

Molecules with an induced dipole moment in a stochastic electric field

Y. B. Band

Department of Chemistry, Department of Physics, Department of Electro-Optics, and the Ilse Katz Center for Nano-Science, Ben-Gurion University, Beer-Sheva 84105, Israel

Y. Ben-Shimol

Department of Communication Systems Engineering, Ben-Gurion University, Beer-Sheva 84105, Israel

(Received 13 June 2013; published 30 October 2013)

The mean-field dynamics of a molecule with an induced dipole moment (e.g., a homonuclear diatomic molecule) in a deterministic and a stochastic (fluctuating) electric field is solved to obtain the decoherence properties of the system. The average (over fluctuations) electric dipole moment and average angular momentum as a function of time for a Gaussian white noise electric field are determined via perturbative and nonperturbative solutions in the fluctuating field. In the perturbative solution, the components of the average electric dipole moment and the average angular momentum along the deterministic electric field direction do not decay to zero, despite fluctuations in all three components of the electric field. This is in contrast to the decay of the average over fluctuations of a magnetic moment in a Gaussian white noise magnetic field. In the nonperturbative solution, the component of the average electric dipole moment and the average angular momentum in the deterministic electric field direction also decay to zero.

DOI: 10.1103/PhysRevE.88.042149

PACS number(s): 05.40.Ca, 03.65.Yz, 02.50.Fz

I. INTRODUCTION

Quantum decoherence arises from the interaction of a system with its environment. This interaction gives rise to the entanglement of the system and the environment and leads to the suppression of interference because the phases of the quantum amplitudes in the system wave function are adversely affected by the entanglement. Here we consider the decoherence of a system that has an induced electric dipole moment, $d_i = \alpha_{ij} E_j$, where α_{ij} is the polarizability tensor and E_j is the j th component of an external electric field that is in contact with an environment (a bath). Examples of such systems include homonuclear diatomic molecules, such as H_2 [1] and N_2 [2], polyatomic molecules with no permanent electric dipole moment (i.e., a molecule which, if fixed in space so that it cannot rotate, has a vanishing electric dipole moment when no external electric field is present), and a mesoscopic or macroscopic system, such as a colloidal particle having no permanent dipole moment [3]. The dynamics of such systems that are in contact with an environment can be represented by evolving the system in an effective electric field, $\mathbf{E}^{(\text{eff})} = \mathbf{E}_0 + \mathbf{E}_B(t)$, where \mathbf{E}_0 is the deterministic electric field (which could be time dependent) and $\mathbf{E}_B(t)$ is the electric field which models the influence of the environment (the bath B) on the dipole moment. The field $\mathbf{E}_B(t)$ can be represented by a vector stochastic process $\boldsymbol{\varepsilon}(t)$, where the nature of the environment determines the type of stochastic process. Averaging over fluctuations corresponds to tracing over the environmental degrees of freedom. This yields a reduced nonunitary dynamics wherein the averaged dipole moment and angular momentum decohere in time. This approach was recently used to treat decoherence of spin systems caused by an environment [4] and the decoherence of systems with a permanent dipole moment [5]. The physical properties of the environment determine the statistical properties of $\mathbf{E}_B(t)$, which in turn determine the type of stochastic process $\boldsymbol{\varepsilon}(t)$. A prototype model for fluctuations

is Gaussian white noise, wherein the random process has vanishing correlation time, but other types of noise are also commonly encountered [4,6–10].

II. CLASSICAL DYNAMICS

Consider a static electric field \mathbf{E} in the direction of the space-fixed z axis and obtain the classical equations of motion of the system. In spherical coordinates, the kinetic energy is $T = \frac{1}{2}I(\dot{\theta}^2 + \sin^2\theta\dot{\phi}^2)$ and the potential energy is $U = -\mathbf{d} \cdot \mathbf{E} = -\frac{\alpha}{2}(\mathbf{n} \cdot \mathbf{E})^2 = -\frac{\alpha E^2}{2} \cos^2\theta$, where θ and ϕ are the polar and azimuthal angles of the diatomic axis, I is the moment of inertia, and \mathbf{n} is the unit vector in the direction of the axis of the system (e.g., the unit vector along the axis of a homonuclear diatomic molecule). For simplicity we assumed that the polarizability α has a component only along the axis of the molecule so that the induced dipole moment is given by $\mathbf{d} = (\alpha/2)(\mathbf{n} \cdot \mathbf{E})\mathbf{n}$. Hence the Lagrangian is

$$\mathcal{L}(\theta, \phi, \dot{\theta}, \dot{\phi}) = T - U = \frac{1}{2}I(\dot{\theta}^2 + \sin^2\theta\dot{\phi}^2) + \frac{\alpha E^2}{2} \cos^2\theta. \quad (1)$$

The Euler-Lagrange equations of motion are

$$\begin{aligned} 0 &= \frac{\partial \mathcal{L}}{\partial \phi} - \frac{d}{dt} \frac{\partial \mathcal{L}}{\partial \dot{\phi}} = -I \frac{d}{dt} (\sin^2\theta \dot{\phi}) \\ \Rightarrow \dot{\phi} &= \frac{\omega}{\sin^2\theta}, \quad \omega = \text{const}, \quad (2) \\ 0 &= \frac{\partial \mathcal{L}}{\partial \theta} - \frac{d}{dt} \frac{\partial \mathcal{L}}{\partial \dot{\theta}} \Rightarrow \ddot{\theta} + \frac{\alpha E^2}{2I} \sin 2\theta - \frac{\omega^2 \sin 2\theta}{2 \sin^4\theta} = 0. \quad (3) \end{aligned}$$

The dynamics are relatively simple since the z component of the angular momentum, $L_z = \frac{\partial \mathcal{L}}{\partial \dot{\phi}} = I \sin^2\theta \dot{\phi} \equiv I\omega$, is conserved because there is no component of torque, $\boldsymbol{\tau} = \mathbf{d} \times \mathbf{E} = \frac{\alpha}{2}(\mathbf{n} \cdot \mathbf{E})(\mathbf{n} \times \mathbf{E})$, along z . The second constant of the

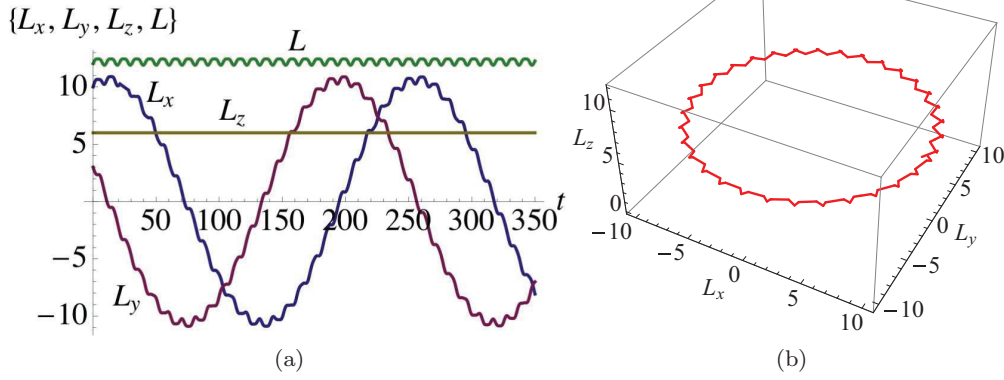


FIG. 1. (Color online) (a) $L_x(t), L_y(t), L_z(t)$, and $L(t) = \sqrt{L_x^2(t) + L_y^2(t) + L_z^2(t)}$ (green curve) for $\mathbf{n}(0) = (\sin(\pi/4) \cos(\pi/4), \sin(\pi/4) \sin(\pi/4), \cos(\pi/4))$ and $(L_x(0), L_y(0), L_z(0)) = (10, 3, 6)$. (b) Parametric plot of $\mathbf{L}(t)$.

motion is the total energy \mathcal{E} ,

$$\mathcal{E} = T + U = \frac{1}{2}I \left(\dot{\theta}^2 + \frac{\omega^2}{\sin^2 \theta} \right) - \frac{\alpha E^2}{2} \cos^2 \theta. \quad (4)$$

Another way of expressing the classical equations of motion is in terms of the angular momentum \mathbf{L} and the unit vector \mathbf{n} ,

$$\dot{\mathbf{n}} = -\frac{1}{I} \mathbf{L} \times \mathbf{n} = -\boldsymbol{\omega} \times \mathbf{n}, \quad (5)$$

$$\dot{\mathbf{L}} = -\alpha(\mathbf{n} \cdot \mathbf{E})(\mathbf{E} \times \mathbf{n}). \quad (6)$$

Figure 1 shows $L_x(t), L_y(t), L_z(t)$, and $L(t) = \sqrt{L_x^2(t) + L_y^2(t) + L_z^2(t)}$ versus time, and Figs. 2 and 3 show $n_x(t), n_y(t)$, and $n_z(t)$ versus time. The dimensionless parameters used in these calculations are $\alpha = 1$, $I = 20$, and $(E_x, E_y, E_z) = (0, 0, 1)$, and the initial conditions are taken as $(L_x(0), L_y(0), L_z(0)) = (10, 3, 6)$, and $\mathbf{n}(0) = (\sin(\pi/4) \cos(\pi/4), \sin(\pi/4) \sin(\pi/4), \cos(\pi/4)) = (1/2, 1/2, 1/\sqrt{2})$. The dynamics is *almost* periodic with a period of about 250 (dimensionless units). This “quasiperiodicity” is a general feature of systems with as many degrees of freedom as conserved dynamical quantities [11,12].

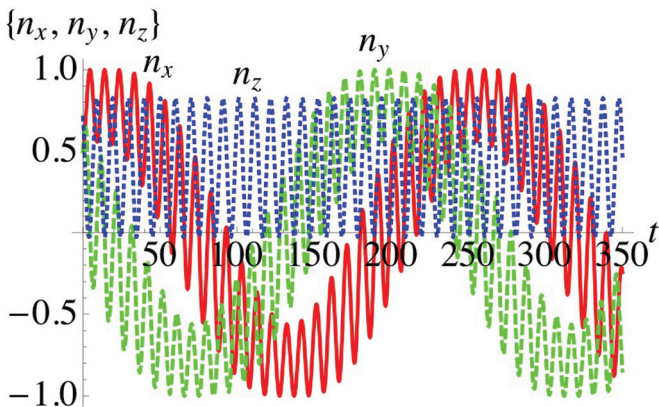


FIG. 2. (Color online) $n_x(t), n_y(t)$, and $n_z(t)$ vs time for $n_x(0) = n_y(0) = 1/2, n_z(0) = 1/\sqrt{2}$ [initial conditions for $\mathbf{L}(t)$ given in Fig. 1 caption].

III. QUANTUM TREATMENT

The Hamiltonian for the system is given by $H = \frac{\hat{L}^2}{2I} - \hat{\mathbf{d}} \cdot \mathbf{E}$; taking the induced dipole moment operator to have a component only along the system axis, $\hat{\mathbf{d}} = (\alpha/2)(\hat{\mathbf{n}} \cdot \mathbf{E})\hat{\mathbf{n}}$, where $\hat{\mathbf{n}}$ is the vector operator of unit length in the direction of the system axis, we obtain [13,14]

$$H = \frac{\hat{L}^2}{2I} - \frac{\alpha}{2}(\hat{\mathbf{n}} \cdot \mathbf{E})^2. \quad (7)$$

The Heisenberg equations of motion for $\hat{\mathbf{n}}$ and $\hat{\mathbf{L}}$, $\dot{\hat{\mathbf{n}}} = \frac{i}{\hbar} [H, \hat{\mathbf{n}}]$ and $\dot{\hat{\mathbf{L}}} = \frac{i}{\hbar} [H, \hat{\mathbf{L}}]$, determine the dynamics. For $\dot{\hat{\mathbf{n}}}$ we find

$$\dot{\hat{\mathbf{n}}} = \frac{i}{2\hbar I} [\hat{L}^2, \hat{\mathbf{n}}] - \frac{i\alpha}{2\hbar} [(\hat{\mathbf{n}} \cdot \mathbf{E})^2, \hat{\mathbf{n}}]. \quad (8)$$

Using the fact that $[\hat{L}_i, \hat{n}_j] = i\hbar \epsilon_{ijk} \hat{n}_k$, we obtain $[\hat{L}^2, \hat{\mathbf{n}}] = 2i\hbar [\hat{\mathbf{L}} \times \hat{\mathbf{n}} + i\hbar \hat{\mathbf{n}}]$. Because $[\hat{n}_i, \hat{n}_j] = 0$ for all i and j , the second term on the right-hand side of Eq. (8) vanishes, and we find

$$\dot{\hat{\mathbf{n}}} = -\frac{1}{I} [\hat{\mathbf{L}} \times \hat{\mathbf{n}} + i\hbar \hat{\mathbf{n}}]. \quad (9)$$

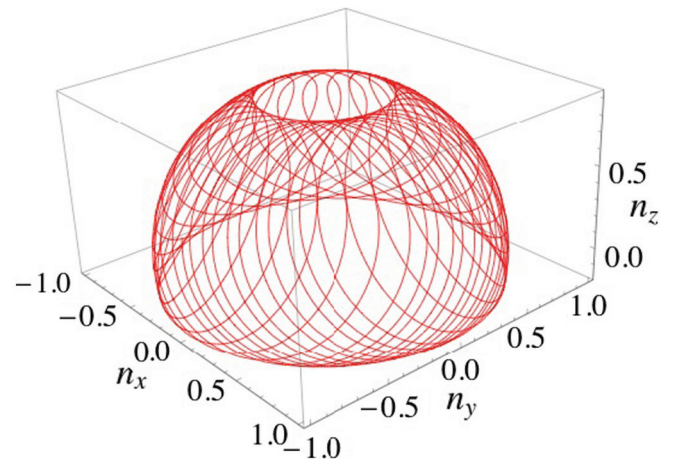


FIG. 3. (Color online) Parametric plot of $\mathbf{n}(t)$ vs time.

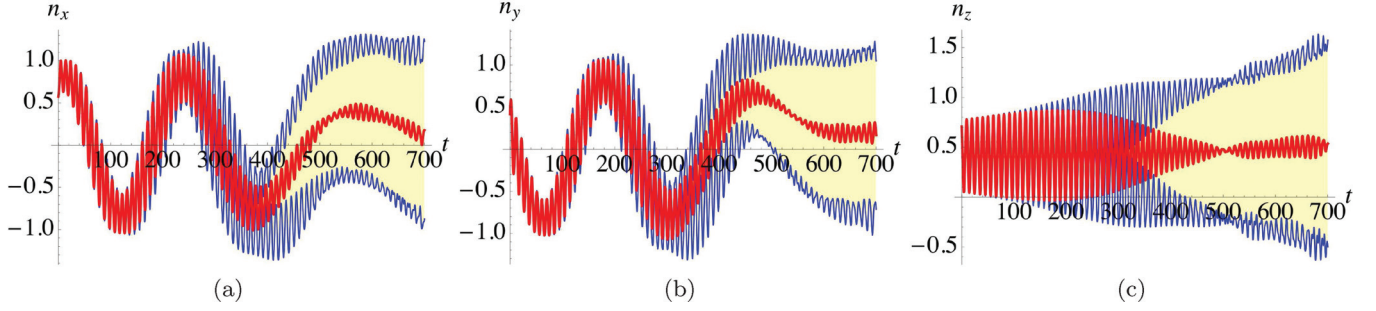


FIG. 4. (Color online) Average and standard deviation of $n_x(t), n_y(t), n_z(t)$ vs time obtained for stochastic dynamics using Eqs. (15) and (16), with $\varepsilon_x(t), \varepsilon_y(t)$, and $\varepsilon_z(t)$ fields taken as Gaussian white noise and volatility $\sigma = 0.02$.

The torque on the molecule due to the presence of the external field, $\hat{\mathbf{L}} = \frac{i}{\hbar}[H, \hat{\mathbf{L}}]$, is given by

$$\dot{\hat{\mathbf{L}}} = -\alpha(\hat{\mathbf{n}} \cdot \mathbf{E})(\mathbf{E} \times \hat{\mathbf{n}}). \quad (10)$$

Since the angular momentum is not conserved, the solution of the Heisenberg equations of motion would require a basis set calculation including many angular momentum states; doing so with a stochastic electric field (see Sec. IV) would be very tedious. Therefore, we develop a mean-field approach.

A. Mean-field dynamics

If the initial angular momentum of the molecule is large compared to \hbar , a semiclassical treatment can be a good approximation. Setting $\hbar = 0$ in Eq. (9) allows a semiclassical solution for the expectation values $\langle \hat{\mathbf{n}}(t) \rangle$ and $\langle \hat{\mathbf{L}}(t) \rangle$. The semiclassical equations are equivalent to the classical solution presented in Sec. II and are valid for an arbitrary direction of \mathbf{E} . The mean-field theory treatment takes the expectation values of Eqs. (9) and (10), replacing the expectation value of the product $\hat{\mathbf{L}} \times \hat{\mathbf{n}}$ by the product of the expectation values [15–18] and taking the limit as $\hbar \rightarrow 0$ on the right-hand side of (9):

$$\langle \dot{\hat{\mathbf{n}}} \rangle = -\frac{1}{I} \langle \hat{\mathbf{L}} \rangle \times \langle \hat{\mathbf{n}} \rangle, \quad (11)$$

$$\langle \dot{\hat{\mathbf{L}}} \rangle = -\alpha(\langle \hat{\mathbf{n}} \rangle \cdot \mathbf{E})(\mathbf{E} \times \langle \hat{\mathbf{n}} \rangle). \quad (12)$$

The nonlinear equations of motion, (11) and (12) [which are the same as Eqs. (5) and (6)], must be solved simultaneously.

IV. STOCHASTIC DYNAMICS

We now consider the dynamics in the presence of a stochastic electric field, so the total electric field is taken to be the sum of a deterministic field and a stochastic field, $\mathbf{E} = \mathbf{E}_0 + \boldsymbol{\varepsilon}(t)$, where $\boldsymbol{\varepsilon}(t)$ is a stochastic process. We solve for the dynamics in two ways. First, we treat the stochastic field perturbatively, by dropping the term in the dynamical equations of motion that is quadratic in $\boldsymbol{\varepsilon}(t)$ and by taking the linear term in $\boldsymbol{\varepsilon}(t)$ to be Gaussian white noise. Then we treat the full (nonperturbative) dynamics, taking $\boldsymbol{\varepsilon}(t)$ to be an Ornstein-Uhlenbeck process.

In what follows, we denote the quantum averages of the unit vector along the axis of the molecule and the angular momentum by $\mathbf{n}(t) \equiv \langle \hat{\mathbf{n}}(t) \rangle$ and $\mathbf{L}(t) \equiv \langle \hat{\mathbf{L}}(t) \rangle$. The averages of these quantities over the stochasticity can be denoted by $\overline{\mathbf{n}(t)}$ and $\overline{\mathbf{L}(t)}$, respectively.

A. Perturbation theory in $\boldsymbol{\varepsilon}(t)$

In Eq. (12), we substitute $\mathbf{E} = \mathbf{E}_0 + \boldsymbol{\varepsilon}(t)$ and expand, keeping only the linear term in $\boldsymbol{\varepsilon}(t)$ and dropping the quadratic term. The resulting equation is of the form of a stochastic differential equation. We need only specify the details of the stochastic electric field $\boldsymbol{\varepsilon}(t)$. We take $\varepsilon_x(t), \varepsilon_y(t)$, and $\varepsilon_z(t)$ to be stochastic processes with zero mean and δ function correlation function $\kappa(t - t')$,

$$\overline{\varepsilon_i(t)} = 0, \quad (13)$$

$$\overline{\varepsilon_i(t)\varepsilon_j(t')} = \kappa(t - t')\delta_{ij} = \varepsilon_0^2 \delta(t - t')\delta_{ij} \quad (14)$$

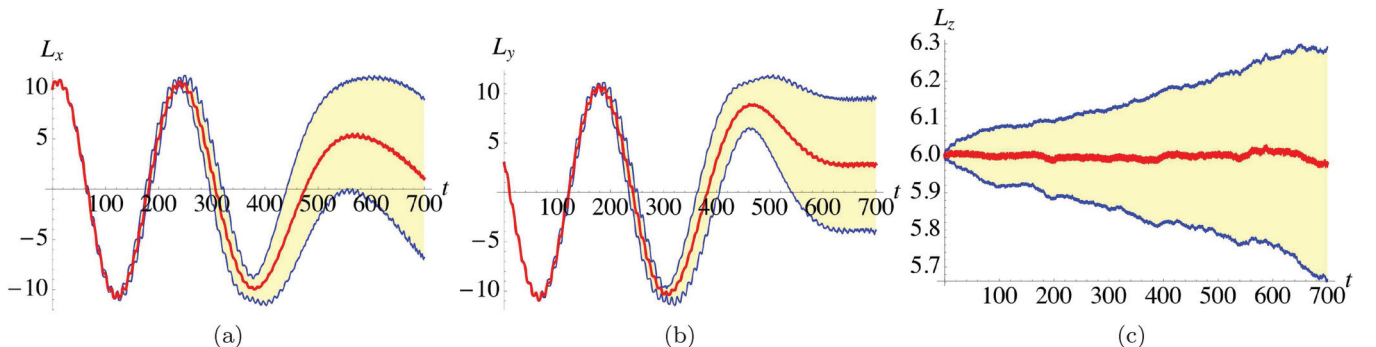


FIG. 5. (Color online) Average and standard deviation of the angular momentum vector ($L_x(t), L_y(t), L_z(t)$) vs time obtained for stochastic dynamics using Eqs. (15) and (16), with $\varepsilon_x(t), \varepsilon_y(t)$, and $\varepsilon_z(t)$ fields taken as Gaussian white noise and volatility $\sigma = 0.02$.

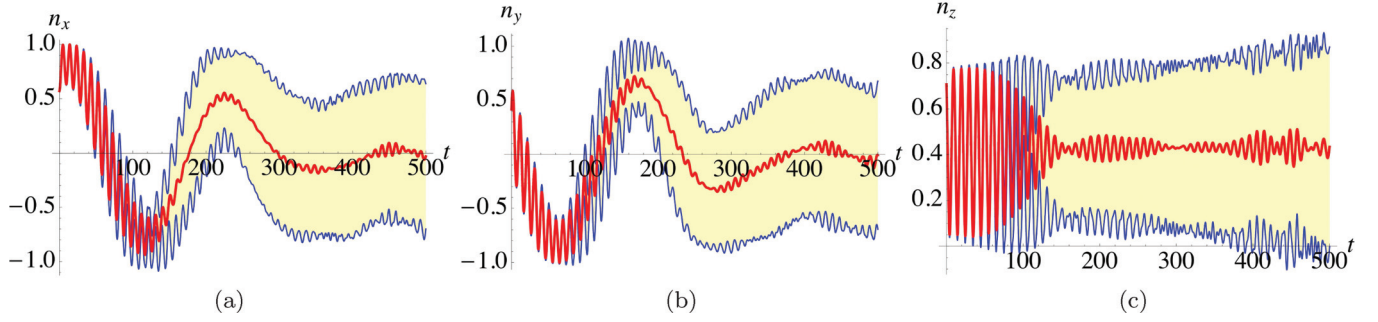


FIG. 6. (Color online) Average and standard deviation of $n_x(t), n_y(t), n_z(t)$ vs time obtained for stochastic dynamics using Eqs. (15) and (16), with $\varepsilon_x(t), \varepsilon_y(t)$, and $\varepsilon_z(t)$ fields taken as Gaussian white noise and volatility $\sigma = 0.1$.

for $i, j = x, y$, and z . We consider a vector Wiener process $\mathbf{W}(t)$, where the white noise $\boldsymbol{\varepsilon}(t)$ can be written as the time derivative of the Wiener process, $\boldsymbol{\varepsilon}(t) = d\mathbf{W}/dt$. More formally, the Wiener process is the integral of the white noise. Equations (11) and (12) form a system of differential equations, which can be written in the standard stochastic differential equation form [6–10],

$$d\mathbf{n}(t) = -\frac{1}{I}\mathbf{L}(t) \times \mathbf{n}(t) dt, \quad (15)$$

$$d\mathbf{L}(t) = -\alpha \{[\mathbf{n}(t) \cdot \mathbf{E}_0][\mathbf{E}_0 \times \mathbf{n}(t)] dt + [\mathbf{n}(t) \cdot \mathbf{E}_0][d\mathbf{W}(t) \times \mathbf{n}(t)] + [\mathbf{n}(t) \cdot d\mathbf{W}(t)][\mathbf{E}_0 \times \mathbf{n}(t)]\}. \quad (16)$$

The stochastic field calculations were carried out using the Mathematica 9.0 built-in command ItoProcess [19] for solving stochastic differential equations, with the stochastic field $\mathbf{W}(t)$ taken as a Wiener process. In the numerical calculations we took $\mathbf{E}_0 = (0, 0, 1)$ (in dimensionless units) and the initial conditions $\mathbf{n}(0)$ and $\mathbf{L}(0)$ as in Figs. 1, 2, and 3. Figures 4 and 5 show the vectors $\mathbf{n}(t) = (n_x(t), n_y(t), n_z(t))$ and $\mathbf{L}(t) = (L_x(t), L_y(t), L_z(t))$ calculated with the stochastic fields, $\varepsilon_x(t), \varepsilon_y(t)$, and $\varepsilon_z(t)$, taken as Gaussian white noise with a small stochastic field strength (volatility), $\varepsilon_0 = 0.02$. The central curves [in red (medium gray)] give the averages $\overline{\mathbf{n}(t)}$ and $\overline{\mathbf{L}(t)}$ in Figs. 4 and 5, and the mean values plus and minus the standard deviations are shown as curves [in blue (dark gray)], with the region between the plus and minus standard deviations shaded [in yellow (light gray)]. Since the initial conditions for the stochastic field is taken to be

$\boldsymbol{\varepsilon}(0) = \mathbf{0}$ and ε_0 is small, the dynamical variables $\mathbf{n}(t)$ and $\mathbf{L}(t)$ start off very much like the variables calculated without stochasticity, but by a time of about 250 (dimensionless time units), decoherence is evident. The decoherence becomes significant for times larger than about 400. The variables $n_x(t), n_y(t)$ and $L_x(t), L_y(t)$ decay to zero at large times, but $n_z(t)$ and $L_z(t)$ “hang up” at finite values. It is clear from Eq. (16) that $d\langle L_z(t) \rangle / dt = 0$ because $d\langle \boldsymbol{\varepsilon}(t) \rangle / dt = 0$ and $[\mathbf{E}_0 \times \mathbf{n}(t)]_z = 0$.

Figures 6 and 7 are similar to Figs. 4 and 5 and show the average and standard deviation of the vectors $\mathbf{n}(t) = (n_x(t), n_y(t), n_z(t))$ and $\mathbf{L}(t) = (L_x(t), L_y(t), L_z(t))$ calculated with a larger value of volatility, $\varepsilon_0 = 0.1$. Now, decoherence sets in at earlier times, becoming significant for times larger than around 150. Again, $n_z(t)$ and $L_z(t)$ hang up at finite values. Since we expect perturbation theory to begin to break down at larger values of volatility, we now carry out nonperturbative calculations.

B. Nonlinear in $\boldsymbol{\varepsilon}(t)$ calculation

In order to treat the equations of motion that are nonlinear in the variable $\boldsymbol{\varepsilon}$, we recast them in the form [recall that we are using the notation $\mathbf{n}(t) \equiv \langle \hat{\mathbf{n}}(t) \rangle$ and $\mathbf{L}(t) \equiv \langle \hat{\mathbf{L}}(t) \rangle$]

$$d\mathbf{n}(t) = -\frac{1}{I}\mathbf{L}(t) \times \mathbf{n}(t) dt, \quad (17)$$

$$d\mathbf{L}(t) = -\alpha \mathbf{n}(t) \cdot [\mathbf{E}_0 + \boldsymbol{\varepsilon}(t)][\mathbf{E}_0 + \boldsymbol{\varepsilon}(t)] \times \mathbf{n}(t) dt, \quad (18)$$

$$d\boldsymbol{\varepsilon}(t) = -\vartheta \boldsymbol{\varepsilon}(t) dt + \sigma d\mathbf{W}(t). \quad (19)$$

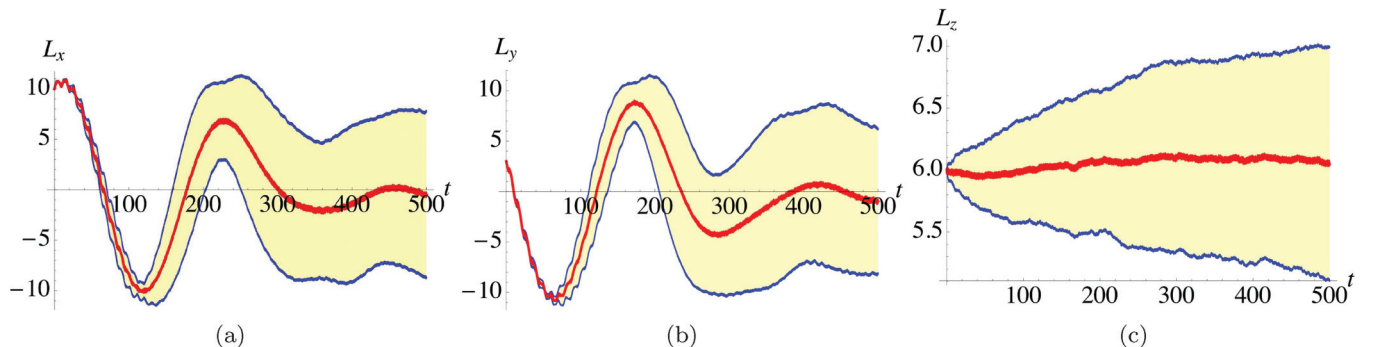


FIG. 7. (Color online) Average and standard deviation of the angular momentum vector $(L_x(t), L_y(t), L_z(t))$ vs time obtained for stochastic dynamics using Eqs. (15) and (16), with $\varepsilon_x(t), \varepsilon_y(t)$, and $\varepsilon_z(t)$ fields taken as Gaussian white noise and volatility $\sigma = 0.1$.

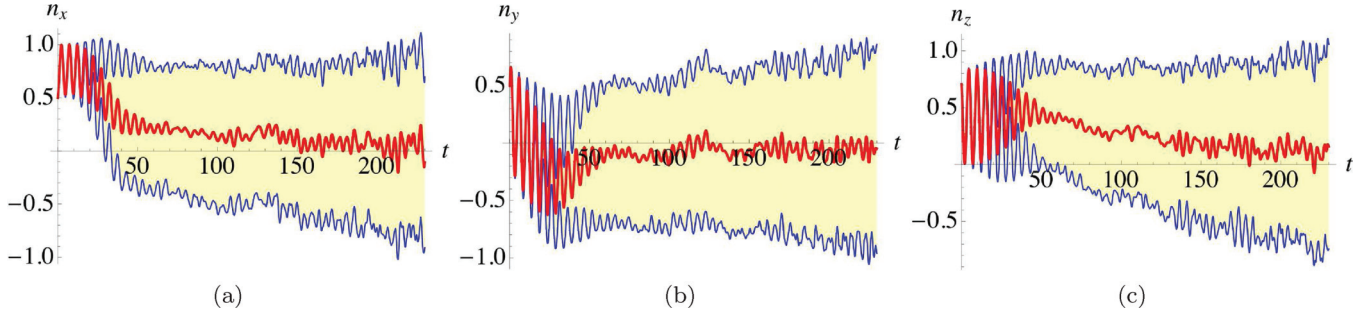


FIG. 8. (Color online) Average and standard deviation of $n_x(t), n_y(t), n_z(t)$ vs time obtained for stochastic dynamics using Eqs. (17)–(19), with $\varepsilon_x(t), \varepsilon_y(t)$, and $\varepsilon_z(t)$ fields taken as Gaussian white noise and volatility $\sigma = 0.1$.

Here ϑ is the mean reversion rate of the Ornstein-Uhlenbeck process ε with long term mean equal to zero, $\mathbf{W}(t)$ is the standard Wiener process with zero mean and volatility 1, and σ is the volatility of the Ornstein-Uhlenbeck process. With nonvanishing ϑ , the functional dependence on time of the variance and correlation function of the process ε is very different from a Wiener process, but with $\vartheta = 0$, the process ε is a Wiener process. Here too we used the Mathematica 9.0 built-in command `ItoProcess` [19] for solving stochastic differential equations (17)–(19). In the numerical calculations we took $\vartheta = 0$ and initial conditions $\varepsilon_i(0) = 0$ for $i = 1, 2, 3$.

The results of the nonlinear calculation for small volatility are similar to the linear calculations for sufficiently small times. This is not surprising because the nonlinear terms in the volatility are quadratically small. But as the volatility increases (the dimensionless parameter is $\sigma/|\mathbf{E}_0|$, but we have set $|\mathbf{E}_0| = 1$), the nonlinear terms begin to contribute significantly, even for small times. The nonlinear terms contribute additional decoherence mechanisms, so we expect faster and additional decoherence. Figures 8 and 9 show the results of the nonlinear calculation with a value of volatility $\sigma = 0.1$. There is a significant difference between the perturbative results shown in Figs. 6 and 7 and the nonperturbative results, even for times as early as $t = 25$, and decoherence is already significant for times beyond $t = 25$. The most significant difference is that the dynamical variables $n_z(t)$ and $L_z(t)$ no longer hang up at finite values but decay to zero at large time. It is not surprising that the hang-up is removed by the additional decoherence resulting from the nonlinear terms.

V. SUMMARY AND CONCLUSION

We introduced a model for treating the dynamics of a molecule with an induced dipole moment in the presence of an external electric field. We showed that the classical dynamics is equivalent to the mean-field quantum dynamics of the system. The dynamics is more complicated than the dynamics of a magnetic dipole moment in a magnetic field, despite the similarity of the Stark Hamiltonian for an electric dipole moment in an electric field, $H_S = -\mathbf{d} \cdot \mathbf{E}$, and Zeeman Hamiltonian for a magnetic dipole moment in a magnetic field, $H_Z = -\boldsymbol{\mu} \cdot \mathbf{B}$. The difference is due to the fact that the electric dipole moment of a molecule is locked along a molecule-fixed direction (as opposed to the magnetic case where the moment is along the angular momentum), and its evolution in an electric field is coupled to but distinct from the rotational motion of the molecule. Hence, modeling the dynamics requires equations of motion for *both* the angular momentum operator $\hat{\mathbf{L}}(t)$ and the operator for the unit vector in the direction of the axis of the molecule $\hat{\mathbf{n}}(t)$. Then, we considered the dynamics in the presence of an external electric field that is a sum of a deterministic field and a stochastically fluctuating field (noise). For simplicity, we took the fluctuations to be Gaussian white noise. The model makes the external noise approximation [6] wherein no back action of the system on the environment is present. A consequence of this assumption is that the system does not come into equilibrium with a thermal environment [4]. This is a good approximation when the back action is weak, as explained in Refs. [4,6]; that is, the influence of the system dipole moment on the dynamics of the bath is negligible

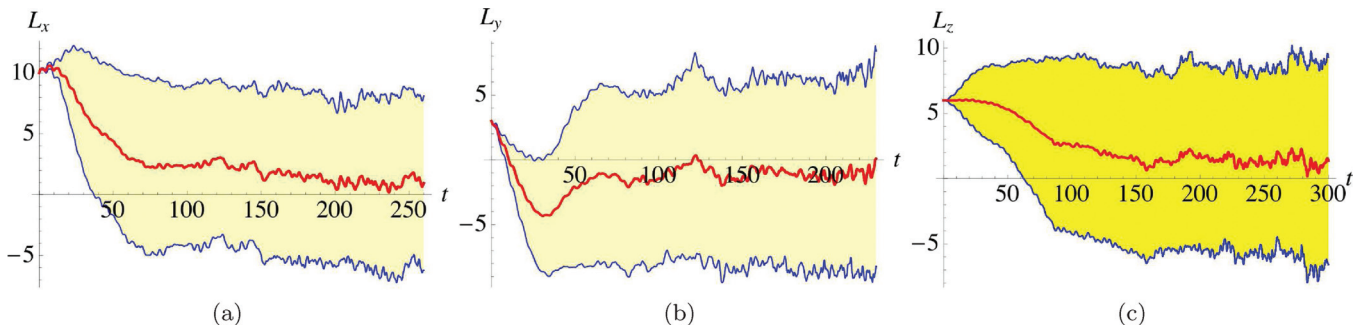


FIG. 9. (Color online) Average and standard deviation of the angular momentum vector ($L_x(t), L_y(t), L_z(t)$) vs time obtained for stochastic dynamics using Eqs. (17)–(19), with $\varepsilon_x(t), \varepsilon_y(t)$, and $\varepsilon_z(t)$ fields taken as Gaussian white noise and volatility $\sigma = 0.1$.

[20]. Using perturbation theory for the stochastic field, the z component of the average induced electric dipole moment, $\langle \hat{\mathbf{n}}(t) \rangle$, and the z component of the average angular momentum, $\langle \hat{\mathbf{L}}(t) \rangle$, do not decay (decohere) to zero, despite fluctuations in all three components of the electric field (but the other components of these vectors do decohere). This is in contrast to the decay of the average over fluctuations of a magnetic moment, which does decohere to zero in a stochastic magnetic field with Gaussian white noise in all three components [4]. In contradistinction to the perturbative analysis, i.e., upon including the term nonlinear in the stochastic field in the equations of motion, we find that decoherence occurs in all three components of $\langle \hat{\mathbf{L}}(t) \rangle$ and $\langle \hat{\mathbf{n}}(t) \rangle$. Moreover, decoherence of the transverse components of these vectors appears significantly earlier than in the perturbation theory solutions. These predictions, obtained under the external noise assumption, should be able to be readily checked experimentally. These predictions should remain valid also for Gaussian colored

noise stochastic processes, as long as the temporal correlation time of the colored noise process τ_c is short compared with the rotation time of the molecule, $\tau_r = I/\langle L \rangle$, and the Stark time scale, $\tau_S = \hbar/(Ed)$. It would be particularly interesting to experimentally verify the full decoherence of all components of the averaged dipole moment and angular momentum in the strong (nonperturbative) stochastic field case and to experimentally determine the conditions for back action to become observable (i.e., obtaining the long-time statistical mechanics distributions).

ACKNOWLEDGMENTS

This work was supported in part by grants from the Israel Science Foundation (Grant No. 2011295) and the James Franck German-Israel Binational Program. We are grateful to John Scales for useful conversations.

-
- [1] E. Ishiguro, T. Arai, M. Mizushima, and M. Kotani, *Proc. Phys. Soc. A* **65**, 178 (1952).
- [2] S. Fleischer, I. S. Averbukh, and Y. Prior, *Phys. Rev. Lett.* **99**, 093002 (2007).
- [3] C. Scherer, *Braz. J. Phys.* **34**, 442 (2004).
- [4] P. Szańkowski, M. Trippenbach, and Y. B. Band, *Phys. Rev. E* **87**, 052112 (2013).
- [5] Y. B. Band, *Phys. Rev. E* **88**, 022127 (2013).
- [6] N. G. Van Kampen, *Stochastic Processes in Physics and Chemistry* (Elsevier, Amsterdam, 2007).
- [7] P. E. Kloeden and E. Platen, *Numerical Solution of Stochastic Differential Equations* (Springer, Berlin, 2011).
- [8] P. E. Kloeden, E. Platen, and H. Schurz, *Numerical Solutions of Stochastic Differential Equations through Computer Experiments* (Springer, Berlin, 2003).
- [9] C. W. Gardiner, *Handbook of Stochastic Methods for Physics, Chemistry and the Natural Sciences*, 3rd ed. (Springer, Berlin, 2004).
- [10] G. Adomian, *Nonlinear Stochastic Operator Equations* (Academic, Orlando, FL, 1986). By nonlinear, the authors mean equations of the form $dx(t) = a(t, x(t))dt + b(t, x(t))dw(t)$, where the functions $a(t, x(t))$ and $b(t, x(t))$ are nonlinear in $x(t)$ and $w(t)$ is a stochastic process.
- [11] L. D. Landau and E. M. Lifshitz, *Mechanics* (Pergamon, Oxford, 1994), Sec. 52.
- [12] G. V. Jose and E. J. Saletan, *Classical Dynamics: A Contemporary Approach* (Cambridge University Press, Cambridge, 1998), Sec. 6.2.
- [13] For simplicity we take a symmetric top, rather than a spherical top Hamiltonian. If we had taken a symmetric top, $\frac{\hat{L}^2}{2I} + C\hat{L}_Z^2$, where $C = \hat{L}^2(I_z^{-1} - I^{-1})/2$, \hat{L}_Z is the angular momentum along the body-fixed Z axis, $\hat{L}_Z = \hat{\mathbf{L}} \cdot \hat{\mathbf{n}}$, and $\hat{\mathbf{n}}$ is the unit vector in the direction of the body-fixed Z axis, the use of angular momentum in the space-fixed frame would be required.
- [14] L. D. Landau and E. M. Lifshitz, in *Quantum Mechanics Non-relativistic Theory*, 2nd ed., pp. 312–316; in *Quantum Mechanics Non-relativistic Theory*, 3rd ed., p. 337, Problem 1 (Pergamon Press, Oxford, England).
- [15] O. Zobay and B. M. Garraway, *Phys. Rev. A* **61**, 033603 (2000).
- [16] J. Liu, L. Fu, B.-Y. Ou, S.-G. Chen, D.-I. Choi, B. Wu, and Q. Niu, *Phys. Rev. A* **66**, 023404 (2002).
- [17] I. Tikhonenkov, E. Pazy, Y. B. Band, M. Fleischhauer, and A. Vardi, *Phys. Rev. A* **73**, 043605 (2006).
- [18] Y. B. Band, I. Tikhonenkov, E. Pazy, M. Fleischhauer, and A. Vardi, *J. Mod. Opt.* **54**, 697 (2007).
- [19] <http://reference.wolfram.com/mathematica/ref/ItoProcess.html>
- [20] Reference [5] describes one method of overcoming the non-back-action approximation when its assumptions are not valid by augmenting the equations of motion for the electric dipole moment with a decay term that ensures that the system comes into thermal equilibrium at long times.