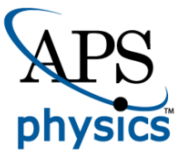

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Reduced Equations of Motion for Quantum Systems Driven by Diffusive Markov Processes

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Reduced equations of motion for quantum systems driven by diffusive Markov processes

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The expansion of a stochastic Liouville equation for the coupled evolution of a quantum system and an Ornstein-Uhlenbeck process into a hierarchy of coupled differential equations is a useful technique that simplifies the simulation of stochastically-driven quantum systems. We expand the applicability of this technique by completely characterizing the class of diffusive Markov processes for which a useful hierarchy of equations can be derived. The expansion of this technique enables the examination of quantum systems driven by non-Gaussian stochastic processes with bounded range. We present an application of this extended technique by simulating Stark-tuned Förster resonance transfer in Rydberg atoms with non-perturbative position fluctuations.

Describing the dynamics of a quantum system coupled to uncontrolled degrees of freedom has been an important problem since the inception of quantum mechanics, e.g., [1]. The accurate description of such *open quantum systems* is particularly vital for the design of quantum technologies, such as quantum computers. Several approximate and exact methods exist for describing the dynamics of open quantum systems, including master equations, surrogate Hamiltonians, and Monte-Carlo numerical simulations.

In this work, we examine quantum systems driven linearly by classical fluctuations. That is, the Hamiltonian for the system is described by $\hat{H}_\Omega(t) = \hat{H}_0 + \Omega(t)\hat{V}$, where $\Omega(t)$ is a time-dependent stochastic variable. This is a sub-class of a more general open quantum system where $\Omega(t)$ is an operator in the Hilbert space of an uncontrolled environment. The replacement of the operator with a scalar variable is an approximation that is valid in certain limits (e.g., in the high temperature limit of a bosonic environment). Scalar fluctuations can also describe noise in a quantum system that is controlled by an effectively classical quantity, such as a gate voltage.

For such stochastic evolution, the dynamics of the system under a *given* trace of the noise is dictated by the von-Neumann (Schrödinger) equation:

$$\frac{\partial}{\partial t}\hat{\rho}(t|\{\Omega(t)\}) = -\frac{i}{\hbar}[\hat{H}_\Omega(t), \hat{\rho}(t|\{\Omega(t)\})]. \tag{1}$$

Here, the notation $\hat{\rho}(t|\{\Omega(t)\})$ is used to explicitly indicate that this density matrix is *conditioned* on a particular realization of the noise process. This equation is formally solved to yield $\hat{\rho}(t|\{\Omega(t)\}) = \hat{\mathcal{U}}(t,0)\hat{\rho}^0\hat{\mathcal{U}}(t,0)^\dagger$, for initial state ρ^0, where $\hat{\mathcal{U}}(t,0) = \mathcal{T}e^{-\frac{i}{\hbar}\int_0^t \hat{H}_\Omega(t)}$ with $\mathcal{T}$ being the time-ordering operator. We are often more interested in the unconditioned evolution of the system state, after the fluctuating quantity has been averaged:

$$\hat{\rho}(t) = \langle \hat{\rho}(t|\{\Omega(t)\})\rangle_{\{\Omega(t)\}}, \tag{2}$$

where the angled brackets denote an expectation value over the stochastic process up to time t. We will refer to differential equations describing the evolution of this averaged quantity as *reduced equations of motion*. In this work we derive reduced equations of motion for a quantum system coupled to a wide family of stochastic processes.

We focus on stochastic processes that are time-continuous, time-homogenous, and Markov, which means that the evolution of the conditional probability distribution for the process evolves as [2–4]

$$\frac{\partial}{\partial t}P(\Omega, t|\Omega', t') = \Gamma_\Omega P(\Omega, t|\Omega', t'), \tag{3}$$

where $P(\Omega, t|\Omega', t')$ is the probability that the stochastic process takes the value Ω at time t given that it took the value Ω' at time t' ($t' \leq t$). Γ_Ω is the forward *generator* of the process and is a differential operator only involving derivatives with respect to Ω. The generator of evolution is in general a complex quantity that results from a *Kramers-Moyal* expansion of the (classical) master equation for the probability distribution of the process [2]. Below, we will restrict ourselves to certain forms of this generator, but for now only the Markov assumption will be used.

In order to derive an expression related to the reduced equations of motion in a simple manner, consider the quantity $\hat{\rho}(t,\Omega) \equiv \hat{\rho}_\Omega(t)P(\Omega,t)$, which is the joint distribution of the quantum-mechanical coordinates and the stochastic variable at time t. $\hat{\rho}_\Omega(t) \equiv \hat{\rho}(t|\Omega(t))$ is the density matrix conditioned on the value of the stochastic process at time t (in contrast to $\hat{\rho}(t|\{\Omega(t)\})$, which is the density matrix conditioned on an entire history of the stochastic process). Evaluating the time derivative of this joint distribution results in the *stochastic Liouville equation*, first derived by Kubo [5]:

$$\frac{\partial\hat{\rho}(t,\Omega)}{\partial t} = \left[-\frac{i}{\hbar}\hat{H}_0^\times - \frac{i}{\hbar}\Omega(t)\hat{V}^\times\right]\hat{\rho}(t,\Omega) + \Gamma_\Omega\hat{\rho}(t,\Omega), \tag{4}$$

where $A^\times B \equiv [A,B]$. Here, we have used the fact that because the stochastic process is Markov, the time evolution of $P(\Omega,t)$ follows the same law as the conditional distribution in Eq. (3) and is generated by Γ_Ω. This simple linear form for the evolution of the joint distribution is possible because both the conditional quantum

density matrix and the probability distribution for the stochastic variable evolve linearly and in a Markov fashion. Because $\hat{\rho}(t,\Omega)$ is a joint distribution, its marginal over the stochastic variable yields the average density matrix, i.e., $\hat{\rho}(t) = \int_{\mathcal{D}} \mathrm{d}\Omega \hat{\rho}(t,\Omega)$, where $\mathcal{D}$ is the range of the stochastic variable [6].

Markov diffusion processes. A wide a class of physical processes can be approximated by truncating the Kramers-Moyal expansion for the generator Γ_Ω of the Markov process at the second term [2]. This results in the *Fokker-Planck* equation for the probability distribution, with the generator being the differential operator [4]: $\Gamma_\Omega^{\mathrm{FP}} \equiv \Phi_\Omega = -\frac{\partial}{\partial\Omega}A(\Omega) + \frac{1}{2}\frac{\partial^2}{\partial\Omega^2}B(\Omega)$, where $A(\Omega)$ and $B(\Omega)$ are real differentiable coefficients, with the restriction that $B > 0$.

For such *diffusion processes*, it is common to define the backward, or adjoint, generator for the process: $\overleftarrow{\Phi}_\Omega = A(\Omega)\frac{\partial}{\partial\Omega} + \frac{1}{2}B(\Omega)\frac{\partial^2}{\partial^2\Omega}$. For a Markov process, Eq. (3) is often called the *forward Kolmogorov equation* and $\frac{\partial}{\partial t'}P(\Omega,t|\Omega',t') = \overleftarrow{\Phi}_{\Omega'}P(\Omega,t|\Omega',t')$ is the *backward Kolmogorov equation*. Let $f_n(\Omega)$ and $\overleftarrow{f}_n(\Omega)$ be eigenfunctions of the forward and backward generators, respectively, i.e., $\Phi_\Omega f_n(\Omega) = -\lambda_n f_n(\Omega)$ and $\overleftarrow{\Phi}_\Omega \overleftarrow{f}_n(\Omega) = -\lambda_n \overleftarrow{f}_n(\Omega)$, with $\lambda_n \geq 0$ [2]. These two eigenfunctions are related through the stationary distribution of the process, explicitly $f_n(\Omega) = P^0(\Omega)\overleftarrow{f}_n(\Omega)$, where P^0 is the stationary distribution of the process Ω [3]. Furthermore, the eigenfunctions of the backward generator form a complete, orthogonal system (on the range $\mathcal{D}$) under the measure induced by the stationary distribution of the process: $\int_{\mathcal{D}} \mathrm{d}\Omega P^0(\Omega)\overleftarrow{f}_n(\Omega)\overleftarrow{f}_m(\Omega) = \delta_{mn}$, which in turn implies the following about the eigenfunctions of the forward generator [3]: $\int_{\mathcal{D}} \mathrm{d}\Omega (P^0(\Omega))^{-1} f_n(\Omega) f_m(\Omega) = \delta_{mn}$

Diffusive hierarchical equations of motion. Using the completeness and orthogonality of the backward and forward generator eigenfunctions [3], we can expand $P(\Omega,t)$ in terms of these eigenfunctions, and consequently, $\hat{\rho}(t,\Omega) \equiv \hat{\rho}_\Omega(t)P(\Omega,t) = \sum_{n=0}^{\infty}\bar{\rho}_n(t)f_n(\Omega)$ Here, $\bar{\rho}_n(t)$ are unnormalized auxiliary density matrices, which do not have the interpretation of being *conditioned* density matrices of the quantum system. Then, expressing the stochastic Liouville equation Eq. (4) in terms of this eigenfunction expansion yields

$$\sum_{n=0}^{\infty}\frac{\partial}{\partial t}\bar{\rho}_n(t)f_n(\Omega) = \sum_{n=0}^{\infty} -\frac{i}{\hbar}H_0^\times \bar{\rho}_n(t)f_n(\Omega) - \frac{i}{\hbar}V^\times\bar{\rho}_n(t)\left[\Omega(t)f_n(\Omega)\right] - \lambda_n\bar{\rho}_n(t)f_n(\Omega). \quad (5)$$

To simplify further, we make the assumption that the eigenfunctions of the backward Markov generator are polynomials in Ω, i.e., $\overleftarrow{f}_n(\Omega)$ is an n^{th} order polynomial in Ω. With this assumption, we utilize the following theorem, typically attributed to Favard [7], which states an important property of orthogonal polynomials:

Theorem (Favard): Let $\{p_n(x)\}, n \geq 0$ be a system of polynomials. This system satisfies a three-term recurrence relation $p_{n+1}(x) = (A_n x + B_n)p_n(x) - C_n p_{n-1}(x)$, (with $p_{-1}(x) = 0$) if and only if it is a system of *orthogonal polynomials*. Here $A_n \neq 0, B_n$ and $C_n \neq 0$ are Ω-independent (real) recurrence coefficients.

Favard's theorem implies that the orthogonal polynomial eigenfunctions of the backward Markov generator (and hence, those of the forward Markov generator) satisfy the three-term recurrence relations:

$$\Omega f_n(\Omega) = \frac{C_n}{A_n}f_{n-1}(\Omega) - \frac{B_n}{A_n}f_n(\Omega) + \frac{1}{A_n}f_{n+1}(\Omega). \quad (6)$$

Using this recurrence relation, the eigenfunction expansion of the stochastic Liouville equation becomes:

$$\sum_{n=0}^{\infty}\frac{\partial}{\partial t}\bar{\rho}_n(t)f_n(\Omega) = \sum_{n=0}^{\infty}\left[-\frac{i}{\hbar}H_0^\times - \lambda_n\right]\bar{\rho}_n(t)f_n(\Omega) - \frac{i}{\hbar}V^\times\bar{\rho}_n(t)\left[\frac{1}{A_n}f_{n+1}(\Omega) + \frac{C_n}{A_n}f_{n-1}(\Omega) - \frac{B_n}{A_n}f_n(\Omega)\right] \quad (7)$$

Multiplying both sides of this equation by $f_m(\Omega)/P^0(\Omega)$ and integrating over Ω results in an equation for each m because of the orthogonality of the functions $f_n(\Omega)$. These equations form a hierarchy of coupled (operator) differential equations, which we refer to as *diffusive hierarchical equations of motion* (DHEOM) [8]:

$$\begin{aligned}
\frac{\partial}{\partial t}\bar{\rho}_0(t) &= -\left[\frac{i}{\hbar}\left(H_0^\times - \frac{B_0}{A_0}V^\times\right) + \lambda_0\right]\bar{\rho}_0(t) - \frac{i}{\hbar}\frac{C_1}{A_1}\,V^\times\bar{\rho}_1(t),\\
\frac{\partial}{\partial t}\bar{\rho}_n(t) &= -\left[\frac{i}{\hbar}\left(H_0^\times - \frac{B_n}{A_n}V^\times\right) + \lambda_n\right]\bar{\rho}_n(t) - \frac{i}{\hbar}\frac{C_{n+1}}{A_{n+1}}\,V^\times\bar{\rho}_{n+1}(t) - \frac{i}{\hbar}\frac{1}{A_{n-1}}V^\times\bar{\rho}_{n-1}(t), \qquad n > 0. \quad (8)
\end{aligned}$$

These equations are useful because they describe the evolution of the system density matrix under the influence of stochastic noise, but without explicit reference to noise variables. In addition, each member of the hierarchy only couples to its two neighbors, i.e., n couples to $n+1$ and $n-1$, making their integration easy. We also need to specify the initial conditions and the prescription for calculating the system density matrix from the solution of the

hierarchy of coupled equations. A physically motivated initial state for the quantity $\hat{\rho}(t,\Omega)$ is $\hat{\rho}(0,\Omega) = \hat{\rho}^0 P^0(\Omega)$, where P^0 is the stationary (equilibrium) distribution of the noise process and $\hat{\rho}^0$ is any system density matrix. Evolution from this initial state describes the response of a quantum system to fluctuations around the bath equilibrium. Now, $\Phi_\Omega P^0 = 0$ by definition, and therefore, $\bar{\rho}_0(0) = \hat{\rho}^0$, and $\bar{\rho}_n(0) = 0 \;\forall\; n \neq 0$. At any time, the system density matrix is defined as the integral of the quantity $\hat{\rho}(t,\Omega)$ over Ω:

$$\hat{\rho}(t) = \int_{\mathcal{D}} d\Omega\; \hat{\rho}(t,\Omega) = \int_{\mathcal{D}} d\Omega \sum_{n=0}^{\infty} \bar{\rho}_n(t) f_n(\Omega) \propto \bar{\rho}_0(t).$$

The proportionality is a result of the following property of the eigenfunctions:

$$\int_{\mathcal{D}} d\Omega\; f_n(\Omega) \propto \int_{\mathcal{D}} d\Omega\; P^0(\Omega) \overleftarrow{f}_n(\Omega) \overleftarrow{f}_0(\Omega) = \delta_{n0},$$

where this proportionality follows from the fact that $\overleftarrow{f}_0$ is a zeroth-order polynomial in Ω. As we shall see below, it is always possible to choose $\overleftarrow{f}_0 = 1$, and therefore, the proportionality above can be converted to an equality. Hence, the auxiliary density matrix $\bar{\rho}_0(t)$ always keeps track of the averaged system density matrix $\hat{\rho}(t)$.

To summarize, we have shown that the dynamics of a quantum system that is linearly driven by a diffusion process with a Markov generator possessing polynomial eigenfunctions can be described by a hierarchy of coupled dynamical equations. The solution to these equations will reproduce the reduced density matrix of the quantum system at any time, with no approximations. However, the above hierarchy of equations is infinite, and therefore, we require some truncation strategy in order to solve them. If the equations can be truncated at some $n = N$ at the expense of bounded error, they can be numerically solved (or analytically solved via a partial fraction expansion if H_0 is time-independent [9]). A general truncation strategy exists if the following conditions hold: (1) for large enough $n = N$, $|\lambda_N| \gg ||H_0^\times - \frac{B_N}{A_N} V^\times||_2$, and (2) $\frac{1}{A_n} \in \omega\left(\frac{C_n}{A_n}\right)$, where $||\cdot||_2$ is the induced 2-norm and $f_n \in \omega(g_n)$ is indicates that f_n dominates g_n asymptotically (in n). In the Supplementary Information, we develop such a general truncation strategy that results in the following *terminator equation* for the hierarchy at a level N satisfying these conditions:

$$\frac{\partial}{\partial t}\bar{\rho}_N(t) = -\left[\frac{i}{\hbar}\left(H_0^\times - \frac{B_N}{A_N}V^\times\right) + \lambda_N\right]\bar{\rho}_N(t) - \frac{1}{\hbar^2}\frac{C_{N+1}}{\lambda_{N+1}A_{N+1}A_N}V^\times V^\times \bar{\rho}_N(t) - \frac{i}{\hbar}\frac{1}{A_{N-1}}V^\times \bar{\rho}_{N-1}(t). \quad (9)$$

We now turn to characterizing the diffusive stochastic processes with generators that have polynomial eigenfunctions. A quantum system driven by each one of these processes will have a dynamical description in terms of the DHEOM derived above. The combination of Bochner's theorem [10], which is stated explicitly in the Supplementary Information, and Favard's theorem implies that there are only three diffusive processes with backward and forward generators that have orthogonal polynomial eigenfunctions: (a) the Ornstein-Uhlenbeck (OU) process, defined on $(-\infty,\infty)$ (b) the square-root process, defined on $(\Omega_{\min},\infty)$ and (c) the Jacobi process, defined on $(\Omega_{\min},\Omega_{\max})$. In the Supplementary Information we list these diffusive processes with their orthogonal polynomial eigenfunctions and other relevant properties (table 1), and also explicitly write the DHEOM that describe the dynamics of a quantum system linearly driven by each of the above three stochastic processes. The Jacobi process is particularly efficient to simulate because the eigenvalues λ_n of its generator scale quadratically with n, making truncation possible at a smaller depth than for the other two processes, where λ_n scales linearly with n. We note that the DHEOM for the OU process was previously derived by Kubo and Tanimura [5, 9], but this more general framework resulting in DHEOMs for all major diffusive Markov processes was heretofore unknown. As explained in the Supplementary Information, the DHEOM method is limited to simulating square-root processes with mean reversion rate $\gamma > 1$, whereas any γ is valid for the other processes. Finally, for completeness the Supplementary Information also presents the DHEOM for the propagator for the dynamical map as opposed to the density matrix.

Application and Discussion. The primary advantage of our general formulation of DHEOM is that it allows for the efficient simulation of quantum systems driven by noise sources with bounded range and non-Gaussian amplitude distribution. As an application, we consider energy transfer between two systems mediated by a dipole-dipole interaction. This interaction is an approximation to the Coulomb coupling of electric charge distributions and is the basis of several common experimental techniques, such as Förster resonance energy transfer (FRET) spectroscopy [11] and Stark-tuned resonant transfer in Rydberg atoms [12, 13]; we focus on this latter system as our example.

The electric dipole-dipole coupling energy scales as $J \propto 1/R^3$, where R is the distance between the coupled molecules or atoms. Consider the case where this interparticle spacing is varying in time as a result of thermal, electromagnetic, or vibrational fluctuations. If the fluctuations in R are treated perturbatively the dipole-

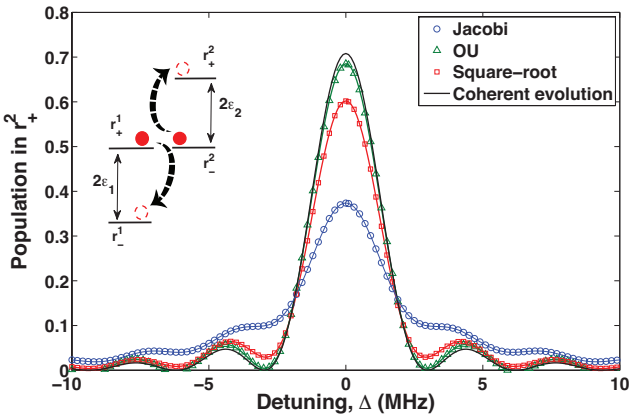

FIG. 1. Average population in the excited state of atom 2 after an interaction time of $T = 1\mu$s as a function of the Stark detuning for different models of motional noise. The black (solid) curve is for coherent evolution with no fluctuation in inter-atom distance. The parameters used are: $\gamma = 1.5, \mu = 1$ for all noise sources, and $\sigma^2 = 0.3$ for the OU process. The square root process is defined in the semi-interval $[0, \infty)$ with $c_1 = 1$, and the Jacobi process in the interval $(1/8, 8)$ with $c = 1$. See Supplementary Information for the interpretation of these parameters.

dipole coupling energy can be expanded linearly around the mean separation, R_0: $J \sim 1/R_0^3 - (3/R_0^2)\delta R(t)$. However, in cases where such a perturbative treatment is not accurate we must consider the energy J as a time-dependent fluctuating quantity in a range $[J_{\min}, J_{\max}]$. To explore the consequences of such a non-perturbative treatment, consider the Hamiltonian of two interacting Rydberg atoms: $H = -\epsilon_1\sigma_z^1 - \epsilon_2\sigma_z^2 + J(t)(\sigma_+^1\sigma_-^2 + \sigma_-^1\sigma_+^2)$, where we restrict the description of the atoms to the two energy levels relevant to the energy transfer, and therefore use Pauli matrices to describe them as effective two-level systems in the basis $\left|r_-^i\right\rangle, \left|r_+^i\right\rangle$ where $r_\pm^i$ are Rydberg states for atom i with the energy gap ϵ_i, i.e., $\sigma_z^i \left|r_\pm^i\right\rangle = \pm\epsilon_i \left|r_\pm^i\right\rangle$. Typically, these levels are chosen so that when an electric field tunes atomic energies by the Stark effect they satisfy $\epsilon_1 = \epsilon_2$ (Förster resonance condition) at some critical field value. For example, in Ref. [13], $\left|r_+^1\right\rangle = \left|37P_{3/2}\right\rangle, \left|r_-^1\right\rangle = \left|37S_{1/2}\right\rangle, \left|r_+^2\right\rangle = \left|38S_{1/2}\right\rangle, \left|r_-^2\right\rangle = \left|37P_{3/2}\right\rangle$, where these are fine states of Rb atoms. We ignore relaxation of these states because for sufficiently cooled Rydberg atoms it can be neglected on the timescales we are considering [14]. We also assume that the relative orientation of the atomic dipoles remains constant. The initial state is the excited state of atom 1: $\rho^0 = \left|r_+^1, r_-^2\right\rangle\left\langle r_+^1, r_-^2\right|$, and the dipole-dipole coupling is assumed to fluctuate as $J(t) \equiv J_0\Omega(t)$, where $\Omega(t)$ is one of the three diffusive noise processes described above. The quantity being observed (e.g., by selective field ionization [13]) is the population in the excited state of atom 2 after an interaction time T. Although position fluctuations were reported to be minor in the experiment of Ref. [13], we use physical parameters compatible with this experiment for concreteness: $J_0/\hbar = 0.5$MHz, and $T = 1\mu$s. Fig. 1 shows the average population of $\left|r_+^2\right\rangle$ at T for different values of the Stark detuning $\Delta \equiv \epsilon_2 - \epsilon_1$. We expect that this transferred population will drop off as $|\Delta|$ increases (as the atoms become less resonant), but the figure shows that the behavior of this drop-off depends heavily on the exact nature of the fluctuations in R, although all processes have the same mean $\mu = 1$ and mean reversion rate $\gamma = 1.5$. The OU and square-root processes predict average populations that have significant oscillations as a function of the detuning, similar to the completely coherent (noiseless) case. In contrast, the Jacobi process driven evolution predicts damped oscillations and smaller transfer populations. We predict that for experiments performed in this regime, where position fluctuations are relevant, the Jacobi process average will be more accurate (and they are certainly more consistent with what is experimentally observed in Ref. [13]). This is because the Jacobi process is a more accurate description than the OU and square-root processes, which can only be approximations in this scenario; realistic trapping conditions impose strict lower and upper bounds on J resulting from the fact that the atoms can only drift within the trap volumes. The OU and square-root simulations predict large transfer populations because rare, unphysical, large magnitude couplings contribute to the averages in these cases while they do not to the Jacobi process average. These results emphasize the advantage of being able to efficiently average over non-Gaussian noise processes with bounded range. To underscore the numerical efficiency of the DHEOM simulations, we note that the calculations producing Fig. 1 were performed 10 times faster using the present theory than conventional Monte Carlo sampling of the noise processes [15]. Furthermore, because of the well-defined truncation strategy, convergence issues and noise are not a concern for DHEOM simulations as in the Monte Carlo approach.

Summary. We have completely characterized the class of diffusively driven quantum systems for which a useful hierarchy of reduced equations of motion can be derived, and explicitly derived these DHEOM. We applied the technique to examine position fluctuation dependence of Stark tuned Förster resonance between Rydberg atoms. We expect that the general techniques developed in this work will be useful for widening the range of open quantum systems that can be simulated efficiently. Use of the DHEOM for Jacobi processes will be particularly useful for modeling quantum systems subject to fluctuations with bounded range, e.g., classical noise in external control fields [16].

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tional Nuclear Security Administration under contract DE-AC04-94AL85000.

[1] W. Pauli (Leipzig Hirzel, 1928) p. 30.

[2] H. Risken, *The Fokker-Planck equation* (Springer-Verlag, 1984).

[3] C. W. Gardiner, *Handbook of stochastic methods* (Springer, 2004).

[4] N. G. van Kampen, *Stochastic processes in physics and chemistry* (North Holland, 2007).

[5] R. Kubo, Adv Chem Phys, **15**, 1 (1969).

[6] It is not immediately obvious that this integral gives the same average density matrix as the expression in Eq. (2) since it prescribes an integral over the variable at time t whereas Eq. (2) prescribes a functional integral over the entire history of the stochastic process. This equivalence of the marginal derived from the solution of Eq. (4) and that given in Eq. (2) is entirely due to the Markovianity of the stochastic process and is proven in some detail in Ref. [17].

[7] W. Schoutens, *Stochastic Processes and Orthogonal Polynomials* (Springer, 2000).

[8] The term *diffusive* is included in this name to avoid confusion with the recently derived hierarchical equations of motion that describe average dynamics resulting from coupling to a fully quantum mechanical description of the environment [18].

[9] Y. Tanimura, J. Phys. Soc. Jpn., **75**, 082001 (2006).

[10] T. S. Chihara, *An introduction to orthogonal polynomials* (Gordon and Breach, 1978).

[11] H. Sahoo, J. Photoch. Photobio. C, **12**, 20 (2011).

[12] C. van Ditzhuijzen, A. Koenderink, J. Hernández, F. Robicheaux, L. Noordam, and H. van den Heuvell, Phys. Rev. Lett., **100**, 243201 (2008).

[13] I. I. Ryabtsev, D. B. Tretyakov, I. I. Beterov, and V. M. Entin, Phys. Rev. Lett., **104**, 073003 (2010).

[14] M. Saffman, T. G. Walker, and K. Molmer, Rev. Mod. Phys., **82**, 2313 (2010).

[15] The DHEOM simulations required 1984 seconds on a standard laptop, while a Monte Carlo simulation (with 500 samplings over the noise processes to obtain reasonable convergence of the average) required 19403 seconds on the same machine. Both codes were unoptimized.

[16] M. Grace, C. Brif, H. Rabitz, I. A. Walmsley, R. L. Kosut, and D. A. Lidar, J. Phys. B: At. Mol. Opt. Phys., **40**, S103 (2007); M. D. Grace, C. Brif, H. Rabitz, D. A. Lidar, I. A. Walmsley, and R. L. Kosut, J. Mod. Opt., **54**, 2339 (2007); M. D. Grace, J. Dominy, R. L. Kosut, C. Brif, and H. Rabitz, New J. Phys., **12**, 015001 (2010); K. C. Young, D. J. Gorman, and K. B. Whaley, http://arxiv.org/abs/1005.5418 (2010).

[17] M. Ban, S. Kitajima, and F. Shibata, Phys. Rev. A, **82**, 022111 (2010).

[18] A. Ishizaki and Y. Tanimura, J. Phys. Soc. Jpn., **74**, 3131 (2005); A. Ishizaki and G. R. Fleming, J. Chem. Phys., **130**, 234111 (2009).