ref. [11] the relaxation problem in the light of the new approach described here. This procedure, based on averaging the stochastic Gilbert equation for the magnetization $\mathbf{M}(t)$ allowed us to derive equations for the time evolution of the averages of magnetization components directly from that equation. The problem of constructing and solving the Fokker-Planck equation was thus bypassed entirely.

To discuss the Langevin equation for a single-domain ferromagnetic particle we first consider Gilbert's equation for the dynamic behavior of the magnetization vector of the particle $\mathbf{M}(t)$ in the presence of thermal agitation[11, 15]:

$$\frac{\mathrm{d}}{\mathrm{d}t}\mathbf{M}(t) = \gamma \mathbf{M}(t) \times \left(\mathbf{H}_{\mathrm{T}}(t) - \eta \frac{\mathrm{d}}{\mathrm{d}t}\mathbf{M}(t)\right) \tag{153}$$

where γ is the "gyromagnetic" ratio and η is a phenomenological damping parameter,

$$\mathbf{H}_{\mathrm{T}} = \mathbf{H}_{\mathrm{ef}} + \mathbf{H}(t) + \mathbf{h}(t) \tag{154}$$

is the total magnetic field, which consists of an axially symmetrical field

$\mathbf{H}_{ef}$ and a small external applied field $\mathbf{H}(t)$ ($\nu(\mathbf{M} \cdot \mathbf{H}) \ll kT$, k is the Boltzman constant, T is the temperature, ν is the volume of the particle) and $\mathbf{h}(t)$ is a random field term as used by Brown.[15] The random field $\mathbf{h}(t)$ has the properties

$$\overline{h_i(t)} = 0 \qquad \overline{h_j(t)h_i(t')} = \frac{2kT\eta}{\nu}\delta(t - t')\delta_{ij} \qquad (i, j = x, y, z) \quad (155)$$

where M_s is the (constant) magnitude of the magnetization, only its direction varies with time, the overbar indicates a statistical average,[3] $\delta(t)$ is the Dirac-delta function, and δ_{ij} is Kronecker's delta, so that we have assumed isotropy of the statistical properties of $h_i(t)$.

The effective field $\mathbf{H}_{ef}$ is due to the effect of the crystalline anisotropy.[33] If $V(\mathbf{M})$ is the free energy per unit volume expressed as a function of $\mathbf{M}$, then[33]

$$\mathbf{H}_{ef}(\mathbf{M}) = -\frac{\partial V}{\partial \mathbf{M}}$$

where

$$\frac{\partial}{\partial \mathbf{M}} = \mathbf{i}\frac{\partial}{\partial M_x} + \mathbf{j}\frac{\partial}{\partial M_y} + \mathbf{k}\frac{\partial}{\partial M_z}$$

If we confine ourselves to an uniaxial potential, so that $\mathbf{H}_{ef}(\mathbf{M})$ has only a $\mathbf{k}$ component, then

$$\mathbf{H}_{ef}(\mathbf{M}) = -\mathbf{k}\frac{\partial}{\partial M_z}V(\mathbf{M}) \qquad (156)$$

In the context of relaxation a single-domain ferromagnetic particle may reverse the direction of its magnetization due to thermal fluctuations inside the particle (spontaneous remagnetization). This is the solid-state mechanism known as Néel relaxation.[11] The Néel relaxation can be considered as a rotational diffusion of the magnetization $\mathbf{M}(t)$ with respect to the body of the particle in the crystalline field, so that it is an analog of the rotational Brownian motion of molecules in nematic liquid crystals.

Gilbert's Eq. (153) may be rearranged explicitly as shown in Ref. 35 to yield that equation in the Landau-Lifshitz form[36]:

$$\frac{\mathrm{d}}{\mathrm{d}t}\mathbf{M}(t) = g'M_s\mathbf{M}(t) \times \mathbf{H}_T(t) + h'(\mathbf{M}(t) \times \mathbf{H}_T(t)) \times \mathbf{M}(t) \quad (157)$$

where $M_s = |\mathbf{M}(t)|$, and the constants g' and h' are

$$g' = \frac{\gamma}{\left(1 + \eta^2\gamma^2 M_s^2\right)M_s} \qquad h' = \frac{\eta\gamma^2}{1 + \eta^2\gamma^2 M_s^2} \tag{158}$$

Using Eq. (154) we can rewrite Eq. (157) as

$$\begin{aligned}\frac{d\mathbf{M}(t)}{dt} &= g'M_s\mathbf{M}(t) \times \mathbf{H}_{ef}(t) - h'\mathbf{M}(t) \times (\mathbf{M}(t) \times \mathbf{H}_{ef}(t)) \\ &\quad + \mathbf{F}(t)H(t) + \mathbf{L}(t)\end{aligned} \tag{159}$$

where

$$\begin{aligned}\mathbf{F}(t) &= g'M_s\mathbf{M}(t) \times \mathbf{e} - h'\mathbf{M}(t) \times (\mathbf{M}(t) \times \mathbf{e}) \qquad \mathbf{e} = \frac{\mathbf{H}(t)}{H(t)} \\ \mathbf{L}(t) &= g'M_s\mathbf{M}(t) \times \mathbf{h}(t) - h'\mathbf{M}(t) \times (\mathbf{M}(t) \times \mathbf{h}(t))\end{aligned} \tag{160}$$

We may further rewrite Eqs. (159) and (160) as

$$\begin{aligned}\frac{d}{dt}M_x(t) &= g'H_{ef}(t)M_sM_y(t) - h'H_{ef}(t)M_x(t)M_z(t) + F_x(t)H(t) \\ &\quad + h'\left(M_s^2 - M_x^2(t)\right)h_x(t) \\ &\quad - \left(g'M_sM_z(t) + h'M_x(t)M_y(t)\right)h_y(t) \\ &\quad + \left(g'M_y(t)M_s - h'M_z(t)M_x(t)\right)h_z(t)\end{aligned} \tag{161}$$

$$\begin{aligned}\frac{d}{dt}M_y(t) &= -g'H_{ef}(t)M_sM_x(t) - h'H_{ef}(t)M_z(t)M_y(t) + F_y(t)H(t) \\ &\quad + h'\left(M_s^2 - M_y^2(t)\right)h_y(t) \\ &\quad - \left(g'M_sM_x(t) + h'M_z(t)M_y(t)\right)h_z(t) \\ &\quad + \left(g'M_sM_z(t) - h'M_x(t)M_y(t)\right)h_x(t)\end{aligned} \tag{162}$$

$$\begin{aligned}\frac{d}{dt}M_z(t) &= h'H_{ef}(t)\left(M_s^2 - M_z^2(t)\right) + F_z(t)H(t) \\ &\quad + h'\left(M_s^2 - M_z^2(t)\right)h_z(t) \\ &\quad - \left(g'M_sM_y(t) + h'M_z(t)M_x(t)\right)h_x(t) \\ &\quad + \left(g'M_sM_x(t) - h'M_z(t)M_y(t)\right)h_y(t)\end{aligned} \tag{163}$$

where $F_i(t)$ and $L_i(t)$ are the projections of the vectors $\mathbf{F}(t)$ and $\mathbf{L}(t)$ onto the x, y, and z axes. These are the Langevin equations for the problem under consideration.

It is evident that Eqs. (161)–(163) have the form of the set of nonlinear stochastic equations:

$$\frac{d}{dt'}M_i(t') = \Phi_i(\{\mathbf{M}(t')\}, t') + g_{ik}(\{\mathbf{M}(t')\}, t')h_k(t') \qquad (i = x, y, z) \tag{164}$$

$$\overline{h_k(t)} = 0 \qquad \overline{h_k(t)h_m(t')} = \delta_{km}\mu\delta(t - t') \tag{165}$$

where summation over k is understood and $\mu = 2kT\eta/\nu$.

Equation (164) contains multiplicative noise terms $g_{ik}(\{\mathbf{M}(t')\}, t')h_k(t')$. This poses an interpretation problem as discussed in Section II.B.

We can use Eq. (39) to evaluate the statistical average of the multiplicative noise terms in Eqs. (161)–(165). In our case the stochastic variables are $\xi_1(t') = M_x(t')$, $\xi_2(t') = M_y(t')$, $\xi_3(t') = M_z(t')$. Thus, on averaging Eqs. (161)–(163) we may obtain the equations of motion for the starting values of the components $M_i = M_i(t)$, $(i = x, y, z)$ of the magnetization, namely[11]

$$\frac{d}{dt}M_x = g'H_{ef}M_sM_y - h'H_{ef}M_xM_z + F_xH(t) + \frac{\mu}{2}\left(g_{jk}\frac{\partial}{\partial M_j}g_{1k}\right) \tag{166}$$

$$\frac{d}{dt}M_y = -g'H_{ef}M_sM_x - h'H_{ef}M_yM_z + F_yH(t) + \frac{\mu}{2}\left(g_{jk}\frac{\partial}{\partial M_j}g_{2k}\right) \tag{167}$$

$$\frac{d}{dt}M_z = h'H_{ef}(M_s^2 - M_z^2) + F_zH(t) + \frac{\mu}{2}\left(g_{jk}\frac{\partial}{\partial M_j}g_{3k}\right) \tag{168}$$

The evaluation of the noise induced drift $(\mu/2)[g_{jk}(\partial/\partial M_j)g_{ik}]$ leads to[11]

$$\frac{\mu}{2}g_{jk}\frac{\partial}{\partial M_j}g_{ik} = -\mu(g'^2 + h'^2)M_s^2M_i = -\tau_N^{-1}M_i \tag{169}$$

where $\tau_N = \nu/(2kTh')$ is the relaxation time for $H_{ef} = 0$, and then to the

equations for the averages $\langle M_i \rangle$:

$$\frac{\mathrm{d}}{\mathrm{d}t}\langle M_x \rangle + \frac{1}{\tau_N}\langle M_x \rangle = g' M_s \langle H_{ef} M_y \rangle - h' \langle H_{ef} M_x M_z \rangle + \langle F_x \rangle H(t) \tag{170}$$

$$\frac{\mathrm{d}}{\mathrm{d}t}\langle M_y \rangle + \frac{1}{\tau_N}\langle M_y \rangle = -g' M_s \langle H_{ef} M_x \rangle - h' \langle H_{ef} M_y M_z \rangle + \langle F_y \rangle H(t) \tag{171}$$

$$\frac{\mathrm{d}}{\mathrm{d}t}\langle M_z \rangle + \frac{1}{\tau_N}\langle M_z \rangle = h' \langle H_{ef}(M_s^2 - M_z^2) \rangle + \langle F_z \rangle H(t) \tag{172}$$

These equations govern the Néel relaxation of a single-domain ferromagnetic particle. They resemble Eqs. (120)–(122) for the Debye relaxation of a nematic liquid crystal. They differ from the nematic equations, however, in so far as they contain precessional terms $g' M_s \langle H_{ef} M_x \rangle$ and $g' M_s \langle H_{ef} M_y \rangle$, which are absent from the nematic equations. The precessional terms are responsible for ferromagnetic resonance which is due to precessional motion of the magnetization $\mathbf{M}(t)$ in the anisotropy field $\mathbf{H}_{ef}$.

Equations (170)–(172) (v. Section VII of Ref. 11) are the first terms in an infinite hierarchy of differential equations. It is obvious that to solve these equations, we must also obtain equations for $\langle H_{ef} M_x \rangle$, $\langle H_{ef} M_y M_z \rangle$, $\langle H_{ef} M_y \rangle$, $\langle H_{ef} M_x M_z \rangle$, $\langle H_{ef} \rangle$, $\langle H_{ef} M_z^2 \rangle$ and so on. This difficulty may also be circumvented by means of the effective eigenvalue method.

On using this method we have shown in Ref. 11 that the longitudinal ($\tau_\parallel$) and transverse ($\tau_\perp$) magnetization relaxation times may be expressed in terms of the equilibrium averages $\langle \cos \vartheta \rangle_0$ and $\langle \cos^2 \vartheta \rangle_0$. We have

$$\tau_\parallel = 2\tau_N \frac{\langle \cos^2 \vartheta \rangle_0 - \langle \cos \vartheta \rangle_0^2}{1 - \langle \cos^2 \vartheta \rangle_0} \tag{173}$$

$$\tau_\perp = 2\tau_N \frac{1 - \langle \cos^2 \vartheta \rangle_0}{1 + \langle \cos^2 \vartheta \rangle_0} \tag{174}$$

Equations (173) and (174) for the relaxation times $\tau_\parallel$ and $\tau_\perp$ are valid for any axially symmetric potential of the crystalline anisotropy. We also note that the term $\langle \cos \vartheta \rangle_0$ in Eq. (176) appears due to the fact that the effective field H_{eff} may also include an applied constant magnetic field $\mathbf{H}_0$.

In the simplest uniaxial case, the crystalline anisotropy energy density is $K \sin^2 \vartheta$ ($K > 0$), where ϑ is the angle between $\mathbf{M}$ and the positive z

axis.[33] In the absence of the external field $\mathbf{H}_0 = 0$, so that for relaxation in the purely crystalline potential,

$$V(\vartheta) = -K\cos^2\vartheta + K \tag{175}$$

which is entirely analogous to the Meier-Saupe nematic potential, we have

$$\langle\cos\vartheta\rangle_0 = 0 \tag{176}$$

and

$$\langle\cos^2\vartheta\rangle_0 = \frac{1}{2\sqrt{\sigma}\,D(\sqrt{\sigma})} - \frac{1}{2\sigma}$$

or

$$\langle\cos^2\vartheta\rangle_0 = \frac{F'(\sigma)}{F(\sigma)} \tag{177}$$

where $\sigma = \nu K/kT$, $D(x)$ is the Dawson integral Eq. (72), and $F(\sigma)$ is defined by Eq. (69). The symbol $\langle f\rangle_0$ now means

$$\langle f\rangle_0 = \frac{\int_0^\pi f(\vartheta)\mathrm{e}^{-\nu V(\vartheta)/kT}\sin\vartheta\,\mathrm{d}\vartheta}{\int_0^\pi \mathrm{e}^{-\nu V(\vartheta)/kT}\sin\vartheta\,\mathrm{d}\vartheta} \tag{178}$$

According to Eqs. (173) and (174) we then have[11]

$$\tau_{\|} = 2\tau_{\mathrm{N}}\frac{\sqrt{\sigma} - D(\sqrt{\sigma})}{D(\sqrt{\sigma})(1+2\sigma) - \sqrt{\sigma}} = 2\tau_{\mathrm{N}}\frac{F'(\sigma)}{F(\sigma) - F'(\sigma)} \tag{179}$$

$$\tau_{\perp} = 2\tau_{\mathrm{N}}\frac{D(\sqrt{\sigma})(2\sigma+1) - \sqrt{\sigma}}{D(\sqrt{\sigma})(2\sigma-1) + \sqrt{\sigma}} = 2\tau_{\mathrm{N}}\frac{F(\sigma) - F'(\sigma)}{F(\sigma) + F'(\sigma)} \tag{180}$$

Equations (179) and (180), that is, the magnetization relaxation times $\tau_{\|}$ and $\tau_{\perp}$ coincide precisely with the dielectric relaxation times of a nematic obtained in Sections II.D and II.E (if we put $\tau_{\mathrm{N}} = \tau_{\mathrm{D}}$). They also coincide with results of Refs. 6 and 11.

Just as in Section II.D, Eq. (179) allows us to calculate from Eq. (172) the longitudinal susceptibility $\chi_{\|}(\omega)$ of the particle, which is given by[6]:

$$\chi_{\|}(\omega) = \frac{\chi'_{\|}(0)}{1 + \mathrm{i}\omega\tau_{\|}} \tag{181}$$

where[11]

$$\chi_{\parallel}(0) = \frac{\nu}{kT}\langle M_z^2\rangle_0 = \frac{\nu M_s^2}{kT}\frac{F'(\sigma)}{F(\sigma)}$$

is the static longitudinal susceptibility. Calculations of the transverse susceptibility $\chi_\perp(\omega)$ (when the probe field $\mathbf{H}(t)$ is applied along the x axis) have been given in the context of the Fokker-Planck equation approach in Ref. 6. We give an equivalent derivation based on the Langevin equation. As shown in Appendix B the average equations of motion when expressed in terms of spherical harmonics are for the dipole component $\langle X_{11}\rangle$

$$\frac{d}{dt}\langle X_{11}\rangle + \frac{1}{\tau_N}\left(1+\frac{\sigma}{5}\right)\langle X_{11}\rangle + \frac{2\sigma}{15\tau_N}\langle X_{31}\rangle - \frac{\sigma}{3\alpha\tau_N}\langle X_{2,-1}\rangle = \frac{\xi(t)}{6\tau_N}\left[2 - \tfrac{1}{2}\langle X_{22}\rangle + \langle P_2\rangle\right] \tag{182}$$

and for the quadrupole component $\langle X_{2,-1}\rangle$

$$\begin{aligned}\frac{d}{dt}\langle X_{2,-1}\rangle &+ \frac{1}{\tau_N}\left(3-\frac{\sigma}{7}\right)\langle X_{2,-1}\rangle + \frac{2\sigma}{5\alpha\tau_N}\langle X_{31}\rangle \\ &+ \frac{3\sigma}{5\alpha\tau_N}\langle X_{11}\rangle + \frac{12\sigma}{35\tau_N}\langle X_{4,-1}\rangle \\ &= \frac{\xi(t)}{\tau_N}\left[\frac{6}{5}\langle P_1\rangle + \frac{3}{10}\langle P_3\rangle - \frac{1}{20}\left(\langle X_{32}\rangle + \langle X_{3,-2}\rangle\right)\right. \\ &\qquad \left. - \frac{1}{2\alpha}\left(1 - \langle P_2\rangle - \frac{1}{2}\langle X_{22}\rangle\right)\right]\end{aligned} \tag{183}$$

where $\alpha = h'/g' = \gamma M_s\eta$ is the dimensionless damping parameter (for all ferromagnets $\alpha \ll 1$ [6]),

$$\xi(t) = \frac{\nu M_s H(t)}{kT}$$

$$\langle X_{nm}\rangle = \langle P_n^{|m|}\cos m\phi\rangle, \langle X_{n,-m}\rangle = \langle P_n^{|m|}\sin m\phi\rangle \qquad m \le n$$

and $P_n^{|m|}$ are the associated Legendre functions of order n. These are the exact average equations of motion.

To determine the frequency dependence of the transverse susceptibility $\chi_\perp(\omega)$ let us now suppose that a small constant magnetic field $\mathbf{H} = \mathbf{i}H$ (where $\mathbf{i}$ denotes the transverse x direction) has been applied to the system for a long time and that it is suddenly switched off at time $t = 0$. Our task is to calculate the one-sided Fourier transform of the magnetization decay function $f(t)$ defined as

$$f_\perp(t) = \frac{M_s\langle X_{11}\rangle}{H} \tag{184}$$

which according to linear response theory[49] allows us to calculate the transverse susceptibility $\chi_\perp(\omega)$ as follows:

$$\chi_\perp(\omega) = f_\perp(0) - \mathrm{i}\omega\int_0^\infty f_\perp(t)\mathrm{e}^{-\mathrm{i}\omega t}\,\mathrm{d}t \tag{185}$$

The effective eigenvalue procedure now reduces Eqs. (182) and (183) for $t > 0$ to

$$\frac{\mathrm{d}}{\mathrm{d}t}\langle X_{11}\rangle + \frac{1}{\tau_\perp}\langle X_{11}\rangle - \frac{\sigma}{3\alpha\tau_N}\langle X_{2,-1}\rangle = 0 \tag{186}$$

$$\frac{\mathrm{d}}{\mathrm{d}t}\langle X_{2,-1}\rangle + \frac{1}{\tau_2}\langle X_{2,-1}\rangle + \frac{3}{\alpha}\left(\frac{1}{\tau_\perp} - \frac{1}{\tau_N}\right)\langle X_{11}\rangle = 0 \tag{187}$$

with initial conditions

$$\langle X_{11}\rangle \cong \frac{\nu M_s H}{kT}\langle \sin^2\vartheta\cos^2\phi\rangle_0 = \frac{\nu M_s H}{2kT}\left(\frac{F(\sigma) - F'(\sigma)}{F(\sigma)}\right) = \frac{\chi'_\perp(0)H}{M_s} \tag{188}$$

$$\langle X_{2,-1}\rangle = 0 \tag{189}$$

where $\chi'_\perp(0)$ is the static transverse susceptibility.

At the derivation of Eq. (187) from Eq. (183) we have taken into account that in accordance with Eqs. (182) and (186)

$$\langle X_{31}\rangle = \frac{15}{2\sigma}\left[\frac{\tau_N}{\tau_\perp} - 1 - \frac{\sigma}{5}\right]\langle X_{11}\rangle$$

The effective relaxation time $\tau_\perp$ has been calculated before[6, 11] and is

given by Eq. (180). The second effective relaxation time τ_2 is evaluated as follows. In the absence of the probe field $\mathbf{H}(t)$, $\langle X_{2,-1}\rangle$ is proportional in the time domain to the equilibrium correlation function $C_{2,-1}(t)$, defined as

$$C_{2,-1}(t) = \langle X_{2,-1}(0) X_{2,-1}(t)\rangle_0$$

Therefore, we have

$$\tau_2 = -\frac{\langle X_{2,-1}\rangle}{(\mathrm{d}/\mathrm{d}t)\langle X_{2,-1}\rangle} = -\frac{C_{2,-1}(0)}{\dot{C}_{2,-1}(0)} \tag{190}$$

The equation for $\dot{C}_{2,-1}(0)$ can be obtained from Eq. (B.13) for the starting value of $M_y H_{\mathrm{ef}} = (2K/3M_s)X_{2,-1}$ at $H(t) = 0$ on multiplying this equation by $X_{2,-1}(0)$ and averaging over the equilibrium distribution function at $t = 0$. The result is

$$\dot{C}_{2,-1}(0) + \frac{1}{\tau_N}\left(3 - \frac{\sigma}{7}\right)C_{2,-1}(0) = -\frac{12\sigma}{35\tau_N}\langle X_{2,-1}(0) X_{4,-1}(0)\rangle_0 \tag{191}$$

Thus, we have from Eqs. (190) and (191)

$$\begin{aligned}\tau_2 &= \tau_N \frac{\langle X_{2,-1}^2\rangle_0}{(3-\sigma/7)\langle X_{2,-1}^2\rangle_0 + (12/35)\sigma\langle X_{4,-1}X_{2,-1}\rangle_0} \\ &= 2\tau_N\left(3 - \sigma + 2\sigma\frac{F''(\sigma) - F'''(\sigma)}{F'(\sigma) - F''(\sigma)}\right) \\ &= 2\tau_N\left(\sigma - 2 - \frac{2\sqrt{\sigma} - 2D(\sqrt{\sigma})}{3\sqrt{\sigma} - (3+2\sigma)D(\sqrt{\sigma})}\right)\end{aligned} \tag{192}$$

Equation (192) coincides precisely with Eq. (34) of Ref. 6.

On applying the one-sided Fourier transform to Eqs. (186) and (187) at $t > 0$ and taking into account Eqs. (188) and (189), we obtain

$$\left(\mathrm{i}\omega + \frac{1}{\tau_\perp}\right)\langle \tilde{X}_{11}\rangle - \frac{\sigma}{3\alpha\tau_N}\langle \tilde{X}_{2,-1}\rangle = \frac{\chi_\perp'(0)H}{M_s} \tag{193}$$

$$\frac{3}{\alpha}\left(\frac{1}{\tau_\perp} - \frac{1}{\tau_N}\right)\langle \tilde{X}_{11}\rangle + \left(\mathrm{i}\omega + \frac{1}{\tau_2}\right)\langle \tilde{X}_{2,-1}\rangle = 0 \tag{194}$$

where the one-sided Fourier transforms $\langle \tilde{X}_{nm} \rangle$ are defined as

$$\langle \tilde{X}_{nm} \rangle = \int_0^\infty e^{-i\omega t} \langle X_{nm} \rangle \, dt$$

On solving these equations and using Eqs. (184) and (185) we obtain

$$\tilde{f}(\omega) = \frac{M_s \langle \tilde{X}_{11} \rangle}{H} = \frac{\chi_\perp(0)(i\omega + 1/\tau_2)}{(i\omega + 1/\tau_2)(i\omega + 1/\tau_\perp) + \dfrac{\sigma}{\tau_N \alpha^2}\left(\dfrac{1}{\tau_\perp} - \dfrac{1}{\tau_N}\right)} \tag{195}$$

$$f(0) = \chi'_\perp(0)$$

Thus, on using Eq. (185) we obtain the transverse susceptibility

$$\frac{\chi_\perp(\omega)}{\chi'_\perp(0)} = \frac{(1 + i\omega\tau_2) + \Delta}{(1 + i\omega\tau_2)(1 + i\omega\tau_\perp) + \Delta} \tag{196}$$

where

$$\Delta = \frac{\sigma\tau_2(\tau_N - \tau_\perp)}{\alpha^2 \tau_N^2}$$

Equation (196) is in full agreement with Eq. (37) of Ref. 6, as may be seen by substituting for τ_2, τ_N, and Δ. For small σ Eq. (196) reduces to Eq. (7.67) of Ref. 11. In the limit of high damping when the dimensionless damping constant $\alpha = \gamma\eta M_s$ becomes very large, Eq. (196) reduces to the purely relaxation equation

$$\chi_\perp(\omega) = \frac{\chi'_\perp(0)}{1 + i\omega\tau_\perp} \tag{197}$$

In the other important limit, $\sigma \to \infty$ (high potential barrier) when $\tau_\perp, \tau_2 \cong \tau_N/\sigma$, Eq. (196) yields the Landau-Lifshitz formula for ferromagnetic resonance[36]:

$$\chi_\perp(\omega) = \gamma M_s \frac{(1 + \alpha^2)\omega_0 + i\alpha\omega}{(1 + \alpha^2)\omega_0^2 - \omega^2 + 2i\alpha\omega\omega_0} \tag{198}$$

where

$$\omega_0 = \frac{\sigma\gamma^2}{\tau_N \alpha} = \gamma H_A \tag{199}$$

is the frequency of precession of the vector **M** in the anisotropy field $H_A = 2K/M$ and $(\alpha\omega_0)^{-1}$ is a characteristic relaxation time. A detailed investigation of the variation with frequency of $\chi_\parallel(\omega)$ and $\chi_\perp(\omega)$ and the conditions of applicability of the results is given in Ref. 6.

I. Calculation of the Dynamic Orientation Factor for the Kerr Effect Response of Noninteracting Dipolar Molecules

The analysis so far has been concerned with the linear dielectric response of a nematic liquid crystal in the presence of an arbitrary uniaxial potential of the crystalline anisotropy and with the analogous magnetic problem. Another application in the theory of dielectrics is to the Kerr effect response following the application of a dc electric field to an assembly of noninteracting dipolar molecules, each having a permanent dipole moment $\boldsymbol{\mu}$. This problem is distinguished from the linear dielectric response[12] in that two effective eigenvalues are needed to characterize the Kerr effect response.

Under the influence of a strong electric field **E**, an isotropic fluid becomes optically anisotropic or birefringent; i.e., the refractive index $n_\parallel$ for a light wave with its electric field parallel to the applied field **E** differs from the refractive index $n_\perp$ for a light wave with its electric field vector perpendicular to this direction. The quantity that characterizes the dynamic Kerr effect is the orientation factor[14]

$$\Phi(t) = \langle P_2(\cos\vartheta(t))\rangle \tag{200}$$

which determines the dynamic birefringence

$$\Delta n = n_\parallel - n_\perp = K_n \Phi(t)$$

where K_n is the Kerr constant. We illustrate the calculation of this quantity by the effective eigenvalue method for the case of a strong dc field **E**, which is applied to an assembly of noninteracting rigid dipolar molecules. This yields the rise transient of the Kerr effect.[14]

Again starting from the vector Langevin equation for the rotational. Brownian motion of a linear molecule in the field **E** [24] (c.f. Eq. (28)),

$$\frac{\mathrm{d}}{\mathrm{d}t}\boldsymbol{\mu}(t) = \frac{1}{\zeta}[\boldsymbol{\lambda}(t) \times \boldsymbol{\mu}(t) + (\boldsymbol{\mu}(t) \times \mathbf{E}) \times \boldsymbol{\mu}(t)] \tag{201}$$

we find by direct averaging of this equation in the manner of the previous section that (for details see Ref. 24, where we have considered dielectric relaxation of a polar fluid under the influence of a constant field **E**)

$$\frac{\mathrm{d}}{\mathrm{d}t}\langle P_1\rangle + \frac{1}{\tau_D}\langle P_1\rangle = \frac{\xi}{3\tau_D}(1 - \langle P_2\rangle) \tag{202}$$

$$\frac{\mathrm{d}}{\mathrm{d}t}\langle P_2\rangle + \frac{3}{\tau_D}\langle P_2\rangle = \frac{3\xi}{5\tau_D}(\langle P_1\rangle - \langle P_3\rangle) \tag{203}$$

where

$$\xi = \frac{\mu E}{kT} \tag{204}$$

is the normalized applied field giving rise to a potential

$$V(\vartheta) = -\mu E \cos\vartheta \tag{205}$$

and

$$\tau_D = \frac{\zeta}{2kT} \tag{206}$$

is the Debye relaxation time.

Assuming that the assembly of rotators is initially unpolarized so that at $t = 0$, $\langle P_1\rangle = \langle P_2\rangle = 0$, we obtain with the help of the Laplace transformation

$$\frac{\mathscr{L}\langle P_n\rangle}{\mathscr{L}\langle P_{n-1}\rangle} = (\xi/2)[n(n+1)/(2n+1)] \times \left\{\tau_D s + n(n+1)/2 + (\xi/2)[n(n+1)/(2n+1)]\frac{\mathscr{L}\langle P_{n+1}\rangle}{\mathscr{L}\langle P_n\rangle}\right\}^{-1} \qquad n = 1, 2 \tag{207}$$

Equation (207) is valid for any n and has been obtained from the underlying Fokker-Planck equation in previous investigations (see, e.g., Refs. 7 and 14). For $n = 1$, Eq. (207) is

$$\frac{\mathscr{L}\langle P_1\rangle}{\mathscr{L}\langle P_0\rangle} = \frac{\xi/3}{s\tau_D + 1 + (\xi/3)(\mathscr{L}\langle P_2\rangle/\mathscr{L}\langle P_1\rangle)} \tag{208}$$

If we now suppose, following the approach of Section II.D, that the term $\mathscr{L}\langle P_2\rangle/\mathscr{L}\langle P_1\rangle$ may be replaced by its final equilibrium value, Eq. (208) may be evaluated in terms of a single effective relaxation time, since we

have[24] (denoting in this instance final equilibrium values by $\langle P_n \rangle_\xi$)

$$\langle P_1 \rangle_\xi = L(\xi) \tag{209}$$

$$\langle P_2 \rangle_\xi = 1 - \frac{3L(\xi)}{\xi} \tag{210}$$

so that

$$\mathscr{L}\langle P_1 \rangle = \frac{\xi}{3s(s\tau_D + \xi/3L(\xi))} \tag{211}$$

so that the rise transient has the effective time constant

$$\tau_{\text{eff}} = \frac{3L(\xi)\tau_D}{\xi} \tag{212}$$

in agreement with the result of Morita.[7] Since we have supposed that in the infinite continued fraction (Eq. (207)) that the second-order Legendre polynomial (describing the dynamic Kerr effect average) has reached its equilibrium value, it is necessary to proceed further down the continued fraction in order to calculate the rise transient of the dynamic Kerr effect. We have from Eq. (207)

$$\frac{\mathscr{L}\langle P_2 \rangle}{\mathscr{L}\langle P_1 \rangle} = \frac{3\xi/5}{s\tau_D + 3(1 + (\xi/5)(\mathscr{L}\langle P_3 \rangle / \mathscr{L}\langle P_2 \rangle))} \tag{213}$$

We now again truncate by supposing that the ratio $\langle P_3 \rangle / \langle P_2 \rangle$ has reached its equilibrium value. The simplest way to calculate this ratio is to recall that at equilibrium

$$\langle P_2 \rangle_\xi = \frac{\xi}{5}\left[\langle P_1 \rangle_\xi - \langle P_3 \rangle_\xi\right] \tag{214}$$

so that

$$\frac{\langle P_1 \rangle_\xi}{\langle P_2 \rangle_\xi} - \frac{5}{\xi} = \frac{\langle P_3 \rangle_\xi}{\langle P_2 \rangle_\xi} \tag{215}$$

And recalling that

$$\frac{\langle P_1 \rangle_\xi}{\langle P_2 \rangle_\xi} = \frac{L(\xi)}{1 - 3L(\xi)/\xi} \tag{216}$$

we have

$$\frac{\langle P_3\rangle_\xi}{\langle P_2\rangle_\xi} = \frac{\xi^2 L(\xi) - 5[\xi - 3L(\xi)]}{\xi[\xi - 3L(\xi)]} \tag{217}$$

so that at this level of truncation

$$\begin{aligned}\frac{\mathscr{L}\langle P_2\rangle}{\mathscr{L}\langle P_1\rangle} &= \frac{3\xi/5}{s\tau_D + \frac{3}{5}\{[\xi^2 L(\xi) - 5(\xi - 3L(\xi))]/(\xi - 3L(\xi))\}} \\ &= \frac{3\xi/5}{s\tau_D + 3\xi^2 L(\xi)/(\xi - 3L(\xi))}\end{aligned} \tag{218}$$

Thus,

$$\mathscr{L}\langle P_2\rangle = \frac{3\xi}{5\tau_D}\frac{\mathscr{L}\langle P_1\rangle}{s + (3/\tau_D)[\xi^2 L(\xi)/(\xi - 3L(\xi))]} \tag{219}$$

and on using Eq. (195)

$$\mathscr{L}\langle P_2\rangle = \frac{\frac{1}{5}(\xi^2/\tau_D^2)}{s(s + \xi/L(\xi)\tau_D)\{s + (3/\tau_D)[\xi^2 L(\xi)/(\xi - 3L(\xi))]\}} \tag{220}$$

Equation (220) shows clearly that two effective eigenvalues λ_1 and λ_2, namely

$$\lambda_1 = \frac{\xi}{L(\xi)\tau_D} \qquad \lambda_2 = \frac{3}{\tau_D}\left(\frac{\xi^2 L(\xi)}{\xi - 3L(\xi)}\right) \tag{221}$$

are needed to describe the transient Kerr effect response due to the application of a strong dc field. This point was first emphasized by Morita and Watanabe[14] who derived Eq. (20) in their review of the dynamic Kerr effect. Calculations performed by them suggest that Eq. (220) provides a satisfactory approximation to the response computed by taking successive convergents of the continued fraction—Eq. (207). The calculation described here is valuable as yet another simple example of a situation where two effective eigenvalues are required to describe the response. The procedure may be continued by iteration to obtain the cubic term in the dielectric response. We note that we are calculating the response to a strong dc field; thus, there is no connection between any of the above formulae and the ac response, unlike the nematic liquid crystal problem.

J. Comparison of Effective and Longest Relaxation Times

It is important to compare the relaxation times yielded by Eqs. (138) and (146) with the numerical and asymptotic results for the largest relaxation time. We consider the simplest potential of uniaxial anisotropy, namely

$$\frac{V}{kT} = \sigma(1 - \cos^2 \vartheta)$$

For a longitudinal applied field Brown[15] has shown that the longest relaxation time in the limit of high potential barriers, $\sigma \gg 1$, is well approximated by

$$\tau_{\parallel} \simeq \tau_D \frac{\sqrt{\pi}}{2} \sigma^{-3/2} \exp \sigma \tag{222}$$

For small potential barriers, $\sigma \ll 1$, on the other hand, one may show by perturbation theory[15] that

$$\tau_{\parallel} \simeq \frac{\tau_D}{1 - \frac{2}{5}\sigma + 48\sigma^2/875} \tag{223}$$

The exact solution for all the relaxation times and relaxation modes may be found by numerical solution of the set of differential-difference equations:

$$\begin{aligned} \frac{2\tau_D}{n(n+1)} \langle \dot{P}_n \rangle + \left[1 - \frac{2\sigma}{(2n-1)(2n+3)} \right] \langle P_n \rangle \\ = \frac{2\sigma}{2n+1} \left[\frac{n-1}{2n-1} \langle P_{n-2} \rangle - \frac{n+2}{2n+3} \langle P_{n+2} \rangle \right] \\ \langle P_n \rangle = \langle P_n(\cos \vartheta(t)) \rangle \qquad t > 0 \end{aligned} \tag{224}$$

These govern the linear response of the system following the removal of a steady dc field at time $t = 0$. The set of Eqs. (224) may be written either by successively averaging the Langevin equation as described here and in Appendix A or by separating the variables in the Fokker-Planck equation, as described in Section III of Ref. 11. The Fokker-Planck equation is [11]

$$2\tau_D \sin \vartheta \frac{\partial W}{\partial t} = \frac{\partial}{\partial \vartheta} \left[\sin \vartheta \left(\frac{v}{kT} \frac{\partial V}{\partial \vartheta} W + \frac{\partial W}{\partial \vartheta} \right) \right] \tag{225}$$

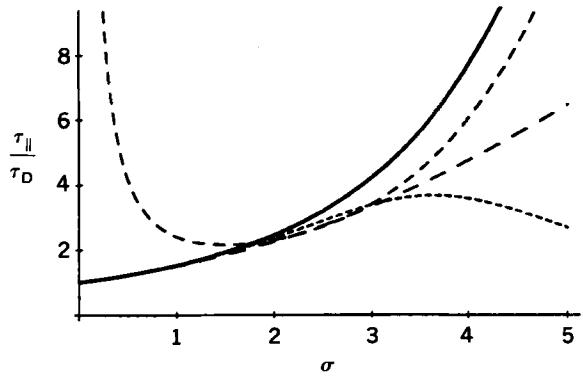


Figure 3. Longest relaxation time (solid line) from Eq. (224), Brown's asymptotic expression Eq. (222) (--- line), and perturbation solution Eq. (223) (------ line) compared with relaxation time rendered by the effective eigenvalue (——— line) Eq. (138) as a function of the barrier height parameter σ.

The differential-difference equations (224) are solved by writing them as

$$\dot{\mathbf{X}} = \mathbf{A}\mathbf{X} \tag{226}$$

where (it will only be necessary to consider the vector of odd averages)

$$\mathbf{X} = (\langle P_1 \rangle, \langle P_3 \rangle, \ldots, \langle P_{2n+1} \rangle)^T \tag{227}$$

and forming the characteristic equation

$$\det(s\mathbf{I} - \mathbf{A}) = 0 \tag{228}$$

The longest relaxation time is proportional to the reciprocal of the lowest root of this equation, the size of the matrix being adjusted until convergence is obtained. Satisfactory results are usually obtained by taking $\mathbf{A}$ as 20×20. In Figs. 3 and 4 we show the results for the longest relaxation time yielded by the three Eqs. (222), (223), and (224) compared with the effective relaxation time rendered by Eq. (138). It is evident that $\tau_{\|}$ as rendered by Eq. (138) provides a poor approximation to the longest relaxation time in the limit of high potential barriers. This is a direct consequence of the exponential character of the longest relaxation time (causing it to dominate all the others in the high σ limit). The effective eigenvalue provides a poor approximation in the high σ limit because it depends on the equilibrium averages, which have *no exponential terms*.

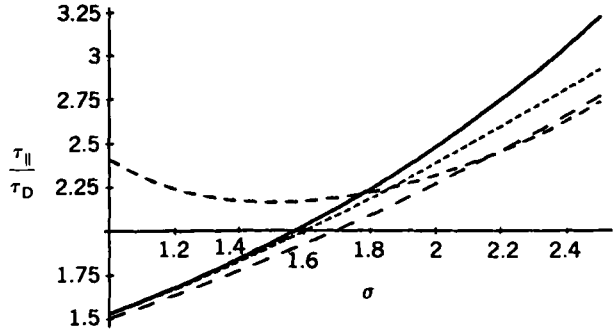


Figure 4. Magnification of Fig. 3.

The relaxation modes of $\langle P_1 \rangle$ may be found from Eq. (224) by assuming that **A** has a linearly independent set of n eigenvectors $(\mathbf{p}_1 \cdots \mathbf{p}_n)$, so that

$$\mathbf{P}^{-1}\mathbf{A}\mathbf{P} = \mathbf{\Lambda} \tag{229}$$

where **P** is the matrix consisting entirely of the n eigenvectors of **A** and

$$\mathbf{\Lambda} = \begin{pmatrix} \lambda_1 & & & \\ & \lambda_2 & & 0 \\ & & \lambda_3 & \\ & 0 & & \ddots \end{pmatrix} \tag{230}$$

whence

$$\mathbf{X}(t) = \mathbf{P} \exp \mathbf{\Lambda} t \mathbf{P}^{-1} \mathbf{X}_0 \tag{231}$$

where

$$\exp \mathbf{\Lambda} t = \begin{pmatrix} e^{\lambda_1 t} & & & \\ & e^{\lambda_2 t} & & 0 \\ 0 & & \ddots & \\ & & & e^{\lambda_n t} \end{pmatrix}$$

and $\mathbf{X}_0$ is the matrix of the initial values of the $\langle P_n \rangle$. The solution, Eq. (231), may then be exhibited in the form

$$\mathbf{X}(t) = b_1 e^{\lambda_1 t}\mathbf{p}_1 + b_2 e^{\lambda_2 t}\mathbf{p}_2 + \cdots + b_n e^{\lambda_n t}\mathbf{p}_n \tag{232}$$

where the b_n are to be determined from the initial conditions. The initial value vector $\mathbf{X}_0$ is determined as follows. We have at time $t = 0$ when the steady field $\mathbf{E}_1(t)$ is switched off

$$\langle P_1(\cos\vartheta)\rangle = \frac{\int_{-1}^{+1} x\, e^{\sigma x^2+\xi_1 x}\, dx}{\int_{-1}^{+1} e^{\sigma x^2+\xi_1 x}\, dx} \tag{233}$$

which in the linear approximation is

$$\begin{aligned}\langle P_1(x)\rangle &= \frac{\int_{-1}^{+1} x\, e^{\sigma x^2}[1+\xi_1 x]\, dx}{\int_{-1}^{+1} e^{\sigma x^2}[1+\xi_1 x]\, dx} = \xi_1 \frac{\int_0^1 x^2 e^{\sigma x^2}\, dx}{\int_0^1 e^{\sigma x^2}\, dx} \\ &= \xi_1\langle x^2\rangle_0 = \frac{\xi_1}{3}[1+2\langle P_2\rangle_0]\end{aligned} \tag{234}$$

In general, we will have

$$\langle P_n(x)\rangle = \frac{\int_{-1}^{+1} P_n(x) e^{\sigma x^2}\, dx}{\int_{-1}^{+1} e^{\sigma x^2}\, dx} + \xi_1 \frac{\int_{-1}^{+1} x P_n(x) e^{\sigma x^2}\, dx}{\int_{-1}^{+1} e^{\sigma x^2}\, dx} \tag{235}$$

Now

$$xP_n(x) = \frac{1}{2n+1}[(n+1)P_{n+1}(x) + nP_{n-1}(x)] \tag{236}$$

so that for even $n = 2k$, say,

$$\langle P_{2k}\rangle = \langle P_{2k}\rangle_0$$

Thus, the even averages will not be represented in the initial value vector $\mathbf{X}_0$. For the averages of odd functions

$$\langle P_{2k+1}\rangle = \xi_1 \frac{\int_{-1}^{+1}[1/(4k+3)][(2k+2)P_{2k+2} + (2k+1)P_{2k}] e^{\sigma x^2}\, dx}{\int_{-1}^{+1} e^{\sigma x^2}\, dx} \tag{237}$$

so that

$$\langle P_{2k+1}\rangle = \frac{\xi_1}{4k+3}[(2k+2)\langle P_{2k+2}\rangle_0 + (2k+1)\langle P_{2k}\rangle_0] \tag{238}$$

Since

$$\begin{aligned}\langle P_2\rangle_0 &= \frac{1}{2}\left[3\langle\cos^2\vartheta\rangle_0 - 1\right] \\ &= \frac{1}{2}\left(\frac{3}{2\sqrt{\sigma}D(\sqrt{\sigma})} - \frac{3}{2\sigma} - 1\right)\end{aligned} \tag{239}$$

We may calculate $\langle P_{2k}\rangle_0$ in terms of Dawson's integral from the recurrence relation (Eq. (224) with $\langle \dot{P}_n\rangle = 0$)

$$\begin{aligned}&\left[1 - \frac{2\sigma}{(2n-1)(2n+3)}\right]\langle P_n\rangle_0 \\ &\quad = \frac{2\sigma}{2n+1}\left[\frac{n-1}{2n-1}\langle P_{n-2}\rangle_0 - \frac{n+2}{2n+3}\langle P_{n+2}\rangle_0\right]\end{aligned} \tag{240}$$

and Eq. (239). Hence we have the elements of the vector $\mathbf{X}_0$. This procedure with the aid of Eq. (232) now yields $\langle P_1\rangle$ in the form

$$\mu\langle P_1\rangle = \frac{\mu^2 E_1}{kT}\sum_{k=1,3,5} a_k\, e^{-\lambda_k t} \tag{241}$$

The amplitudes a_k are given in Table I and are in close agreement with those given by Martin et al.,[16] who used the Sturm-Liouville method to

TABLE I

σ	k	a_k
0.1	1	0.342
	3	0.93×10^{-5}
	5	0.843×10^{-10}
2	1	0.5275
	3	0.36×10^{-2}
	5	0.146×10^{-4}
5	1	0.754
	3	0.9×10^{-2}
	5	0.31×10^{-3}
8	1	0.856
	3	0.52×10^{-2}
	5	0.7×10^{-3}

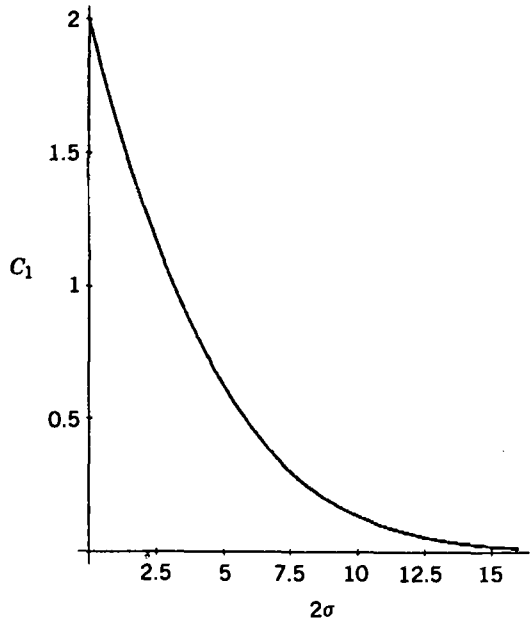


Figure 5. Eigenvalue c_1 as a function of the nematic potential parameter $A = 2\sigma$.

calculate the a_k. The effective eigenvalue

$$\lambda_{\text{eff}} = \frac{\Sigma_k a_k \lambda_k}{\Sigma_k a_k} \tag{242}$$

may now be evaluated from the numerical results presented here. We write in order to utilize the presentation of Martin et al.[16]

$$2\tau_D \lambda_{\text{eff}} = \frac{\Sigma_{k=1,3,5} c_k a_k}{\Sigma_{k=1,3,5} a_k} \tag{243}$$

Here we have supposed that $\lambda_k = c_k(2\tau_D)^{-1}$. For $\sigma = 5$ we have from Fig. 2 of Martin et al.[16] (see also Figs. 5–7)

$$c_1 \simeq 0.1 \qquad c_3 \simeq 13 \qquad c_5 = 31$$

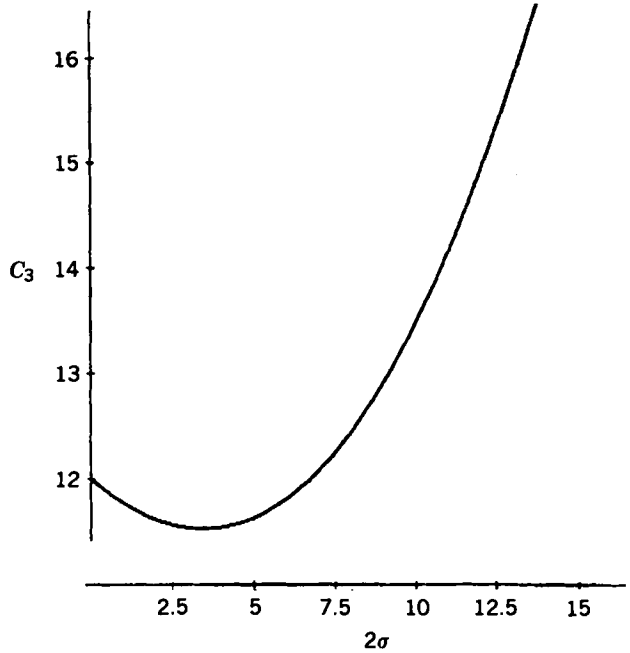


Figure 6. Same as Fig. 5 for c_3.

so that

$$2\tau_D\lambda_{\text{eff}} = \frac{0.754 \times 0.1 + 0.9 \times 10^{-2} \times 13 + 0.32 \times 10^{-4} \times 31}{0.754 + 0.9 \times 10^{-2} + 0.32 \times 10^{-4}} = \frac{0.0754 + \mathbf{0.117} + 0.000992}{0.764} \tag{244}$$

showing that the contribution to λ_{eff} due to the *second* eigenvalue (shown boldface) is large. The result is also in agreement with λ_{eff} as computed from Eq. (138). The reciprocal of the correlation time, on the other hand, is

$$\lambda_{c0} = \frac{1}{\int_0^\infty C(t)\,dt} = \frac{\Sigma_k a_k}{2\tau_D \Sigma_k a_k c_k^{-1}} \tag{245}$$

$$= \frac{1}{2\tau_D} \frac{0.754 + 0.9 \times 10^{-2} + 0.32 \times 10^{-4}}{0.754/0.1 + 0.009/13 + (0.32 \times 10^{-4})/31} \tag{246}$$

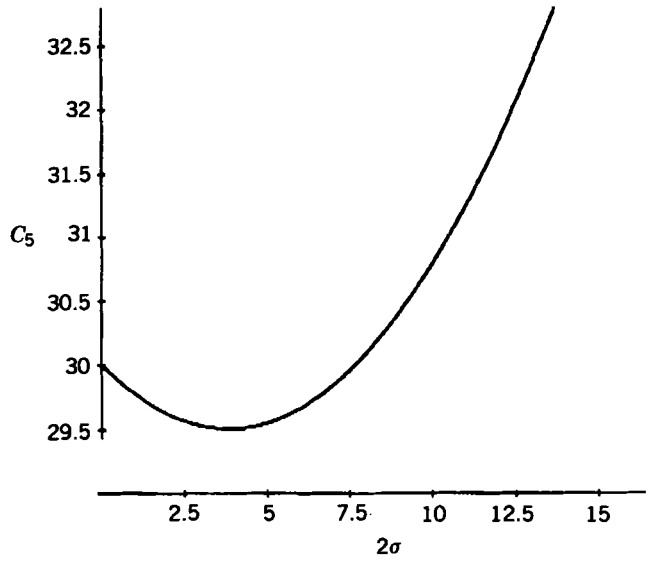


Figure 7. Same as Fig. 5 for c_5.

showing that the contribution of the higher order eigenvalues to the correlation time is minute, so that for the parallel case the relaxation is closely described by a single mechanism, with relaxation time given by Eq. (222) for $\sigma \geq 1.5$. For $\sigma < 1.5$ the effective relaxation time given by Eq. (138) provides an acceptable description. In the transverse case with

$$\frac{V}{kT} = \sigma(1 - \cos^2 \vartheta) \tag{247}$$

an asymptotic formula for the longest relaxation time has been given by Storonkin[30]:

$$\frac{\tau_\perp}{\tau_D} = \sigma^{-1}(1 + \sigma^{-1} + 3\sigma^{-2}) \tag{248}$$

Here there is no exponential behavior of the relaxation time, and the asymptotic formula agrees with the result for $\tau_\perp$ as rendered by λ_{eff} for large σ.

The Fokker-Planck equation following the removal of a transverse field is [11]

$$2\tau_D \frac{\partial W}{\partial t} = \frac{1}{\sin\vartheta}\frac{\partial}{\partial\vartheta}\left(\sin\vartheta\frac{\partial W}{\partial\vartheta}\right) + \frac{1}{\sin^2\vartheta}\frac{\partial^2 W}{\partial\phi^2} + 2\sigma\sin\vartheta\cos\vartheta\frac{\partial W}{\partial\vartheta} + 2\sigma(3\cos^2\vartheta - 1)W \tag{249}$$

which on assuming that

$$W(\vartheta,\phi,t) = \sum_{n=0}^{\infty}\sum_{m=-n}^{n} a_{nm}(t)P_n^{|m|}(\cos\vartheta)e^{im\phi} \qquad |m| \le n \tag{250}$$

results in the set of differential-difference equations [11]:

$$2\tau_D\frac{da_{nm}(t)}{dt} = \left[-n(n+1) + 2\sigma\frac{n(n+1) - 3m^2}{(2n+3)(2n-1)}\right]a_{nm}(t) + 2\sigma\left[\frac{(n-|m|)(n+1)(n-|m|-1)}{(2n-1)(2n-3)}a_{n-2,m} - \frac{n(n+|m|+1)(n+|m|+2)}{(2n+3)(2n+5)}a_{n+2,m}\right] \tag{251}$$

with

$$\left\langle P_n^{|m|}(x)e^{-im\phi}\right\rangle = \frac{1}{2n+1}\frac{(n+|m|)!}{(n-|m|)!}\frac{a_{n,-m}}{a_{00}} \tag{252}$$

$$\left\langle P_n^{|m|}(x)e^{im\phi}\right\rangle = \frac{1}{2n+1}\frac{(n+|m|)!}{(n-|m|)!}\frac{a_{nm}}{a_{00}} \tag{253}$$

$$\left\langle P_n^{|m|}(x)\cos m\phi\right\rangle = \frac{1}{2n+1}\frac{(n+|m|)!}{(n-|m|)!}\frac{a_{nm}+a_{n,-m}}{2a_{00}} \tag{254}$$

$$\left\langle P_n^{|m|}(x)\sin m\phi\right\rangle = \frac{1}{2n+1}\frac{(n+|m|)!}{(n-|m|)!}\frac{a_{nm}-a_{n,-m}}{2ia_{00}} \tag{255}$$

and

$$a_{n,-m} = a_{nm}^* \tag{256}$$

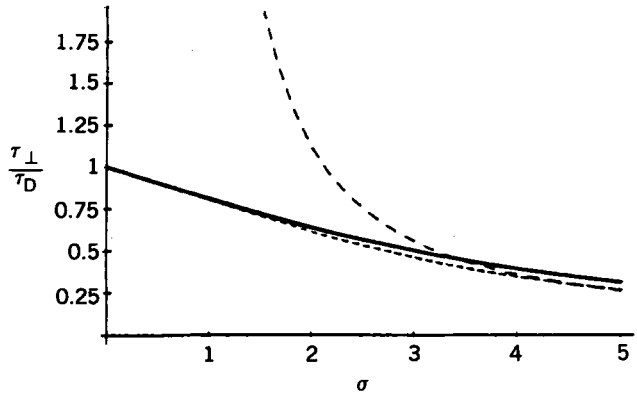


Figure 8. Numerical solution for longest relaxation time for a transverse applied field from Eq. (251) (solid line) compared with Storonkin's asymptotic solution Eq. (248) (dashed line) and compared with the relaxation time rendered by the effective eigenvalue Eq. (146) as a function of σ. Note the close agreement of the effective eigenvalue with the numerical solution for all σ values.

The longest relaxation time of Eq. (251) (shown in Fig. 8) may be found just as for $m = 0$ by forming the characteristic equation

$$\det(s\mathbf{I} - \mathbf{A}) = 0$$

The effective relaxation time is compared with Eq. (248) and that yielded by the exact numerical solution for the longest relaxation time in Fig. 8, showing that the effective relaxation time yields a very close approximation to the longest relaxation time for all values of σ in this case. We would like to thank John Waldron for the numerical solutions presented in this section.

III. TWO-DIMENSIONAL ROTATIONAL BROWNIAN MOTION IN *N*-FOLD COSINE POTENTIALS

A comprehensive numerical study of the rotational Brownian motion of the two-dimensional rotator in N-fold cosine potentials in both time and frequency domains has been given by Reid,[37] with particular reference to the behavior of the dielectric dispersion and absorption spectra. He compares the spectra computed from the model with those obtained from experimental observations of rotator-phase furan and CH_2Cl_2. There is reasonable agreement with experiment. Further, he finds that (unlike the free rotator or the harmonic potential version) this model can reproduce

both relaxation and resonant (when inertial effects are included) behavior. This is due to the use of a periodic rather than a parabolic potential. Such a potential allows the flipping of rotators to neighboring wells, thus permitting both relaxation and oscillatory behavior[37] in the same model. We apply the effective eigenvalue method to this model in the noninertial limit. Such a model has also been studied (again in the noninertial limit) by Lauritzen and Zwanzig[38] in connection with site models of dielectric relaxation in molecular crystals. They converted the solution to a Sturm-Liouville problem. The governing Sturm-Liouville equation is the particular form of the Hill equation[39] known as Ince's equation,[40] which they integrated using Runge-Kutta methods. They then determined the eigenvalues by the trial and error method. They also obtained using asymptotic methods a closed-form expression for the lowest eigenvalue for high potential barriers, which is the analog of that given by Brown[15] for rotation in three dimensions.

A. The Langevin Equation for Rotation in Two Dimensions: Application to the Dielectric Relaxation of an Assembly of Two-Dimensional Rotators

In this section we deal with the rotational motion of a two-dimensional rotator with dipole moment $\boldsymbol{\mu}$ in the N-fold cosine potential

$$V(\theta) = -V_0 \cos N\theta \tag{257}$$

where θ is the angle between the dipole vector $\boldsymbol{\mu}$ and the z axis. The quantities of interest are the longitudinal and transverse components of the polarization $P_{\parallel}(t)$ and $P_{\perp}(t)$, respectively, where we apply a small additional time-dependent field $\mathbf{F}(t)$ along either the z axis or the x axis. These polarization components are determined by

$$P_{\parallel}(t) = \mu N\left[\langle \cos\theta(t)\rangle - \langle \cos\theta\rangle_{eq}\right] \tag{258}$$

$$P_{\perp}(t) = \mu N\langle \sin\theta(t)\rangle \tag{259}$$

where $\langle \cos\theta\rangle_{eq}$ is the equilibrium average of $\cos\theta$ in the absence of the field $F(t)$, and N is the number of dipoles per unit volume. The one-sided Fourier transforms of $P_{\parallel}(t)$ and $P_{\perp}(t)$ are proportional to the components of the complex susceptibility $\chi_{\parallel}(\omega)$ and $\chi_{\perp}(\omega)$, respectively.

The Langevin equation for a dipole $\boldsymbol{\mu}$ to rotate about an axis normal to the xz plane is[37]

$$I\ddot{\theta}(t) + \zeta\dot{\theta}(t) + NV_0 \sin N\theta(t) - \frac{\partial}{\partial\theta}(\boldsymbol{\mu}(t)\cdot\mathbf{F}(t)) = \lambda(t) \tag{260}$$

In Eq. (260), I is the moment of inertia of the rotator about the axis of rotation, and $\zeta\dot{\theta}$ and $\lambda(t)$ are the frictional and white noise torques due to the Brownian motion of the surroundings. The random torque $\lambda(t)$ has the property

$$\overline{\lambda(t)} = 0 \qquad \overline{\lambda(t)\lambda(t')} = 2\zeta kT\delta(t - t') \tag{261}$$

To specialize Eq. (260) to the step-on field we write

$$\mathbf{F}(t) = \mathbf{F}_0 U(t) \tag{262}$$

where $U(t)$ is the unit step function, and F_0 is the amplitude we require to calculate, for this model, the statistical averages in Eqs. (258) and (259) when the inertial effects are ignored.

The problem that presents itself when treating the model using the Langevin equation in the form of Eq. (260) is that it is not apparent how that equation may be linearized to yield the solution for small $\mu F_0/kT$. This difficulty may be circumvented by rewriting Eq. (260) as an equation of motion for

$$p = \cos\theta \tag{263}$$

so that

$$\dot{\theta} = -\dot{p}(1 - p^2)^{-1/2} = -\dot{p}(\sin\theta)^{-1} \tag{264}$$

$$\ddot{\theta} = -\ddot{p}(1 - p^2)^{-1/2} - \dot{p}^2 p(1 - p^2)^{-3/2} \tag{265}$$

$$= -\ddot{p}(\sin\theta)^{-1} - \dot{\theta}^2 p(\sin\theta)^{-1} \tag{266}$$

The Langevin equation (260) with the field $\mathbf{F}$ applied along the z axis and with this change of variable becomes

$$\begin{aligned} &I\frac{d^2}{dt^2}\cos\theta(t) + \zeta\frac{d}{dt}\cos\theta(t) + I\dot{\theta}^2(t)\cos\theta(t) \\ &\quad + \frac{NV_0}{2}[\cos(N+1)\theta(t) - \cos(N-1)\theta(t)] \\ &\quad = \mu F_0(1 - \cos^2\theta(t)) - \sin\theta(t)\lambda(t) \end{aligned} \tag{267}$$

which is the Langevin equation for the motion of the instantaneous dipole moment. To solve it we first form its statistical average over a large

number of rotators

$$I\frac{d^2}{dt^2}\langle\cos\theta\rangle + \zeta\frac{d}{dt}\langle\cos\theta\rangle + \langle I\dot{\theta}^2(t)\cos\theta\rangle + \frac{NV_0}{2}[\langle\cos(N+1)\theta\rangle - \langle\cos(N-1)\theta\rangle] = \mu F_0(1 - \langle\cos^2\theta\rangle) \tag{268}$$

We have stipulated in Eq. (268) that

$$\langle\sin\theta(t)\lambda(t)\rangle = 0 \tag{269}$$

We also note that Eq. (268) may be written as

$$I\frac{d^2}{dt^2}\langle\cos\theta\rangle + \zeta\frac{d}{dt}\langle\cos\theta\rangle + \langle I\dot{\theta}^2\cos\theta\rangle + \frac{NV_0}{2}[\langle\cos(N+1)\theta\rangle - \langle\cos(N-1)\theta\rangle] = \frac{1}{2}\mu F_0[1 - \langle\cos 2\theta\rangle] \tag{270}$$

We note further that when the field **F** is applied along the x axis the quantity of interest $q = \sin\theta(t)$ obeys the similar equation

$$I\frac{d^2}{dt^2}\langle\sin\theta\rangle + \zeta\frac{d}{dt}\langle\sin\theta\rangle + \langle I\dot{\theta}^2\sin\theta\rangle + \frac{NV_0}{2}[\langle\sin(N+1)\theta\rangle - \langle\sin(N-1)\theta\rangle] + \mu F_0[\langle\sin^2\theta\rangle - 1] = 0 \tag{271}$$

The remaining terms in Eqs. (270) and (271) that cause difficulty are

$$\langle I\dot{\theta}^2\sin\theta\rangle \quad \text{and} \quad \langle I\dot{\theta}^2\cos\theta\rangle \tag{272}$$

Since the noninertial response pertains to the situation where

$$t \gg \frac{I}{\zeta} \tag{273}$$

which implicitly means[41] that a Maxwellian distribution of angular velocities has been achieved, we may now write

$$\langle I\dot{\theta}^2 \cos\theta \rangle = kT\langle \cos\theta \rangle \tag{274}$$

and

$$\langle I\dot{\theta}^2 \sin\theta \rangle = kT\langle \sin\theta \rangle \tag{275}$$

since the orientation and the angular velocity variables, when equilibrium of the angular velocities has been reached, are decoupled from each other, as far as the time behavior of the orientations is concerned.[42, 43] In the noninertial limit

$$I\langle \ddot{p} \rangle = 0 \tag{276}$$

in Eqs. (270) and (271), so that finally

$$\frac{\mathrm{d}}{\mathrm{d}t}\langle \cos\theta \rangle + \frac{kT}{\zeta}\langle \cos\theta \rangle = \frac{NV_0}{2\zeta}\left[\langle \cos(N-1)\theta \rangle - \langle \cos(N+1)\theta \rangle\right] + \frac{1}{2\zeta}\mu F(t)\left[1 - \langle \cos 2\theta \rangle\right] \tag{277}$$

and

$$\frac{\mathrm{d}}{\mathrm{d}t}\langle \sin\theta \rangle + \frac{kT}{\zeta}\langle \sin\theta \rangle = \frac{NV_0}{2\zeta}\left[\langle \sin(N-1)\theta \rangle - \langle \sin(N+1)\theta \rangle\right] + \frac{\mu}{\zeta}F(t)\left[1 - \langle \sin^2\theta \rangle\right] \tag{278}$$

B. General Expressions for the Longitudinal and Transverse Effective Relaxation Times

Equations (277) and (278) are the first terms in the infinite hierarchy of differential-difference equations, which describe the ensemble averages $\langle \cos n\theta \rangle$ and $\langle \sin n\theta \rangle$. We have mentioned that the standard approach to calculating the longitudinal and transverse relaxation times is accomplished by rewriting the infinite hierarchy as a set of ordinary differential equations of the form

$$\dot{\mathbf{X}} = \mathbf{A}\mathbf{X} + \mathbf{B}U(t) \tag{279}$$

where **A** is the transition matrix and **B** is the driving force matrix, and truncating at a given size of **A**. The longest relaxation time is then the reciprocal of the lowest root of the characteristic equation

$$\det\{s\mathbf{I} - \mathbf{A}\} = 0 \tag{280}$$

where s denotes the complex frequency.

The disadvantage of this method is that it is, in general, impossible to obtain a closed-form expression for the longest relaxation time. This difficulty may be circumvented in certain cases (see Section II.J) by means of the effective eigenvalue method.

We first consider the parallel equation of motion, Eq. (277), and recall that, just as in Section II, we are usually interested only in the response linear in F. We therefore assume that in Eq. (277)

$$\langle \cos n\theta \rangle = \langle \cos n\theta \rangle_{eq} + \langle \cos n\theta \rangle_1 \tag{281}$$

where the subscript eq denotes the equilibrium ensemble average in the absence of a perturbing constant field F and subscript 1 denotes the portion of the ensemble average that is linear in F. On substituting this equation into Eq. (277) we find that

$$\frac{\mathrm{d}}{\mathrm{d}t}\langle \cos\theta \rangle_1 + \frac{kT}{\zeta}\langle \cos\theta \rangle_1 = \frac{NV_0}{2\zeta}\left[\langle \cos(N-1)\theta \rangle_1 - \langle \cos(N+1)\theta \rangle_1\right] + \frac{1}{2\zeta}\mu F(t)\left[1 - \langle \cos 2\theta \rangle_0\right] \tag{282}$$

whence the effective eigenvalue method leads us to

$$\lambda_{\mathrm{eff}}^{\parallel} = -\frac{(\mathrm{d}/\mathrm{d}t)\langle \cos\theta \rangle_1}{\langle \cos\theta \rangle_1} = \frac{(kT/\zeta)\langle \cos\theta \rangle_1 + (NV_0/2\zeta)\left[\langle \cos(N+1)\theta \rangle_1 - \langle \cos(N-1)\theta \rangle_1\right]}{\langle \cos\theta \rangle_1} \tag{283}$$

We now have at $t = 0$

$$\langle \cos\theta \rangle_1 = \langle \cos\theta \rangle_0 - \langle \cos\theta \rangle_{eq} \tag{284}$$

At equilibrium

$$\begin{aligned}\langle\cos\theta\rangle_{eq} &= \frac{\int_0^{2\pi}\cos\theta\, e^{(V_0\cos N\theta+\mu F_0\cos\theta)/kT}\,d\theta}{\int_0^{2\pi} e^{(V_0\cos N\theta+\mu F_0\cos\theta)/kT}\,d\theta}\\ &\cong \langle\cos\theta\rangle_0 + \frac{\mu F_0}{kT}\left[\langle\cos^2\theta\rangle_0 - \langle\cos\theta\rangle_0^2\right]\\ &= \langle\cos\theta\rangle_0 + \frac{\mu F_0}{2kT}\left[1 + \langle\cos 2\theta\rangle_0 - 2\langle\cos\theta\rangle_0^2\right]\end{aligned} \tag{285}$$

Further, at $t = 0$

$$\begin{aligned}&\frac{NV_0}{2}\left[\langle\cos(N+1)\theta\rangle_1 - \langle\cos(N-1)\theta\rangle_1\right]\\ &\quad= \frac{NV_0}{2}\left[\langle\cos(N+1)\theta\rangle_0 - \langle\cos(N-1)\theta\rangle_0\right]\\ &\quad- \frac{NV_0}{2}\left[\langle\cos(N+1)\theta\rangle_{eq} - \langle\cos(N-1)\theta\rangle_{eq}\right]\\ &\quad= -NV_0\left[\langle\sin N\theta\sin\theta\rangle_0 - \langle\sin N\theta\sin\theta\rangle_{eq}\right]\end{aligned} \tag{286}$$

At equilibrium

$$\begin{aligned}NV_0\langle\sin N\theta\sin\theta\rangle_{eq} &\cong NV_0\langle\sin N\theta\sin\theta\rangle_0\\ &\quad+ \frac{NV_0\mu F_0}{kT}\langle\sin N\theta\sin\theta(\cos\theta - \langle\cos\theta\rangle_0)\rangle_0\end{aligned} \tag{287}$$

The last term on the right side of Eq. (287) can be simplified as follows:

$$\begin{aligned}&\frac{NV_0\mu F_0}{kT}\langle\sin N\theta\sin\theta(\cos\theta - \langle\cos\theta\rangle_0)\rangle_0\\ &\quad= \frac{\mu F_0}{2kT}\left\langle(\sin 2\theta - 2\sin\theta\langle\cos\theta\rangle_0)\frac{\partial}{\partial\theta}V(\theta)\right\rangle_0\\ &\quad= \mu F_0\left[\langle\cos 2\theta\rangle_0 - \langle\cos\theta\rangle_0^2\right]\end{aligned} \tag{288}$$

Thus, taking account of Eqs. (285)–(288) we obtain

$$\lambda_{\text{eff}}^{\parallel} = \frac{kT}{\zeta} - \frac{2kT}{\zeta}\frac{\langle\cos 2\theta\rangle_0 - \langle\cos\theta\rangle_0^2}{1 + \langle\cos 2\theta\rangle_0 - 2\langle\cos\theta\rangle_0^2} = \frac{kT}{\zeta}\frac{1 - \langle\cos 2\theta\rangle_0}{1 + \langle\cos 2\theta\rangle_0 - 2\langle\cos\theta\rangle_0^2} \tag{289}$$

so that the effective longitudinal relaxation time $\tau_{\parallel} = \lambda_{\text{eff}}^{\parallel}$ is given by

$$\tau_{\parallel} = \tau_{\text{D}}\frac{1 + \langle\cos 2\theta\rangle_0 - 2\langle\cos\theta\rangle_0^2}{1 - \langle\cos 2\theta\rangle_0} \tag{290}$$

where

$$\tau_{\text{D}} = \frac{\zeta}{kT} \tag{291}$$

is the Debye relaxation time for planar rotators.[12]

To calculate the transverse relaxation time $\tau_{\perp}$ we consider the same problem as above, but this time the step change in the field is applied parallel to the x axis so that we need to determine the behavior of $\langle\mu_x\rangle$. We assume as before that

$$\langle\sin n\theta\rangle = \langle\sin n\theta\rangle_{\text{eq}} + \langle\sin n\theta\rangle_1$$

which leads us to the linearized equation for $\langle\sin\theta\rangle_1$;

$$\frac{\text{d}}{\text{d}t}\langle\sin\theta\rangle_1 + \frac{kT}{\zeta}\langle\sin\theta\rangle_1 = \frac{NV_0}{2\zeta}\left[\langle\sin(N-1)\theta\rangle_1 - \langle\sin(N+1)\theta\rangle_1\right] + \frac{\mu}{\zeta}F(t)\left[1 - \langle\sin^2\theta\rangle_0\right] \tag{292}$$

Likewise, we find that the eigenvalue equation is

$$\lambda_{\text{eff}}^{\perp} = \frac{(kT/\zeta)\langle\sin\theta\rangle_1 + (NV_0/2\zeta)\left[\langle\sin(N+1)\theta\rangle_1 - \langle\sin(N-1)\theta\rangle_1\right]}{\langle\sin\theta\rangle_1} \tag{293}$$

Noting that

$$\langle \sin\theta \rangle_0 = 0 \tag{294}$$

we obtain in the linear approximation of $\mu F_0/kT$ that at $t = 0$

$$\langle \sin\theta \rangle_1 = -\langle \sin\theta \rangle_{eq} = -\frac{\mu F_0}{2kT}(1 - \langle \cos 2\theta \rangle_0) \tag{295}$$

and similarly

$$\frac{NV_0}{2\zeta}[\langle \sin(N+1)\theta \rangle_1 - \langle \sin(N-1)\theta \rangle_1] = -\frac{\mu F_0}{\zeta}\langle \cos 2\theta \rangle_0 \tag{296}$$

Thus,

$$\lambda_{eff}^{\perp} = \frac{kT}{\zeta}\frac{1 + \langle \cos 2\theta \rangle_0}{1 - \langle \cos 2\theta \rangle_0} \tag{297}$$

so that the effective transverse relaxation time is given by

$$\tau_{\perp} = \tau_D \frac{1 - \langle \cos 2\theta \rangle_0}{1 + \langle \cos 2\theta \rangle_0} \tag{298}$$

C. Effective Dielectric Relaxation Times in a Twofold Cosine Potential: Calculation of $\tau_{\parallel}$

Having determined general formulae for the effective relaxation time in an N-fold cosine potential, we now calculate the relaxation times for a dipole rotating in a $\cos 2\theta$ potential. This potential is of interest for the Brownian motion of a particle in an uniaxial crystalline potential created by its neighbors. The formulas for $\tau_{\parallel}$ and $\tau_{\perp}$ (Eqs. (290) and (298), respectively) are of course still valid.

Equation (290) will hold for the effective relaxation time for any potential of the form $\cos N\theta$, so that once again

$$\tau_{\parallel} = \tau_D \frac{1 + \langle \cos 2\theta \rangle_0 - 2\langle \cos\theta \rangle_0^2}{1 - \langle \cos 2\theta \rangle_0} \tag{299}$$

In Eq. (299) the quantities $\langle \cos n\theta \rangle_0$ are given by

$$\langle \cos \theta \rangle_0 = \frac{\int_0^{2\pi} \cos\theta \, e^{\alpha \cos 2\theta} \, d\theta}{\int_0^{2\pi} e^{\alpha \cos 2\theta} \, d\theta} = 0 \tag{300}$$

$$\langle \cos 2\theta \rangle_0 = \frac{\int_0^{2\pi} \cos 2\theta \, e^{\alpha \cos 2\theta} \, d\theta}{\int_0^{2\pi} e^{\alpha \cos 2\theta} \, d\theta} \tag{301}$$

where

$$\alpha = \frac{V_0}{kT} \tag{302}$$

From the theory of Bessel functions[44] we know that

$$e^{\alpha \cos 2\theta} = \sum_{m=-\infty}^{\infty} I_m(\alpha) e^{im2\theta} \tag{303}$$

and

$$I_{-m}(\alpha) = I_m(\alpha) \tag{304}$$

where $I_m(\alpha)$ is the modified Bessel function of the first kind of integer order. Equation (301) thus becomes

$$\begin{aligned} \langle \cos 2\theta \rangle_0 &= \frac{\sum_{m=-\infty}^{\infty} \int_0^{2\pi} I_m(\alpha) e^{im2\theta} \left[(e^{i2\theta} + e^{-i2\theta})/2 \right] d\theta}{\sum_{m=-\infty}^{\infty} \int_0^{2\pi} I_m(\alpha) e^{im2\theta} \, d\theta} \\ &= \frac{I_{-1}(\alpha) + I_1(\alpha)}{2I_0(\alpha)} \end{aligned} \tag{305}$$

By using Eq. (304), Eq. (305) simplifies to

$$\langle \cos 2\theta \rangle_0 = \frac{I_1(\alpha)}{I_0(\alpha)} \tag{306}$$

Therefore,

$$\tau_{\parallel} = \tau_D \frac{I_0(\alpha) + I_1(\alpha)}{I_0(\alpha) - I_1(\alpha)} \tag{307}$$

For small values of α (low potential barrier) we have

$$\langle \cos 2\theta \rangle_0 \cong \frac{\int_0^{2\pi} \cos 2\theta [1 + \alpha \cos 2\theta] \, d\theta}{\int_0^{2\pi} [1 + \alpha \cos 2\theta] \, d\theta} = \frac{\alpha}{2} \tag{308}$$

and hence

$$\tau_{\parallel} \cong \tau_D \frac{1 + \alpha/2}{1 - \alpha/2} \cong \tau_D (1 + \alpha) \tag{309}$$

For large values of α we evaluate $\langle \cos 2\theta \rangle_0$ in the following way: The asymptotic expansion of $I_n(\alpha)$ for large values of α is[44]

$$\begin{aligned} I_n(\alpha) = {} & \frac{e^{\alpha}}{\sqrt{2\pi\alpha}} \sum_{k=0}^{\infty} \frac{(-1)^k}{(2\alpha)^k} \frac{\Gamma(n + k + \frac{1}{2})}{k! \Gamma(n - k + \frac{1}{2})} \\ & + \frac{e^{-\alpha \pm (n + \frac{1}{2})i\pi}}{\sqrt{2\pi\alpha}} \sum_{k=0}^{\infty} \frac{(-1)^k}{(2\alpha)^k} \frac{\Gamma(n + k + \frac{1}{2})}{k! \Gamma(n - k + \frac{1}{2})} \end{aligned} \tag{310}$$

For $\alpha \to \infty$ only the first term is of significance. On noting that[44]

$$\Gamma\left(n + \frac{1}{2}\right) = \frac{\sqrt{\pi}}{(2^n)} (2n - 1)!! \tag{311}$$

$$\Gamma\left(\frac{1}{2} - n\right) = (-1)^n \frac{2^n \sqrt{\pi}}{(2n - 1)!!} \tag{312}$$

we obtain

$$I_0(\alpha) = \frac{e^{\alpha}}{\sqrt{2\pi\alpha}} \left[1 + \frac{1}{8\alpha} + \cdots\right] \tag{313}$$

Similarly,

$$\begin{aligned} I_1(\alpha) &= \frac{e^{\alpha}}{\sqrt{2\pi\alpha}} \sum_{k=0}^{\infty} \frac{(-1)^k}{(2\alpha)^k} \frac{\Gamma(\frac{3}{2} + k)}{k! \Gamma(\frac{3}{2} - k)} \\ &= \frac{e^{\alpha}}{\sqrt{2\pi\alpha}} \left[\frac{\Gamma(\frac{3}{2})}{\Gamma(\frac{3}{2})} - \frac{\Gamma(\frac{5}{2})}{(2\alpha)\Gamma(\frac{1}{2})} + \frac{1}{2(2\alpha)^2} \frac{\Gamma(\frac{7}{2})}{\Gamma(-\frac{1}{2})} + \cdots\right] \\ &= \frac{e^{\alpha}}{\sqrt{2\pi\alpha}} \left[1 - \frac{3}{8\alpha} + \cdots\right] \end{aligned} \tag{314}$$

Using the expressions for $I_0(\alpha)$ and $I_1(\alpha)$ in Eq. (307) we get

$$\begin{aligned}\tau_{\parallel} &= \tau_D \frac{(1 + 1/8\alpha + \cdots) + (1 - 3/8\alpha - \cdots)}{(1 + 1/8\alpha + \cdots) - (1 - 3/8\alpha - \cdots)} \\ &= \tau_D \frac{2 - 1/4\alpha - \cdots}{1/2\alpha + \cdots} \cong 4\alpha\tau_D\end{aligned} \tag{315}$$

The arguments of Section II.J again applies to this result since in the limit of high potential barriers[38] the longest relaxation time is:

$$\tau_{\parallel} \simeq \tau_D \frac{\pi}{4\alpha} \exp \alpha \tag{316}$$

D. Calculation of $\tau_{\perp}$

The relaxation time $\tau_{\perp}$ is given by Eq. (298):

$$\tau_{\perp} = \tau_D \frac{1 - \langle \cos 2\theta \rangle_0}{1 + \langle \cos 2\theta \rangle_0}$$

On taking account of Eq. (306) we have

$$\tau_{\perp} = \tau_D \frac{I_0(\alpha) - I_1(\alpha)}{I_0(\alpha) + I_1(\alpha)} \tag{317}$$

On comparing with the results for $\tau_{\parallel}$, we see that

$$\tau_{\parallel}\tau_{\perp} = (\tau_D)^2$$

Therefore,

$$\tau_{\perp} = \frac{(\tau_D)^2}{\tau_{\parallel}}$$

Using the expression for $\tau_{\parallel}$ we then obtain for $\alpha \gg 1$

$$\tau_{\perp} = \frac{\tau_D}{4\alpha} \tag{318}$$

Similarly, for small values of α we obtain

$$\tau_{\perp} = \tau_{D} \frac{1 - \alpha/2}{1 + \alpha/2} \cong \tau_{D}(1 - \alpha) \tag{319}$$

Thus, the results obtained are in full agreement with those for the three-dimensional counterpart of this model—the Meier-Saupe model.

At the time of going to Press, we, together with J. T. Waldron, have shown from the definition of the correlation time given in Eq. (12), that the exact solutions for the longitudinal and transverse correlation times are

$$T_{\parallel} = \frac{\tau}{2\alpha}(e^{2\alpha} - 1)\left[1 + \sum_{p=1}^{\infty} \frac{(-1)^p}{(2p+1)} \frac{f_{2p+1}(0)}{f_1(0)} \frac{I_{p+1/2}(\alpha)}{I_{1/2}(\alpha)}\right] \tag{320}$$

and

$$T_{\perp} = \frac{\tau}{2\alpha}(1 - e^{-2\alpha})\left[1 + \sum_{p=1}^{\infty} \frac{(-1)^p}{(2p+1)} \frac{g_{2p+1}}{g_1(0)} \frac{I_{p+1/2}(\alpha)}{I_{1/2}(\alpha)}\right] \tag{321}$$

where

$$\frac{f_{2p+1}(0)}{f_1(0)} = \frac{I_{p+1}(\alpha) + I_p(\alpha)}{I_1(\alpha) + I_0(\alpha)}$$

and

$$\frac{g_{2p+1}(0)}{g_1(0)} = \frac{I_{p+1}(\alpha) - I_p(\alpha)}{I_1(\alpha) - I_0(\alpha)}$$

Eq. (320) reduces to Eq. (316) in the limit of very high potential barriers while Eq. (321) reduces to Eq. (318) in the same limit as has been shown by Crothers using the method of steepest descents.

IV. THE BROWNIAN MOTION IN A TILTED COSINE POTENTIAL

We recall the relaxation problems to which this model may be applied before describing the solution procedure. Among these we mention the current–voltage characteristics of a Josephson tunneling junction,[23, 45] the mobility of superionic conductors,[3] cycle slips in second-order phase-locked loops,[46] the ring laser gyro,[19, 20, 47] etc. We consider the Josephson junction

as a definite example. A related problem, although easier than the junction problem, is to calculate the relaxation times for a dipolar molecule in a $\cos N\theta$ potential. This has been considered in Section III. The Josephson junction represents the Brownian motion of a particle in a tilted cosine potential, while the dielectric problem represents the motion of a particle in a level cosine potential. We note that setting the tilt equal to zero in the Josephson problem and applying the effective eigenvalue procedure yields the longitudinal relaxation time for dielectric relaxation of an assembly of two-dimensional rotators in a cosine potential, in other words, the relaxation in a constant electric field $\mathbf{E}_0$ applied in the z direction. The transverse relaxation time is that associated with a small field applied perpendicular to the z direction, that is, the x direction.

A. Application to the Josephson Junction

The main aim of this section is to evaluate the effective eigenvalues for the Josephson junction and to demonstrate the accuracy of the effective eigenvalue method by comparing the exact and approximate calculations of the junction impedance for an externally applied small-signal alternating current in the presence of noise. We recall from the introduction that the Josephson tunneling junction is made up of two superconductors separated from each other by a thin layer of oxide. We label ψ_R, and ψ_L the wave functions for the right and left superconductors, respectively. The Josephson tunneling junction can be represented by the so called RCSJ (resistance–capacitance shunted junction) model, as shown in Fig. 1[23, 45, 50] where

$$\phi = \phi_R - \phi_L \tag{322–332}$$

ϕ_R and ϕ_L are the phase angles associated with the wave functions ψ_R and ψ_L respectively, C and R are the capacitance and resistance of the junction, the term $I \sin \phi$ is the phase-dependent current generator representing the Josephson supercurrent due to the Cooper pairs tunneling through the junction, and I_{dc} is the bias current applied to the junction. Since the junction operates at temperatures above absolute zero there is a white noise current $L(t)$ superimposed on the bias current satisfying the conditions

$$\overline{L(t_1)L(t_2)} = \frac{2kT}{R}\delta(t_1 - t_2) \tag{333}$$

$$\overline{L(t)} = 0 \tag{334}$$

The overbar denotes the statistical average, $\delta(t)$ is the Dirac delta function, and t_1 and t_2 are distinct times.

The current balance equation for the junction is

$$I_{dc} + L(t) = C\frac{dV(t)}{dt} + \frac{V(t)}{R} + I\sin\phi(t) \tag{335}$$

We note that $(\hbar/2e)\dot{\phi}(t)$ is the time-dependent voltage across the junction,[23] where $\hbar = h/2\pi$ (h is the Planck constant) and e is the charge of an electron. Thus,[23]

$$V(t) = \left(\frac{\hbar}{2e}\right)\dot{\phi}(t) \tag{336}$$

Substitution of Eq. (336) in Eq. (335) yields

$$\left(\frac{\hbar}{2e}\right)^2 C\ddot{\phi}(t) + \frac{1}{R}\left(\frac{\hbar}{2e}\right)^2\dot{\phi}(t) + \left(\frac{\hbar}{2e}\right)I\sin\phi(t) = \frac{\hbar}{2e}(I_{dc} + L(t)) \tag{337}$$

Equation (337) has the same form as that for a Brownian particle of mass $(\hbar/2e)^2C$ moving in the tilted cosine potential[22, 23]:

$$U(\phi) = -\frac{\hbar}{2e}(I_{dc}\phi + I\cos\phi) \tag{338}$$

On further simplification Eq. (337) yields

$$C\ddot{\phi}(t) + \frac{1}{R}\dot{\phi}(t) + \frac{2eI}{\hbar}\sin\phi(t) = \frac{2e}{\hbar}(I_{dc} + L(t)) \tag{339}$$

This is the Langevin equation of the Josephson junction.

B. Reduction of the Averaged Langevin Equation to a Set of Differential-Difference Equations

To proceed we change the variable in Eq. (339) by writing

$$r^n = e^{-in\phi} \qquad n = \ldots, -1, 0, 1, \ldots \tag{340}$$

Consequently,

$$\frac{\mathrm{d}}{\mathrm{d}t}r^n = -\mathrm{i}n\dot{\phi}\,\mathrm{e}^{-\mathrm{i}n\phi} \tag{341}$$

so that

$$\dot{\phi} = \frac{\mathrm{i}\dot{r}^n}{nr^n} \tag{342}$$

Similarly,

$$\ddot{r}^n = -n^2\dot{\phi}^2 r^n - \mathrm{i}n\ddot{\phi}r^n \tag{343}$$

so that

$$\ddot{\phi} = \frac{\mathrm{i}\left(\ddot{r}^n + n^2\phi^2 r^n\right)}{nr^n} \tag{344}$$

Substituting expressions (342) and (344) into Eq. (339) and using the relation

$$\sin\phi = \frac{\mathrm{e}^{\mathrm{i}\phi} - \mathrm{e}^{-\mathrm{i}\phi}}{2\mathrm{i}}$$

we obtain

$$\frac{C\mathrm{i}\left(\ddot{r}^n(t) + n^2\dot{\phi}^2(t)r^n(t)\right)}{nr^n(t)} + \frac{\mathrm{i}\dot{r}^n(t)}{Rnr^n(t)} - \frac{2el}{\hbar}\left(\frac{r(t) - r(t)^{-1}}{2\mathrm{i}}\right) = \frac{2e}{\hbar}\left(I_{\mathrm{dc}} + L(t)\right) \tag{345}$$

or

$$C\frac{\mathrm{d}^2 r^n(t)}{\mathrm{d}t^2} + \frac{1}{R}\frac{\mathrm{d}r^n(t)}{\mathrm{d}t} + n^2C\dot{\phi}^2(t)r^n(t) = \frac{enI}{\hbar}\left(r^{n-1}(t) - r^{n+1}(t)\right) - \frac{\mathrm{i}2en}{\hbar}r^n(t)\left(I_{\mathrm{dc}} + L(t)\right) \tag{346}$$

On averaging and utilizing the properties of Eq. (334), Eq. (346) becomes

$$C\frac{d^2}{dt^2}\langle r^n\rangle + \frac{1}{R}\frac{d}{dt}\langle r^n\rangle + n^2C\langle\dot{\phi}^2 r^n\rangle = \frac{enI}{\hbar}(\langle r^{n-1}\rangle - \langle r^{n+1}\rangle) - \frac{i2enI_{dc}}{\hbar}\langle r^n\rangle \tag{347}$$

We shall consider Eq. (347) in the diffusion (noninertial or low-frequency) limit, where we can neglect the capacitative term $C\ddot{\phi}(t)$. The Maxwell-Boltzmann distribution[22]

$$W_0(\dot{\phi}) = C_0 \exp\left[-\left(\frac{\hbar}{2e}\right)^2 \frac{C\dot{\phi}^2}{kT}\right] \tag{348}$$

with C_0 the normalizing constant, has then set in so that the phase angle ϕ and its derivative $\dot{\phi}$ are decoupled from each other as far as the time behavior of $\phi(t)$ is concerned. Thus, on setting $C = 0$ in Eq. (347) and noting that

$$\langle\dot{\phi}^2 r^n\rangle = \langle\dot{\phi}^2\rangle_0\langle r^n\rangle$$

where

$$\langle\dot{\phi}^2\rangle_0 = \int_{-\infty}^{\infty}\dot{\phi}^2 C_0 \exp\left[-\left(\frac{\hbar}{2e}\right)^2\frac{C\dot{\phi}^2}{kT}\right]d\dot{\phi} = \left(\frac{2e}{\hbar}\right)^2\frac{kT}{C}$$

we have

$$\frac{d}{dt}\langle r^n\rangle + \frac{1}{\tau_0}\left(n^2 + \frac{inx\gamma}{2}\right)\langle r^n\rangle = \frac{n\gamma}{4\tau_0}(\langle r^{n-1}\rangle - \langle r^{n+1}\rangle) \tag{349}$$

where

$$x = \frac{I_{dc}}{I} \tag{350}$$

which is the ratio of bias current amplitude to supercurrent amplitude (bias or tilt parameter),

$$\gamma = \frac{\hbar I}{ekT} \tag{351}$$

is the ratio of Josephson coupling energy to thermal energy (barrier height parameter), and

$$\tau_0 = \left(\frac{\hbar}{2e}\right)^2 \frac{1}{kTR} \tag{352}$$

is the Debye relaxation time. Note that the quantity $(\hbar/2e)^2(1/R)$ has the meaning of a damping coefficient.

Equation (349) is a well-known result,[3] which may be obtained from the underlying Fokker-Planck equation. The relevant Fokker-Planck equation in the noninertial limit is[3, 45]

$$\zeta \frac{\partial W}{\partial t} = \frac{\partial}{\partial \phi}\left(W \frac{\partial}{\partial \phi} U\right) + kT \frac{\partial^2 W}{\partial \phi^2} \tag{353}$$

where W is the transition probability of the phase,

$$\zeta = \left(\frac{\hbar}{2e}\right)^2 \frac{1}{R}$$

is the damping coefficient, and

$$U(\phi) = -\frac{\hbar}{2e}(I \cos \phi + I_{dc}\phi)$$

is the tilted cosine potential.

The distribution function W must be periodic in such a way that it can be expanded in a Fourier series as[3]

$$W(\phi, t) = \sum_{p=-\infty}^{\infty} a_p(t) e^{ip\phi} \tag{354}$$

Substituting Eq. (354) into Eq. (353) and using the orthogonality properties of the circular functions we find that the coefficients $a_p(t)$ satisfy

$$\frac{d}{dt} a_p(t) + \frac{kT}{\zeta}\left(p^2 + \frac{ip\hbar I_{dc}}{2ekT}\right) a_p(t) = \frac{p\hbar I}{4e\zeta}\left(a_{p-1}(t) - a_{p+1}(t)\right) \tag{355}$$

It can be easily shown that the $a_p(t)$, Eq. (354), are related to $\langle r^p \rangle$ by

$$a_p(t) = \frac{1}{2\pi}\langle e^{-ip\phi}\rangle = \frac{1}{2\pi}\langle r^p\rangle \tag{356}$$

Thus, Eq. (355) coincides precisely with Eq. (349).

We have found the average of Eq. (349) by noting that $\langle r^n(t)L(t)\rangle$ will vanish throughout, because in the inertial Langevin equation, $r^n(t)$ and $L(t)$ are statistically independent. However, this is not true for the noninertial Langevin equation[11]:

$$\frac{d}{dt}\phi(t) + \frac{2eR}{\hbar} I \sin\phi(t) = \frac{2eR}{\hbar}(I_{dc} + L(t)) \tag{357}$$

On transforming Eq. (357) as above we obtain

$$\frac{d}{dt}r^n(t) = \frac{enIR}{\hbar}(r^{n-1}(t) - r^{n+1}(t)) - \frac{i2enR}{\hbar} r^n(t)(I_{dc} + L(t)) \tag{358}$$

Just as in Section II, the multiplicative noise term $r^n(t)L(t)$ contributes a noise-induced drift term to the average. The multiplicative noise term poses an interpretation problem in averaging Eq. (358), as discussed in Section II.B. We recall that, taking the Langevin equation for a stochastic variable $\xi(t)$ as

$$\frac{d}{dt}\xi(t) = h(\xi(t), t) + g(\xi(t), t)L(t) \tag{359}$$

with

$$\overline{L_j(t)} = 0 \qquad \overline{L(t)L(t')} = 2\delta(t - t') \tag{360}$$

and interpreting it as a Stratonovich stochastic equation,[11] we have

$$\begin{aligned} \dot{x} &= \lim_{\tau\to 0}\left\{\frac{1}{\tau}\overline{(\xi(t+\tau) - x)}\right\}\Big|_{\xi(t)=x} \\ &= h(x,t) + g(x,t)\frac{\partial}{\partial x}g(x,t) \end{aligned} \tag{361}$$

where $\xi(t + \tau)$, $\tau > 0$ is a solution of Eq. (395), which at time t has the

sharp value $\xi(t) = x$. It should be noted that the quantity x in Eq. (359) is itself a random variable with probability density function $W(x, t)$ defined such that $W(x, t)\,\mathrm{d}x$ is the probability of finding x in the interval $(x, x + \mathrm{d}x)$. Thus, on averaging Eq. (361) over $W(x, t)$ we obtain

$$\frac{\mathrm{d}}{\mathrm{d}t}\langle x\rangle = \langle h(x,t)\rangle + \left\langle g(x,t)\frac{\partial}{\partial x}g(x,t)\right\rangle \tag{362}$$

where the angular braces mean averaging over $f(x, t)$.

We may use the above results to evaluate the average of the multiplicative noise term in Eq. (358). We have

$$g(r^n) = -\frac{\mathrm{i}2enRr^n}{\hbar}$$

$$g(r^n)\frac{\partial}{\partial r^n}g(r^n) = -\left(\frac{2neR}{\hbar}\right)^2 r^n \tag{363}$$

and

$$\frac{\mathrm{d}}{\mathrm{d}t}r^n = \frac{eInR}{\hbar}(r^{n-1} - r^{n+1}) - \frac{Rin2eI_{\mathrm{dc}}r^n}{\hbar} - kTR^2\left(\frac{2en}{\hbar}\right)^2 r^n \tag{364}$$

Thus, the average equation of motion is

$$\frac{\mathrm{d}}{\mathrm{d}t}\langle r^n\rangle = \frac{ReIn}{\hbar}(\langle r^{n-1}\rangle - \langle r^{n+1}\rangle) - \frac{Rin2eI_{\mathrm{dc}}}{\hbar}\langle r^n\rangle - n^2kTR^2\left(\frac{2e}{\hbar}\right)^2\langle r^n\rangle \tag{365}$$

which is Eq. (349).

C. Calculation of the DC Current–Voltage Characteristics

Ambegaokar and Halperin[45] computed the dc current–voltage characteristic (when the capacitance is neglected) by solving the time independent Smoluchowski Eq. (353) associated with the Langevin equation (339). Having noted that the problem is formally the same as that of the translational Brownian motion of a particle in a tilted cosine potential, where the assumption of zero capacitance corresponds to ignoring inertial effects in the mechanical analogy, they found that (for details, see Ref. 3,

p. 289; for a further discussion of mechanical analogs see Ref. 50)

$$\nu = -\frac{4\pi}{\gamma}\left\{(e^{-\pi\gamma x} - 1)\int_0^{2\pi}\int_0^{2\pi}\frac{f(\phi)}{f(\phi')}\,d\phi\,d\phi' + \int_0^{2\pi}\int_0^{\phi'}\frac{f(\phi)}{f(\phi')}\,d\phi\,d\phi'\right\}^{-1} \tag{366}$$

where

$$\nu = \frac{V}{IR} \tag{367}$$

and

$$f(\phi) = \exp\left(\tfrac{1}{2}\gamma(x\phi + \cos\phi)\right) \tag{368}$$

They plotted these results using a numerical method to evaluate the integrals and also gave simple approximations for extreme cases:

$\gamma \to 0$ and $\nu = x$, i.e., $V/R = I_{dc}$ (369)
This corresponds to Ohm's law.

$\gamma \to \infty$ and $x < 1$; then $\nu = 0$
This is pure superconduction.

$\gamma \to \infty$ and $x > 1$; then $\nu = (x^2 - 1)^{1/2}$, i.e., $V = RI(x^2 - 1)^{1/2}$
This is Ohm's law with a correction factor.

There are two major problems associated with the method of Ambegaokar and Halperin:

1. There is no simple form for the $I - V$ curve. The integrals in Eq. (366) have to be evaluated using a numerical method.
2. There is no obvious way of extending their results to the case where the input current contains a time-dependent part.

These difficulties may be overcome using a continued fraction method.[3] We may implement this method by recalling that for the stationary case we now have instead of Eq. (365)

$$\langle r^p\rangle_0\left(p^2 + \frac{i\gamma p x}{2}\right) = \frac{p\gamma}{4}(\langle r^{p-1}\rangle_0 - \langle r^{p+1}\rangle_0) \tag{370}$$

where the symbol $\langle\ \rangle_0$ means averaging over the stationary distribution

function $W_0(\phi)$, which is given by[3]

$$W_0(\phi) = C_0 \, \mathrm{e}^{-U(\phi)/kT}\left[1 - \frac{(1 - \mathrm{e}^{-\pi\hbar I_{\mathrm{dc}}/kT})\int_0^{\phi} \mathrm{e}^{U(\phi')/kT}\,\mathrm{d}\phi'}{\int_0^{2\pi} \mathrm{e}^{-U(\phi')/kT}\,\mathrm{d}\phi'}\right] \tag{371}$$

where C_0 is the normalizing constant defined in such a way that

$$\int_0^{2\pi} W_0(\phi)\,\mathrm{d}\phi = 1$$

We note also that the function $W_0(\phi)$ satisfies the condition[3]

$$W_0(\phi) = W_0(\phi + 2\pi)$$

Following the calculation of Risken,[3] we introduce the quantity

$$S_p = \frac{\langle r^p \rangle_0}{\langle r^{p-1} \rangle_0} \tag{372}$$

So, on dividing by $\langle r^{p-1} \rangle_0$, Eq. (370) becomes

$$S_p\left(p^2 + \mathrm{i}\frac{x\gamma p}{2}\right) = \frac{p\gamma}{4}(1 - S_p S_{p+1})$$

or

$$S_p = \frac{0.5}{\dfrac{2p}{\gamma} + \mathrm{i}x + 0.5S_{p+1}} \tag{373}$$

For $p = 1$ we have

$$S_1 = \frac{0.5}{\dfrac{2}{\gamma} + \mathrm{i}x + 0.5S_2} \tag{374}$$

We can write S_2 from Eq. (373) as

$$S_2 = \frac{0.5}{\dfrac{4}{\gamma} + \mathrm{i}x + 0.5S_3} \tag{375}$$

and so on.

Substituting this into Eq. (374) and similarly for S_3, S_4, etc. we obtain the well-known result[3]

$$S_1 = \cfrac{0.5}{\cfrac{2}{\gamma} + ix + \cfrac{0.25}{\cfrac{4}{\gamma} + ix + \cfrac{0.25}{\cfrac{6}{\gamma} + ix + \cfrac{0.25}{\cfrac{8}{\gamma} + ix \cdots}}}}} \tag{376}$$

To calculate the current–voltage characteristics using Eq. (376) we first recall that the current balance equation is

$$I_{dc} + L(t) = C\frac{dV(t)}{dt} + \frac{1}{R}V(t) + I\sin\phi(t) \tag{377}$$

If we ignore capacitive effects and take expected values at the stationary state, we then obtain

$$I_{dc} - I\langle\sin\phi\rangle_0 - \frac{\langle V\rangle_0}{R} = 0 \tag{378}$$

or

$$\frac{I_{dc}}{I} - \langle\sin\phi\rangle_0 - \frac{\langle V\rangle_0}{IR} = 0 \tag{379}$$

which is

$$x - \langle\sin\phi\rangle_0 = \nu \tag{380}$$

and is the equation that determines the dc current–voltage characteristics. We may find $-\langle\sin\phi\rangle_0$ merely by extracting the imaginary part of S_1 from Eq. (376), since

$$S_1 = \langle e^{-i\phi}\rangle_0 = \langle\cos\phi\rangle_0 - i\langle\sin\phi\rangle_0 \tag{381}$$

The plots yielded by this equation appear to be in agreement with those of Ambegaokar and Halperin.[45] The results are shown in Fig. 9. Equation (380) also allows us to write an expression for the resistance R_J of the

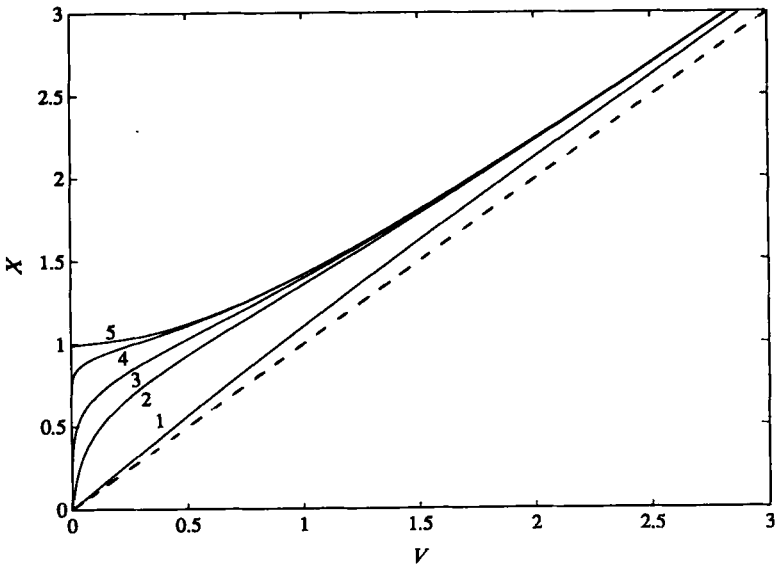


Figure 9. The dc current–voltage characteristics of a Josephson junction in the presence of noise. Curves 1–5 correspond to values of γ (ratio of Josephson coupling energy to thermal energy) = 1, 5, 10, 50, and 100, respectively. Note that for $\gamma = 0$ (dashed line) the behavior corresponds to Ohm's law.

junction arising from the superconductivity. We have from Eq. (378)

$$RI_{\mathrm{dc}}\left(1 - \frac{I}{I_{\mathrm{dc}}}\langle \sin\phi\rangle_0\right) = \langle V\rangle_0$$

so that

$$R_j = R\left(1 - x^{-1}\langle \sin\phi\rangle_0\right) \tag{382}$$

D. Exact Calculation of the Linear Response to an Applied Alternating Current

If we suppose that the current is now $I_{\mathrm{dc}} + I_m \exp(-\mathrm{i}\omega t)$, we may no longer ignore the term $\mathrm{d}\langle r^n\rangle/\mathrm{d}t$ in Eq. (349), since

$$\frac{\mathrm{d}\langle r^n\rangle}{\mathrm{d}t} \neq 0$$

We also suppose that $\hbar I_m/2ekT \ll 1$ so that we can make the perturbation expansion (we may use the exponential form for the ac current since we seek only the linear response)

$$\langle r^n \rangle = \langle r^n \rangle_0 + \tilde{A}_n(\omega) I_m \exp(-i\omega t)/I + \cdots \tag{383}$$

On substituting Eq. (383) into Eq. (355) we obtain for the linear response

$$\left(-i\omega\tau_0 + n^2 + \frac{in\gamma}{2}x\right)\tilde{A}_n(\omega) = \frac{\gamma n}{4}\left(\tilde{A}_{n-1}(\omega) - \tilde{A}_{n+1}(\omega)\right) - \frac{in\gamma}{2}\langle r^n \rangle_0 \tag{384}$$

with $A_0(\omega) = 0$. Eq. (384) may be written as a three-term recurrence relation:

$$Q_n^- \tilde{A}_{n-1}(\omega) + \hat{Q}_n \tilde{A}_n(\omega) + Q_n^+ \tilde{A}_{n+1}(\omega) = -B_n \tag{385}$$

where

$$Q_n^- = -\frac{1}{2} \qquad Q_n^+ = \frac{1}{2} \qquad \hat{Q}_n = \left[\frac{-2i\omega\tau_0}{\gamma n} + \frac{2n}{\gamma} + ix\right]$$

$$B_n = i\langle r^n \rangle_0$$

Equation (385) can readily be solved for the case $B_n = 0$. Denoting the solution of the homogeneous equation by $\tilde{r}_n(\omega)$ and introducing the quantity $\tilde{S}_n(\omega)$ defined as

$$\tilde{S}_n(\omega) = \frac{\tilde{r}_n(\omega)}{\tilde{r}_{n-1}(\omega)} \tag{386}$$

we have the continued fraction solution

$$\tilde{S}_n(\omega) = \cfrac{0.5}{-\dfrac{2i\omega\tau_0}{\gamma n} + \dfrac{2n}{\gamma} + ix + 0.5\tilde{S}_{n+1}(\omega)} \tag{387}$$

In particular for $n = 1$ we obtain

$$\tilde{S}_1(\omega) = \cfrac{0.5}{i\left(x - \frac{2\omega\tau_0}{\gamma}\right) + \frac{2}{\gamma} + \cfrac{0.25}{i\left(x - \frac{2\omega\tau_0}{2\gamma}\right) + \frac{4}{\gamma} + \cfrac{0.25}{i\left(x - \frac{2\omega\tau_0}{3\gamma}\right) + \cdots}}}$$

All other $\tilde{r}_n(\omega)$ can be calculated from Eq. (384) by iteration. On comparing Eqs. (373), (376) and (386), (387) it is obvious that

$$\tilde{S}_n(0) = S_n \equiv \langle r^n \rangle_0 \tag{388}$$

The inhomogeneous Eq. (385) can also be solved by the continued fraction method as described in Ref. 3. If we truncate the system after the Nth term and omit the equations with $n \geq N + 1$, we obtain by inserting the ansatz

$$\tilde{A}_{n+1}(\omega) = R_n^+ \tilde{A}_n(\omega) + C_{n+1} \qquad \text{for } n \geq 0,\ \tilde{A}_0(\omega) = C_0 \tag{389}$$

the following recurrence relations:

$$R_{N-1}^+ = -\frac{Q_N^-}{\hat{Q}_N} \qquad C_N = -\frac{B_N}{\hat{Q}_N} \tag{390}$$

$$R_n^+ = -\frac{Q_{n+1}^-}{\hat{Q}_{n+1} + Q_{n+1}^+ R_{n+1}^+} \qquad \text{for } 0 \leq n \leq N-2 \tag{391}$$

$$C_n = -\frac{B_n + Q_n^+ C_{n+1}}{\hat{Q}_n + Q_n^+ R_n^+} \qquad \text{for } 0 \leq n \leq N-1 \tag{392}$$

Thus, all $\tilde{A}_n(\omega)$ with $n \geq 1$ can now be obtained by iteration. Finally, $\tilde{A}_0(\omega)$ follows from Eq. (385) with $n = 0$; i.e., $\tilde{A}_0(\omega) = C_0$. This method can be used for numerical calculations.[20] The exact calculation of the linear junction impedance based on a numerical solution of the hierarchy of the differential-difference equations (384) obtained from the Fokker-Planck equation has been given in Ref. 53 (see also the discussion of this results in Ref. 54).

However, there is another representation of the exact solution. One may show on successive elimination of the variables in the inhomogeneous

Eq. (384) that the solution is[22]

$$\tilde{A}_1(\omega) = -2i\Big[\tilde{S}_1(\omega)\langle r\rangle_0 - \tilde{S}_1(\omega)\tilde{S}_2(\omega)\langle r^2\rangle_0 + \tilde{S}_1(\omega)\tilde{S}_2(\omega)\tilde{S}_3(\omega)\langle r^3\rangle_0 - \cdots - (-1)^{n+1}\langle r^n\rangle_0 \prod_{k=1}^{n} \tilde{S}_k(\omega) + \cdots\Big]$$

where the $\tilde{S}_n(\omega)$ are given by Eq. (387).

On taking into account Eqs. (372) and (388), the above equation becomes

$$\tilde{A}_1(\omega) = -2i\tilde{S}_1(\omega)\tilde{S}_1(0\big(1 - \tilde{S}_2(\omega)\tilde{S}_2(0)\big(1 - \tilde{S}_3(\omega)\tilde{S}_3(0)(1 - \cdots)\big)\big) \tag{393}$$

We can show in the same way that the response of $\langle r^{-1}\rangle$ where

$$\langle r^{-1}\rangle = \langle r^{-1}\rangle_0 + \tilde{A}_{-1}(\omega) I_m \, e^{-i\omega t}/I + \cdots$$

is given by

$$\begin{aligned}\tilde{A}_{-1}(\omega) &= 2i\tilde{S}_1^*(-\omega)\tilde{S}_1^*(0)\big(1 - \tilde{S}_2^*(-\omega)\tilde{S}_2^*(0)\big(1 - \tilde{S}_3^*(-\omega)\tilde{S}_3^*(0) \\ &\qquad \times\big(1 - \tilde{S}_4^*(-\omega)\tilde{S}_4^*(0)(1 - \cdots)\big)\big)\big) \\ &= A_1^*(-\omega)\end{aligned} \tag{394}$$

where the asterisk indicates the complex conjugate. Equations (393) and (394) allow us to calculate $\langle \sin\phi - \langle \sin\phi\rangle_0\rangle$ and $\langle \cos\phi - \langle \cos\phi\rangle_0\rangle$ and thus the linear ac response

$$\langle \sin\phi - \langle \sin\phi\rangle_0\rangle = D_s(\omega)\frac{I_m \, e^{-i\omega t}}{I} \tag{395}$$

$$\langle \cos\phi - \langle \cos\phi\rangle_0\rangle = D_c(\omega)\frac{I_m \, e^{-i\omega t}}{I} \tag{396}$$

where

$$D_s(\omega) = \frac{i}{2}\left[\tilde{A}_1(\omega) - \tilde{A}_{-1}(\omega)\right] \tag{397}$$

$$D_c(\omega) = \frac{1}{2}\left[\tilde{A}_1(\omega) + \tilde{A}_{-1}(\omega)\right] \tag{398}$$

Equations (393)–(396) are very convenient for numerical calculations.

In particular one may use Eq. (397) to evaluate the impedance $Z(\omega)$ offered by the junction. To accomplish this we recall that the averaged current balance equation in the presence of the ac is

$$I_{dc} + I_m e^{-i\omega t} - I\langle \sin\phi \rangle - \frac{\langle V \rangle}{R} = 0$$

We have supposed that

$$\langle \sin\phi \rangle = \langle \sin\phi \rangle_0 + \langle \sin\phi \rangle_1$$
$$\langle V \rangle = \langle V \rangle_0 + \langle V \rangle_1$$

where the subscript 0 on the angular braces denotes the average in the absence of the ac, and the subscript 1 denotes the portion of the average that is linear in I_m. Thus, on dividing across the current balance equation by the supercurrent amplitude I so that

$$x + \frac{I_m}{I} e^{-i\omega t} - \left[\langle \sin\phi \rangle_0 + \langle \sin\phi \rangle_1\right] - \left(\langle \nu \rangle_0 + \langle \nu \rangle_1\right) = 0$$

and recalling that

$$x - \langle \sin\phi \rangle_0 - \langle \nu \rangle_0 = 0$$

we have

$$\langle \nu \rangle_1 = \frac{I_m}{I} e^{-i\omega t} - \langle \sin\phi \rangle_1$$

so that on reverting to the original variables

$$\langle V \rangle_1 = R\left[1 - D_s(\omega)\right] I_m e^{-i\omega t} = Z(\omega) I_m e^{-i\omega t}$$

where

$$Z(\omega) = R[1 - D_s(\omega)] \tag{399}$$

is the impedance of the junction.

E. The Effective Eigenvalues for the Josephson Junction

Let us suppose that a strong dc current I_{dc} had been applied to the junction in the infinite past and that at $t = 0$, I_{dc} is incremented by a small current $U(t)\Delta$, where $U(t)$ is the unit step function so that the total current is $I_t = I_{dc} + U(t)\Delta$. Now we are only interested in the response linear in Δ. We therefore assume that

$$\langle r^n \rangle = \langle r^n \rangle_{eq} + \langle r^n \rangle_1 \tag{400}$$

where the subscript 1 denotes the portion of the statistical average which is linear in Δ and the subscript eq denotes the statistical average in the stationary state computed using the stationary distribution function $W_0(\phi)$ of Eq. (371) where

$$U(\phi) = -\frac{\hbar[\cos\phi + (I_{dc} + \Delta)\phi]}{2e} \tag{401}$$

As $t \to \infty$ we have

$$\lim_{t\to\infty} \langle r^n \rangle = \langle r^n \rangle_{eq} \qquad \lim_{t\to\infty} \langle r^n \rangle_1 = 0$$

On substituting Eq. (400) into Eq. (349) we obtain

$$\left(n^2 + \frac{in\hbar}{2ekT}(I_{dc} + \Delta)\right)\langle r^n \rangle_{eq} + \frac{n\hbar I}{4ekT}\left(\langle r^{n+1} \rangle_{eq} - \langle r^{n-1} \rangle_{eq}\right) = 0 \tag{402}$$

and

$$\frac{d}{dt}\langle r^n \rangle_1 + \frac{1}{\tau_0}\left(n^2 + \frac{in\hbar}{2ekT}I_{dc}\right)\langle r^n \rangle_1 = \frac{n\hbar I}{4\tau_0 ekT}\left(\langle r^{n-1} \rangle_1 - \langle r^{n+1} \rangle_1\right) - \frac{i\Delta n\hbar}{2e\tau_0 kT}\langle r^n \rangle_{eq} U(t) \tag{403}$$

Equation (403) represents a three-term recurrence relation driven by a forcing function, namely the $U(t)$ term. To determine the effective eigenvalue we consider the unforced part of Eq. (403) and reduce it to an eigenvalue problem:

$$\frac{\mathrm{d}}{\mathrm{d}t}\langle r\rangle_1 + \lambda_{\mathrm{eff}}^{+}\langle r\rangle_1 = 0 \tag{404}$$

In this case

$$\langle r\rangle_1 = \langle \mathrm{e}^{-\mathrm{i}\phi}\rangle_1 \tag{405}$$

is a complex variable, so that λ_{eff} is also complex:

$$\lambda_{\mathrm{eff}}^{+} = \lambda = \lambda' + \mathrm{i}\lambda'' \tag{406}$$

The real part of $\lambda_{\mathrm{eff}}^{+}$ when inverted will give the effective relaxation time, while the imaginary part will give the frequency of oscillation.

Equation (404) yields

$$\lambda_{\mathrm{eff}}^{+} = -\frac{(\mathrm{d}/\mathrm{d}t)\langle r\rangle_1}{\langle r\rangle_1} \tag{407}$$

The effective eigenvalue method suggests that Eq. (407) may be replaced by its initial value (i.e., its value at $t = 0$). We therefore have

$$\lambda_{\mathrm{eff}}^{+} = -\frac{(\mathrm{d}/\mathrm{d}t)\langle r\rangle_1}{\langle r\rangle_1} \qquad t = 0 \tag{408}$$

On substituting Eq. (403) into Eq. (408) for $n = 1$, we obtain

$$\lambda_{\mathrm{eff}}^{+} = \frac{(1/\tau_0)(1 + \mathrm{i}\hbar I_{\mathrm{dc}}/2ekT)\langle r\rangle_1 + (eI/\hbar)R\langle r^2\rangle_1}{\langle r\rangle_1} \qquad t = 0 \tag{409}$$

Equation (409) may be further simplified to

$$\lambda_{\mathrm{eff}}^{+} = \frac{1}{\tau_0} + \frac{\mathrm{i}2eR}{\hbar} I_{\mathrm{dc}} + \frac{\langle r^2\rangle_1}{\langle r\rangle_1}\frac{eIR}{\hbar} \qquad t = 0 \tag{410}$$

Now from Eq. (400) we have

$$\langle r^n\rangle_1 = \langle r^n\rangle - \langle r^n\rangle_{eq} \tag{411}$$

with

$$\lim_{t\to 0}\langle r^n\rangle_1 = \langle r^n\rangle_0 - \langle r^n\rangle_{eq} \tag{412}$$

Equation (410) with the aid of the Eqs. (411) and (412) simplifies further to

$$\lambda^{+}_{eff} = \frac{1}{\tau_0} + \frac{i2eR}{\hbar} I_{dc} + \left[\frac{\langle r^2\rangle_{eq} - \langle r^2\rangle_0}{\langle r\rangle_{eq} - \langle r\rangle_0}\right]\frac{eIR}{\hbar} \tag{413}$$

where the averages $\langle r\rangle_{eq}$ and $\langle r^2\rangle_{eq}$ are over the stationary distribution $W_0(\phi)$ of Eq. (371) with the perturbed potential of Eq. (401). However, remembering that we are interested only in the linear response of Δ, we can express $\langle r^n\rangle_{eq}$ as

$$\langle r^n\rangle_{eq} = \langle r^n\rangle_0 + \Delta\frac{\partial}{\partial\Delta}\langle r^n\rangle_0 + O(\Delta^2) \tag{414}$$

Thus, we need to evaluate only $(\partial/\partial\Delta)\langle r\rangle_0$ and $(\partial/\partial\Delta)\langle r^2\rangle_0$, which may be done as follows.

On using Eq. (414) in Eq. (402), we find that the linear approximation in Δ is given by the following set of equations:

$$\left(\frac{2n}{\gamma} + ix\right)\langle r^n\rangle_0 + \frac{1}{2}\left(\langle r^{n+1}\rangle_0 - \langle r^{n-1}\rangle_0\right) = 0 \tag{415}$$

$$\left(\frac{2n}{\gamma} + ix\right)\frac{\partial}{\partial\Delta}\langle r^n\rangle_0 + \frac{1}{2}\left(\frac{\partial}{\partial\Delta}\langle r^{n+1}\rangle_0 - \frac{\partial}{\partial\Delta}\langle r^{n-1}\rangle_0\right) = -i22\langle r^n\rangle_0 \tag{416}$$

The solution of Eq. (415) may again be given in terms of an infinite

continued fraction (see Section IV.C). We have from Eq. (415)

$$\frac{\langle r^n\rangle_0}{\langle r^{n-1}\rangle_0} = \cfrac{0.5}{\cfrac{2n}{\gamma} + ix + \cfrac{1}{2}\cfrac{\langle r^{n+1}\rangle_0}{\langle r^n\rangle_0}} \tag{417}$$

Thus, noting that

$$\langle r^0\rangle_0 = 1$$

we obtain Eq. (376), viz.,

$$\langle r\rangle_0 = \cfrac{0.5}{\cfrac{2}{\gamma} + ix + \cfrac{0.25}{\cfrac{4}{\gamma} + ix + \cfrac{0.25}{\cfrac{6}{\gamma} + ix + \cfrac{0.25}{\cfrac{8}{\gamma} + ix + \cdots}}}} \tag{418}$$

The other quantities $\langle r^n\rangle_0$ with $n \geq 2$ can be obtained from the recurrence relation of Eq. (415) by iteration; for example,

$$\langle r^2\rangle_0 = 1 - 2\left(\frac{2}{\gamma} + ix\right)\langle r\rangle_0 \tag{419}$$

On substituting $\langle r^n\rangle_0$ into Eq. (416) and successively eliminating the variables we obtain

$$\begin{aligned}\frac{\partial}{\partial\Delta}\langle r\rangle_0 &= -\frac{2i}{I}\Big[S_1\langle r\rangle_0 - S_1S_2\langle r^2\rangle_0 + S_1S_2S_3\langle r^3\rangle_0 \\ &\qquad - \cdots + (-1)^{n-1}\prod_{k=1}^{n} S_k\langle r^n\rangle_0 + \cdots\Big] \qquad (420)\\ &= -\frac{2i}{I}S_1^2\big(1 - S_2^2\big(1 - S_3^2\big(1 - S_4^2(1 - \cdots)\big)\big)\big)\end{aligned}$$

where

$$S_k = \cfrac{0.5}{\cfrac{2k}{\gamma} + ix + \cfrac{0.25}{\cfrac{2(k+1)}{\gamma} + ix + \cfrac{0.25}{\cfrac{2(k+2)}{\gamma} + ix + \cdots}}} \tag{421}$$

Noting that Eq. (417) allows us to express S_n in terms of $\langle r^n \rangle_0$ as

$$\langle r^n \rangle_0 = \langle r^{n-1} \rangle_0 S_n \tag{422}$$

we obtain

$$\begin{aligned} \frac{\partial}{\partial \Delta} \langle r \rangle_0 &= -\frac{2i}{I} \left[\langle r \rangle_0^2 - \langle r^2 \rangle_0^2 + \langle r^3 \rangle_0^2 - \langle r^4 \rangle_0^2 + \cdots \right] \\ &= \frac{2i}{I} \sum_{n=1}^{\infty} (-1)^n \langle r^n \rangle_0^2 \end{aligned} \tag{423}$$

The quantity $(\partial/\partial\Delta)\langle r^2 \rangle_0$ can be obtained from Eq. (416) at $n = 1$, whence

$$\begin{aligned} \frac{\partial}{\partial \Delta} \langle r^2 \rangle_0 &= -\frac{2i}{I} \langle r \rangle_0 - 2\left(\frac{2}{\gamma} + ix \right) \frac{\partial}{\partial \Delta} \langle r \rangle_0 \\ &= -\frac{2i}{I} \langle r \rangle_0 + \frac{4i}{I} \left(\frac{2}{\gamma} + ix \right) \\ &\quad \times \left[\langle r \rangle_0^2 - \langle r^2 \rangle_0^2 + \langle r^3 \rangle_0^2 - \langle r^4 \rangle_0^2 + \cdots \right] \end{aligned} \tag{424}$$

On substituting Eqs. (423) and (424) into Eq. (413) we obtain

$$\begin{aligned} \lambda_{\text{eff}}^{+} &= \frac{1}{\tau_0} + \frac{i2RI_{\text{dc}}}{\hbar} \\ &\quad + \frac{eIR}{\hbar} \frac{\langle r \rangle_0 - 2(2/\gamma + ix)\left[\langle r \rangle_0^2 - \langle r^2 \rangle_0^2 + \langle r^3 \rangle_0^2 - \cdots \right]}{\langle r \rangle_0^2 - \langle r^2 \rangle_0^2 + \langle r^3 \rangle_0^2 - \cdots} \end{aligned} \tag{425}$$

$$= \frac{1}{\tau_0} + \frac{i2RI_{\text{dc}}}{\hbar} - \frac{2eIR}{\hbar} \left(\frac{2}{\gamma} + ix \right) + \frac{eIR}{\hbar} \frac{\langle r \rangle_0}{\sum_{n=1}^{\infty} (-1)^{n+1} \langle r^n \rangle_0^2} \tag{426}$$

Noting that $1/\tau_0 = kTR(2e/\hbar)^2$, $x = I_{dc}/I$ and $\gamma = \hbar I/ekT$, Eq. (426) reduces to

$$\lambda_{\text{eff}}^{+} = \frac{eIR}{\hbar}\frac{\langle r\rangle_0}{\sum_{n=1}^{\infty}(-1)^{n+1}\langle r^n\rangle_0^2} = \frac{\gamma}{4\tau_0}\frac{\langle r\rangle_0}{\sum_{n=1}^{\infty}(-1)^{n+1}\langle r^n\rangle_0^2} \tag{427}$$

Thus, we have expressed the complex effective eigenvalue $\lambda_{\text{eff}}^{+} = \lambda' + i\lambda''$ in terms of the equilibrium averages $\langle r^n\rangle_0$ only. However, for numerical calculations it is more convenient to express λ_{eff}^{+} in terms of S_n. On using Eq. (422) we obtain after some algebra

$$\lambda_{\text{eff}}^{+} = \frac{\gamma}{4\tau_0 S_1\left(1 - S_2^2\left(1 - S_3^2\left(1 - S_4^2(1 - \cdots)\right)\right)\right)} \tag{428}$$

We can show in the same way that the effective eigenvalue λ_{eff}^{-} for $\langle r^{-1}\rangle$ is related to λ_{eff}^{+} by

$$\lambda_{\text{eff}}^{-} = (\lambda_{\text{eff}}^{+})^* = \lambda^* = \lambda' - i\lambda'' \tag{429}$$

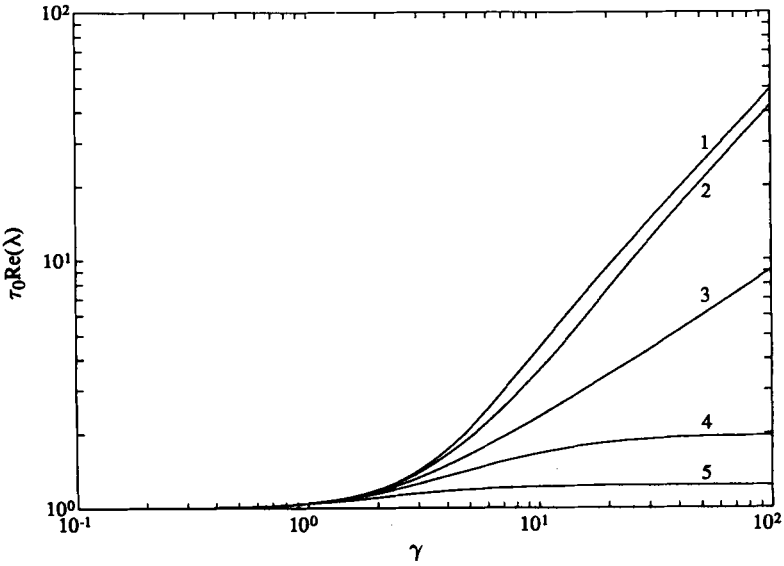


Figure 10. The real part of the effective eigenvalue for the Josephson junction $\tau_0\lambda'$ versus γ. 1, x (bias current) = 0.1; 2, $x = 0.5$; 3, $x = 1.0$; 4, $x = 1.5$; and 5, $x = 2.5$.

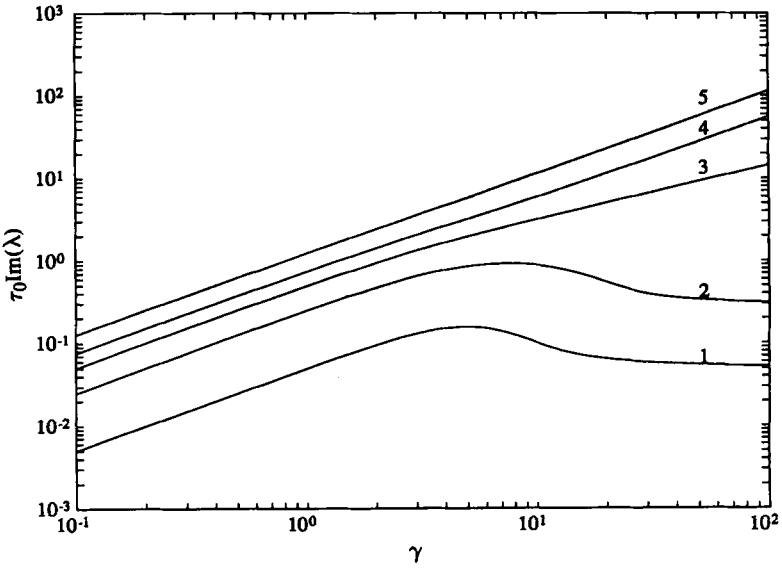


Figure 11. The same as Fig. 10 for $\tau_0\lambda''$ versus γ.

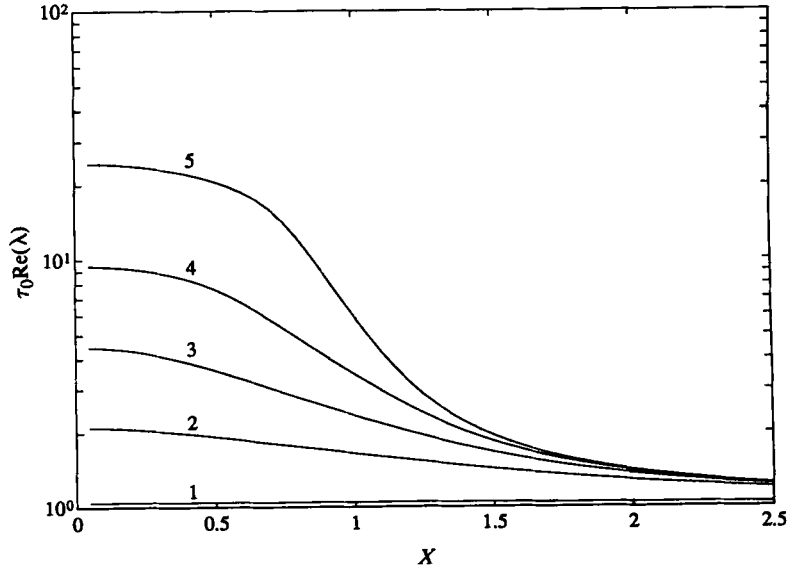


Figure 12. The real part of $\tau_0\lambda'$ versus x. 1, $\gamma = 1$; 2, $\gamma = 5$; 3, $\gamma = 10$; 4, $\gamma = 20$; and 5, $\gamma = 50.0$.

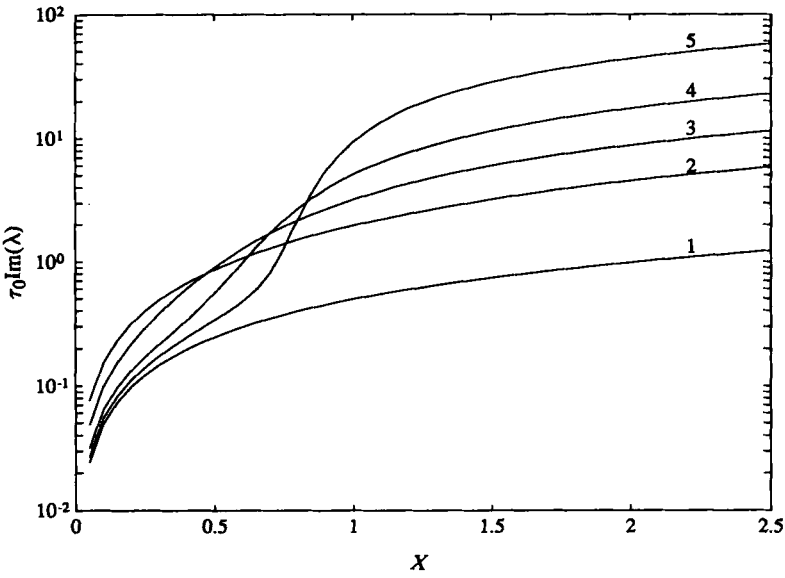


Figure 13. The same as in Fig. 12 for $\tau_0\lambda''$ versus x.

The behavior of the real ($\tau_0\lambda'$) and imaginary ($\tau_0\lambda''$) parts of the normalized effective eigenvalue $\tau_0\lambda_{\mathrm{eff}}^{+}$ as a function of the barrier height γ and bias parameters x is illustrated in Figs. 10–13. As we shall see in Section IV.F, Eq. (428) accurately represents the behavior of the frequency of oscillations ($\mathrm{Im}\{\lambda_{\mathrm{eff}}^{+}\}$) and the spectrum broadening ($\mathrm{Re}\{\lambda_{\mathrm{eff}}^{+}\}$).

F. The Linear Response to an Alternating Current Calculated Using the Effective Eigenvalues

Having determined the effective eigenvalues $\lambda_{\mathrm{eff}}^{+}$ and $\lambda_{\mathrm{eff}}^{-}$, we may calculate the impedance of the junction as follows. We recall that the leading members of the hierarchy of differential-difference equations are

$$\frac{\mathrm{d}}{\mathrm{d}t}\langle r^{-1}\rangle + \frac{1}{\tau_0}\left(1 - \frac{\mathrm{i}\hbar I_{\mathrm{dc}}}{2ekT}\right)\langle r^{-1}\rangle = \frac{\hbar I}{4ekT\tau_0}(1 - \langle r^{-2}\rangle) + \frac{\mathrm{i}2eR}{\hbar} I_m \mathrm{e}^{-\mathrm{i}\omega t}\langle r^{-1}\rangle \quad (430)$$

$$\frac{\mathrm{d}}{\mathrm{d}t}\langle r\rangle + \frac{1}{\tau_0}\left(1 + \frac{\mathrm{i}\hbar I_{\mathrm{dc}}}{2ekT}\right)\langle r\rangle = \frac{\hbar I}{4ekT\tau_0}(1 - \langle r^{2}\rangle) - \frac{\mathrm{i}2eR}{\hbar}\langle r\rangle I_m \mathrm{e}^{-\mathrm{i}\omega t} \quad (431)$$

Our closure procedure reduces these equations to the coupled ordinary differential equations of the first order:

$$\frac{\mathrm{d}}{\mathrm{d}t}\langle r^{-1}\rangle_1 + \lambda_{\mathrm{eff}}^{-}\langle r^{-1}\rangle_1 = \frac{\mathrm{i}2eR}{\hbar}\langle r^{-1}\rangle_0 I_m \mathrm{e}^{-\mathrm{i}\omega t} \tag{432}$$

$$\frac{\mathrm{d}}{\mathrm{d}t}\langle r\rangle_1 + \lambda_{\mathrm{eff}}^{+}\langle r\rangle_1 = -\frac{\mathrm{i}2eR}{\hbar}\langle r\rangle_0 I_m \mathrm{e}^{-\mathrm{i}\omega t} \tag{433}$$

The steady-state solutions of Eqs. (432) and (433) are

$$\begin{aligned}\langle r^{-1}\rangle_1 &= \frac{\mathrm{i}2eR\langle r^{-1}\rangle_0}{\hbar(\lambda_{\mathrm{eff}}^{-} - \mathrm{i}\omega)} I_m \mathrm{e}^{-\mathrm{i}\omega t} \\ &= \frac{\mathrm{i}\gamma S_1^*}{2\tau_0(\lambda_{\mathrm{eff}}^{-} - \mathrm{i}\omega)} \frac{I_m \mathrm{e}^{-\mathrm{i}\omega t}}{I} \\ \langle r\rangle_1 &= -\frac{\mathrm{i}2eR\langle r\rangle_0}{\hbar(\lambda_{\mathrm{eff}}^{+} - \mathrm{i}\omega)} I_m \mathrm{e}^{-\mathrm{i}\omega t} \\ &= -\frac{\mathrm{i}\gamma S_1}{2\tau_0(\lambda_{\mathrm{eff}}^{+} - \mathrm{i}\omega)} \frac{I_m \mathrm{e}^{-\mathrm{i}\omega t}}{I}\end{aligned}$$

Here we have used Eq. (429) and $\langle r\rangle_0^* = \langle r^{-1}\rangle_0 = S_1^*$. Thus,

$$\begin{aligned}\langle \sin\phi - \langle\sin\phi\rangle_0\rangle &= \frac{\langle r^{-1}\rangle_1 - \langle r\rangle_1}{2\mathrm{i}} \\ &= \frac{D_{\mathrm{s}}(\omega) I_m \mathrm{e}^{-\mathrm{i}\omega t}}{I}\end{aligned} \tag{434}$$

and

$$\begin{aligned}\langle \cos\phi - \langle\cos\phi\rangle_0\rangle &= \frac{\langle r^{-1}\rangle_1 + \langle r\rangle_1}{2} \\ &= \frac{D_{\mathrm{c}}(\omega) I_m \mathrm{e}^{-\mathrm{i}\omega t}}{I}\end{aligned}$$

where

$$D_{\mathrm{s}}(\omega) = \frac{\gamma}{4\tau_0}\left[\frac{S_1^*}{\lambda' - \mathrm{i}(\omega + \lambda'')} + \frac{S_1}{\lambda' - \mathrm{i}(\omega - \lambda'')}\right] \tag{435}$$

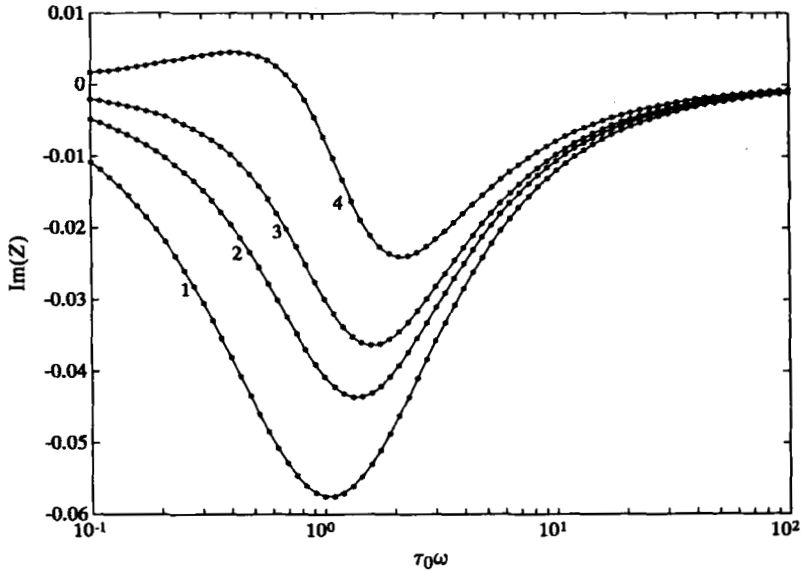


Figure 14. Comparison of the exact (solid lines) and approximate (stars) solutions for the imaginary part of the normalized ($R = 1$) impedance Im{Z} versus $\tau_0\omega$. 1, $\gamma = 1$, $x = 0.1$; 2, $x = 0.75$; 3, $x = 1.0$; and 4, $x = 1.5$.

and

$$D_c(\omega) = \frac{i\gamma}{4\tau_0}\left[\frac{S_1^*}{\lambda' - i(\omega + \lambda'')} - \frac{S_1}{\lambda' - i(\omega - \lambda'')}\right]$$

Thus, the impedance of the junction $Z(\omega)$ in Eq. (399) is given by

$$\begin{aligned} Z(\omega) &= R[1 - D_s(\omega)] \\ &= R\left\{1 - \frac{\gamma}{4\tau_0}\left[\frac{S_1^*}{\lambda' - i(\omega + \lambda'')} + \frac{S_1}{\lambda' - i(\omega - \lambda'')}\right]\right\} \end{aligned} \tag{436}$$

We now compare the impedance $Z(\omega)$ from Eq. (436) with the exact solution (Eqs. (397) and (399)). The results of the calculations are shown in Figs. 14–19. It is apparent from these figures that the effective eigenvalue method gives perfect correspondence to the exact solution for a wide range of the bias parameter x and barrier height parameter γ. Thus, it allows us to represent the impedance of the junction $Z(\omega)$ by the simple analytic formula of Eq. (436), which describes[48] a resonance with natural angular frequency λ''.

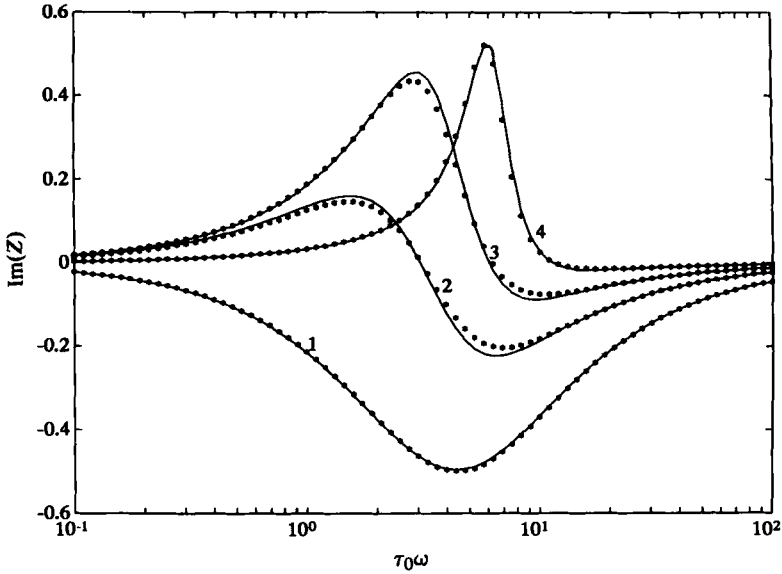


Figure 15. The same as in Fig. 14 for $\gamma = 10$.

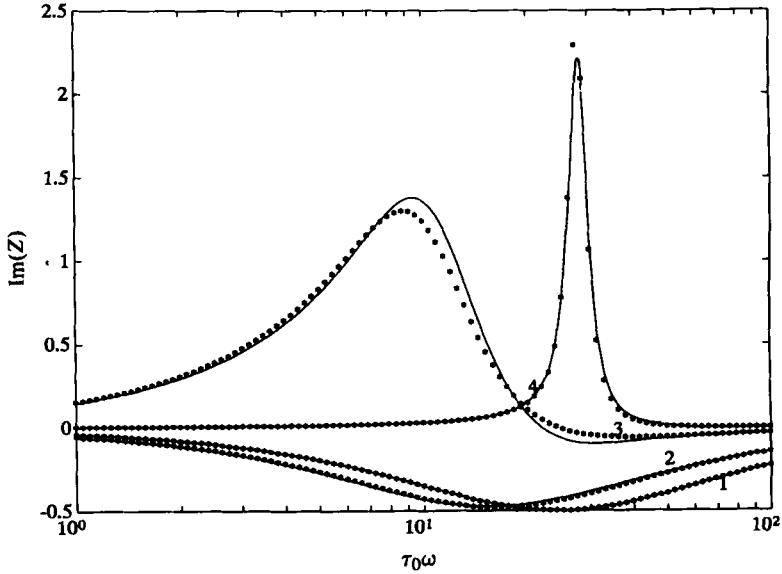


Figure 16. The same as in Fig. 14 for $\gamma = 50$ (the low noise case).

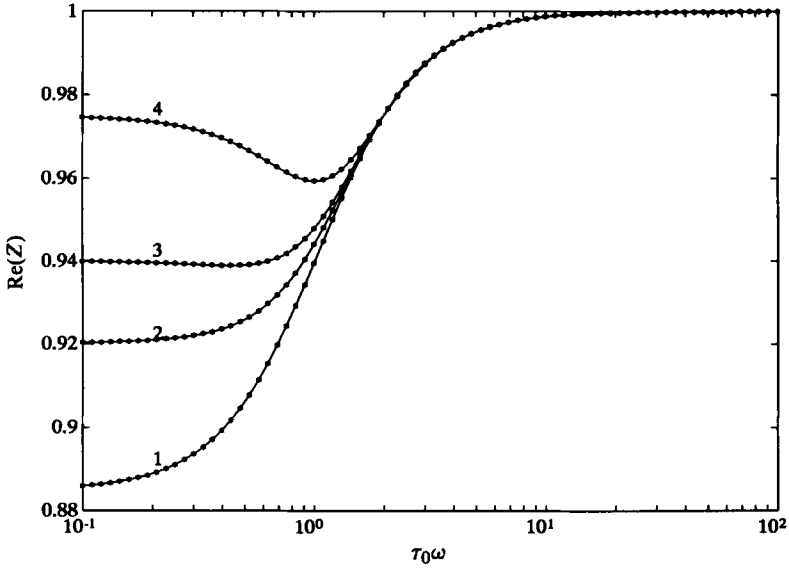


Figure 17. Comparison of the exact (solid lines) and approximate (stars) solutions for the real part of the normalized impedance Re{Z} versus $\tau_0\omega$; $\gamma = 1$. Key as in Fig. 14.

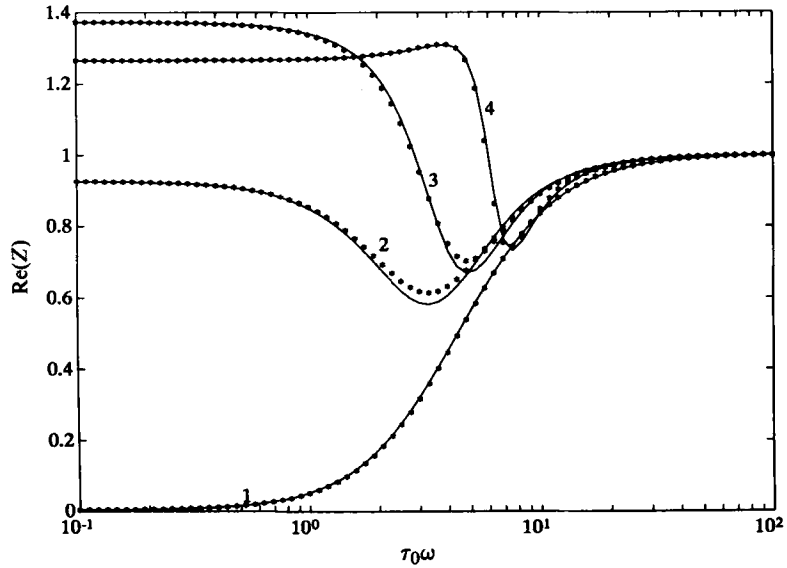


Figure 18. The same as in Fig. 17 for $\gamma = 10$.

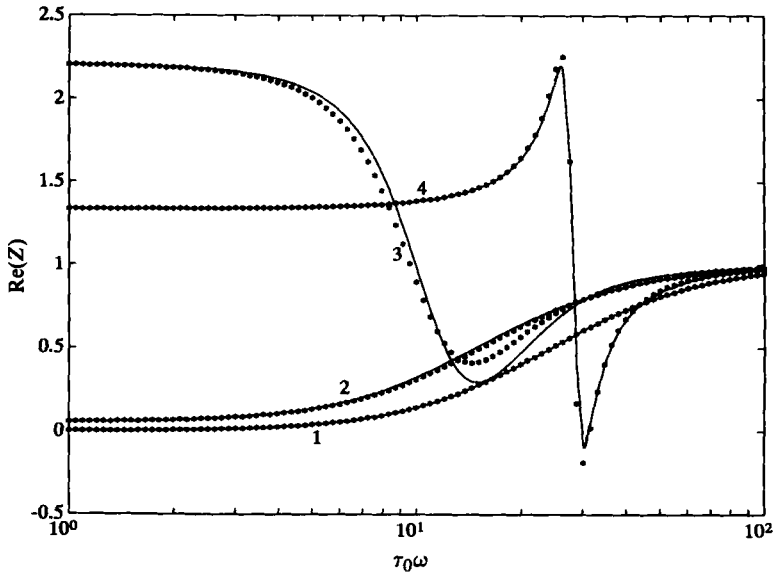


Figure 19. The same as in Fig. 17 for $\gamma = 50$.

Approximate equations for the impedance of similar form to the above equation have been derived in Ref. 54 (Eqs. (3.11) and (3.14)). However, the equations have a narrower range of applicability than ours as they confined in one case (Eq. (3.11)) to large ω and in the other case (Eq. (3.14)) to small fluctuations.

It is of interest to compare the results we have obtained with those for the noiseless case. In the absence of noise one can calculate the impedance analytically by finding the differential impedance at the bias point.[55] The derivation and discussion of the noiseless case is given elsewhere.[54-56] In our notation the result is as follows for values of the bias parameter $x \leq 1$:

$$\frac{Z(\omega)}{R} = \frac{\Omega\left(\Omega - i\sqrt{1 - x^2}\right)}{1 + \Omega^2 - x^2} \tag{437}$$

and for $x > 1$:

$$\frac{Z(\omega)}{R} = 1 + \frac{\sqrt{x^2 - 1}}{\left(x + \sqrt{x^2 - 1}\right)\left(x^2 - 1 - \Omega^2\right)} + \frac{i\pi}{2\left(x + \sqrt{x^2 - 1}\right)}\delta\left(\Omega - \sqrt{x^2 - 1}\right) \tag{438}$$

where

$$\Omega = \frac{2\omega\tau_0}{\gamma} \tag{439}$$

The above equations have a simple physical interpretation. If $x < 1$ the junction behaves like an inductance, then

$$L_j = \frac{\hbar/2eI}{\sqrt{1-x^2}}$$

in parallel with the resistance R, yielding the admittance

$$Y(\omega) = \frac{1}{R} - \frac{1}{i\omega L}$$

which gives the impedance $Z(\omega) = Y^{-1}(\omega)$ from Eq. (437).[55] If $x > 1$ the impedance is entirely real with a singularity at $x_s = \sqrt{1+\Omega^2}$. This singularity vanishes in the presence of noise, as is evident from Figs. 14–19. Such behavior is even more pronounced in Figs. 20–23. There we

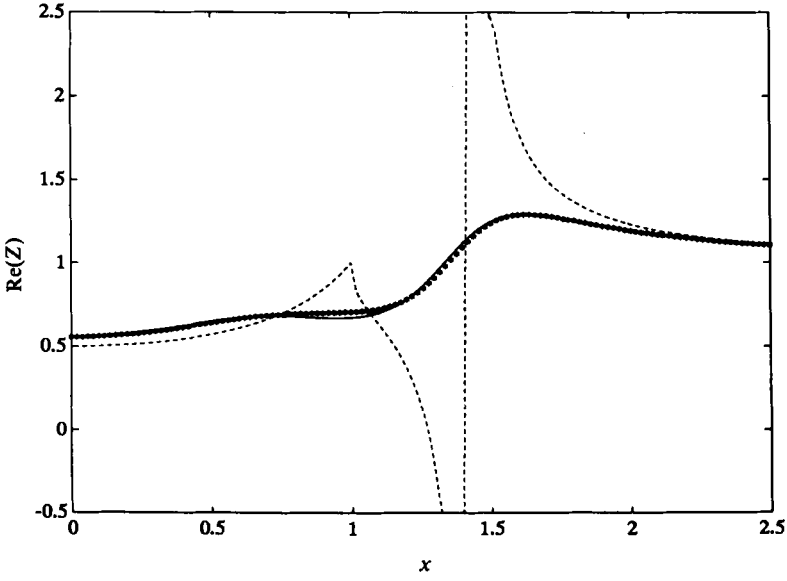


Figure 20. Comparison of the exact (solid line), approximate (stars), and noise-less solutions (dashed line) for the real part of the normalized impedance Re{Z} versus x at $\gamma = 10$.

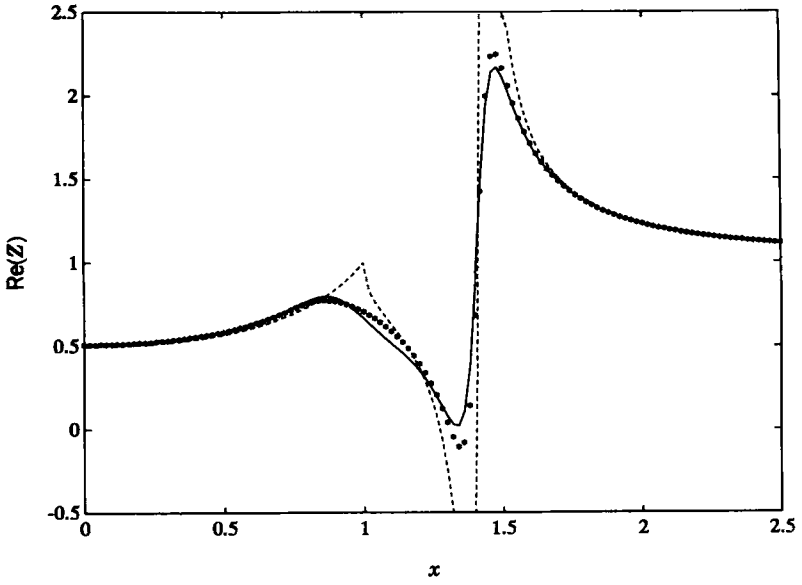


Figure 21. The same as in Fig. 20 for $\gamma = 50$.

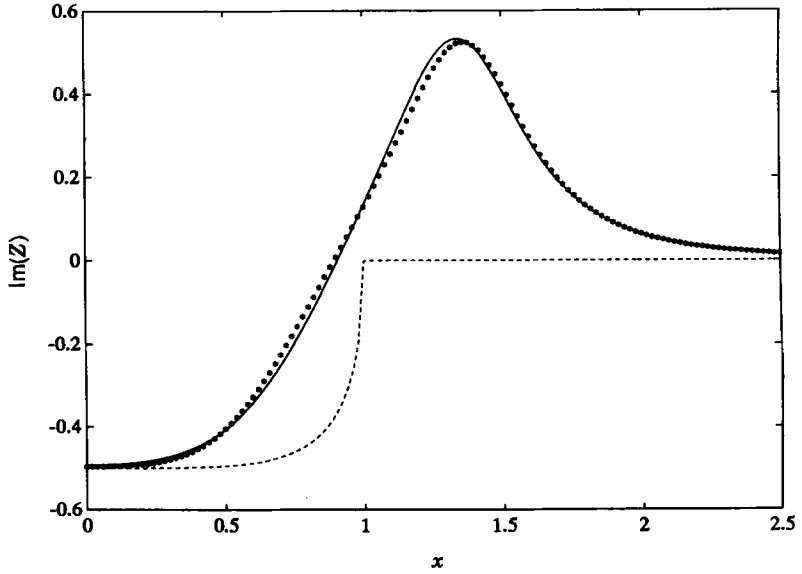


Figure 22. The same as in Fig. 20 for $\mathrm{Im}\{Z\}$ at $\gamma = 10$.

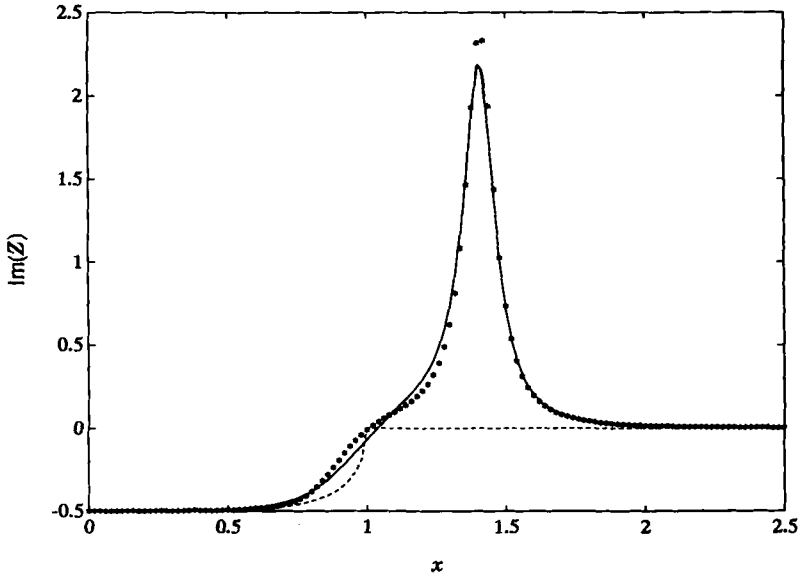


Figure 23. The same as in Fig. 20 for Im{Z} at $\gamma = 50$.

have plotted the normalized impedance $Z(\omega)/R$ as a function of the bias parameter x and have compared it with the noiseless case. It is apparent from Figs. 15 and 17 that for weak noise (large γ) Eqs. (437) and (438) yield a satisfactory description of the impedance excluding the region in the vicinity of the singular point $x_s = \sqrt{1 + \Omega^2}$. However, at $\gamma = 10$, there is a striking difference between the solutions with and without noise. This is particularly apparent in Fig. 20, where close to the singular point x_s the noiseless solution (in contrast to that including noise) possesses a negative real part, which is an indication of amplification or oscillation, which may occur if the junction is inserted in an appropriate microwave circuit.[56]

G. Calculation of the Beat Signal Spectrum of the Ring Laser Gyro

In the previous sections we have considered the Josephson junction as an example of the Brownian motion in a tilted cosine potential. However, the above results can also be applied to an analogous system, namely the ring laser gyro.[19-21, 47] We recall that the dynamical behavior of the ring laser gyro operating in the steady state is described by a similar Langevin equation to that of the Josephson junction[19, 47]:

$$\dot{\phi}(t) - b \sin \phi(t) = a + \Gamma(t) \tag{440}$$

where ϕ is the relative phase between the clockwise and counterclockwise modes of the laser,

$$a = -\frac{8\pi}{\lambda L}(\mathbf{A} \cdot \mathbf{\Omega}) \tag{441}$$

$|\mathbf{A}|$ is the area covered by the optical path of length L along the ring cavity, $\mathbf{\Omega}$ is the rotation rate of the gyro, and λ is the wavelength of the laser; b is the backscattering coefficient and $\Gamma(t)$ is the noise source with properties

$$\overline{\Gamma(t)} = 0 \qquad \overline{\Gamma(0)\Gamma(t)} = 2D\delta(t) \tag{442}$$

The Langevin equation (357) for the Josephson junction has the same mathematical form as Eq. (440) with appropriate change of parameters, viz.,

$$\frac{2eIR}{\hbar} \rightarrow -b \tag{443}$$

$$\frac{2eRI_{dc}}{\hbar} \rightarrow a \tag{444}$$

$$kTR\left(\frac{2e}{\hbar}\right)^2 \rightarrow D \tag{445}$$

Thus, on redefining the parameters x, γ, and τ_0 as

$$x = -\frac{a}{b} \tag{446}$$

$$\gamma = -\frac{2b}{D} \tag{447}$$

$$\tau_0 = \frac{1}{D} \tag{448}$$

we obtain equations for the ring laser gyro in the same form as those for the Josephson junction. However, both x and γ are now negative quantities.

For the ring laser gyro the quantity of interest is the spectrum of the beat signal,[19] defined as

$$\begin{aligned}\alpha(\omega) &= \int_{-\infty}^{\infty} \langle \cos\phi(0)\cos\phi(t)\rangle_0 \, e^{i\omega t}\, dt \\ &= 2\,\mathrm{Re}\left\{ \int_0^{\infty} \langle \cos\phi(0)\cos(t)\rangle_0 \, e^{i\omega t}\, dt \right\}\end{aligned} \tag{449}$$

where

$$C(t) = \langle \cos\phi(0)\cos\phi(t)\rangle_0 \tag{450}$$

is the beat signal autocorrelation function $\cos\phi(t)$, which is a measure of the total detected intensity.[19, 47] A numerical method of exact calculation of $\alpha(\omega)$ has been given in Ref. 20.

Another representation of the exact solution can be obtained in the following manner. On multiplying the both sides of Eq. (364) by $\cos\phi(0)$ and averaging the equation so obtained over the stationary distribution function $W_0(\phi(0))$, we arrive at the set of differential-difference equations:

$$\frac{d}{dt}\psi_n(t) + \frac{1}{\tau_0}\left(n^2 + \frac{inx\gamma}{2}\right)\psi_n(t) = \frac{n\gamma}{4\tau_0}\left(\psi_{n-1}(t) - \psi_{n+1}(t)\right) \tag{451}$$

where

$$\psi_n(t) = \langle \cos\phi(0) r^n(t)\rangle_0 \tag{452}$$

It is obvious that $C(t)$ from Eq. (450) is related to $\psi_1(t)$ by

$$C(t) = \mathrm{Re}\{\psi_1(t)\} \tag{453}$$

Let us introduce instead of $\psi_n(t)$ the true correlation functions $C_n(t)$, defined as

$$\begin{aligned}C_n(t) &= \psi_n(t) - \psi_n(\infty) \\ &= \langle \cos\phi(0) r^n(t)\rangle_0 - \langle \cos\phi(0)\rangle_0 \langle r^n(0)\rangle_0\end{aligned} \tag{454}$$

Then we obtain from Eq. (451)

$$\frac{d}{dt}C_n(t) + \frac{1}{\tau_0}\left(n^2 + \frac{inx\gamma}{2}\right)C_n(t) = \frac{n\gamma}{4\tau_0}\left(C_{n-1}(t) - C_{n+1}(t)\right) \tag{455}$$

It should be noted that now

$$C_0(t) = 0 \tag{456}$$

in contrast to the calculations previously described. We have taken into account here that $\psi_n(\infty)$ satisfies the recurrence relation

$$\left(n^2 + \frac{inx\gamma}{2}\right)\psi_n(\infty) = \frac{n\gamma}{4}\left(\psi_{n-1}(\infty) - \psi_{n+1}(\infty)\right) \tag{457}$$

With the help of the one-sided Fourier transform we may now derive from Eq. (455) the usual three-term recurrence relation:

$$\left(-i\omega\tau_0 + n^2 + \frac{inx\gamma}{2}\right)\tilde{C}_n(\omega) = \frac{\gamma n}{4}\left(\tilde{C}_{n-1}(\omega) - \tilde{C}_{n+1}(\omega)\right) + \tau_0 C_n(0) \tag{458}$$

where

$$\tilde{C}_n(\omega) = \int_0^\infty e^{i\omega t} C_n(t)\, dt \tag{459}$$

Equation (458) has the same form as Eq. (384) and hence has a solution of similar form:

$$\begin{aligned}\tilde{C}_1(\omega) = \frac{4\tau_0}{\gamma}\Bigg(&\tilde{S}_1(\omega)C_1(0) - \frac{1}{2}\tilde{S}_1(\omega)\tilde{S}_2(\omega)C_2(0) \\ &+ \frac{1}{3}\tilde{S}_1(\omega)\tilde{S}_2(\omega)\tilde{S}_3(\omega)C_3(0) - \cdots \\ &+ \frac{1}{n}(-1)^{n+1}C_n(0)\prod_{i=1}^{n}\tilde{S}_i(\omega) + \cdots\Bigg) \\ = \frac{4\tau_0}{\gamma}\tilde{S}_1(\omega)C_1(0)\Bigg(1 - \frac{\tilde{S}_2(\omega)C_2(0)}{2C_1(0)}\Bigg(1 - \frac{2\tilde{S}_3(\omega)C_3(0)}{3C_2(0)}&\\ \times\left(1 - \frac{3\tilde{S}_4(\omega)C_4(0)}{4C_3(0)}(1 - \cdots)\right)\Bigg)\Bigg)&\end{aligned} \tag{460}$$

where as before $\tilde{S}_n(\omega)$ is determined by Eq. (387). The quantity $C_n(0)/C_{n-1}(0)$ appearing in Eq. (460) can be expressed in terms of S_1 and

S_n as follows. We have

$$\begin{aligned} C_n(0) &= \tfrac{1}{2}\langle r^{n+1}(0) + r^{n-1}(0)\rangle_0 - \mathrm{Re}(S_1)\langle r^n(0)\rangle_0 \\ &= \tfrac{1}{2}S_1S_2\cdots S_{n-1}(1 + S_nS_{n+1}) - \mathrm{Re}(S_1)S_1S_2\cdots S_n \end{aligned} \tag{461}$$

Hence, we have for $n \geq 2$

$$\frac{C_n(0)}{C_{n-1}(0)} = \frac{S_{n-1}(1 + S_nS_{n+1}) - 2S_{n-1}S_n\,\mathrm{Re}(S_1)}{1 + S_{n-1}S_n - 2S_{n-1}\,\mathrm{Re}(S_1)} \tag{462}$$

or on taking into account Eq. (373),

$$\frac{C_n(0)}{C_{n-1}(0)} = \frac{1 - S_n(\mathrm{i}x + 2n/\gamma) - S_n\,\mathrm{Re}(S_1)}{S_n + \mathrm{i}x + 2(n-1)/\gamma - \mathrm{Re}(S_1)} \qquad n \geq 2 \tag{463}$$

We also note that

$$C_1(0) = \frac{1}{2}(S_2S_1 + 1) - S_1\,\mathrm{Re}(S_1) = 1 - S_1\left(\mathrm{i}x + \frac{2}{\gamma}\right) - S_1\,\mathrm{Re}(S_1) \tag{464}$$

We can show in the same way that

$$\begin{aligned} \tilde{C}_{-1}(\omega) = \frac{4\tau_0}{\gamma}\tilde{S}_1^*(-\omega)C_1^*(0)\Bigg(1 - \frac{\tilde{S}_2^*(-\omega)C_2^*(0)}{2C_1^*(0)}\Bigg(1 - \frac{2\tilde{S}_3^*(-\omega)C_3^*(0)}{3C_2^*(0)} \\ \times\left(1 - \frac{3\tilde{S}_4^*(-\omega)C_4^*(0)}{4C_3^*(0)}(1 - \cdots)\right)\Bigg)\Bigg) \end{aligned} \tag{465}$$

Thus, on using Eqs. (453), (454), (459), (460), and (465), we obtain the beat signal spectrum as

$$\alpha(\omega) = 2\pi\,\mathrm{Re}^2(S_1)\delta(\omega) + \mathrm{Re}\{T(\omega) + T^*(-\omega)\} \tag{466}$$

where the function $T(\omega)$ is given by

$$T(\omega) = \frac{4\tau_0}{\gamma}\tilde{S}_1(\omega)C_1(0)\left(1 - \frac{\tilde{S}_2(\omega)C_2(0)}{2C_1(0)}\left(1 - \frac{2\tilde{S}_3(\omega)C_3(0)}{3C_2(0)} \times \left(1 - \frac{3\tilde{S}_4(\omega)C_4(0)}{4C_3(0)}(1 - \cdots)\right)\right)\right) \tag{467}$$

Equations (466) and (467) are very convenient for numerical calculations and constitute a simpler algorithm than Eq. (389), which has been used in Ref. 20 to evaluate $\alpha(\omega)$. Equations (466) and (467) allow the exact calculation of $\alpha(\omega)$.

We shall now show how the effective eigenvalue method can be used to evaluate $\alpha(\omega)$. To implement it we need, just as in the application to ferromagnetic resonance, to keep equations for $C_1(t)$ and $C_2(t)$ of Eq. (455). Thus, the effective eigenvalue method requires that $C_1(t)$ and $C_2(t)$ obey the coupled equations

$$\frac{\mathrm{d}}{\mathrm{d}t}C_1(t) + \lambda_1 C_1(t) + \frac{\gamma}{4\tau_0}C_2(t) = 0 \tag{468}$$

$$\frac{\mathrm{d}}{\mathrm{d}t}C_2(t) + \lambda_2 C_2(t) - \frac{\gamma}{2\tau_0}C_1(t) = 0 \tag{469}$$

where $\lambda_1 = (1 + \mathrm{i}x\gamma/2)/\tau_0$ and λ_2 is the effective eigenvalue to be determined.

According to Eq. (455), at $n = 2$, λ_2 is determined as

$$\lambda_2 = -\frac{\dot{C}_2(0) - (\gamma/2\tau_0)C_1(0)}{C_2(0)} \tag{470}$$

where

$$\dot{C}_2(0) = -\frac{1}{\tau_0}(4 + \mathrm{i}x\gamma)C_2(0) - \frac{\gamma}{2\tau_0}(C_3(0) - C_1(0)) \tag{471}$$

and $C_2(0), C_3(0)$ are given by Eq. (461).

On substituting Eq. (471) into Eq. (470) and using Eq. (462), we obtain

$$\lambda_2 = \frac{1}{\tau_0}\left[4 + \mathrm{i}x\gamma + \frac{\gamma S_2}{2}\left(\frac{S_4S_3 + 1 - 2S_3\,\mathrm{Re}(S_1)}{1 + S_2[(S_3 - 2\,\mathrm{Re}(S_1)]}\right)\right] \tag{472}$$

The solution of Eqs. (468) and (469) is easily found using the one-sided Fourier transform. The result is (where we emphasis that $C_1(0)$, $C_2(0)$, λ_1 and λ_2 are complex)

$$\tilde{C}_1(\omega) = \frac{C_1(0)(\lambda_2 - i\omega) - \gamma C_2(0)/4\tau_0}{(\lambda_1 - i\omega)(\lambda_2 - i\omega) + \gamma^2/8\tau_0^2} \tag{473}$$

where

$$\tilde{C}_1(\omega) = \int_0^\infty e^{i\omega t} C_1(t)\, dt \tag{474}$$

Thus, we may now calculate in a simple analytical form from Eqs. (449), (453), and (473) the beat signal spectrum:

$$\alpha(\omega) = 2\pi \operatorname{Re}^2(S_1)\delta(\omega) + \operatorname{Re}\{\tilde{C}_1(\omega) + \tilde{C}_1^*(-\omega)\} \tag{475}$$

The spectrum $\alpha(\omega)$ yielded by Eqs. (466) and (474) contains two parts, a coherent δ-function spectrum and an incoherent broad spectrum.[20] The incoherent part of the laser-gyro beat signal spectrum is presented in Figs. 24–26 for different values of the parameters a/b and γ/b. Our exact

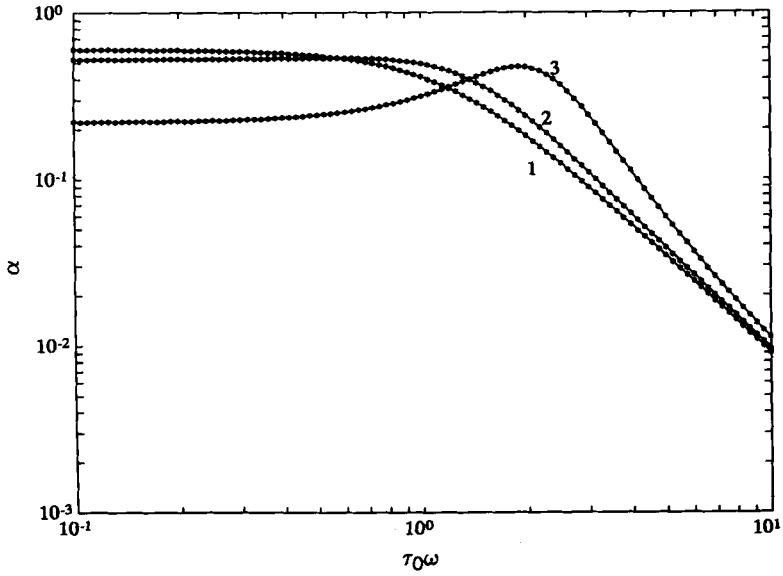


Figure 24. Comparison of the exact (solid lines) and approximate (stars) solutions for "incoherent" part of the beat signal spectrum α of the ring laser gyro versus $\tau_0\omega$. 1, $\gamma = -2$, $x = -0.5$; 2, $x = -0.9$; 3, $x = -2.0$.

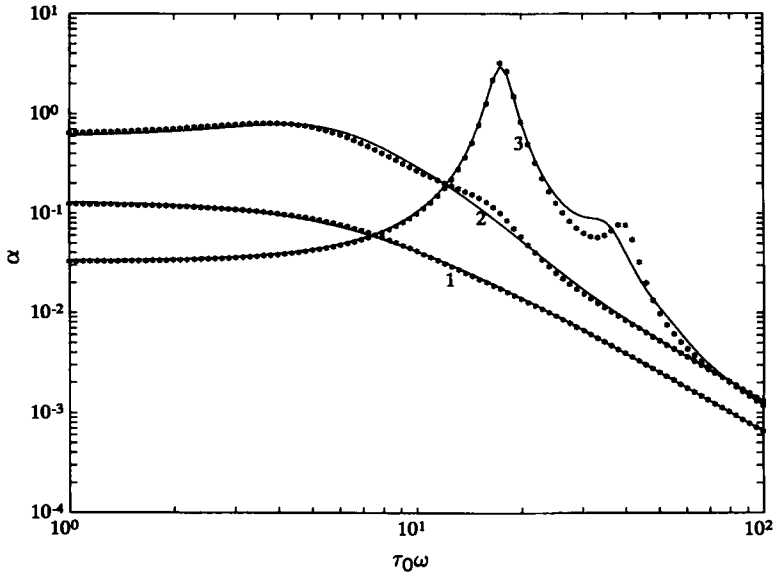


Figure 25. The same as in Fig. 24 for $\gamma = -20$.

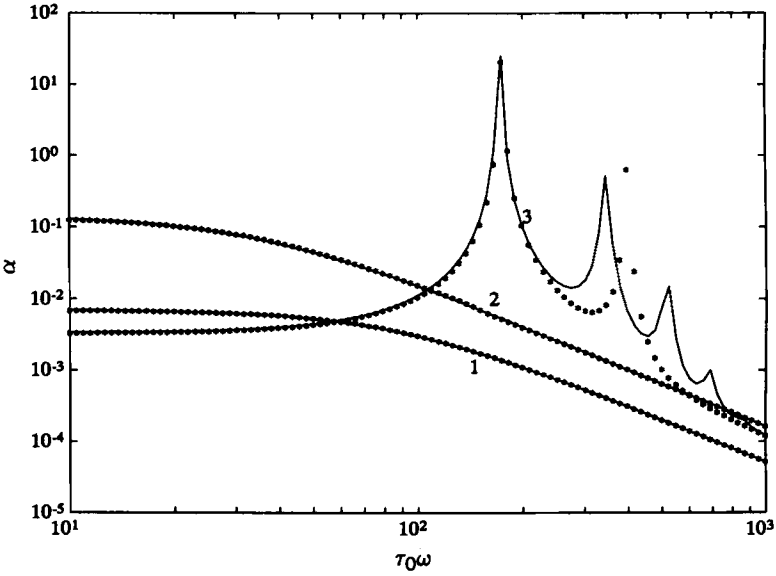


Figure 26. The same as in Fig. 24 for $\gamma = -200$ (the low noise case).

results coincide with those given in Ref. 20. One can see by inspection of these figures that the effective eigenvalue method gives a good quantitative description of the main features of the spectrum $\alpha(\omega)$ in all regions of interest:[19-21,47] $|a| < b$ (the so-called locked region), $|a| \lesssim b$, and $|a| > b$ (the unlocked region). As expected at small damping ($D \ll 1$) the approximate solution gives only a qualitative description of the second-harmonic contribution to $\alpha(\omega)$ and does not describe higher harmonics at all (see Fig. 26). This is due to our truncation of the hierarchy (455) at the second level. The effective eigenvalue method may, however, be applied in this instance as well if we truncate the hierarchy (455) at a higher level. To give a qualitative description of the second-harmonic band, for instance, one would truncate the hierarchy at the third level and calculate the effective eigenvalue for $C_3(t)$.

V. CONCLUSIONS

The purpose of this review was to demonstrate how the effective eigenvalue method allied with the Langevin equation may be applied with much success to a variety of stochastic problems in the theory of the Brownian movement. The most noteworthy feature of the method is that it yields closed-form expressions, which in many cases may accurately describe the low-frequency relaxation behavior of the system in question.

The method yields formulae for the transverse and longitudinal components of the dynamic susceptibility of nematic liquid crystals entirely in terms of the equilibrium order parameter. These are valid for any axially symmetric potential. This is not at all obvious in the context of the Fokker-Planck equation. Furthermore, the transverse and longitudinal orientation factors in the low barrier height limit are connected by a simple general relation. The same considerations hold for magnetic relaxation. In addition, it is possible to easily deduce the formula[6] describing ferromagnetic resonance in the simplest uniaxial potential of the crystalline anisotropy. We emphasize that the effective eigenvalue cannot describe the longitudinal relaxation process for large potential barriers due to the exponential nature of the lowest eigenvalue, which causes the longest relaxation time to dominate the relaxation process. On the other hand, in the transverse case (which in the magnetic problem is crucial to the discussion of ferromagnetic resonance) the lowest eigenvalue does not exhibit exponential behavior and the system is accurately described by the effective eigenvalue for all σ values. It should be noted that the asymptotic formula of Brown [15] namely

$$\frac{\tau_{\parallel}}{\tau_D} = \frac{\sqrt{\pi}}{2}\sigma^{-3/2}e^{\sigma} \tag{476}$$

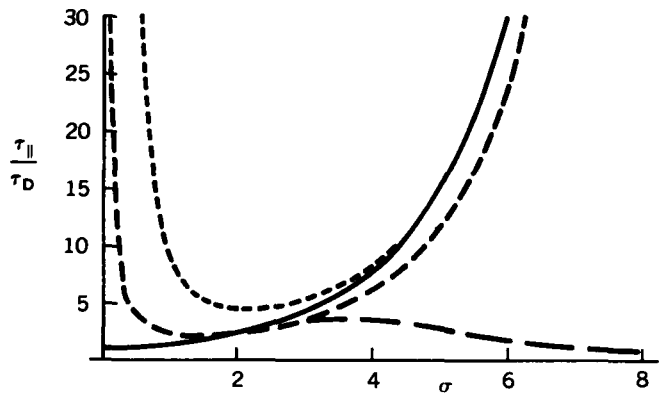


Figure 27. Numerical solution for the longest relaxation time for a longitudinal applied field (solid line) as computed from Eq. (224) compared with solution yielded by perturbation theory (largest dashes) Eq. (223) and Brown's asymptotic solution Eq. (476) (next largest dashing) and Brown's solution as modified by Storonkin Eq. (477).

may be substantially improved for large σ by incorporating the additional terms in σ^{-1} given in the asymptotic expansion of Storonkin [30]. His formula reads

$$\frac{\tau_\parallel}{\tau_D} = \frac{\sqrt{\pi}}{2}\sigma^{-3/2}e^{\sigma}\left(1 + \frac{1}{\sigma} + \frac{7}{4\sigma^2} + \cdots\right) \tag{477}$$

which agrees completely with the exact solution for large σ (see Fig. 27 and Table II) unlike Eq. (476).

At the time of going to Press we have shown in conjunction with D. S. F. Crothers and J. T. Waldron using the definition of the correlation time given in Eq. (12) and Eq. (224) that the solution for the longitudinal

TABLE II

Brown's formula Eq. (476) compared with Storonkin's formula Eq. (477) and the exact solution. Eq. (477) renders the exact asymptotic behaviour. The exact solution is obtained by numerical solution of Eq. (224) for the lowest eigenvalue.

σ	Exact Solution	Storonkin	Brown
10	694.428	689.882	617.291
20	5.07188×10^{6}	5.06856×10^{6}	4.807×10^{6}
42	5.80373×10^{15}	5.80335×10^{15}	5.662×10^{15}
52	9.23455×10^{19}	9.23424×10^{19}	9.054×10^{19}
60	2.21503×10^{23}	2.21500×10^{23}	2.177×10^{23}

correlation time is

$$\frac{\tau_{\|}}{\tau} = M\left(1, \tfrac{5}{2}, \sigma\right) + \frac{3/2}{M\left(\frac{3}{2}, \frac{5}{2}, \sigma\right)} \sum_{n=1}^{\infty} \left[\frac{\left(n + \frac{3}{4}\right)\Gamma\left(n + \frac{1}{2}\right)\Gamma\left(n + \frac{3}{2}\right)}{(n+1)\Gamma\left(2n + \frac{5}{2}\right)^2} \right.$$

$$\left. \times (-\sigma^2)^n M\left(n + \tfrac{3}{2}, 2n + \tfrac{5}{2}, \sigma\right) M\left(\left(n + 1, 2n + \tfrac{5}{2}, \sigma\right)\right] \right. \quad (478)$$

where $M(a, b, \sigma)$ is Kummer's function of given a, b [44]. In addition Crothers has shown from Eq. (478) by the method of the steepest descents that in the limit of high σ

$$\frac{\tau_{\|}}{\tau} = \frac{\sqrt{\pi}}{2} \sigma^{-3/2} e^{\sigma} \left(1 + \frac{1}{\sigma}\right) \quad (479)$$

which agrees with Eq. (477) when truncated at the second term.

The foregoing problems concern the linear relaxation behavior. We have demonstrated using the Kerr effect relaxation as an example how the method may be extended to nonlinear relaxation problems.

On applying the effective eigenvalue technique to the model of Brownian motion in a tilted cosine potential we find that it yields simple analytic formulae for the response of a Josephson junction to a weak ac current, which agree closely with the exact solution. This has the merit that we now have analytic formulae for the impedance in the presence of noise, the spectrum broadening, and the resonant frequency. The inclusion of noise has a profound effect on the operation of the junction, because it removes[57, 58] the resonance singularity of the noiseless case. In addition to the derivation of these approximate formulae, we have shown that the exact linear response of the Josephson junction to a weak ac current is determined by the continued fraction $S_n(\omega)$ (as is apparent from Eqs. (393) and (394)). This representation of the exact solution has the advantage that it can easily be adapted for iteration to determine the nonlinear response, in particular the important frequency-dependent dc component of that response. In addition, the connection of the linear ac response with the dc current voltage characteristics when expressed as a continued fraction is also apparent. Substantially the same considerations apply to the ring laser gyro where the exact solution is more complicated because the desired quantity is the autocorrelation function of the beat signal and its spectrum. The effective eigenvalue method radically simplifies the solution of this problem, because it yields an analytic expression for the

beat signal spectrum analogous to the ferromagnetic resonance spectrum described above. The solutions just described may also be applied with appropriate changes of notation to the analogous stochastic problems listed in Refs. 3 and 22, namely the phase-locked loop, superionic conductors, etc.

If the method can be extended to systems that are governed by a matrix continued fraction[3] (to which the solution of many relaxation problems occurring in phase space may be reduced), then it would be possible to use it to include inertial effects. We believe that the synergetic approach that we have adopted is fruitful, because it demonstrates the basic similarity of the relaxation problems that may be treated using the theory of the Brownian motion.

APPENDIX A: DETERMINATION OF THE DIFFERENTIAL-DIFFERENCE EQUATION FOR $\langle X_{31} \rangle$ FROM THE LANGEVIN EQUATION

The comparison of the analytic expressions for the effective relaxation times $\tau_{\parallel}$ and $\tau_{\perp}$ for the nematic potential $V(\vartheta) = -V_0 \cos^2 \vartheta$ with the results obtained by solving the appropriate secular equation

$$\det(s\mathbf{I} - \mathbf{A}) = 0 \tag{A.1}$$

requires a knowledge of the differential-difference equation for second-, third-, and higher-order spherical harmonics. We illustrate, for example, how the differential-difference equation for $\langle X_{31} \rangle$ may be obtained by averaging the noninertial Langevin equation. We recall that for the Meier-Saupe mean field $\mathbf{E}_0 = \mathbf{k} E_0 \cos \vartheta$, the Langevin equations are

$$\dot{\mu}_x(t) = \frac{1}{\zeta}\big(\lambda_y(t)\mu_z(t) - \lambda_z(t)\mu_y(t)\big) - \frac{\mu_x(t)\mu_z^2(t)}{\mu\zeta} E_0 \tag{A.2}$$

$$\dot{\mu}_y(t) = \frac{1}{\zeta}\big(\lambda_z(t)\mu_x(t) - \lambda_x(t)\mu_z(t)\big) - \frac{\mu_y(t)\mu_z^2(t)}{\mu\zeta} E_0 \tag{A.3}$$

$$\dot{\mu}_z(t) = \frac{1}{\zeta}\big(\lambda_x(t)\mu_y(t) - \lambda_y(t)\mu_x(t)\big) + \frac{\mu^2\mu_z(t) - \mu_z^3(t)}{\mu\zeta} E_0 \tag{A.4}$$

We now make the transformation

$$\mu_x(t) \to x(t) \tag{A.5}$$

$$\mu_y(t) \to y(t) \tag{A.6}$$

$$\mu_x(t)\mu_z^2(t) \to z(t) \tag{A.7}$$

so that Eqs. (A.2)–(A.4) become

$$\dot{x}(t) = \frac{1}{\zeta}\left(\lambda_y(t)\sqrt{\frac{z(t)}{x(t)}} - \lambda_z(t)y(t)\right) - \frac{z(t)}{\mu\zeta}E_0 \tag{A.8}$$

$$\dot{y}(t) = \frac{1}{\zeta}\left(\lambda_z(t)x(t) - \lambda_x(t)\sqrt{\frac{z(t)}{x(t)}}\right) - \frac{y(t)}{\mu\zeta}\frac{z(t)}{x(t)}E_0 \tag{A.9}$$

$$\dot{z}(t) = \frac{1}{\zeta}\left[2\lambda_x(t)y(t)\sqrt{\frac{z(t)}{x(t)}} + \lambda_y(t)\left(\sqrt{\frac{z^3(t)}{x^3(t)}} - 2\sqrt{x^3(t)z(t)}\right)\right.$$

$$\left. -\lambda_z(t)\frac{y(t)z(t)}{x(t)}\right] + \frac{z(t)}{\zeta\mu}\left(2\mu^2 - 3\frac{z(t)}{x(t)}\right)E_0 \tag{A.10}$$

We now apply formula (39) for the noise-induced drift terms to Eq. (A.10), just as in Section II.B. The relevant components of the tensor g_{ij} in Eq. (36) are

$$g_{11} = 0 \qquad g_{12} = \frac{\sqrt{z/x}}{\zeta} \qquad g_{13} = \frac{-y}{\zeta}$$

$$g_{21} = \frac{-\sqrt{z/x}}{\zeta} \qquad g_{22} = 0 \qquad g_{23} = \frac{x}{\zeta} \tag{A.11}$$

$$g_{31} = \frac{2y\sqrt{z/x}}{\zeta} \qquad g_{32} = \frac{1}{\zeta}\left(\sqrt{\frac{z^3}{x^3}} - 2\sqrt{x^3z}\right) \qquad g_{33} = -\frac{yz}{\zeta x}$$

Hence, after tedious algebra we obtain

$$kT\zeta\left(g_{kj}\frac{\partial}{\partial x_k}g_{3j}\right) = \frac{2kT}{\zeta}(x\mu^2 - 6z) \tag{A.12}$$

This is the noise-induced part of z. On averaging over the distribution $W(\{\mu\}, t)$ at time t the deterministic contribution to z is

$$\left\langle \frac{\mu_x\mu_z^2}{\mu\zeta}(2\mu^2 - 3\mu_z^2)E_0 \right\rangle \tag{A.13}$$

Thus, the complete equation averaged over $W(\{\mu\}, t)$ is

$$\frac{\mathrm{d}}{\mathrm{d}t}\langle z\rangle + \frac{12kT}{\zeta}\langle z\rangle = \frac{2kT}{\zeta}\langle \mu_x\rangle\mu^2 + \frac{E_0}{\mu\zeta}\left\langle \mu_x\mu_z^2(2\mu^2 - 3\mu_z^2)\right\rangle \tag{A.14}$$

We note that

$$X_{31} = \frac{3}{2}(5\mu_z^2 - 1)\mu_x = \frac{3}{2}\left(5\frac{z}{\mu^3} - X_{11}\right) \tag{A.15}$$

so that

$$\langle \dot{X}_{31}\rangle = \frac{15}{2\mu^3}\langle \dot{z}\rangle - \frac{3}{2}\langle \dot{X}_{11}\rangle \tag{A.16}$$

where $\langle \dot{X}_{11}\rangle$ is given by Eq. (51). Therefore, Eq. (A.14) becomes

$$\frac{\mathrm{d}}{\mathrm{d}t}\langle X_{31}\rangle + \frac{6}{\tau_{\mathrm{D}}}\langle X_{31}\rangle\left(1 - \frac{\sigma}{30}\right) = \frac{48\sigma}{35\tau_{\mathrm{D}}}\langle X_{11}\rangle - \frac{4\sigma}{7\tau_{\mathrm{D}}}\langle X_{51}\rangle \tag{A.17}$$

where the spherical harmonic X_{51} is defined as[25]

$$X_{51} = \tfrac{1}{8}\sin\vartheta\cos\phi(315\cos^4\vartheta + 210\cos^2\vartheta + 15)$$

Thus, the calculation shows how the hierarchy of equations may be formed.

APPENDIX B: DERIVATION OF THE EQUATION FOR $\langle H_{ef} M_y \rangle$ FROM THE STOCHASTIC LANDAU-LIFSHITZ-GILBERT EQUATION

To calculate the transverse magnetic susceptibility $\chi_\perp(\omega)$ we first require the equation of motion for $\langle H_{ef} M_y \rangle$. The effective field $H_{ef}(\{\mathbf{M}\})$ for the mean field potential

$$V(\{\mathbf{M}\}) = -K\cos^2\vartheta + K$$

is from Eq. (175)

$$\mathbf{H}_{ef} = -\mathbf{k}\frac{\partial V}{\partial M_z} = \mathbf{k}\frac{2K}{M_s^2}M_z \tag{B.1}$$

Thus, on noting that

$$\frac{\mathrm{d}}{\mathrm{d}t}\left(H_{ef}(\{\mathbf{M}(t)\})M_y(t)\right) = \frac{2K}{M_s^2}\left(M_z(t)\frac{\mathrm{d}}{\mathrm{d}t}M_y(t) + M_y(t)\frac{\mathrm{d}}{\mathrm{d}t}M_z(t)\right) \tag{B.2}$$

we can derive the Langevin equation for $H_{ef}M_y$ from Eqs. (162) and (163). On introducing new variables

$$X(t) = M_x(t) \tag{B.3}$$

$$Y(t) = M_s H_{ef}(\{\mathbf{M}(t)\}M_y(t)/2K \tag{B.4}$$

$$Z(t) = M_z(t) \tag{B.5}$$

we obtain from Eqs. (161)–(163)

$$\begin{aligned}\frac{\mathrm{d}}{\mathrm{d}t}X(t) &= 2g'KY(t) - \frac{2h'K}{M_s^2}X(t)Z^2(t) + F_x(t)H(t)\\ &\quad + h'\big(M_s^2 - X^2(t)\big)h_x(t)\\ &\quad - \left(g'M_sZ(t) + h'M_s\frac{X(t)Y(t)}{Z(t)}\right)h_y(t)\\ &\quad + \left(g'M_s^2\frac{Y(t)}{Z(t)} - h'X(t)Z(t)\right)h_z(t)\end{aligned} \tag{B.6}$$

$$\begin{aligned}\frac{\mathrm{d}}{\mathrm{d}t}Y(t) &= -2g'\frac{K}{M_s^2}Z^2(t)X(t) - 4h'\frac{K}{M_s^2}Z^2(t)Y(t) + 2h'KY(t)\\ &\quad + \left[F_y(t)\frac{1}{M_s}Z(t) + \frac{Y(t)}{Z(t)}F_z(t)\right]H(t)\\ &\quad + \left[g'\left(Z^2(t) - M_s^2\frac{Y^2(t)}{Z^2(t)}\right) - 2h'X(t)Y(t)\right]h_x(t)\\ &\quad + \left[h'M_s\left(Z(t) - \frac{2Y^2(t)}{Z(t)}\right) + g'M_s\frac{X(t)Y(t)}{Z(t)}\right]h_y(t)\\ &\quad + \left[\frac{h'Y(t)}{Z(t)}\big(M_s^2 - 2Z^2(t)\big) - g'X(t)Z(t)\right]h_z(t)\end{aligned} \tag{B.7}$$

$$\begin{aligned}\frac{\mathrm{d}}{\mathrm{d}t}Z(t) &= \frac{2h'K}{M_s^2}Z(t)\big(M_s^2 - Z^2(t)\big) + F_z(t)H(t)\\ &\quad + h'\big(M_s^2 - Z^2(t)\big)h_z(t) - \left(g'M_s^2\frac{Y(t)}{Z(t)} + h'X(t)Z(t)\right)h_x(t)\\ &\quad + \big(g'M_sX(t) - h'M_sY(t)\big)h_y(t)\end{aligned} \tag{B.8}$$

We may now apply Eq. (39) to calculate the noise induced drift terms in Eqs. (B.6)–(B.8), just as in Section II.B and Appendix A. The tensor g_{ik} is given by Equation B.9.

$$g_{ik} \equiv \begin{cases} h'(M_s^2 - X^2) & -\left(g'M_s Z + \frac{h'M_s XY}{Z}\right) & \left(\frac{g'M_s^2 Y}{Z} - h'XZ\right) \\ \left[g'\left(Z^2 - \frac{M_s^2 Y^2}{Z^2}\right) - 2h'XY\right] & \left[h'M_s\left(Z - \frac{2Y^2}{Z}\right) + \frac{g'M_s XY}{Z}\right] & \left[\frac{h'Y}{Z}(M_s^2 - 2Z^2) - g'XZ\right] \\ -\left(\frac{g'M_s^2 Y}{Z} + h'XZ\right) & (g'M_s X - h'M_s Y) & h'(M_s^2 - Z^2) \end{cases} \tag{B.9}$$

The quantity of interest, namely

$$g_{jk}\frac{\partial}{\partial x_j}g_{2k}$$

is

$$\begin{aligned} g_{jk}\frac{\partial}{\partial x_j}g_{2k} = {} & g_{11}\frac{\partial}{\partial x_1}g_{21} + g_{21}\frac{\partial}{\partial x_2}g_{21} + g_{31}\frac{\partial}{\partial x_3}g_{21} \\ & + g_{12}\frac{\partial}{\partial x_1}g_{22} + g_{22}\frac{\partial}{\partial x_2}g_{22} + g_{32}\frac{\partial}{\partial x_3}g_{22} \\ & + g_{13}\frac{\partial}{\partial x_1}g_{23} + g_{23}\frac{\partial}{\partial x_2}g_{23} + g_{33}\frac{\partial}{\partial x_3}g_{23} \end{aligned} \tag{B.10}$$

where $x_1 = X$, $x_2 = Y$, and $x_3 = Z$, can be evaluated after considerable algebra. The result is

$$g_{jk}\frac{\partial}{\partial x_j}g_{2k} = -6\left(g'^2 + h'^2\right)M_s^2 Y \tag{B.11}$$

Thus, on returning to the old variables we obtain the equation for the starting value of $H_{ef}M_y$ at time t

$$\begin{aligned} \frac{M_s}{2K}\frac{d}{dt}\left(H_{ef}M_y\right) = {} & -g'H_{ef}M_zM_x - \frac{2h'M_s^2H_{ef}M_y}{M_s} + h'M_sM_yH_{ef} \\ & + \left(F_y\frac{M_z}{M_s} + \frac{M_s}{2K}\frac{H_{ef}M_y}{M_z}\right)H(t) \\ & - \frac{3\mu}{2K}\left(h'^2 + g'^2\right)M_s^3H_{ef}M_y \end{aligned} \tag{B.12}$$

or

$$\begin{aligned} & \frac{d}{dt}\left(H_{ef}M_y\right) + \left(\frac{3}{\tau_N} - \frac{\nu K}{kT}\right)M_yH_{ef} \\ & \quad = -H_{ef}^2\left(g'M_sM_x + 2h'M_zM_y\right) + \left(F_y + \frac{F_zM_y}{M_z}\right)H_{ef}H(t) \end{aligned} \tag{B.13}$$

where

$$\tau_N^{-1} = \mu\left(h'^2 + g'^2\right)M_s^2$$

In the case when the ac probe field $\mathbf{H}(t)$ is applied along the x axis

$$F_y + \frac{F_z M_y}{M_z} = h'\left(M_s^2 - M_y^2 - M_x M_y\right) - \frac{g' M_s M_y^2}{M_z} \tag{B.14}$$

On introducing the dimensionless variables

$$u_x = \frac{M_x}{M_s} = \sin\vartheta\cos\phi \qquad u_y = \frac{M_y}{M_s} = \sin\vartheta\sin\phi \qquad u_z = \frac{M_z}{M_s} = \cos\vartheta \tag{B.15}$$

and

$$\xi(t) = \frac{\nu M_s H(t)}{kT} \qquad \alpha = \frac{h'}{g'} \qquad \sigma = \frac{\nu K}{kT} \tag{B.16}$$

we obtain from Eqs. (170) and (B.13) and (B.14) (having been averaged) the equations governing ferromagnetic resonance:

$$\frac{\mathrm{d}}{\mathrm{d}t}\langle u_x\rangle + \frac{1}{\tau_N}\langle u_x\rangle = \frac{\sigma}{\alpha\tau_N}\langle u_z u_y\rangle - \frac{\sigma}{\tau_N}\langle u_x u_z^2\rangle + \frac{\xi(t)}{2\tau_N}\left(1 - \langle u_x^2\rangle\right) \tag{B.17}$$

$$\begin{aligned}\frac{\mathrm{d}}{\mathrm{d}t}\langle u_z u_y\rangle + \frac{1}{\tau_N}(3-\sigma)\langle u_z u_y\rangle &= -\frac{\sigma}{\alpha\tau_N}\langle u_x u_z^2\rangle - \frac{2\sigma}{\tau_N}\langle u_z^3 u_y\rangle \\ &\quad + \frac{\xi(t)}{2\tau_N}\left[\left\langle u_z\left(1 - u_x^2 - u_x u_y\right)\right\rangle - \frac{1}{\alpha}\langle u_y^2\rangle\right]\end{aligned} \tag{B.18}$$

Equations (B.17) and (B.18) can also be written in terms of spherical

harmonics X_{nm} as

$$\frac{d}{dt}\langle X_{11}\rangle + \frac{1}{\tau_N}\left(1+\frac{\sigma}{5}\right)\langle X_{11}\rangle = -\frac{2\sigma}{15\tau_N}\langle X_{31}\rangle + \frac{\sigma}{3\alpha\tau_N}\langle X_{2,-1}\rangle + \frac{\xi(t)}{6\sigma_N}\left[2-\frac{1}{2}\langle X_{22}\rangle + \langle P_2\rangle\right] \quad \text{(B.19)}$$

$$\frac{d}{dt}\langle X_{2,-1}\rangle + \frac{1}{\tau_N}\left(3-\frac{\sigma}{7}\right)\langle X_{2,-1}\rangle = -\frac{2\sigma}{5\alpha\tau_N}\langle X_{31}\rangle - \frac{3\sigma}{5\alpha\tau_N}\langle X_{11}\rangle - \frac{12\sigma}{35\tau_N}\langle X_{4,-1}\rangle + \frac{\xi(t)}{\tau_N}\left[\frac{6}{5}\langle P_1\rangle + \frac{3}{10}\langle P_3\rangle - \frac{1}{20}(\langle X_{32}\rangle + \langle X_{3,-2}\rangle) - \frac{1}{2\alpha}\left(1-\langle P_2\rangle - \frac{1}{2}\langle X_{22}\rangle\right)\right] \quad \text{(B.20)}$$

where

$$\langle X_{nm}\rangle = \langle P_n^{|m|}\cos m\phi\rangle \qquad \langle X_{n,-m}\rangle = \langle P_n^{|m|}\sin m\phi\rangle \qquad m \le n$$

P_n is the Legendre polynomial of order n and $P_n^{|m|}$ is the associated Legendre function. In the absence of the probe field $\mathbf{H}(t)$, Eqs. (B.19) and (B.20) can be derived from the hierarchy of differential-difference equations obtained in Refs. 11 and 51 from the underlying Fokker-Planck equation. The above procedure shows how this hierarchy is formed.

APPENDIX C: A FORTRAN PROGRAM FOR CALCULATING THE IMPEDANCE OF A JOSEPHSON JUNCTION

This simple program has been used to calculate the impedance of a Josephson junction. In the program:

X = bias parameter x.
G = barrier height parameter γ.
KM = level of truncation of the continued fractions. (KM depends on x and γ).
A(AE) = normalized impedance $Z(\omega)/R$ for the exact (approximate) solutions respectively.
w = normalized frequency $\omega\tau_0$.

```
      SUBROUTINE MCAW(A,AE,w)
      REAL X,G,KMI,G1
      COMPLEX A,E,S,AE,Z,A1,E1,S1,Z1,XG,w1
```

```
      COMMON KM,X,G
      XG=X*G*(0.0,1.0)
      w1=(0.0,-2.0)*w/KM
      G1=0.5*G
      Z=G1/(2.0*KM+XG)
      E=G1/(XG+w1+2.0*KM)
      E1=G1/(w1-XG+2.0*KM)
      A=1.0-Z*E
      A1=1.0-CONJG(Z)*E1
      S=1.0-Z*Z
C     CALCULATION OF CONTINUED FRACTIONS
      DO 10 I=1,KM-1
       KMI=2.0*(KM-I)
       w1=(0.0,-4.0)*w/KMI
       Z=G1/(XG+KMI+Z*G1)
       E=G1/(XG+w1+KMI+E*G1)
       E1=G1/(w1-XG+KMI+E1*G1)
       S=1.0-Z*Z*S
       A=1-Z*E*A
       A1=1-CONJG(Z)*E1*A1
    10ENDDO
       S=0.5*G1*Z/(1.0-S)
       S1=CONJG(S)
       Z1=CONJG(Z)
       A=-1.0+A1+A
       AE=1.0-0.5*G1*(Z/((0.0,-1.0)*w+S)+Z1/
         ((0.0,-1.0)*w+S1))
       RETURN
       END
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