

## Non-Markovian stochastic Schrödinger equation

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We report a study of a stochastic Schrödinger equation corresponding to the Redfield master equation with slipped initial conditions, which describes the dynamics of a slow subsystem weakly coupled to a fast thermal bath. Using the projection-operator method by Feshbach, we derive a non-Markovian stochastic Schrödinger equation of the generalized Langevin type, which simulates the time evolution of the quantum wavefunctions of the subsystem driven by the fluctuating bath. For  $\delta$ -correlated baths, the non-Markovian stochastic Schrödinger equation reduces to the previously derived Markovian one. Numerical methods are proposed to simulate the time evolution under our non-Markovian stochastic Schrödinger equation. These methods are illustrated with the spin-boson model.

### I. INTRODUCTION

The coupling of a small system to a larger system has been repetitively studied since the pioneering works of Einstein, Langevin, and others on Brownian motion [1, 2]. In classical dynamics, two interrelated and equivalent descriptions are commonly used: on one hand, a master equation of the Chapman-Kolmogorov type ruling the time evolution of the probability density of the subsystem variables, such as the Fokker-Planck equation for the Brownian motion; on the other hand, a Langevin-type stochastic differential equation ruling the time evolution of individual and random trajectories of the subsystem [3]. Both descriptions are equivalent in the sense that the master equation can be derived from the corresponding stochastic differential equation. We notice that the original Langevin stochastic differential equation describes a Markovian time evolution. Recently, non-Markovian classical time evolutions have been studied with generalized Langevin equations [4]. These generalized Langevin equations are particularly important for reactions in condensed phases [5].

Stochastic schemes have also been recently developed for open quantum systems [3, 6–10]. In many problems, a small quantum system is coupled to a larger one which plays the role of a bath or reservoir. This situation happens in particular for spins or effective two-level subsystems embedded in condensed phases, as well as for reactive subsystems in liquids for instance such as charge-transfer (CT) reactions where the intrinsic time scale of CT processes should be comparable to the bath relaxation. For such open quantum systems, several master equations have been derived for the density matrix describing the subsystem by different methods since Pauli's seminal work [11].

In particular, Lindblad has derived in the seventies the most general Markovian master equation which explicitly preserves positivity during the whole time evolution [12]. Recently, stochastic Schrödinger equations of the Langevin type have been proposed which corresponds to the Lindblad master equation in a similar way as the Langevin stochastic equation corresponds to the Fokker-Planck equation [3, 6, 7, 10]. These stochastic Schrödinger equations have been introduced with the aim to simulate the quantum jumps observed in trapped atoms or molecules continuously excited by laser [6, 7, 10]. In such open quantum systems, the stochastic Schrödinger equation is supposed to rule the stochastic time evolution of the quantum wavefunction describing an atom or molecule which undergoes random fluctuations caused by the coupling to the environment. Besides the conceptual clarity provided by such stochastic Schrödinger equations, they also provide a great advantage in numerical simulations where their time integration only involves a number of variables equal to the dimension  $N$  of the quantum state space of the subsystem. If the corresponding master equation had to be integrated, the number of variables should include all the  $N^2$  elements of the density matrix of the subsystem, which may become computationally prohibitive as the dimension  $N$  increases.

However, the Lindblad master equation is applicable only to subsystems coupled to the so-called  $\delta$ -correlated baths such that the time correlation functions of the coupling operators to the bath are proportional to the Dirac delta distribution. A master equation which is valid for more general baths than the  $\delta$ -correlated ones was derived by Redfield in the context of NMR [13]. This master equation, that is called today the Redfield equation, describes the time evolution of a slow subsystem weakly coupled to a fast bath characterized by general time correlation functions, and it has been applied to many systems. Nevertheless, a stochastic Schrödinger equation corresponding to the Redfield master equation has not yet been found.

The aim of the present paper is to obtain such a stochastic Schrödinger equation which could apply to subsystems coupled to general baths which are not necessarily  $\delta$ -correlated. For this goal, we first introduce a concept of “subsystem wavefunction” to describe a subsystem of the total system. Thereafter, we apply the Feshbach projection-operator method to the Schrödinger equation of the total system in order to obtain a stochastic Schrödinger equation for the subsystem wavefunctions. It should be noticed that the Feshbach projection-operator method is applied to the Schrödinger equation ruling the wavefunction and, for this reason, it differs from the Mori-Zwanzig projection-operator method that is instead applied to the Liouvillian master equation ruling the density matrix. The Feshbach projection-operator method has often been used in the scattering theory to obtain an effective Hamiltonian with an optical potential for the study of reactive collisions, not only in nuclear physics [14] but also in the study of unimolecular reactions [15]. When the Feshbach projection-operator method is extended from scattering systems to large many-body systems which include a small subsystem continuously interacting with a thermal bath, the term which contains the initial conditions can no longer be neglected and turns out to describe the stochastic forcing of the bath against the small subsystem. This observation allows us to derive a stochastic Schrödinger equation which turns out to be non-Markovian and valid for general baths characterized by specific correlations functions. Moreover, the Redfield master equation with slipped initial conditions can be derived from the non-Markovian stochastic Schrödinger equation. Besides, in the case of  $\delta$ -correlated baths, the non-Markovian stochastic Schrödinger equation reduces to the previously known Markovian stochastic Schrödinger equation, while the Redfield master equation reduces to the Lindblad master equation. The methods are illustrated for the example of the spin-boson model.

The plan of the paper is as follows. In Sec. II, we introduce the concept of “subsystem wavefunction” for a small subsystem coupled to a large bath and we set up the statistical framework as well as the interaction representations. In Sec. III, we apply the Feshbach projection-operator method to the Schrödinger equation of the total system. A perturbative expansion in the coupling parameter is performed in Sec. IV. The stochastic forcing is derived in Sec. V and the non-Markovian stochastic Schrödinger equation is finally established in Sec. VI. In Sec. VII, we show how the Redfield master equation with slipped initial conditions is obtained as a consequence of the non-Markovian stochastic Schrödinger equation. In Sec. VIII, we further show that our non-Markovian stochastic Schrödinger equation reduces to the previously known one in the limit of a  $\delta$ -correlated bath. Numerical methods for the simulation of our stochastic Schrödinger equation are given in Sec. IX. The methods are applied to the spin-boson model in Sec. X. Conclusions are finally drawn in Sec. XI.

## II. STATISTICAL ENSEMBLE OF SUBSYSTEM WAVEFUNCTIONS

In order to derive a stochastic Schrödinger equation, we need to introduce a concept of “subsystem wavefunction” to describe a subsystem interacting with a reservoir, forming together a total system. For this purpose, we need to address the well-known problem of the nonseparability of two coupled subsystems in quantum mechanics. However, the time evolution can be formulated in the simplest way at the level of the total system that is composed of a small subsystem and a bath.

We suppose that a subsystem of Hamiltonian  $H_s$  is coupled to a bath of Hamiltonian  $H_b$  by an interaction potential  $\lambda V$ , where  $\lambda$  is the coupling parameter. The total Hamiltonian of this system is thus

$$H = H_0 + \lambda V = H_s + H_b + \lambda V . \quad (1)$$

The potential is assumed of the form

$$V = V^\dagger = \sum_{\alpha} S_{\alpha} B_{\alpha} , \quad (2)$$

where  $S_{\alpha}$  and  $B_{\alpha}$  are respectively the subsystem and bath operators. These operators can always be redefined as Hermitian operators [16], so that we may assume in the following that  $S_{\alpha}^\dagger = S_{\alpha}$  and  $B_{\alpha}^\dagger = B_{\alpha}$  [17].

We denote by  $x_s$  and  $x_b$  the coordinates of the subsystem and bath degrees of freedom, respectively. The wavefunction of the total system belongs to the Hilbert space that is the direct product of the Hilbert space of the subsystem  $\mathcal{H}_s$  with that of the bath  $\mathcal{H}_b$

$$\Psi(x_s, x_b) \in \mathcal{H} = \mathcal{H}_s \otimes \mathcal{H}_b . \quad (3)$$

According to the principles of quantum mechanics, the wavefunction of the total system obeys the time-dependent Schrödinger equation

$$i \partial_t \Psi(x_s, x_b; t) = H \Psi(x_s, x_b; t) = \left( H_s + H_b + \lambda \sum_{\alpha} S_{\alpha} B_{\alpha} \right) \Psi(x_s, x_b; t), \quad (4)$$

where  $H$  is the total Hamiltonian (1)-(2).

We consider a complete and orthonormal basis set in the Hilbert space of the bath

$$\{\chi_n(x_b) = \langle x_b | n \rangle \in \mathcal{H}_b\} : \quad \langle m | n \rangle = \delta_{mn}, \quad \sum_n |n\rangle \langle n| = I_b, \quad (5)$$

where  $\chi_n(x_b)$  denote the basis functions and  $|n\rangle$  the corresponding basis kets. For instance, the basis functions can be chosen as the eigenfunctions of the bath Hamiltonian

$$H_b \chi_n(x_b) = \epsilon_n \chi_n(x_b), \quad (6)$$

where  $\{\epsilon_n\}$  are the associated eigenenergies of the bath.

The total wavefunction can be decomposed in this basis as

$$\Psi(x_s, x_b; t) = \sum_n \phi_n(x_s; t) \chi_n(x_b), \quad (7)$$

where  $\{\phi_n(x_s; t)\}$  are the coefficients of this linear decomposition, which are given by

$$\phi_n(x_s; t) = \int dx_b \chi_n^*(x_b) \Psi(x_s, x_b; t). \quad (8)$$

Since the basis functions only depend on the bath degrees of freedom, the coefficients of this decomposition take into account the dependence of the total wavefunction on the subsystem degrees of freedom. Therefore, the coefficients are functions of the subsystem coordinates,  $x_s$ , as well as of the time  $t$ . The total wavefunction is normalized to unity so that

$$\int dx_s dx_b |\Psi(x_s, x_b; t)|^2 = \sum_n \int dx_s |\phi_n(x_s; t)|^2 = 1. \quad (9)$$

However, we notice that the coefficients  $\phi_n$  themselves are not normalized to unity. Indeed, the square of their norm

$$p_n(t) = \int dx_s |\phi_n(x_s; t)|^2 = \|\phi_n(t)\|^2, \quad (10)$$

gives the probability for the bath to be observed in the corresponding basis function  $\chi_n(x_b)$ .

In order to interpret further these coefficients, let us consider the average value of an observable  $A_s$  of the subsystem,

$$\begin{aligned} \langle \Psi | A_s | \Psi \rangle &= \int dx_s dx_b \Psi^* A_s \Psi \\ &= \sum_{mn} \left( \int dx_s \phi_m^* A_s \phi_n \right) \left( \int dx_b \chi_m^* \chi_n \right) \\ &= \sum_n \int dx_s \phi_n^* A_s \phi_n = \sum_n \langle \phi_n | A_s | \phi_n \rangle \\ &= \text{tr}_s \rho_s A_s, \end{aligned} \quad (11)$$

where we have used the orthonormality of the basis functions of the bath (5). Hence, we observe that the average of a subsystem observable over the pure state given by the total wavefunction  $\Psi$  can be expressed as an average over the statistical mixture represented by the subsystem density matrix

$$\rho_s \equiv \sum_n |\phi_n\rangle \langle \phi_n|, \quad (12)$$

which follows the time evolution of the coefficients of the decomposition (7) according to

$$\langle x_s | \rho_s(t) | x'_s \rangle = \sum_n \phi_n(x_s; t) \phi_n(x'_s; t)^*. \quad (13)$$

Eq. (12) can be rewritten in terms of the probabilities (10) as

$$\rho_s(t) \equiv \sum_n |\check{\phi}_n(t)\rangle p_n(t) \langle \check{\phi}_n(t)|, \quad (14)$$

where we have introduced the normalized wavefunctions  $\check{\phi}_n(x_s; t) = \phi_n(x_s; t) / \|\phi_n(t)\|$ . In this form, we observe that the wavefunctions  $\check{\phi}_n(x_s; t)$  form a statistical ensemble of wavefunctions describing the subsystem. The probability that each one of these wavefunctions occurs in the statistical ensemble is given by the probability (10) to observe the bath in the corresponding basis function  $\chi_n$ . In this way, we notice that the subsystem cannot be described by a single wavefunction but by an ensemble of wavefunctions, which has its origin in the property of nonseparability of quantum mechanics. Therefore, the ensemble of subsystem wavefunctions has a natural statistical and probabilistic interpretation if the bath is a large subsystem.

The next step in our derivation of the stochastic Schrödinger equation is to find the equations which rule the time evolution of each wavefunction of our statistical ensemble. Inserting the decomposition (7) into the Schrödinger equation (4), taking the scalar product of both sides with each basis eigenfunction (5)-(6), and using their orthonormality, we obtain the following set of coupled equations for the coefficients

$$i \partial_t \phi_m(x_s; t) = H_s \phi_m(x_s; t) + \epsilon_m \phi_m(x_s; t) + \lambda \sum_{\alpha} \sum_n \langle m|B_{\alpha}|n\rangle S_{\alpha} \phi_n(x_s; t). \quad (15)$$

We can introduce a *partial* interaction picture with the definitions

$$\tilde{\phi}_n(x_s; t) \equiv e^{i\epsilon_n t} \phi_n(x_s; t), \quad (16)$$

and

$$B_{\alpha}(t) \equiv e^{iH_b t} B_{\alpha} e^{-iH_b t}, \quad (17)$$

in order to transform the equations (15) into

$$i \partial_t \tilde{\phi}_m(x_s; t) = H_s \tilde{\phi}_m(x_s; t) + \lambda \sum_{\alpha} \sum_n \langle m|B_{\alpha}(t)|n\rangle S_{\alpha} \tilde{\phi}_n(x_s; t). \quad (18)$$

Furthermore, we have the *complete* interaction picture with the definitions

$$\varphi_n(x_s; t) \equiv e^{iH_s t} e^{i\epsilon_n t} \phi_n(x_s; t), \quad (19)$$

and

$$S_{\alpha}(t) \equiv e^{iH_s t} S_{\alpha} e^{-iH_s t}, \quad (20)$$

in order to transform the equations (15) into

$$i \partial_t \varphi_m(x_s; t) = \lambda \sum_{\alpha} \sum_n \langle m|B_{\alpha}(t)|n\rangle S_{\alpha}(t) \varphi_n(x_s; t). \quad (21)$$

These coupled differential equations give the time evolution of the coefficients in the linear decomposition (7).

### III. THE FESHBACH PROJECTION-OPERATOR METHOD

The idea behind the derivation of a stochastic Schrödinger equation is that the different coefficients  $\phi_n(x_s; t)$  behave in a random way not only because of their mutual interaction under the evolution given by Eqs. (15) but also because of the large number of these coefficients. Indeed, the bath being a large subsystem, its density of energy levels is very high so that the energy spectrum is very dense. Since each eigenenergy of the bath is associated with a coefficient in the decomposition (7), we may understand that the time evolution of a typical coefficient, such as  $\phi_l(x_s; t)$  taken among all these coefficients, is affected by a very large set of coefficients which are coupled to it by the coupling matrix elements  $\langle l|B_{\alpha}|n\rangle$ .

In this regard, we may look after an equation for a typical coefficient among all the others. A method of choice to carry out this program is the projection-operator method by Feshbach where the Schrödinger equation is decomposed

into two subequations using the projection operators  $P$  and  $Q$  acting in the Hilbert space (3) of the total system [14, 15]. These projection operators should obey the usual properties

$$\begin{aligned} P^2 &= P = P^\dagger, & Q^2 &= Q = Q^\dagger, \\ QP &= PQ = 0, & P + Q &= I. \end{aligned} \quad (22)$$

We notice that the method can be developed for each one of the four equivalent forms (4), (15), (18), and (21) of the Schrödinger equation. However, let us here carry out the discussion for the generic form (4). In this case, the projection operators can be chosen as

$$P \equiv I_s \otimes |l\rangle \langle l|, \quad Q \equiv I_s \otimes \sum_{n(\neq l)} |n\rangle \langle n|, \quad (23)$$

or equivalently

$$(P\Psi)(x_s, x_b) \equiv \phi_l(x_s) \chi_l(x_b), \quad (24)$$

$$(Q\Psi)(x_s, x_b) \equiv \sum_{n(\neq l)} \phi_n(x_s) \chi_n(x_b). \quad (25)$$

Applying these projection operators to the Schrödinger equation, we obtain the following two coupled subequations

$$\begin{cases} i \partial_t P\Psi = PHP\Psi + PHQ\Psi, \\ i \partial_t Q\Psi = QHQ\Psi + QHP\Psi. \end{cases} \quad (26)$$

In order to obtain an equation for  $\phi_l$ , i.e., for  $P\Psi$ , we first need to solve the second equation for  $Q\Psi$  and to insert its solution into the first equation. The second equation is an inhomogeneous linear equation for  $Q\Psi$ , the solution of which is given by the method of variation of the constants of integration of the corresponding homogeneous linear equation. The solution is thus

$$Q\Psi(t) = e^{-iQHQt} Q\Psi(0) - i \int_0^t d\tau e^{iQHQ(\tau-t)} QHP P\Psi(\tau), \quad (27)$$

where  $Q\Psi(0)$  is the initial condition for all the coefficients  $\{\phi_n(0)\}_{n(\neq l)}$  except the coefficient  $\phi_l(0)$  that is ruled by the first equation for  $P\Psi$ . We now insert the solution (27) of the second equation into the first of Eqs. (26) to obtain finally the equation

$$\begin{aligned} i \partial_t P\Psi(t) &= PHP P\Psi(t) + PHQ e^{-iQHQt} Q\Psi(0) \\ &\quad - i \int_0^t d\tau PHQ e^{iQHQ(\tau-t)} QHP P\Psi(\tau). \end{aligned} \quad (28)$$

We have therefore obtained a closed equation for  $P\Psi(t)$  which is the coefficient  $\phi_l(x_s; t)$  of the total wavefunction.

The three terms in the r. h. s. of Eq. (28) can be interpreted as follows: The first one is nothing but the Hamiltonian operator  $H_s$  of the subsystem. The second one depends on the initial condition  $Q\Psi(0)$  of all the other coefficients but  $P\Psi$ . This term will be found in the following to be the stochastic forcing against the subsystem due to the fluctuations of the bath. The third one represents the damping of the subsystem wavefunction coefficient,  $P\Psi$  or  $\phi_l$ , due to its coupling to the other coefficients  $Q\Psi$ . This damping is thus caused by the interaction of the subsystem with the bath. It should be noticed that this damping term turns out to be non-Markovian.

In the scattering theory of reactive collisions, a similar projection-operator method has been developed since Feshbach pioneering work [14]. This technique has been applied by Nordholm and Rice to the study of unimolecular reactions [15]. In this context, the system is divided into a region  $\mathcal{B}$  of the potential energy surface where the quasi-bound states of the molecule are expected to be localized and another region  $\mathcal{F}$  containing the quasi-free states of the dissociating fragments of the molecule. In this situation, the projection operators project onto these two regions,  $P = P_{\mathcal{B}}$  and  $Q = P_{\mathcal{F}}$ , respectively. For the study of dissociation, i.e., half-collisions, it is natural to consider an initial condition which is localized in the quasi-bound region so that  $Q\Psi(0) = P_{\mathcal{F}}\Psi(0) = 0$ . Therefore, the second term in Eq. (28) is often disregarded in the applications of the Feshbach method to scattering systems. For these systems, the third term gives the optical potential and provides together with the first term an effective Hamiltonian which is non-Hermitian [14].

However, in the case of a small subsystem interacting with its environment, it is very exceptional that the bath is observed in one of its eigenstates for a typical initial condition of the total system. Therefore, the second term in

the r.h.s. of Eq. (28) cannot be supposed to vanish and Eq. (28) is inhomogeneous. Consequently, the second term in the equation for  $P\Psi(t)$  appears as a time-dependent forcing term due to the coupling to the bath. It should be noticed that the forcing is fast if the bath has a faster dynamics than the subsystem. Moreover, we may expect that this time-dependent forcing becomes random for an arbitrarily large bath defined in a thermodynamic limit. This reasoning leads us to the conclusion that the second term in the r.h.s. of Eq. (28) corresponds to the stochastic forcing in the equation for  $\phi_l(x_s; t)$ .

In order to reach a specific stochastic differential equation, we shall first rederive Eq. (28) in the complete interaction picture (19)-(21). For simplicity, we assume that the average values of the coupling operators  $B_\alpha$  vanish for the eigenstate  $\chi_l$ :  $\langle l|B_\alpha|l\rangle = 0$ . Otherwise, the subsystem Hamiltonian can be redefined as  $H'_s = H_s + \lambda \sum_\alpha \langle l|B_\alpha|l\rangle S_\alpha$ , with a corresponding redefinition of the interaction potential into  $V' = V - \sum_\alpha \langle l|B_\alpha|l\rangle S_\alpha$ . Accordingly, Eqs. (21) can be separated into two coupled equations as in Eqs. (26):

$$\begin{cases} i \partial_t \varphi_l = \lambda \sum_\alpha S_\alpha(t) \mathbf{B}_{\alpha l}(t)^\dagger \cdot \Phi \\ i \partial_t \Phi = \lambda \mathbf{W}(t) \cdot \Phi + \lambda \sum_\alpha \mathbf{B}_{\alpha l}(t) S_\alpha(t) \varphi_l, \end{cases} \quad (29)$$

where  $\varphi_l$  represents  $P\Psi$  in the complete interaction picture, while  $\Phi$  represents  $Q\Psi$  and is a column vector gathering all the coefficients  $\varphi_n$  but  $\varphi_l$

$$\Phi(x_s; t) \equiv \{\varphi_n(x_s; t)\}_{n(\neq l)}. \quad (30)$$

In Eqs. (29), we have also defined the following column vector and matrix

$$\mathbf{B}_{\alpha l}(t) \equiv \{\langle m|B_\alpha(t)|l\rangle\}_{m(\neq l)}, \quad (31)$$

$$\mathbf{W}(t) \equiv \left[ \sum_\beta \langle m|B_\beta(t)|n\rangle S_\beta(t) \right]_{m,n(\neq l)}. \quad (32)$$

Using the same integration over time as in Eqs. (26) to (28), we obtain the following closed equation

$$\begin{aligned} i \partial_t \varphi_l(t) &= \lambda \sum_\alpha S_\alpha(t) \mathbf{B}_{\alpha l}(t)^\dagger \cdot \mathbf{U}(t) \cdot \Phi(0) \\ &- i \lambda^2 \int_0^t d\tau \sum_{\alpha\beta} S_\alpha(t) \mathbf{B}_{\alpha l}(t)^\dagger \cdot \mathbf{U}(t-\tau) \cdot \mathbf{B}_{\beta l}(\tau) S_\beta(\tau) \varphi_l(\tau), \end{aligned} \quad (33)$$

where we have introduced an evolution operator  $\mathbf{U}(t)$ , which is the solution of

$$i \partial_t \mathbf{U}(t) = \lambda \mathbf{W}(t) \cdot \mathbf{U}(t), \quad (34)$$

issued from the initial condition  $\mathbf{U}(0) = \mathbf{I}$ . We emphasize that no approximation has been made until now so that Eq. (33) is strictly equivalent to the original Schrödinger equation for the total system. We notice that Eq. (33) is equivalent to the non-Markovian equation (28) because the former is nothing but the rewriting of the latter in the interaction picture. The first term in the r.h.s. of Eq. (33) represents the forcing against the coefficient  $\varphi_l$  by the bath, while the second term represents the damping due to the bath.

#### IV. PERTURBATIVE EXPANSION IN THE COUPLING PARAMETER $\lambda$

Since our purpose is to derive a stochastic Schrödinger equation which corresponds to the Redfield master equation that is obtained within the second-order perturbation theory, we shall here perform a perturbative expansion of the non-Markovian equation (33) and truncate the expansion at the second order in the coupling parameter  $\lambda$ , which is a weak-coupling approximation.

The perturbative expansion of the evolution operator that is a solution of Eq. (34) is obtained as

$$\mathbf{U}(t) = \mathbf{I} - i \lambda \int_0^t dt_1 \mathbf{W}(t_1) - \lambda^2 \int_0^t dt_1 \int_0^{t_1} dt_2 \mathbf{W}(t_1) \cdot \mathbf{W}(t_2) + \mathcal{O}(\lambda^3). \quad (35)$$

If we insert this into Eq. (33) and truncate at the second order, we get

$$i \partial_t \varphi_l(t) = f_l(t) - i \lambda^2 \int_0^t d\tau \sum_{\alpha\beta} S_\alpha(t) \langle l|B_\alpha(t)B_\beta(\tau)|l\rangle S_\beta(\tau) \varphi_l(\tau) + \mathcal{O}(\lambda^3), \quad (36)$$

with the forcing term given by

$$\begin{aligned}
f_l(t) &= \lambda \sum_{\alpha} \sum_{m(\neq l)} S_{\alpha}(t) \langle l|B_{\alpha}(t)|m\rangle \varphi_m(0) \\
&- i \lambda^2 \int_0^t d\tau \sum_{\alpha\beta} \sum_{m(\neq l)} S_{\alpha}(t) \langle l|B_{\alpha}(t)B_{\beta}(\tau)|m\rangle S_{\beta}(\tau) \varphi_m(0) + \mathcal{O}(\lambda^3).
\end{aligned} \tag{37}$$

In the previous steps to get Eqs. (36) and (37), we have used the assumption  $\langle l|B_{\alpha}|l\rangle = 0$  so that

$$\sum_{n(\neq l)} \langle l|B_{\alpha}(t)|n\rangle \langle n|B_{\beta}(\tau)|m\rangle = \langle l|B_{\alpha}(t)B_{\beta}(\tau)|m\rangle. \tag{38}$$

## V. STATISTICAL TYPICALITY AND THE STOCHASTIC NOISES

In order to obtain a stochastic differential equation, we need to assume that the coefficient  $\varphi_l(t)$  represents statistically each one of the coefficients  $\varphi_n(t)$  of the linear decomposition (7) of the total wavefunction in the interaction picture. In this sense, we should assume that all the coefficients  $\varphi_n(t)$  behave similarly and form a statistical ensemble for which the coefficient  $\varphi_l(t)$  is a typical representative coefficient. This assumption shall be called the *assumption of statistical typicality*. We notice that there is no reason to believe that such an assumption should always be valid. For special systems with a special choice of the basis function  $\chi_l(x_b)$ , this assumption would not be valid, but it must be reasonable to assume its validity for a majority of basis functions  $\chi_l$ .

The assumption of statistical typicality can be justified if the bath subsystem is classically chaotic. Namely, according to a conjecture by Berry [18], which has been mathematically proved for certain classically hyperbolic systems [19], the average of a bath operator  $A_b$  over a typical eigenstate  $\chi_l$  is essentially equivalent to a classical average over the microcanonical statistical ensemble at the energy equal to the eigenenergy  $\epsilon_l$ . This behavior has its origin in the property that typical eigenfunctions are statistically irregular at high quantum numbers. Moreover, since the bath is a large subsystem, this microcanonical average is essentially equivalent to a canonical average as recently argued by Srednicki [20]. Therefore, we may expect that the average over a typical eigenstate  $\chi_l$  of the bath is approximately equal to the thermal average at the temperature of the bath:

$$\langle l|A_b|l\rangle \approx \text{tr}_b \frac{e^{-\beta H_b}}{Z_b} A_b \equiv \text{tr}_b \rho_b^{\text{eq}} A_b, \tag{39}$$

where  $Z_b = \text{tr}_b \exp(-\beta H_b)$ . The inverse temperature  $\beta = (k_B T)^{-1}$  should be fixed by the knowledge of the bath energy  $\epsilon_l$  and the condition that the subsystem is very small with respect to the bath [20].

As a consequence of the assumption of statistical typicality of the eigenfunction  $\chi_l$ , the damping term in Eq. (36) involves the correlation functions of the bath operators,

$$\langle l|B_{\alpha}(t)B_{\beta}(\tau)|l\rangle \approx \text{tr}_b \frac{e^{-\beta H_b}}{Z_b} B_{\alpha}(t)B_{\beta}(\tau) \equiv C_{\alpha\beta}(t - \tau), \tag{40}$$

so that the form of the damping term should be essentially independent of the particular coefficient that has been chosen among the ensemble of coefficients. It must be emphasized that the time correlation functions  $C_{\alpha\beta}(t)$  in Eq. (40) are precisely those which appear in the Redfield master equation [16].

The forcing term (37) can be treated similarly. In order to obtain the typical behavior of the forcing term, we need to make assumptions on the initial condition of the total wavefunction because the initial condition enters as an essential ingredient in the forcing (37). The initial wavefunction of the total system  $\Psi(x_s, x_b; t = 0)$  is assumed to be a pure state which should be equivalent to the direct product of a pure state for the initial state of the subsystem and a canonical mixed state for the bath

$$\langle \Psi(0)|A|\Psi(0)\rangle \approx \text{tr} \left[ |\psi(0)\rangle \langle \psi(0)| \otimes \frac{e^{-\beta H_b}}{Z_b} \right] A, \tag{41}$$

where  $\psi(0)$  is the subsystem wavefunction at the initial time  $t = 0$  and with the normalization  $\|\psi(0)\| = 1$ . If Eq. (41) is expanded in the basis of the eigenfunctions (6) of the bath, the following expression is obtained

$$\langle \Psi(0)|A|\Psi(0)\rangle \approx \sum_m \frac{e^{-\beta \epsilon_m}}{Z_b} \int dx_s \int dx_b \psi^*(x_s; 0) \chi_m^*(x_b) A \chi_m(x_b) \psi(x_s; 0). \tag{42}$$

Since the total wavefunction admits the decomposition (7) in the basis  $\{\chi_m(x_b)\}$ , the approximate equality (42) can be established under the assumption that the initial condition of the total wavefunction is given by

$$\Psi(x_s, x_b; 0) \approx \psi(x_s; 0) \sum_m \sqrt{\frac{e^{-\beta\epsilon_m}}{Z_b}} e^{i\theta_m} \chi_m(x_b), \quad (43)$$

where  $\{\theta_m\}$  form a set of independent random phases, each of which is uniformly distributed in the interval  $[0, 2\pi[$ . The assumption (43) implies that the initial conditions of the coefficients (8) of the decomposition (7) take the form

$$\phi_m(x_s; 0) = \varphi_m(x_s; 0) \approx \psi(x_s; 0) \sqrt{\frac{e^{-\beta\epsilon_m}}{Z_b}} e^{i\theta_m}, \quad (44)$$

which also gives the initial conditions in the interaction pictures (16) and (19).

An important consequence of Eq. (44) is that all the initial coefficients are proportional to the same initial wavefunction of the subsystem. In particular, the chosen coefficient  $\varphi_l$  is also proportional to the same initial wavefunction because Eq. (44) also holds for the special coefficient with  $m = l$ . Hence, we find that

$$\varphi_m(x_s; 0) \approx \varphi_l(x_s; 0) e^{-\beta(\epsilon_m - \epsilon_l)/2} e^{i(\theta_m - \theta_l)}. \quad (45)$$

This result is very important to obtain a *closed* stochastic differential equation which depends only on the coefficient  $\varphi_l$ . Indeed, according to our assumptions, the forcing term (37) becomes

$$\begin{aligned} f_l(t) &\approx \lambda \sum_{\alpha} \sum_{m(\neq l)} S_{\alpha}(t) \langle l|B_{\alpha}(t)|m\rangle e^{-\beta(\epsilon_m - \epsilon_l)/2} e^{i(\theta_m - \theta_l)} \varphi_l(0) \\ &- i \lambda^2 \int_0^t d\tau \sum_{\alpha\beta} \sum_{m(\neq l)} S_{\alpha}(t) \langle l|B_{\alpha}(t)B_{\beta}(\tau)|m\rangle S_{\beta}(\tau) e^{-\beta(\epsilon_m - \epsilon_l)/2} e^{i(\theta_m - \theta_l)} \varphi_l(0) + \mathcal{O}(\lambda^3) \\ &\approx \lambda \sum_{\alpha} \sum_{m(\neq l)} S_{\alpha}(t) \langle l|B_{\alpha}(t)|m\rangle e^{-\beta(\epsilon_m - \epsilon_l)/2} e^{i(\theta_m - \theta_l)} \varphi_l(t) \\ &\equiv \lambda \sum_{\alpha} \eta_{\alpha}(t) S_{\alpha}(t) \varphi_l(t), \end{aligned} \quad (46)$$

where we have supposed that the second term in the perturbative expansion gives an approximation for the time evolution of the coefficient  $\varphi_l(t)$ . This approximation can be justified by the fact that, in the interaction picture, the coefficient has a slow time evolution in the weak-coupling approximation where the coupling parameter  $\lambda$  is small with respect to the ratio between the subsystem Hamiltonian and the coupling potential energy.

In view of Eq. (46), the stochastic forcing due to the interaction term  $\lambda S_{\alpha} B_{\alpha}$  can be identified as the noise

$$\eta_{\alpha}(t) \equiv \sum_{m(\neq l)} \langle l|B_{\alpha}(t)|m\rangle e^{-\beta(\epsilon_m - \epsilon_l)/2} e^{i(\theta_m - \theta_l)}. \quad (47)$$

In order to characterize these stochastic noises we calculate their mean value as well as their correlation functions. The mean values vanish according to the random phase assumption

$$\overline{\eta_{\alpha}(t)} = 0, \quad \text{because} \quad \overline{e^{i\theta_m}} = 0, \quad (48)$$

for all  $m$ . For the same reason, the following correlation functions vanish

$$\overline{\eta_{\alpha}(t)\eta_{\beta}(t')} = 0, \quad \text{because} \quad \overline{e^{i(\theta_m + \theta_n)}} = 0, \quad (49)$$

for all  $m$  and  $n$ . Moreover, the random phase approximation implies that the other correlation functions are

$$\overline{\eta_{\alpha}^*(t)\eta_{\beta}(t')} = e^{\beta\epsilon_l} \sum_n \langle l|B_{\beta}(t')|n\rangle e^{-\beta\epsilon_n} \langle n|B_{\alpha}(t)|l\rangle, \quad \text{because} \quad \overline{e^{i(\theta_m - \theta_n)}} = \delta_{mn}. \quad (50)$$

These correlation functions depend on the chosen eigenstate  $\chi_l$  as follows

$$\overline{\eta_{\alpha}^*(t)\eta_{\beta}(t')|l} = \frac{Z_b}{e^{-\beta\epsilon_l}} \langle l|B_{\beta}(t')\rho_b^{\text{eq}}B_{\alpha}(t)|l\rangle. \quad (51)$$



In order to obtain a typical value for these correlation functions, we perform a thermal average to get

$$\sum_l \frac{e^{-\beta\epsilon_l}}{Z_b} \overline{\eta_\alpha^*(t)\eta_\beta(t')}|_l = \text{tr}_b \rho_b^{\text{eq}} B_\alpha(t) B_\beta(t') = C_{\alpha\beta}(t-t'), \quad (52)$$

which are also the time correlation functions (40). As a consequence, we infer that the typical value for the correlation functions (51) are typically given by these bath correlation functions

$$\overline{\eta_\alpha^*(t)\eta_\beta(t')} \approx C_{\alpha\beta}(t-t'). \quad (53)$$

Moreover, if the bath is large enough the noise (47) is given by a very large sum of complex oscillating terms. Such sums are known to lead to random variables of Gaussian type by the central limit theorem [21]. This result justifies that the stochastic noises (47) should be taken as Gaussian noises which are fully characterized by the mean values (48) and the correlation functions (49) and (53).

## VI. THE NON-MARKOVIAN STOCHASTIC DIFFERENTIAL EQUATION

Collecting together the previous approximations about the forcing and the damping terms of (36), we obtain the following stochastic differential equation for a typical coefficient  $\varphi_l(x_s; t)$  of the decomposition (7) in the complete interaction picture (19)

$$\begin{aligned} i \partial_t \varphi_l(t) = & \lambda \sum_\alpha \eta_\alpha(t) S_\alpha(t) \varphi_l(t) \\ & - i \lambda^2 \int_0^t d\tau \sum_{\alpha\beta} C_{\alpha\beta}(t-\tau) S_\alpha(t) S_\beta(\tau) \varphi_l(\tau) + \mathcal{O}(\lambda^3). \end{aligned} \quad (54)$$

The last step is to obtain the equation for the evolution from an initial condition which is the statistical mixture

$$\rho_s(0) = \sum_k |\psi_k(0)\rangle p_k \langle\psi_k(0)|, \quad (55)$$

defined in terms of the normalized subsystem wavefunctions  $\psi_k(0)$  and their probabilities  $\{p_k\}$  such that  $\sum_k p_k = 1$ . In this case, we should consider the following statistical ensemble of coefficients

$$\varphi_{kl}(x_s; t) \approx \psi_{Ik}(x_s; t) \sqrt{\frac{e^{-\beta\epsilon_l}}{Z_b}} e^{i\theta_l}, \quad (56)$$

where  $l$  is the index that already appears in Eq. (54), while the new index  $k$  specifies the member of the statistical mixture (55). Since Eq. (54) is homogeneous in the coefficient  $\varphi_l(t)$  we may multiply both sides by the inverse of the factor that accompanies  $\psi_{Ik}(t)$  in (56) and concludes that the subsystem wavefunction  $\psi_{Ik}(t)$  obeys the same stochastic equation (54). Moreover, if we perform a transformation from the complete interaction picture (19) back to the partial interaction picture (16), we obtain the following stochastic equation for the subsystem wavefunctions  $\psi = \psi_k = e^{-iH_s t} \psi_{Ik}$

$$\begin{aligned} i \partial_t \psi(t) = & H_s \psi(t) + \lambda \sum_\alpha \eta_\alpha(t) S_\alpha \psi(t) \\ & - i \lambda^2 \int_0^t d\tau \sum_{\alpha\beta} C_{\alpha\beta}(t-\tau) S_\alpha e^{-iH_s(t-\tau)} S_\beta \psi(\tau) + \mathcal{O}(\lambda^3). \end{aligned} \quad (57)$$

Finally, if we replace the time variable  $\tau$  by  $t-\tau$  in the integral our stochastic Schrödinger equation takes the equivalent form

$$\begin{aligned} i \partial_t \psi(t) = & H_s \psi(t) + \lambda \sum_\alpha \eta_\alpha(t) S_\alpha \psi(t) \\ & - i \lambda^2 \int_0^t d\tau \sum_{\alpha\beta} C_{\alpha\beta}(\tau) S_\alpha e^{-iH_s \tau} S_\beta \psi(t-\tau) + \mathcal{O}(\lambda^3), \end{aligned} \quad (58)$$

where the Gaussian noises  $\eta_\alpha(t)$  satisfy

$$\overline{\eta_\alpha(t)} = 0, \quad \overline{\eta_\alpha(t)\eta_\beta(t')} = 0, \quad \text{and} \quad \overline{\eta_\alpha^*(t)\eta_\beta(t')} = C_{\alpha\beta}(t-t') = C_{\beta\alpha}^*(t'-t). \quad (59)$$

Eq. (58) shows explicitly that the time evolution of the wavefunction is determined by its own past. Therefore, this stochastic differential equation is *non-Markovian*, which has its origin in the non-Markovian character of Eq. (28) given by the Feshbach projection-operator method.

Since the bath correlation functions decay to zero for infinitely large baths, the memory of the past is limited to a time interval of the order of the bath relaxation time  $t_b$ . Therefore, the integral  $\int_0^t$  can be replaced by  $\int_0^\infty$  in the stochastic equation (58) if  $t \gg t_b$ . We notice that the same replacement can be carried out in any case if we assume that  $\psi(t) = 0$  for  $t < 0$ .

## VII. THE MASTER EQUATION OF THE STOCHASTIC SCHRÖDINGER EQUATION

Because the wavefunctions  $\psi_k(t) = \psi(t)$  evolve in time according to a stochastic equation, the reduced density matrix of the subsystem issued from the initial condition (55) should be given at each instant of time by the statistical average

$$\rho_s(t) = \sum_k p_k \frac{\overline{|\psi_k(t)\rangle\langle\psi_k(t)|}}{\overline{\langle\psi_k(t)|\psi_k(t)\rangle}}, \quad (60)$$

where the denominator is introduced to guarantee the normalization. Because of the stochastic noise, the wavefunctions  $\psi_k(t)$  form a statistical ensemble. The average over this statistical ensemble is denoted by the overline. Besides, we have also to average over the statistical mixture of initial wavefunctions  $\psi_k(0)$  with their probabilities  $p_k$ . This further average is carried out with the sum over the index  $k$  of the initial wavefunctions.

In this section, we show that the reduced density matrix (60) evolves in time according to the Redfield master equation with the slippage of initial conditions, as a consequence of the non-Markovian stochastic Schrödinger equation (58). We carry out the proof in the interaction picture

$$\psi_I(x_s; t) \equiv e^{iH_s t} \psi(x_s; t). \quad (61)$$

Moreover, for simplicity, we suppose that the initial state (55) is pure so that we may drop the average over the statistical mixture in (60). In the interaction picture, the stochastic equation (57) becomes

$$i \partial_t \psi_I(t) = \lambda N(t) \psi_I(t) - i \lambda^2 \int_0^t d\tau K(t, \tau) \psi_I(\tau) + \mathcal{O}(\lambda^3), \quad (62)$$

where

$$N(t) \equiv \sum_\alpha \eta_\alpha(t) S_\alpha(t), \quad (63)$$

represents the noise operator of the stochastic forcing term and

$$K(t, \tau) \equiv \sum_{\alpha, \beta} C_{\alpha\beta}(t-\tau) S_\alpha(t) S_\beta(\tau), \quad (64)$$

is the memory kernel of the damping term. The time integral of Eq. (62) up to the second order in the coupling parameter gives

$$\begin{aligned} \psi_I(t) &= \psi_I(0) - i \lambda \int_0^t dt_1 N(t_1) \psi_I(0) \\ &\quad - \lambda^2 \int_0^t dt_1 \int_0^{t_1} dt_2 N(t_1) N(t_2) \psi_I(0) \\ &\quad - \lambda^2 \int_0^t dt_1 \int_0^{t_1} dt_2 K(t_1, t_2) \psi_I(0) + \mathcal{O}(\lambda^3). \end{aligned} \quad (65)$$

We consider the projector

$$X_I(t) \equiv |\psi_I(t)\rangle\langle\psi_I(t)|, \quad (66)$$

onto a state  $\psi_I(t)$  of the statistical ensemble due to the noise. The average of the projectors (66) over the noise enters into the definition (60) of the reduced density matrix. According to Eq. (65), the time evolution of this average is given by

$$\begin{aligned}
\overline{X_I(t)} &= X_I(0) + \lambda^2 \int_0^t dt_1 \int_0^{t_1} dt_2 \overline{N(t_1)X_I(0)N^\dagger(t_2)} \\
&- \lambda^2 \int_0^t dt_1 \int_0^{t_1} dt_2 \overline{N(t_1)N(t_2)} X_I(0) \\
&- \lambda^2 \int_0^t dt_1 \int_0^{t_1} dt_2 X_I(0) \overline{N^\dagger(t_2)N^\dagger(t_1)} \\
&- \lambda^2 \int_0^t dt_1 \int_0^{t_1} dt_2 K(t_1, t_2) X_I(0) \\
&- \lambda^2 \int_0^t dt_1 \int_0^{t_1} dt_2 X_I(0) K^\dagger(t_1, t_2) + \mathcal{O}(\lambda^4). \tag{67}
\end{aligned}$$

The average of the term in  $\lambda^3$  vanishes because the averages of all the odd moments vanish for a Gaussian noise, whereupon the next correction to Eq. (67) is of order  $\lambda^4$ .

The second term in the r.h.s. of (67) can be evaluated by using the property

$$\int_0^t dt_1 \int_0^t dt_2 f(t_1, t_2) = \int_0^t dt_1 \int_0^{t_1} dt_2 [f(t_1, t_2) + f(t_2, t_1)]. \tag{68}$$

Another important property is that

$$\overline{N(t_1)N(t_2)} = \overline{N^\dagger(t_2)N^\dagger(t_1)} = 0, \tag{69}$$

which follows from Eq. (49). Accordingly, the third and fourth terms vanish in the r.h.s. of Eq. (67).

First of all, we may infer from Eq. (67) that the wavefunction  $\psi_I(t)$  remains normalized on average up to the third order in the coupling parameter  $\lambda$ , during its time evolution under the stochastic Schrödinger equation (58). Indeed, if  $\langle \psi_I(0)|\psi_I(0) \rangle = 1$ , the averaged norm of the wavefunction is given by the trace of Eq. (67), which becomes

$$\begin{aligned}
\overline{\langle \psi_I(t)|\psi_I(t) \rangle} &= 1 + \lambda^2 \int_0^t dt_1 \int_0^{t_1} dt_2 \overline{\langle N^\dagger(t_2)N(t_1) \rangle} \\
&+ \lambda^2 \int_0^t dt_1 \int_0^{t_1} dt_2 \overline{\langle N^\dagger(t_1)N(t_2) \rangle} \\
&- \lambda^2 \int_0^t dt_1 \int_0^{t_1} dt_2 \overline{\langle K(t_1, t_2) \rangle} \\
&- \lambda^2 \int_0^t dt_1 \int_0^{t_1} dt_2 \overline{\langle K^\dagger(t_1, t_2) \rangle} + \mathcal{O}(\lambda^4) \tag{70}
\end{aligned}$$

after using the properties (68) and (69) and the notation  $\langle \cdot \rangle = \langle \psi_I(0)| \cdot |\psi_I(0) \rangle$ . According to the definitions of the noises (59) and the operators (63) and (64), we observe a cancellation between the different second-order terms of Eq. (70), and obtain the result

$$\overline{\langle \psi_I(t)|\psi_I(t) \rangle} = 1 + \mathcal{O}(\lambda^4). \tag{71}$$

As a consequence, the denominator [that guarantees the normalization of the reduced density matrix (60)] has an influence only beyond the third order in  $\lambda$ . Therefore, the denominator may be discarded in a calculation limited to the second order so that the reduced density matrix is given by

$$\rho_{sI}(t) = \overline{X_I(t)} + \mathcal{O}(\lambda^4), \tag{72}$$

in terms of Eq. (67).

Using again the properties (68) and (69) and defining the new integration variables  $\tau = t_1 - t_2$  and  $T = t_1$ , Eqs. (72) and (67) imply that the reduced density matrix evolves in time according to

$$\begin{aligned}
\rho_{\text{sI}}(t) &= \rho_{\text{sI}}(0) + \lambda^2 \int_0^t dT \int_0^T d\tau \sum_{\alpha\beta} C_{\alpha\beta}^*(\tau) S_\alpha(T) \rho_{\text{sI}}(0) S_\beta(T-\tau) \\
&+ \lambda^2 \int_0^t dT \int_0^T d\tau \sum_{\alpha\beta} C_{\alpha\beta}(\tau) S_\beta(T-\tau) \rho_{\text{sI}}(0) S_\alpha(T) \\
&- \lambda^2 \int_0^t dT \int_0^T d\tau \sum_{\alpha\beta} C_{\alpha\beta}(\tau) S_\alpha(T) S_\beta(T-\tau) \rho_{\text{sI}}(0) \\
&- \lambda^2 \int_0^t dT \int_0^T d\tau \rho_{\text{sI}}(0) \sum_{\alpha\beta} C_{\alpha\beta}^*(\tau) S_\beta(T-\tau) S_\alpha(T) + \mathcal{O}(\lambda^4). \tag{73}
\end{aligned}$$

Differentiating with respect to the time and returning to the original picture, we finally obtain that the reduced density matrix obeys

$$\begin{aligned}
\frac{d\rho_{\text{s}}(t)}{dt} &= -i [H_{\text{s}}, \rho_{\text{s}}(t)] + \lambda^2 \int_0^t d\tau \sum_{\alpha\beta} C_{\alpha\beta}^*(\tau) S_\alpha \rho_{\text{s}}(t) S_\beta(-\tau) \\
&+ \lambda^2 \int_0^t d\tau \sum_{\alpha\beta} C_{\alpha\beta}(\tau) S_\beta(-\tau) \rho_{\text{s}}(t) S_\alpha \\
&- \lambda^2 \int_0^t d\tau \sum_{\alpha\beta} C_{\alpha\beta}(\tau) S_\alpha S_\beta(-\tau) \rho_{\text{s}}(t) \\
&- \lambda^2 \int_0^t d\tau \rho_{\text{s}}(t) \sum_{\alpha\beta} C_{\alpha\beta}^*(\tau) S_\beta(-\tau) S_\alpha + \mathcal{O}(\lambda^4), \tag{74}
\end{aligned}$$

which is precisely the non-Markovian master equation derived at the second order of perturbation theory (see Ref. [16]).

In the long-time limit, the solutions of the non-Markovian master equation (74) are known to obey the Redfield master equation [17]

$$\frac{d\rho_{\text{s}}^{\text{R}}}{dt} = -i [H_{\text{s}}, \rho_{\text{s}}^{\text{R}}] + \lambda^2 \sum_{\alpha} \left( T_{\alpha} \rho_{\text{s}}^{\text{R}} S_{\alpha} + S_{\alpha}^{\dagger} \rho_{\text{s}}^{\text{R}} T_{\alpha}^{\dagger} - S_{\alpha} T_{\alpha} \rho_{\text{s}}^{\text{R}} - \rho_{\text{s}}^{\text{R}} T_{\alpha}^{\dagger} S_{\alpha}^{\dagger} \right) + \mathcal{O}(\lambda^4), \tag{75}$$

with the operators

$$T_{\alpha} \equiv \sum_{\beta} \int_0^{\infty} d\tau C_{\alpha\beta}(\tau) \exp(-iH_{\text{s}}\tau) S_{\beta} \exp(iH_{\text{s}}\tau). \tag{76}$$

Moreover, we have shown in Ref. [16] that the non-Markovian master equation (74) also implies that the initial conditions to be used in the Redfield master equation (75) must be slipped by the slippage superoperator

$$\rho_{\text{s}}^{\text{R}}(0) = \mathcal{S} \rho_{\text{s}}(0). \tag{77}$$

Accordingly, we can conclude that the non-Markovian stochastic Schrödinger equation (58) corresponds to the Redfield master equation (75) with the slipped initial conditions (77), which was the announced result to be proved.

We notice that the slippage (77) takes into account the memory effects in the fast early relaxation of the time evolution as described by Suarez, Silbey, and Oppenheim [22]. These effects should indeed be expected because of the non-Markovian character of the basic master equation (74). Even if the Redfield equation looks like a Markovian equation, the fact that its initial conditions need to be slipped, reflects the non-Markovian character of the time evolution that the Redfield equation is supposed to follow. Therefore, it is natural that a non-Markovian stochastic Schrödinger equation is found in correspondence with the Redfield equation with slipped initial conditions.

Moreover, we remark that the reduced density matrix (60) is positive if the initial probabilities  $p_k$  are positive so that the reduced density matrix remains positive if each wavefunction evolves in time, in particular, under the stochastic Schrödinger equation (58). As a consequence of this property, the positivity of the reduced density matrix

(60) should be preserved in a simulation based on the stochastic Schrödinger equation (58). On the other hand, the non-Markovian master equation (74) should preserve positivity at least up to the second order in  $\lambda$ , because this non-Markovian master equation is derived from the stochastic Schrödinger equation within the second order of perturbation theory. Furthermore, the previous deduction of the slippage superoperator (77) shows that our stochastic Schrödinger equation can reproduce the effect of the slippage of initial conditions of the Redfield equations thanks to its non-Markovian character. These results suggest that slipping the initial conditions of the Redfield equation plays an important role to preserve positivity, although they do not constitute a proof of such a conjecture [22].

### VIII. $\delta$ -CORRELATED BATHS

In this section, we consider the special case where the time correlation functions (40) of the bath coupling operators are proportional to the Dirac  $\delta$ -distributions

$$C_{\alpha\beta}(\tau) = 2 D_{\alpha\beta} \delta(\tau) , \quad (78)$$

where  $D_{\alpha\beta} = D_{\beta\alpha}^*$  is a Hermitian matrix of constants characterizing the dissipative coupling to the bath. In Ref. [16], we have already shown that the Redfield master equation reduces to the Lindblad master equation and that no slippage is required in this limit.

At the stochastic level, we recover the Markovian stochastic Schrödinger equation corresponding to the Lindblad master equation [3]. For  $\delta$ -correlated baths, the stochastic Schrödinger equation (58) reduces to the Markovian equation

$$\begin{aligned} i \partial_t \psi(t) = & H_s \psi(t) + \lambda \sum_{\alpha} \eta_{\alpha}(t) S_{\alpha} \psi(t) \\ & - i \lambda^2 \sum_{\alpha\beta} D_{\alpha\beta} S_{\alpha} S_{\beta} \psi(t) + \mathcal{O}(\lambda^3) , \end{aligned} \quad (79)$$

with the Gaussian noises

$$\overline{\eta_{\alpha}(t)} = 0 , \quad \overline{\eta_{\alpha}(t)\eta_{\beta}(t')} = 0 , \quad \text{and} \quad \overline{\eta_{\alpha}^*(t)\eta_{\beta}(t')} = 2 D_{\alpha\beta} \delta(t-t') . \quad (80)$$

This stochastic equation of the Langevin type has been previously obtained by van Kampen [3]. We notice that the norm of the wavefunction is only preserved on average for this stochastic equation, which is consistent with the interpretation of the wavefunction as a typical coefficient of the linear decomposition (7) of the total wavefunction, up to a constant prefactor. According to this interpretation, the norm should not be preserved at each instant of time but only on average according to Eq. (9). We remark that this behavior is in contrast to the situation for the stochastic equation by Gisin and Percival where the norm is not allowed to fluctuate [9].

### IX. NUMERICAL METHODS

In this section, we describe the numerical algorithms for the simulation of the stochastic Schrödinger equation (58). Firstly, the driving Gaussian noises  $\eta_{\alpha}(t)$  defined by (59) have to be numerically synthesized. Secondly, the stochastic differential equation (58) itself has to be integrated. For simplicity, we assume that the coupling to the thermal bath is carried out by a single term with  $\alpha = \beta = 1$ .

#### A. Numerical synthesis of the driving noise

The driving force is a complex colored Gaussian noise which is characterized by

$$\overline{\eta(t)} = 0 , \quad \overline{\eta(t)\eta(0)} = 0 , \quad \text{and} \quad \overline{\eta^*(t)\eta(0)} = C(t) = C^*(-t) . \quad (81)$$

Such a noise can be synthesized with

$$\eta(t) \equiv \int_{-\infty}^{+\infty} d\tau R(\tau) \frac{\xi'(t-\tau) + i \xi''(t-\tau)}{\sqrt{2}} , \quad (82)$$

in terms of two independent real Gaussian white noises

$$\overline{\xi'(t)} = \overline{\xi''(t)} = 0, \quad \overline{\xi'(t)\xi''(0)} = 0, \quad \overline{\xi'(t)\xi'(0)} = \overline{\xi''(t)\xi''(0)} = \delta(t), \quad (83)$$

and a response function which should vanish for negative times in order to satisfy causality:

$$R(t) = 0, \quad \text{for } t < 0. \quad (84)$$

Moreover, the convolution of the response function with its complex conjugate should be equal to the correlation function of the noise (81)

$$C(t) = \int_0^\infty d\tau R^*(t+\tau) R(\tau). \quad (85)$$

If the Fourier transform of the response function is defined by

$$G(\omega) \equiv \int \frac{dt}{\sqrt{2\pi}} R(t) e^{-i\omega t}, \quad (86)$$

we find that the power spectrum of the noise is given by

$$|G(\omega)|^2 = \int \frac{dt}{2\pi} C(t) e^{i\omega t}. \quad (87)$$

If we suppose that the function (86) – or one of its approximations – can be analytically continued towards complex values of its argument  $\omega$ , the complex singularities of  $G(\omega)$  such as its poles or its branch cuts must lie in the upper half plane ( $\text{Im}\omega > 0$ ) in order for the causality condition (84) to be satisfied.

Numerical algorithms have been proposed to approximate such a function from the sole knowledge of the correlation function  $C(t)$  or its Fourier transform (87). If the power spectrum vanishes at zero frequency such as  $|G(\omega)|^2 \sim \omega^{2q}$  with  $q$  integer, the approximation

$$G(\omega) \simeq \frac{\sqrt{b} \omega^q}{\sum_{k=0}^M a_k z^k}, \quad (88)$$

may be assumed where we have introduced the quantity  $z = \exp(i\omega \Delta t)$  defined with a fixed time interval  $\Delta t$  and  $\{a_k\}_{k=0}^M$  and  $b$  are unknown coefficients. An algorithm to compute these unknown coefficients is described in Appendix A.

## B. Numerical integration of the stochastic Schrödinger equation

We propose an integration scheme of the first order in the time step  $\delta$ . The integration is performed in the interaction picture. For a single driving noise, the stochastic equation (62) has the form

$$\partial_t \psi_{\text{I}}(t) = -i \lambda \eta(t) S(t) \psi_{\text{I}}(t) - \lambda^2 \int_0^t d\tau C(\tau) S(t) S(t-\tau) \psi_{\text{I}}(t-\tau), \quad (89)$$

with  $S(t) = e^{iH_s t} S e^{-iH_s t}$ . If the time is discretized and if we require that the norm is preserved on average at the second order in the coupling parameter  $\lambda$ , Eq. (89) leads to the following iterative equation

$$\psi_{\text{I},n+1} \simeq \left( 1 - i \lambda \delta \eta_n S_n - \frac{\lambda^2 \delta^2}{2} C_0 S_n^2 \right) \psi_{\text{I},n} - \lambda^2 \delta^2 \sum_{m=1}^n C_m S_n S_{n-m} \psi_{\text{I},n-m}, \quad (90)$$

with the notations

$$\psi_{\text{I},n} = \psi_{\text{I}}(n\delta), \quad S_n = S(n\delta), \quad (91)$$

and the discretized correlation function

$$C_m = \begin{cases} \overline{\eta_m^* \eta_0}, & \text{for } m = 0, 1, 2, \dots, N, \\ 0, & \text{for } m = N+1, N+2, \dots, \end{cases} \quad (92)$$

where  $t_{\max} = N\delta$  is the time beyond which the correlation function is so small that it may be cutoff. In Eq. (90), the noise  $\eta_n = \eta(n\delta)$  should be generated by

$$\eta_n = \frac{1}{\sqrt{2\delta}} \left[ \frac{R_0}{2}(G'_n + iG''_n) + R_1(G'_{n-1} + iG''_{n-1}) + \dots \right. \\ \left. \dots + R_{N-1}(G'_{n-N+1} + iG''_{n-N+1}) + \frac{R_N}{2}(G'_{n-N} + iG''_{n-N}) \right], \quad (93)$$

where  $R_n = R(n\delta)$  is the discretized response function and  $\{G'_n, G''_n\}_{n=-\infty}^{+\infty}$  are independent real Gaussian random variables with zero mean and unit variance. The values (92) of the discretized correlation function should thus be fixed by using the definition (93) of the noise.

Then, the quantum state in the original picture of Eq. (58) is retrieved by

$$\psi_n = \psi(n\delta) = e^{-iH_s n\delta} \psi_{I,n}. \quad (94)$$

According to Eq. (60), the expected value of an observable  $A$  at the time  $t = n\delta$  can be obtained as the average over the statistical ensemble of trajectories  $\{\psi_n^{(l)}\}_{l=1}^L$  generated by the stochastic iterative equation (90) as

$$\langle A \rangle_t = \text{tr}_s \rho_s(t) A \simeq \frac{\sum_{l=1}^L \langle \psi_n^{(l)} | A | \psi_n^{(l)} \rangle}{\sum_{l=1}^L \langle \psi_n^{(l)} | \psi_n^{(l)} \rangle}, \quad (95)$$

for a large enough number  $L$  of trajectories. As we have shown in Sec. VII, the average (95) should give a result which is equivalent to a calculation based on the non-Markovian master equation (74).

Quantum jumps can also be simulated by considering individual trajectories of the statistical ensemble generated by the stochastic Schrödinger equation. The quantum fluctuations of an observable would appear in the average over an individual wavefunction defined by

$$\langle A \rangle_t^{(l)} = \frac{\langle \psi_n^{(l)} | A | \psi_n^{(l)} \rangle}{\langle \psi_n^{(l)} | \psi_n^{(l)} \rangle}, \quad (96)$$

at the time  $t = n\delta$ .

## X. APPLICATION TO THE SPIN-BOSON MODEL

In this section, we apply our stochastic Schrödinger equation to the dynamics of a two-level subsystem or of a spin  $s = \frac{1}{2}$  coupled to a thermal bath of harmonic oscillators or bosons, as described by the spin-boson model [22, 23]. We use the same notations as in the previous paper [16]. The total Hamiltonian is given by

$$H = -\frac{\Delta}{2} \sigma_z + H_b + \lambda \sigma_x B, \quad (97)$$

in terms of the Hamiltonian  $H_b$  of the bath composed of uncoupled harmonic oscillators and of the Pauli matrices  $\sigma_x$ ,  $\sigma_y$ , and  $\sigma_z$ . The operator  $B$ , which couples the spin to the bosons, is characterized by the spectral strength

$$J(\omega) = \frac{\omega^3}{\omega_c^2} \exp(-\omega/\omega_c), \quad (98)$$

where  $\omega_c$  is a cutoff frequency [22, 23].

### A. Numerical synthesis of the driving noise

Here below, we have chosen the inverse temperature  $\beta = 10$ , the splitting energy  $\Delta = 0.1$ , the cutoff frequency  $\omega_c = 1$ , and the coupling parameter  $\lambda = 0.1$ .

The correlation function of the noise generated by the thermal bath is given in terms of the spectral strength (98) by

$$\begin{aligned} C(t) &= \text{tr}_b \rho_b^{\text{eq}} B(t) B(0) \\ &= \int_0^{+\infty} d\omega J(\omega) \left( \coth \frac{\beta\omega}{2} \cos \omega t - i \sin \omega t \right). \end{aligned} \quad (99)$$

At non-zero temperature, this correlation function decays asymptotically as

$$C(t) = -\frac{12}{\beta\omega_c^3 t^4} + i \frac{24}{\omega_c^3 t^5} + \mathcal{O}\left(\frac{1}{t^6}\right), \quad \text{for } t \rightarrow \infty. \quad (100)$$

Consequently, we expect that the response function also decays to zero for  $t \rightarrow \infty$ .

The power spectrum of the noise is obtained by Eq. (87) as

$$\begin{aligned} |G(\omega)|^2 &= \begin{cases} \frac{1}{2} \left( \coth \frac{\beta\omega}{2} + 1 \right) J(\omega), & \text{for } \omega > 0, \\ -\frac{1}{2} \left( \coth \frac{\beta\omega}{2} + 1 \right) J(-\omega), & \text{for } \omega < 0, \end{cases} \\ &= \frac{\omega^3 \exp(-|\omega|/\omega_c)}{\omega_c^2 [1 - \exp(-\beta\omega)]}, \end{aligned} \quad (101)$$

which vanishes as  $|G(\omega)|^2 \sim \omega^2$  at zero frequency.

In order to generate the noise of the thermal bath, a response function  $R(t)$  has been computed with the algorithm described in Subsec. IX.A. The Fourier transform  $G(\omega)$  of the response function is approximated by Eq. (88) with  $q = 1$  because of Eq. (101). The coefficients of the approximation (88) are obtained by the algorithm of Appendix A with the parameter  $\Delta t = \pi/20$ . The response function has been discretized according to  $R_n = R(n\delta)$  with the time step  $\delta = 0.05$ . Its numerical approximation extends over  $N = 600$  time steps up to the maximum time  $t_{\text{max}} = N\delta = 30$ , after which it is supposed to vanish. The response function computed in this way is depicted in Fig. 1a.

Then, the Gaussian noise (82) has been generated by Eq. (93). In order to check whether this noise produces the expected correlation function (99), it has been statistically calculated with a long time sequence according to  $C(t) = \overline{\eta^*(t)\eta(0)}$ . Fig. 1b compares the correlation function thus computed to that of Eq. (99), showing in agreement that the response function of Fig. 1a is appropriate.

Figure 2a depicts a typical trajectory of the noise  $\eta(t) = \text{Re } \eta(t) + i \text{Im } \eta(t)$  generated by Eq. (93). We observe that the noise fluctuates on the short time scale of the bath relaxation, which is given by the inverse of the cutoff frequency of the spectral strength (98):  $t_b \sim 1/\omega_c = 1$ .

## B. Numerical integration of the stochastic Schrödinger equation

The algorithm (90) of numerical integration has been applied to the spin-boson model as follows. Since the subsystem Hamiltonian is  $H_s = (-\Delta/2)\sigma_z$ , the wavefunction in the interaction picture is related to the original wavefunction by

$$\psi_n = \psi(t) = \psi(n\delta) = e^{-iH_s n\delta} \psi_I(n\delta) = \begin{pmatrix} e^{i\Delta n\delta/2} & 0 \\ 0 & e^{-i\Delta n\delta/2} \end{pmatrix} \psi_{I,n}, \quad (102)$$

at the time  $t = n\delta$ , where  $\delta = 0.05$  is the time step of integration. In the interaction picture, the coupling operator of the subsystem to the bath is given in terms of the Pauli matrix  $\sigma_x$  by

$$S_n = \sigma_x(t) = \sigma_x(n\delta) = \begin{pmatrix} 0 & e^{-i\Delta n\delta} \\ e^{i\Delta n\delta} & 0 \end{pmatrix}, \quad (103)$$

in Eq. (90).

The integration of the stochastic Schrödinger equation has been carried out from the initial condition

$$\psi(0) = \begin{pmatrix} \frac{\sqrt{3}}{2} e^{-i\pi/4} \\ \frac{1}{2} e^{i\pi/4} \end{pmatrix}, \quad (104)$$

that corresponds to the point  $[x(0) = 0, y(0) = \sqrt{3}/2, z(0) = 1/2]$  in the Bloch representation defined with the three real variables

$$x = \langle \sigma_x \rangle, \quad y = \langle \sigma_y \rangle, \quad \text{and} \quad z = \langle \sigma_z \rangle. \quad (105)$$



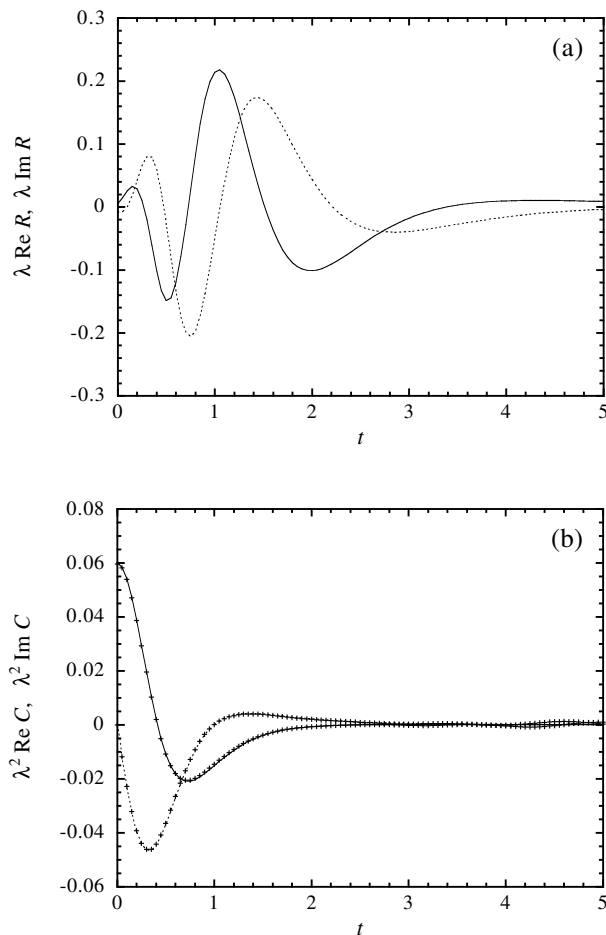


FIG. 1: Dynamical properties of the noise of the thermal bath of harmonic oscillators in the spin-boson model with the inverse temperature  $\beta = 10$ , the cutoff frequency  $\omega_c = 1$ , and the coupling parameter  $\lambda = 0.1$ : (a) Response function versus the time  $t$ . The solid (resp. dashed) line depicts the real (resp. imaginary) part of  $\lambda R(t)$  calculated by Eq. (86) with the approximation (88) obtained by the method of Appendix A. (b) Correlation function versus the time  $t$ . The solid (resp. dashed) line depicts the real (resp. imaginary) part of  $\lambda^2 C(t)$  calculated by Eq. (99). The pluses are the results of a statistical computation of the correlation function using a long time sequence of the noise.

We notice that the variables  $x$  and  $y$  describe the quantum coherence, while the variable  $z$  describes the population inversion between the two levels.

Figure 2b depicts an individual trajectory of the stochastic Schrödinger equation represented by the three variables (105) where the average is defined by Eq. (96). We observe that, due to the coupling to the thermal bath, these variables fluctuate around the trajectories they would follow without the coupling. The  $x$  and  $y$  variables fluctuate around large-amplitude oscillations of period  $T = 2\pi/\Delta$ , while the population variable  $z$  fluctuates around a monotonous trajectory. Moreover, it is observed that the fluctuations of the Bloch variables become smoother than those of the noise itself depicted in Fig. 2a because the stochastic Schrödinger equation is integrated over the noise. Nevertheless, a careful observation of Fig. 2 reveals that the fluctuations of the noise and the Bloch variables are closely correlated.

Figure 3 depicts the same Bloch variables as Fig. 2b but over a much longer time scale of  $10^5$  time units. On this long time scale, the spin has a chance to flip several times due to its stochastic driving by the thermal bath. These flips cause the population variable  $z$  to jump from positive to negative values and vice versa. At these quantum jumps, the oscillations of the quantum-coherence variables  $x$  and  $y$  are interrupted, as seen in Fig. 3. The statistical average of  $z$  is known to obey one of the Redfield equations [16]

$$\dot{z}^R = -g(\infty) (z^R - z_{\text{eq}}) \quad \text{for} \quad t \gg t_b, \quad (106)$$

with  $g(\infty) = 1.23 \times 10^{-4}$  and

$$z_{\text{eq}} = \frac{1 - \exp(-\beta\Delta)}{1 + \exp(-\beta\Delta)} = 0.462, \quad (107)$$

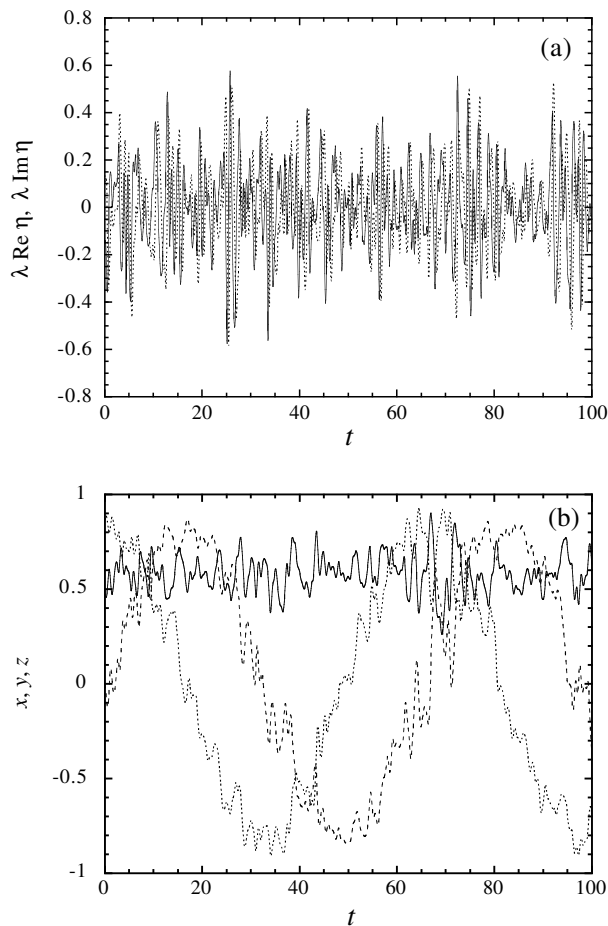


FIG. 2: (a) The Gaussian noise generated by the response function of Fig. 1a according to Eq. (93). The solid (resp. dashed) line depicts the real (resp. imaginary) part of  $\lambda\eta(t)$  versus the time  $t$ . (b) Individual trajectory of the non-Markovian stochastic Schrödinger equation integrated with Eq. (90) for the spin-boson model with the splitting energy  $\Delta = 0.1$ , the inverse temperature  $\beta = 10$ , the cutoff frequency  $\omega_c = 1$ , and the coupling parameter  $\lambda = 0.1$ . The trajectory is represented in terms of the three Bloch variables (105) versus the time  $t$ . ( $x$ ,  $y$ , and  $z$  correspond to the dashed, dotted, and solid lines, respectively.)

for  $\beta = 10$ ,  $\Delta = 0.1$ ,  $\omega_c = 1$ , and  $\lambda = 0.1$ . As a consequence of Eq. (106), the mean time between two flips is approximately equal to  $g(\infty)^{-1} = 8130$  time units, in agreement with the behavior seen in Fig. 3c. However, we notice that the variable  $z$  fluctuates continuously so that sudden flips might not be defined accurately. In Fig. 3c, we also observe that the variable  $z$  is more often positive than negative in accordance with its positive equilibrium value (107).

Figure 4a compares the averages over a statistical ensemble of  $10^4$  trajectories of the stochastic Schrödinger equation with the prediction of the non-Markovian master equation of the spin-boson model given by Eqs. (42)-(43) in the previous paper [16]. The remarkable agreement shows that a simulation with the stochastic Schrödinger equation can give those results equivalent to the quantum master equation, as announced.

Most remarkably, the stochastic Schrödinger equation can simulate the slippage of initial conditions on the time scale of the bath relaxation, as observed in Fig. 4b which depicts the short transient dynamics of the population variable  $z$  for an average over  $2 \times 10^5$  trajectories. In Fig. 4b, we see also that the slippage of initial conditions is indeed completed within the time scale of the bath relaxation,  $t_b \sim 1/\omega_c = 1$ . After this slippage, the trajectory follows the prediction of the Redfield equation (106). The remarkable ability of our stochastic Schrödinger equation to simulate the slippage is directly related to its non-Markovian character.

Besides, it must be noticed that the simulation with the non-Markovian stochastic Schrödinger equation does not violate the positivity thanks to the definition (60) of the reduced density matrix. It is this interesting property of the present stochastic scheme that is in contrast with the deterministic dynamics by the Redfield equation without the slippage of initial conditions. In this regard, the numerical results strongly support the conclusion that the positivity

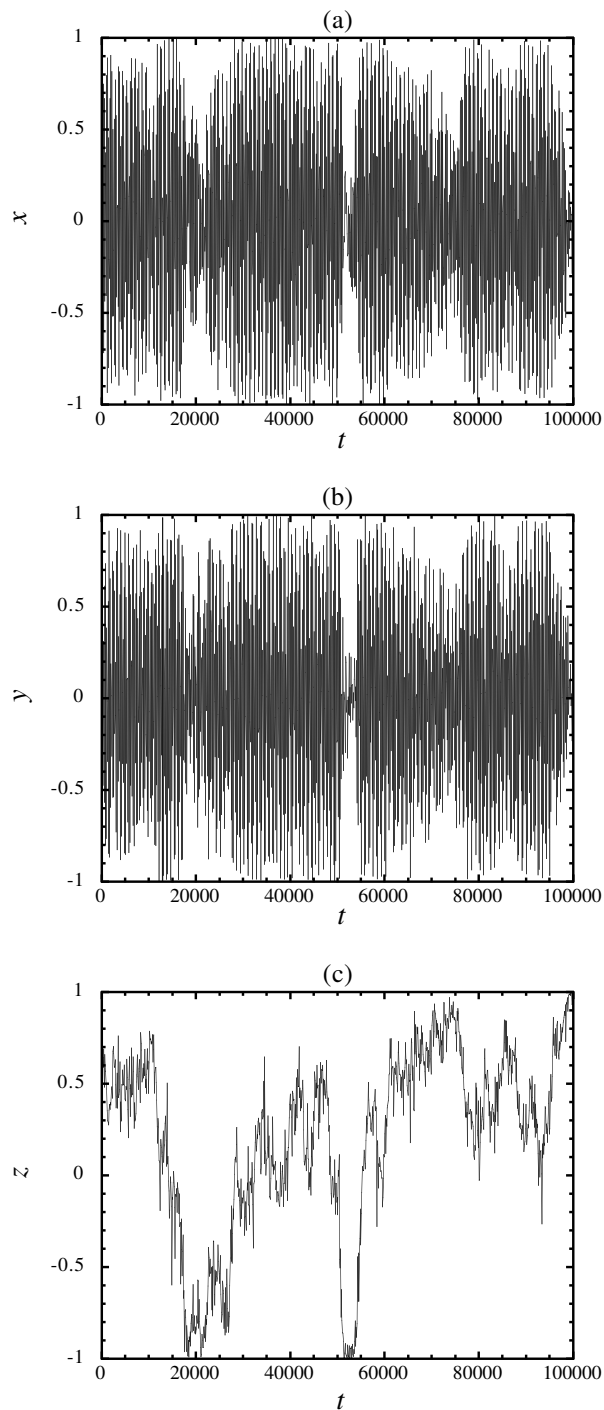


FIG. 3: The same individual trajectory as in Fig. 2b, but depicted over a much longer time. The trajectory is represented in terms of the three Bloch variables (105): (a) the variable  $x$ , (b)  $y$ , and (c)  $z$ .

preservation by the Redfield master equation should require a slippage of initial conditions.

## XI. CONCLUSIONS

In this paper, we have constructed the non-Markovian stochastic Schrödinger equation (58)-(59) describing the time evolution of the wavefunction of a small subsystem weakly coupled to a general bath which is not necessarily

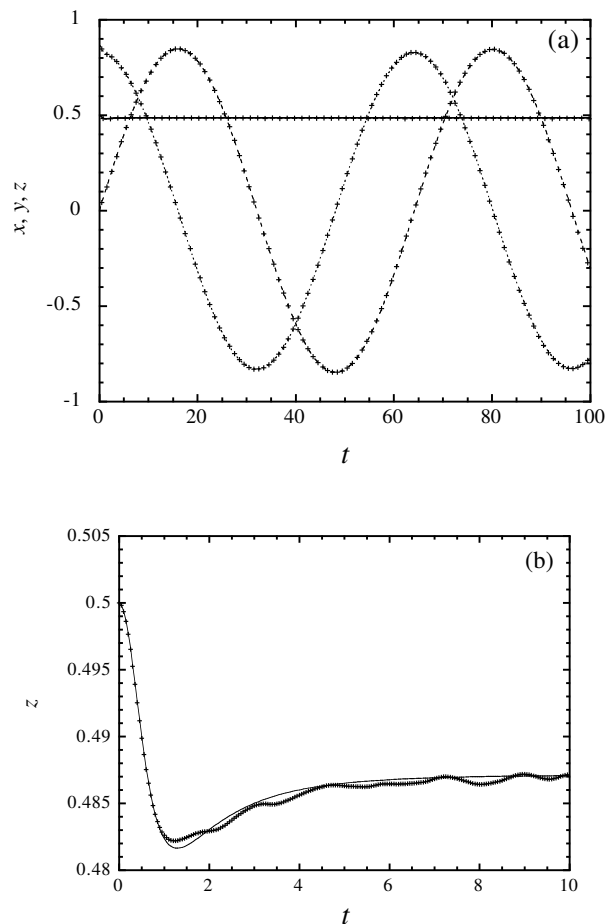


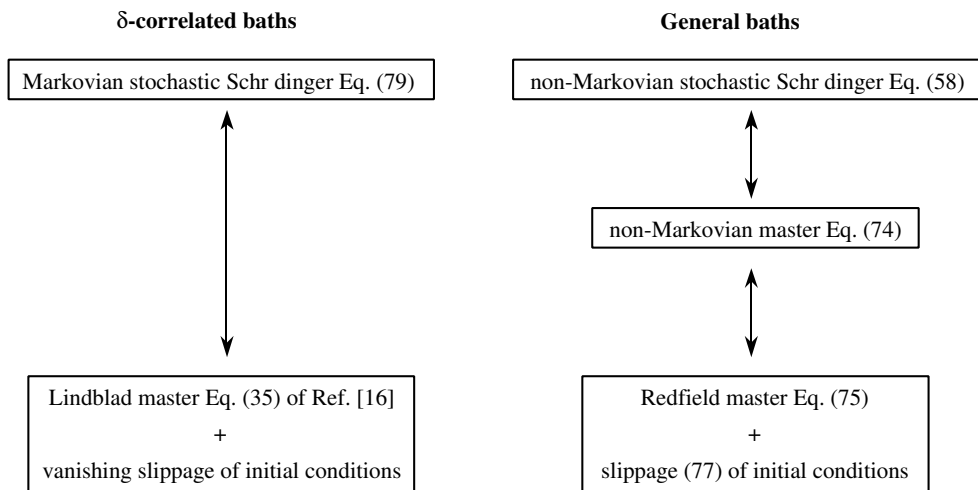
FIG. 4: For the spin-boson model under the same conditions as in Figs. 1, 2 and 3, comparison between a simulation using the non-Markovian stochastic Schrödinger equation (58) and the prediction of the non-Markovian master equation given by Eqs. (42)-(43) of the previous paper [16]. The dashed line is the variable  $x$  of the non-Markovian master equation, the dotted line is  $y$ , and the solid line is  $z$ . (a) The average (pluses) is taken over a statistical ensemble of  $10^4$  individual trajectories like the one of Fig. 2b. (b) The average (pluses) is here over a statistical ensemble of  $2 \times 10^5$  individual trajectories and only the variable  $z$  is depicted to show the slippage of initial conditions over the bath relaxation time  $t_b \sim 1/\omega_c = 1$ .

$\delta$ -correlated. Our purpose was to extend the stochastic Schrödinger equations previously proposed for the Lindblad master equation [10] to a much more general one for the Redfield master equation [13]. Indeed, the Lindblad master equation is restricted to  $\delta$ -correlated baths, while the Redfield master equation with slipped initial conditions can be applied to more general baths characterized by specific correlation functions.

The projection-operator method by Feshbach has been used in order to derive the non-Markovian stochastic Schrödinger equation (58)-(59) under the assumptions that the coupling is weak enough and the bath is large enough. We have started from the idea that the stochastic Schrödinger equation rules the time evolution of a typical coefficient of the linear decomposition of the total wavefunction on the basis of the energy eigenfunctions of the bath Hamiltonian. We have used the assumption that the bath is classically chaotic, so that the bath eigenfunctions are typically irregular and satisfy the Berry conjecture [20]. This assumption allows us to show that the typical behavior of one of these coefficients is ruled by a non-Markovian stochastic equation. For large enough baths, we may assume that the stochastic forcing should show Gaussian character, although it can be generalized to non-Gaussian baths as well.

It has been proved that the non-Markovian stochastic Schrödinger equation (58)-(59) implies that the corresponding reduced density matrix of the subsystem evolves in time according to the non-Markovian master equation (74) and, therefore, according to the Redfield master equation (75) with the slipped initial conditions (77). It should be noticed that the positivity is automatically preserved at the stochastic level because the reduced density matrix is defined by (60) with wavefunctions obeying the stochastic equation. This result emphasizes the role of the slippage of initial conditions in order for the Redfield master equation to satisfy positivity.

For  $\delta$ -correlated baths, the non-Markovian stochastic equation reduces to the Markovian stochastic Schrödinger

Table I. Comparison between  $\delta$ -correlated baths and general baths

equation (79)-(80) previously obtained by van Kampen [3]. The situation is summarized in Table I.

Furthermore, we have developed numerical algorithms in order to simulate the time evolution generated by our non-Markovian stochastic Schrödinger equation. A method is proposed to synthesize the colored Gaussian noises of the thermal bath and a first-order numerical integrator of the non-Markovian stochastic equation is given by Eq. (90).

The application of these numerical methods is illustrated with the spin-boson model. Very good agreement is accomplished between the results obtained by the present stochastic simulation and the prediction of the quantum master equations. This demonstrates, in particular, that the non-Markovian character of our stochastic Schrödinger equation is essential to reproduce the slippage of initial conditions described elsewhere [16, 22].

The stochastic scheme could reduce the computational effort in the simulation of the dynamics of a subsystem coupled to a bath, especially when the number  $N$  of levels of the subsystem becomes too large. Instead of integrating the  $N^2 - 1$  real variables for the reduced density matrix  $\rho_s$ , we only need to integrate  $2N$  real variables for the wavefunction  $\psi = \text{Re} \psi + i \text{Im} \psi$ . Indeed, if the dimension of the quantum state space of the subsystem is equal to  $N$ , the stochastic differential equation (58) requires the simultaneous integration of  $2N$  real variables which are the real and imaginary parts of the components of  $\psi$  in a basis of the quantum state space. On the other hand, the non-Markovian master equation (74) or the Redfield equation (75) requires the integration of  $N^2 - 1$  real variables. For large subsystems with  $N \gg 1$ , the stochastic scheme may thus turn out to provide a great advantage. Of course, the replacement of a deterministic equation like the master equation (74) by a stochastic one like (58) requires an ensemble average over  $M$  wavefunctions of the subsystem according to (60). Nevertheless, the  $M$  time integrations of the  $2N$  variables of the stochastic equations can be carried out sequentially, which should be computationally advantageous, e.g., via parallelization.

Moreover, the stochastic scheme provides the possibility to simulate quantum jumps and fluctuations by using individual trajectories of the stochastic differential equations. In this regard, the stochastic Schrödinger equation provides a model for the process of quantum fluctuations underlying the smooth evolution of the quantum master equations. Finally, it can be said that our non-Markovian stochastic Schrödinger equation (58)-(59) can be applied to the study of quantum jumps in more general systems than the previous schemes [10], which are restricted to Markovian stochastic processes.

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## APPENDIX A

Let us consider the problem of finding an approximate solution of the form

$$F(\omega) \simeq \frac{\sqrt{b}}{\sum_{k=0}^M a_k z^k}, \quad \text{with} \quad z = \exp(i\omega\Delta t), \quad (108)$$

for a function  $F(\omega)$  defined by

$$|F(\omega)|^2 = \int \frac{dt}{2\pi} \Upsilon(t) e^{i\omega t}. \quad (109)$$

If we discretize the integral in the r.h.s. of the last equation, we obtain

$$\frac{b}{|\sum_{k=0}^M a_k z^k|^2} \simeq \sum_{j=-M}^{+M} u_j z^j, \quad (110)$$

with  $u_j = \Upsilon(j\Delta t)\Delta t/2\pi = u_{-j}^*$ .

It has been shown [24] that the unknown coefficients  $\{a_k\}$  and  $b$  can be obtained by solving the following set of linear equations

$$\sum_{k=0}^M u_{j-k} \frac{a_k}{b} = \delta_{j0}, \quad (j = 0, 1, 2, \dots, M). \quad (111)$$

The coefficient  $b$  may be taken to be real under the assumption that  $a_0 = 1$ .

Thanks to this numerical algorithm, the function  $F(\omega)$  is approximated by a function with poles in the upper half of the complex plane  $\omega$  so that the condition (84) is satisfied. The response function  $R(t)$  is then given by the inverse Fourier transform of  $G(\omega) = \omega^q F(\omega)$ . We remark that the approximation must be good enough in order for the noise to have the expected statistical properties (81).

Besides, we should notice that the function  $G(\omega)$  is only determined up to an arbitrary phase function because  $G(\omega)\exp[i\phi(\omega)]$  corresponds to the same power spectrum (87) as  $G(\omega)$  and thus to the same correlation function. Therefore, the response function  $R(t)$  is not uniquely determined from its correlation function. However, all these response functions generate equivalent Gaussian noises corresponding to the same correlation function  $C(t)$ .

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