

## Exact $c$ -Number Representation of Non-Markovian Quantum Dissipation

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The reduced dynamics of a quantum system interacting with a linear heat bath finds an exact representation in terms of a stochastic Schrödinger equation. All memory effects of the reservoir are transformed into noise correlations and mean-field friction. The classical limit of the resulting stochastic dynamics is shown to be a generalized Langevin equation, and conventional quantum state diffusion is recovered in the Born-Markov approximation. The non-Markovian exact dynamics, valid at arbitrary temperature and damping strength, is exemplified by an application to the dissipative two-state system.

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Irreversible quantum processes are important in almost every field of condensed-matter physics and chemistry. In phenomena as different as diffusion of light particles in solids and light harvesting in biological systems, the dissipation of energy, the destruction of phase coherence, and the generation of entropy play a key role. The question of how such processes can be described by the stochastic propagation of quantum states has recently attracted increasing attention [1–5]. For *classical* systems with linear dissipation, Langevin equations provide a theoretical (and numerical) tool to accurately describe the interaction of a system with a complex thermal reservoir in comparably simple terms of stochastic forces and memory friction. For open *quantum* systems, no such simple and exact approach in terms of an equation of motion has been established so far. The dynamical quantity of interest is the reduced density matrix, obtained by tracing out the reservoir degrees of freedom from the dynamics. Significant information about correlations between system and reservoir is lost in this operation; hence no deterministic differential equation of motion can be derived for the reduced density matrix without approximations. Traditional approaches treat these correlations perturbatively, yielding approximate equations of motion such as Redfield or Master equations. While these have been used very successfully in the fields of quantum optics and magnetic resonance, there are many condensed-matter problems where they are qualitatively wrong. Large coupling constants and long correlation time scales both need to be treated nonperturbatively. This can be accomplished in a formally exact manner by path integrals for open quantum systems [6–9], but an analytic evaluation of these functional integrals is usually restricted to special cases and approximations, e.g., the semiclassical limit. Furthermore, present exact numerical techniques such as quantum Monte Carlo (QMC) methods [10,11] have to cope with a sign problem arising from the fact that probability amplitudes with varying phases rather than probabilities have to be added in a quantum computation.

Quantum state diffusion (QSD) [1–3] has been established as an alternative theory of quantum dissipation in the

perturbative regime. Through the *stochastic* propagation of *pure* quantum states it yields an intuitively appealing picture of the behavior of individual quantum trajectories in open systems and permits effective numerical calculations. Generalizing this formalism to the case of nonperturbative, non-Markovian dynamics holds the promise to overcome most of the above-mentioned limitations of currently known techniques. This approach has recently been taken by Diósi, Strunz, and Gisin [4], but at the price of incorporating non-Markovian retardation effects in a memory functional. In practice this means that a general solution of the resulting equation of motion, even numerically, is almost as elusive as that of the underlying path integral.

In this Letter we derive and discuss stochastic Schrödinger equations for open systems which allow the treatment of linear dissipation of arbitrary strength and correlation time scales. Without approximation, all effects of the system-reservoir interaction are recast in the form of  $c$ -number noise and friction forces with a suggestive physical interpretation: The ensemble of phase-space points described by a classical Langevin equation is generalized to a stochastic ensemble of quantum states. Both descriptions are linked by a correspondence principle for open systems.

Any general theory of quantum dissipation has to start from an open system embedded in a large reservoir system, whose degrees of freedom are treated fully quantum mechanically, but can later be eliminated from the dynamics. The Hamiltonian of such a model consists of system and reservoir terms and an interaction potential,

$$H = H_0 + H_R + H_I. \quad (1)$$

Let us consider a thermal correlation function

$$\langle AB(t) \rangle = \text{tr} \{ e^{-\beta H} A e^{(i/\hbar)Ht} B e^{-(i/\hbar)Ht} \} / \text{tr} e^{-\beta H}, \quad (2)$$

where  $A$  and  $B$  are system operators. The time evolution operators can be joined with the “imaginary-time” propagator  $e^{-\beta H}$  to form a propagator with complex time argument which describes the time evolution of a quantum system along the contour  $C$  depicted in Fig. 1. This time

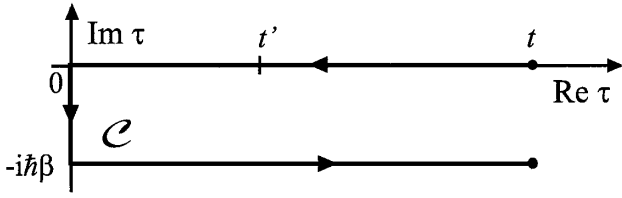


FIG. 1. Contour for propagators in thermal correlation functions.

evolution can be expressed by a path integral over the configuration space of the system

$$\langle AB(t) \rangle \propto \int \mathcal{D}[\mathbf{q}] e^{(i/\hbar)S_0[\mathbf{q}]} I[\mathbf{q}] \langle \mathbf{q}_0^+ | A | \mathbf{q}_0^- \rangle \times \langle \mathbf{q}_t^+ | B | \mathbf{q}_t^- \rangle, \quad (3)$$

where a normalization factor has been suppressed for simplicity. The action functional

$$S_0[\mathbf{q}] = \int_C d\tau L(\mathbf{q}, \dot{\mathbf{q}}) \quad (4)$$

depends in the usual way on the classical Lagrange function. The measure  $d\tau$  is the complex time differential on  $C$ , and  $\dot{\mathbf{q}} = \frac{d\mathbf{q}}{d\tau}$ . In the generalized influence functional

$$I[\mathbf{q}] \equiv \left\langle T_C \exp\left(-\int_C d\tau \frac{i}{\hbar} H_I(\mathbf{q})\right) \right\rangle_\beta, \quad (5)$$

the thermal average of the contour-ordered exponential is taken with respect to the unperturbed reservoir. This exact result can be simplified [8,12] by performing a cumulant expansion of the expectation value in  $I[\mathbf{q}]$ . The most commonly discussed case is that of *linear dissipation*, equivalent to truncating the cumulant expansion after the second order. This procedure is exact for a model reservoir of harmonic oscillators, but also applies to any microscopic model whose collective response to a perturbation  $H_I$  is dominated by its linear term.

In the following we restrict ourselves to linear dissipation, for which  $I[\mathbf{q}]$  is a Gaussian functional determined by the correlation function  $\langle H_I(\mathbf{q}(\tau)) H_I(\mathbf{q}(\tau')) \rangle$ . After an expansion of  $H_I(\mathbf{q})$  in terms of an arbitrary set of reservoir operators  $X_j$ ,

$$H_I = \sum_j f_j(\mathbf{q}) X_j, \quad (6)$$

the correlation matrix  $L_{jk}(\tau - \tau') = \langle X_j(\tau) X_k(\tau') \rangle_\beta$ , characterizing the isolated reservoir, completely determines the influence functional

$$I[\mathbf{q}] = \exp\left(-\frac{1}{\hbar^2} \sum_{j,k} \int_C d\tau \int_{\tau' \prec \tau} d\tau' f_j(\mathbf{q}(\tau)) \times L_{jk}(\tau - \tau') f_k(\mathbf{q}(\tau'))\right). \quad (7)$$

Here “ $\prec$ ” denotes the order relation induced by the contour orientation.

The aim of the present work is to transform the exact description of open quantum systems by the influence functional technique into an equivalent stochastic propagation of the quantum states of the open system. We first show that the influence functional  $I[\mathbf{q}]$  can be constructed as a noise average of the form

$$I[\mathbf{q}] = \left\langle \exp\left(\frac{i}{\hbar} \sum_j \int_C d\tau z_j(\tau) f_j(\mathbf{q}(\tau))\right) \right\rangle_W, \quad (8)$$

where the average is taken with a Gaussian probability measure  $W[z_j]$ . If the stochastic covariance matrix of the complex-valued noise coordinates  $z_j(\tau)$  matches the quantum correlation matrix of the coupling operators  $X_j$ ,

$$\langle z_j(\tau) z_k(\tau') \rangle_W = \begin{cases} \frac{1}{2} L_{jk}(\tau - \tau'), & \tau \geq \tau', \\ \frac{1}{2} L_{jk}(\tau' - \tau), & \tau < \tau', \end{cases} \quad (9)$$

Eqs. (7) and (8) constitute a formal identity between Gaussian functionals. Noting that Eq. (9) does not fully determine the noise statistics, we find that correlations of the type  $\langle z_j(\tau) z_k^*(\tau') \rangle$  can be chosen such that the covariance matrix of all real-valued noise components represents a positive quadratic form; i.e., Gaussian noise  $\{z_j\}$  with the desired properties exists.

For each sample of the noise, the propagation is now governed by a time-local action functional; i.e., the path-integral dynamics can be translated into an equivalent Schrödinger equation. The propagation along the contour  $C$  is governed by the Hamiltonian  $H_0$  and a stochastic potential term,

$$i\hbar |\dot{\psi}\rangle = H_0 |\psi\rangle - \sum_j z_j(\tau) f_j(\mathbf{q}) |\psi\rangle. \quad (10)$$

This remarkably simple equation shows that linear dissipation can be described exactly by a linear QSD theory containing no memory terms.

In order to compare this new finding with previous results, we need to choose the Feynman-Vernon influence functional [6], for which the noise vanishes on the imaginary axis. For simplicity, we discuss a one-dimensional coupling  $H_I = -qx$ , where  $x$  is a reservoir operator with correlation function  $L(\tau - \tau')$ , associated with a single noise coordinate  $z(\tau)$ . The propagation along the two real-time segments of the contour can be restated in the form of a stochastic Liouville equation for the reduced density matrix,

$$i\hbar \frac{d\rho}{dt} = [H_0, \rho] - z_1 q \rho + z_2^* \rho q, \quad (11)$$

where  $z_1(t) = z(t)$  and  $z_2(t) = z(t - i\hbar\beta)$ , and where the sample  $\rho$  of the reduced density matrix is separable,  $\rho = |\psi_1\rangle\langle\psi_2|$ . Hence Eq. (11) is just a compact notation for two stochastic Schrödinger equations for  $|\psi_1\rangle$  and  $|\psi_2\rangle$ .

The noise forces may be represented as the sum of statistically independent terms,  $z_{1,2} = z + v_{1,2}$ . The terms  $v_1$  and  $v_2$  have identical statistics, but are uncorrelated. Performing an average over  $v_{1,2}$  takes us back to the partial decomposition of the influence functional given by Diósi,

Strunz, and Gisin [4]. Using the same representation, the stochastic Schrödinger equation of Markovian QSD can be recovered. The same limits and approximations used to derive Lindblad-type Master equations and their QSD counterparts from a system-reservoir model, i.e., weak damping and time coarse graining, can be applied to Eq. (11). Coarse graining is done at an intermediate time scale which is short compared to the relaxation time of the damped system but long compared to its oscillation periods and the relaxation times of the reservoir. In this approximation the dynamics is sensitive only to segments of the noise spectrum of  $z$  which are centered around the natural transition frequencies of the undamped system. In the Born-Markov limit the coarse-graining time scales to zero, and each of these segments naturally transforms into a distinct Markovian noise force coupled to a Lindblad operator associated with the respective quantum transition. Finally, the work of Stockburger and Mak [5] using real noise and earlier work concentrating on strictly Ohmic damping [13] can be recovered from the present decomposition of the Feynman-Vernon influence functional by applying the stochastic decomposition only to the real part of  $L(\tau - \tau')$  while explicitly evaluating memory effects induced by the imaginary part.

As in the linear versions of conventional QSD,  $\text{tr}\rho$  is not conserved by the time evolution of Eqs. (10) and (11). A transparent physical interpretation of the noise forces and stochastic samples arises in a modified nonlinear theory for which the trace of the reduced density matrix is preserved not only in the stochastic average, but for each sample.

It will be advantageous to introduce new noise variables  $\xi = (z_1 + z_2^*)/2$  and  $\hbar\nu = z_1 - z_2^*$ , with resulting covariances

$$\langle \xi(t)\xi(t') \rangle_W = \text{Re}L(t - t'), \quad (12)$$

$$\begin{aligned} \langle \xi(t)\nu(t') \rangle_W &= (2i/\hbar)\Theta(t - t')\text{Im}L(t - t') \\ &= -i\chi_R(t - t'), \end{aligned} \quad (13)$$

$$\langle \nu(t)\nu(t') \rangle_W = 0, \quad (14)$$

where  $\chi_R$  is the response function of the reservoir. The noise force  $\xi(t)$  can be chosen real, and this case will be discussed here for simplicity. For comparison with phenomenological friction models, one conventionally augments the coupling Hamiltonian by a counterterm in order to make the coupling translationally invariant. The additional term is a quadratic potential modification  $\mu q^2/2$  with  $\mu = \int_0^\infty dt \chi_R(t)$ , which eliminates the static response of the reservoir. Equation (11) is thus transformed into

$$i\hbar\dot{\rho} = [H_0, \rho] + \frac{\mu}{2}[q^2, \rho] - \xi[q, \rho] - \frac{\hbar}{2}\nu\{q, \rho\}. \quad (15)$$

For the normalized density matrix sample  $\hat{\rho} = \rho/\text{tr}\rho$ , this translates into the quasilinear equation of motion

$$\begin{aligned} i\hbar\dot{\hat{\rho}} &= [H_0, \hat{\rho}] + \frac{\mu}{2}[q^2, \hat{\rho}] - \xi[q, \hat{\rho}] \\ &\quad - \frac{\hbar}{2}\nu\{q - \bar{q}, \hat{\rho}\} \end{aligned} \quad (16)$$

with  $\bar{q} = \text{tr}q\hat{\rho}$ . When averaging the normalized density matrix  $\hat{\rho}$ , the factor

$$\text{tr}\rho = \exp\left(i \int_0^t dt' \bar{q}(t')\nu(t')\right) \quad (17)$$

needs to be incorporated into the integration measure. Because the original integration measure  $W[\xi, \nu, \nu^*]$  is Gaussian, the new measure  $W_t[\xi, \nu, \nu^*]$  including the factor  $\text{tr}\rho$  can be rewritten by “completing the square” in the exponent of the Gaussian functional, making the exponent a quadratic form of shifted variables, and yielding  $W_t[\xi, \nu, \nu^*] = W[\xi_t, \nu_t, \nu_t^*]$  with

$$\xi_t(t') = \xi(t') + \int_0^t ds \chi_R(t' - s)\bar{q}(s), \quad (18)$$

$$\nu_t(t') = \nu(t'), \quad (19)$$

$$\nu_t^*(t') = \nu^*(t') - i \int_0^t ds \langle \nu^*(t')\nu(s) \rangle_W \bar{q}(s). \quad (20)$$

Using  $W[\xi_t, \nu_t, \nu_t^*]$  as integration measure allows us to interpret the *shifted* variables as noise to be used for stochastic integration. The Jacobian determinant of the variable change is unity: Because  $\nu_t = \nu$ , and because  $\bar{q}$  is independent of  $\nu^*$ , it simplifies to

$$\begin{vmatrix} \frac{\delta \xi_t}{\delta \xi} & \frac{\delta \xi_t}{\delta \nu} & \frac{\delta \xi_t}{\delta \nu^*} \\ \frac{\delta \nu_t}{\delta \xi} & \frac{\delta \nu_t}{\delta \nu} & \frac{\delta \nu_t}{\delta \nu^*} \\ \frac{\delta \nu_t^*}{\delta \xi} & \frac{\delta \nu_t^*}{\delta \nu} & \frac{\delta \nu_t^*}{\delta \nu^*} \end{vmatrix} = \begin{vmatrix} \frac{\delta \xi_t}{\delta \xi} & \frac{\delta \xi_t}{\delta \nu} & 0 \\ 0 & 1 & 0 \\ \frac{\delta \nu_t^*}{\delta \xi} & \frac{\delta \nu_t^*}{\delta \nu} & 1 \end{vmatrix} = \left| \frac{\delta \xi_t}{\delta \xi} \right|. \quad (21)$$

Using the fact that both the bath response and  $\bar{q}[\xi, \nu]$  are causal, one finds that  $|\delta \xi_t/\delta \xi|$  is represented by a triangular matrix with unit diagonal elements, i.e.,  $|\delta \xi_t/\delta \xi| = 1$ .

After the variable change (with subscripts  $t$  now dropped) the equation of motion is rewritten as

$$\begin{aligned} i\hbar\dot{\hat{\rho}} &= [H_0, \hat{\rho}] - \xi[q, \hat{\rho}] + m\gamma(t)\bar{q}(0)[q, \hat{\rho}] \\ &\quad + m \int_0^t dt' \gamma(t - t')\dot{\bar{q}}(t')[q, \hat{\rho}] \\ &\quad + \frac{\mu}{2}[(q - \bar{q})^2, \hat{\rho}] - \frac{\hbar}{2}\nu\{q - \bar{q}, \hat{\rho}\}. \end{aligned} \quad (22)$$

Here we have integrated by parts, using the relation  $m\dot{\gamma}(t) = -\chi_R(t)$  between the reservoir response function and the friction kernel  $\gamma(t)$ . Remarkably, the mere choice of a constraint for  $\text{tr}\hat{\rho}$  has taken us from the abstract mathematical decomposition (8) to an equation of motion with a number of terms looking quite familiar; as in classical dissipation, the dynamics is governed by  $c$ -number force terms representing noise and a mean-field version of classical friction. Additional terms ascertain that the noise-averaged  $\hat{\rho}$  contains the effect of all system-reservoir correlations.

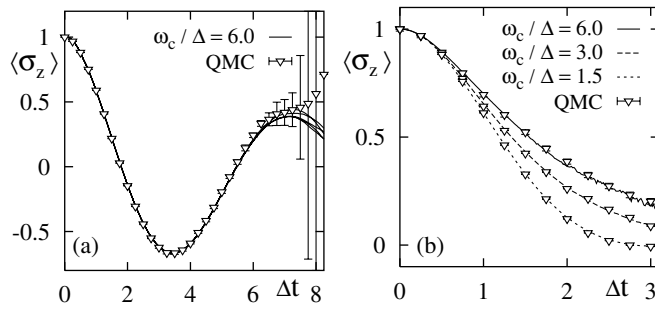


FIG. 2. Stochastic and quantum Monte Carlo simulations of a symmetric spin-boson system with  $\chi_R(t) \propto t \exp(-\omega_c t)$ , dissipation constant [8]  $\alpha = 0.1$ . (a) Coherent dynamics at zero temperature. [Five repeated simulations of Eq. (15) are shown to indicate statistical errors.] (b) Incoherent dynamics at  $T = 5\hbar\Delta/k_B$ , simulated using Eq. (22).

Further examining this analogy, we find a “sample-by-sample” correspondence between quantum mechanical and classical stochastic dynamics. The time evolution of Eq. (22) can be described in a reduced Heisenberg picture, which is defined by interpreting expectation values of the form  $\text{tr}\{A^\dagger \hat{\rho}\}$  as a scalar product  $(A, \hat{\rho})$  of two operators. This allows the introduction of Heisenberg operators  $A(t)$  through the propagating superoperator  $\mathcal{U}(t)$  and its adjoint,

$$(A, \hat{\rho}(t)) = (A, \mathcal{U}(t)\hat{\rho}_0) = (\mathcal{U}^\dagger(t)A, \hat{\rho}_0) = (A(t), \rho_0). \quad (23)$$

In the classical limit, terms proportional to  $q - \bar{q}$  in Eq. (22) can be neglected, and the equations of motion governing the evolution of Heisenberg operators  $A(t)$  become those of a classical stochastic system. For a potential model, these reduce [14] to the generalized Langevin equation

$$m\ddot{q}(t) = -V'(q(t)) - m \int_0^t dt' \gamma(t-t')\dot{q}(t') + \xi(t). \quad (24)$$

The limit  $\hbar \rightarrow 0$  turns our stochastic ensemble of quantum states gradually into an ensemble of phase-space points with classical thermal noise and memory friction; i.e., the quantum dynamics defined in Eq. (22) obeys a stochastic correspondence principle.

Figure 2 shows numerical results for the symmetric spin-boson system [15], where  $q$  is the Pauli matrix  $\sigma_z$  and  $H_0 = (\hbar\Delta/2)\sigma_x$ , compared to a QMC algorithm which directly samples a path integral [10]. For the coherent dynamics shown in Fig. 2a, the QMC method does not fully converge even if run longer than our simulation. Because of an only marginally ergodic Metropolis random walk, the accuracy of the QMC method degrades significantly with increasing  $t$ . Problems of this kind

never appear in our algorithm because our samples are statistically independent by construction. The incoherent dynamics shown in Fig. 2b, a forte of the QMC method, is treated equally well by both approaches.

We have derived exact stochastic equations of motion for the reduced density matrix in linear form (10) and trace-conserving form (22). In the latter, a close correspondence with classical Langevin equations is evident. Examples of numerical results show that the stochastic equations of motion presented here provide a promising alternative to currently available exact methods based directly on the path integral representation. Given the freedom of choice for the noise covariances left undetermined by the physics of quantum dissipation, future improved numerical methods are to be anticipated.

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