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Abstract – It is shown that non-Markovian master equations for an open system which are local in time can be unravelled through a piecewise deterministic quantum jump process in its Hilbert space. We derive a stochastic Schrödinger equation that reveals how non-Markovian effects are manifested in statistical correlations between different realizations of the process. Moreover, we demonstrate that possible violations of the positivity of approximate master equations are closely connected to singularities of the stochastic Schrödinger equation, which could lead to important insights into the structural characterization of positive non-Markovian equations of motion.

Relaxation and decoherence phenomena in open quantum systems [1] can often be modelled with sufficient accuracy by a quantum Markov processes in which the open system's density matrix is governed by a relatively simple quantum Markovian master equation with Lindblad structure [2,3]. However, non-Markovian quantum systems featuring strong memory effects play an increasingly important role in many fields of physics such as quantum optics [4], solid-state physics [5], and quantum information science [6]. Further applications include non-Markovian extensions of quantum process tomography, quantum control [7], and quantum transport [8].

The non-Markovian quantum dynamics of open systems is characterized by pronounced memory effects, finite revival times and non-exponential behavior of damping and decoherence, resulting from long-range correlation functions and from the dynamical relevance of large correlations and entanglement in the initial state. As a consequence the theoretical treatment of non-Markovian quantum dynamics is generally extremely demanding, both from the analytical and from the computational point of view [9]. Even if one is able to derive an appropriate non-Markovian master equation or some other mathematical formulation of the dynamics, the numerical simulation of such processes turns out to be a very difficult and time-consuming task, especially for high-dimensional Hilbert spaces.

From classical physics it is known that Monte Carlo techniques provide efficient tools for the numerical simulation of complex systems. This fact was one of the motivations to introduce the Monte Carlo wave function technique [10–12] which provides efficient quantum simulation techniques in the regime of Markovian dynamics. Several generalizations of the Monte Carlo approach to non-Markovian dynamics have been developed which are based on suitable extensions of the underlying reduced system's Hilbert space [13–16].

Recently, an efficient alternative simulation algorithm for the treatment of non-Markovian open system dynamics has been proposed [17] that does not require any extension of the state space. The purpose of the present paper is to develop a mathematical formulation of this algorithm in terms of a stochastic Schrödinger equation (SSE) in the open system's Hilbert space. We demonstrate that this formulation gives rise to a new type of piecewise deterministic quantum jumps process (PDP).

Quantum master equations are often derived from an underlying microscopic theory by employing some approximation scheme. An appropriate scheme is the time-convolutionless (TCL) projection operator technique which leads to a time-local first-order differential equation for the density matrix [18–20]. It will be shown that TCL master equations allow a stochastic unravelling of the form developed here. Generally, the use of a certain approximation technique may lead to violations of the positivity of the master equation. We demonstrate that positivity violations are closely linked to singularities of the SSE at

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which the stochastic process breaks down. Hence, a great advantage of the present stochastic formulation consists in the fact that it naturally prevents the generation of unphysical solutions and that it could thus lead to important insights into the structural characterization of positive non-Markovian evolution equations.

The most general structure of the TCL master equation is given by[1]

$$\frac{\mathrm{d}}{\mathrm{d}t}\rho(t) = \mathcal{L}_t\rho(t) = -i[H(t),\rho(t)] + \mathcal{D}_t\rho(t), \qquad (1)$$

where

$$\mathcal{D}_t\rho = \sum_m \Delta_m(t)\left[C_m(t)\rho C_m^\dagger(t) - \frac{1}{2}\left\{C_m^\dagger(t)C_m(t),\rho\right\}\right].$$

The time-dependent generator $\mathcal{L}_t$ consists of a commutator term describing the unitary part of the evolution and a dissipator $\mathcal{D}_t$. The latter involves a summation over the various decay channels labelled by m with corresponding time-dependent decay rates $\Delta_m(t)$ and arbitrary time-dependent system operators $C_m(t)$.

In the simplest case the rates Δ_m as well as the Hamiltonian H and the operators C_m are assumed to be time-independent. Equation (1) then represents a master equation in Lindblad form [2,3] which generates a semigroup of completely positive dynamical maps known as quantum Markov process. For arbitrary time-dependent operators $H(t)$ and $C_m(t)$, and for $\Delta_m(t) \geqslant 0$ the generator of the master equation (1) is still in Lindblad form for each fixed time t and leads to a two-parameter family of completely positive dynamical transformations [21] which may be referred to as time-dependent quantum Markov process [22]. An entirely different situation emerges if one or several of the $\Delta_m(t)$ become temporarily negative which expresses the presence of strong memory effects in the reduced system dynamics. The process is then said to be non-Markovian. Of course, the physical interpretation of the master equation requires that it preserves the positivity of the density matrix ρ. The formulation of general mathematical and physical conditions that guarantee the preservation of positivity is, however, an unsolved problem of central importance in the field of non-Markovian quantum dynamics. We emphasize that the emergence of temporarily negative $\Delta_m(t)$ in the master equation is a natural phenomenon in the non-Markovian regime which does in general not imply that the complete positivity of the corresponding quantum-dynamical map is violated. An example is discussed in ref. [1] where the exact non-Markovian master equation of an analytically solvable model is constructed.

The fundamental difference between Markovian and time-dependent Markovian processes on the one hand and non-Markovian processes on the other hand can also be seen very clearly if one attempts to apply the standard stochastic formulations to the master equation (1).

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[1]This statement is a direct consequence of Lemma 2.3 of ref. [2]; for various alternative forms see [1,16].

For both a Markovian and a time-dependent Markovian dynamics the standard unravelling through a stochastic quantum jump process can indeed be applied. This means that in both cases one can formulate an appropriate PDP for the reduced system's state vector $|\psi(t)\rangle$ in such a way that the expectation value

$$\rho(t) = \mathrm{E}[|\psi(t)\rangle\langle\psi(t)|] = \int \mathrm{d}\psi\, P[|\psi\rangle,t]\,|\psi\rangle\langle\psi| \qquad (2)$$

satisfies the master equation (1). Here, we have expressed the expectation value E in terms of an integration over the Hilbert space of states of the open quantum system with the unitarily invariant volume element $\mathrm{d}\psi \equiv D\psi D\psi^*$, and introduced the corresponding probability density functional $P[|\psi\rangle,t]$ which is defined as the probability density of finding at time t the state vector $|\psi\rangle$ [1]. However, the essential feature of a non-Markovian dynamics is the temporary appearance of negative decay rates. The use of the standard unravellings unavoidably leads in this case to negative jump probabilities, which clearly indicates the decisive difference between Markovian and non-Markovian quantum processes.

To account for the sign of the decay rates we decompose $\Delta_m(t)$ into a positive and a negative part defined by $\Delta_m^\pm(t) = \frac{1}{2}[|\Delta_m(t)| \pm \Delta_m(t)]$. The master equation (1) can then be written in the form

$$\begin{aligned}\frac{\mathrm{d}}{\mathrm{d}t}\rho =\ & -i[H(t),\rho] \\ & + \sum_k \Delta_k^+(t)\left[C_k(t)\rho C_k^\dagger(t) - \frac{1}{2}\left\{C_k^\dagger(t)C_k(t),\rho\right\}\right] \\ & - \sum_l \Delta_l^-(t)\left[C_l(t)\rho C_l^\dagger(t) - \frac{1}{2}\left\{C_l^\dagger(t)C_l(t),\rho\right\}\right].\end{aligned}$$
$$(3)$$

In order to better distinguish the positive and the negative channels we label the former by an index k and the latter by an index l. Note that $\Delta_k^+(t) \geqslant 0$ and $\Delta_l^-(t) \geqslant 0$ and that for Markovian or time-dependent Markovian processes we have $\Delta_l^-(t) = 0$.

We can now formulate the central result of this paper. Namely, the stochastic Schrödinger equation given by

$$\begin{aligned}\mathrm{d}|\psi(t)\rangle =\ & -iG(t)|\psi(t)\rangle\mathrm{d}t \\ & + \sum_k\left[\frac{C_k(t)|\psi(t)\rangle}{||C_k(t)|\psi(t)\rangle||} - |\psi(t)\rangle\right]\mathrm{d}N_k^+(t) \\ & + \sum_l\int\mathrm{d}\psi'\left[|\psi'\rangle - |\psi(t)\rangle\right]\mathrm{d}N_{l,\psi'}^-(t)\end{aligned}$$
$$(4)$$

yields an unravelling of the master equation (3) through a non-Markovian piecewise deterministic process. The first term on the r.h.s. of eq. (4) represents the normalized deterministic drift of the process which is generated by

$$\begin{aligned}G(t) =\ & H(t) - \frac{i}{2}\sum_m \Delta_m(t) \\ & \times\left[C_m^\dagger(t)C_m(t) - \langle\psi(t)|C_m^\dagger(t)C_m(t)|\psi(t)\rangle\right].\end{aligned}$$

The instantaneous and random quantum jumps are described by the second and the third line of eq. (4). The quantities $\mathrm{d}N_k^+(t)$ and $\mathrm{d}N_{l,\psi'}^-(t)$ are random Poisson increments satisfying the relations

$$\mathrm{d}N_k^+(t)\mathrm{d}N_{k'}^+(t) = \delta_{kk'}\mathrm{d}N_k^+(t),$$

$$\mathrm{d}N_{l,\psi'}^-(t)\mathrm{d}N_{l',\psi''}^-(t) = \delta_{ll'}\delta\left(|\psi'\rangle - |\psi''\rangle\right)\mathrm{d}N_{l,\psi'}^-(t), \quad (5)$$

$$\mathrm{d}N_k^+(t)\mathrm{d}N_{l,\psi'}^-(t) = 0,$$

and having expectation values

$$\mathrm{E}[\mathrm{d}N_k^+(t)] = \Delta_k^+(t)\langle\psi(t)|C_k^\dagger(t)C_k(t)|\psi(t)\rangle\mathrm{d}t, \quad (6)$$

$$\mathrm{E}[\mathrm{d}N_{l,\psi'}^-(t)] = \Delta_l^-(t)\frac{P\left[|\psi'\rangle,t\right]}{P\left[|\psi\rangle,t\right]}\langle\psi'|C_l^\dagger(t)C_l(t)|\psi'\rangle$$
$$\times\delta\left(|\psi(t)\rangle - \frac{C_l(t)|\psi'\rangle}{||C_l(t)|\psi'\rangle||}\right)\mathrm{d}t. \quad (7)$$

Here, the delta-functional on Hilbert space is defined by $\int \mathrm{d}\psi\,\delta(|\psi\rangle - |\psi_0\rangle)F[|\psi\rangle] = F[|\psi_0\rangle]$, where $F[|\psi\rangle]$ is an arbitrary smooth functional.

The physical meaning of the properties in (5) is that there cannot be two or more jumps simultaneously in a given realization of the process and in a given moment of time. Suppose first that the dynamics is Markovian or time-dependent Markovian. We then have $\mathrm{d}N_{l,\psi'}^-(t) = 0$ and the SSE (4) reduces to the stochastic differential equation of the standard PDP unravelling. According to the second line of eq. (4) the quantum jumps are represented by an instantaneous change of the state vector,

$$|\psi(t)\rangle \longrightarrow \frac{C_k(t)|\psi(t)\rangle}{||C_k(t)|\psi(t)\rangle||},$$

and by virtue of eq. (6) this jump occurs at the rate

$$\Gamma_+ = \Delta_k^+(t)\langle\psi(t)|C_k^\dagger(t)C_k(t)|\psi(t)\rangle. \quad (8)$$

The term in the third line of the SSE (4) describes the negative channels which are crucial for the unravelling of non-Markovian dynamics. The corresponding jumps are given by instantaneous transitions from the actual state $|\psi(t)\rangle$ to some state $|\psi'\rangle$. To account for all possible target states of the negative channel jumps we perform in this term an integration over $|\psi'\rangle$. According to the delta functional in eq. (7) the target state $|\psi'\rangle$ of the possible jumps is related to the source state $|\psi\rangle$ by $|\psi\rangle = C_l|\psi'\rangle/||C_l|\psi'\rangle||$. Hence, negative jumps correspond to a reversal of certain positive jumps, obtained by interchanging the role of source and target state. The quantity $\mathrm{d}N_{l,\psi'}^-(t)$ is the Poisson increment for the negative jumps via channel l. From eq. (7) we infer that the state vector $|\psi\rangle$ can perform a jump to a state vector in some volume element $\mathrm{d}\psi'$ of Hilbert space around $|\psi'\rangle$ with the rate

(for simplicity we omit the time arguments)

$$\Gamma_- = \Delta_l^- \frac{P\left[|\psi'\rangle\right]\mathrm{d}\psi'}{P\left[|\psi\rangle\right]\mathrm{d}\psi}\langle\psi'|C_l^\dagger C_l|\psi'\rangle\delta\left(|\psi\rangle - \frac{C_l|\psi'\rangle}{||C_l|\psi'\rangle||}\right)\mathrm{d}\psi.$$

In an ensemble of realizations of the process the quantity $P[|\psi'\rangle]\mathrm{d}\psi'/P[|\psi\rangle]\mathrm{d}\psi$ can be interpreted as N'/N, where N' is the number of realizations in volume element $\mathrm{d}\psi'$ and N is the number of realizations in element $\mathrm{d}\psi$. Then we can identify $\delta(|\psi\rangle - C_l|\psi'\rangle/||C_l|\psi'\rangle||)\mathrm{d}\psi = 1$. Hence, the negative channel jumps from $|\psi\rangle$ to $|\psi'\rangle$ occur at the rate

$$\Gamma_- = \Delta_l^- \frac{N'}{N}\langle\psi'|C_l^\dagger C_l|\psi'\rangle. \quad (9)$$

The comparison with the rate (8) for positive jumps shows two crucial differences. First, the rates for the positive jumps is proportional to the expectation value of $C_k^\dagger C_k$ in the source state, while the rates for the negative jumps is proportional to the expectation value of $C_l^\dagger C_l$ in the target state. Hence, again the role of source and target state have been interchanged. Second, the negative jump rates carry an additional factor of N'/N, the ratio of the number of ensemble members in the target state to the number of members in the source state. Note that due to the presence of this factor the SSE (4) is not a stochastic differential equation in the usual sense because the expectation values of the random increments (7) depend explicitly on the full probability density. To determine these increments at a certain time t one has to know the probability density $P[|\psi\rangle, t]$. Within a numerical simulation this is achieved by propagating simultaneously an ensemble of realization from which $P[|\psi\rangle, t]$ can then be estimated self-consistently. As an important consequence and as a result of the non-Markovian character of the dynamics we thus find certain correlations between different realizations of the process.

It may seem at first sight that the correlations between the realizations require that a huge number of realizations of the process has to be generated simultaneously in order to obtain a sufficiently accurate estimate for the probability density. However, when the realizations of the process are generated on a computer there is no need to have N_i identical copies of the state $|\psi_i\rangle$ to obtain $P[|\psi_i\rangle]$. It is sufficient to have only a single copy of $|\psi_i\rangle$ and to keep track of the corresponding integer number N_i. This allows to optimize the numerical implementation of the process and to perform simulations in an efficient way [17].

To prove that the expectation value (2) for the process obtained from the SSE (4) indeed satisfies the master equation (3) we start from

$$\mathrm{d}(|\psi\rangle\langle\psi|) = |\mathrm{d}\psi\rangle\langle\psi| + |\psi\rangle\langle\mathrm{d}\psi| + |\mathrm{d}\psi\rangle\langle\mathrm{d}\psi|.$$

Taking the expectation value of this relation, expressing the increments $|\mathrm{d}\psi\rangle$ through the SSE (4), and using the

properties (5) we find

$$
\begin{aligned}
\mathrm{d}\rho = &- i[H,\rho]\mathrm{d}t - \sum_m \frac{\Delta_m}{2}\left\{C_m^\dagger C_m, \rho\right\}\mathrm{d}t \\
&+ \sum_m \Delta_m \mathrm{E}\left[\langle\psi|C_m^\dagger C_m|\psi\rangle|\psi\rangle\langle\psi|\right]\mathrm{d}t \\
&+ \mathrm{E}\left[\sum_k \left(\frac{C_k|\psi\rangle\langle\psi|C_k^\dagger}{\|C_k|\psi\rangle\|^2} - |\psi\rangle\langle\psi|\right)\mathrm{d}N_k^+\right] \\
&+ \mathrm{E}\left[\sum_l \int \mathrm{d}\psi'\left(|\psi'\rangle\langle\psi'| - |\psi\rangle\langle\psi|\right)\mathrm{d}N_{l,\psi'}^-\right].
\end{aligned}
$$

Using here the expectation values of the increments from eq. (7) one immediately obtains the required master equation (3). Hence, we have proven the validity of SSE (4) which is the central result of the paper. It is worth mentioning that while there exists stochastic Schrödinger equations of diffusion type for non-Markovian systems [23,24], to the best of our knowledge our SSE is the first representation through a stochastic quantum jump process in the reduced system's Hilbert space.

It is important to note that the expectation value for $\mathrm{d}N_{l,\psi'}^-(t)$ in eq. (7) is not well defined when the denominator becomes equal to zero, *i.e.* $P[|\psi\rangle, t] = 0$, where $|\psi\rangle = C_l|\psi'\rangle/\|C_l|\psi'\rangle\|$, or alternatively $N = 0$ in eq. (9). The stochastic process breaks down at this point since there exists an open negative channel but there are no realizations which are in the source state of the corresponding jump. The formulation of general conditions on the structure of the master equation (1) that ensure the absence of such singularities in the corresponding SSE (4) seems to be a difficult problem. However, it is quite easy to demonstrate that a breakdown of the process necessarily takes place if the master equation violates positivity. In fact, within the stochastic formulation developed here the density matrix $\rho(t)$ of the open system is given by the expectation value (2) which represents, by the very construction, a positive matrix. Therefore, if the master equation violates positivity at some point of time the stochastic dynamics must necessarily cease to exist. The present method thus signals the point of violation of positivity of the density matrix.

To prove this statement we denote the state space, *i.e.*, the set of all density matrices of the open system by $\mathcal{S}$. Let us assume that the master equation (1) violates positivity. Hence, there is an initial state $\rho(0)$ and a corresponding solution $\rho(t)$ of the master equation which leaves the state space $\mathcal{S}$ after some point of time $t = t_0$. At this point $\rho(t_0) = \rho_0$ reaches the boundary of $\mathcal{S}$. Let $\lambda(t) = \langle\varphi(t)|\rho(t)|\varphi(t)\rangle$ be the lowest eigenvalue of $\rho(t)$ with corresponding eigenvector $|\varphi(t)\rangle$. Lying on the boundary, ρ_0 must have at least one zero eigenvalue with corresponding eigenvector $|\varphi_0\rangle = |\varphi(t_0)\rangle$, *i.e.*, we have $\lambda(t_0) = \langle\varphi_0|\rho_0|\varphi_0\rangle = 0$. Hence, an appropriate condition implying the violation of positivity is given by the inequality

$\dot{\lambda}(t_0) < 0$ (see footnote [2]). The Hellman-Feynman theorem yields

$$
\dot{\lambda}(t_0) = \langle\varphi_0|\dot{\rho}(t_0)|\varphi_0\rangle = \langle\varphi_0|\mathcal{L}_{t_0}\rho_0|\varphi_0\rangle,
$$

and we find the following condition for the violation of positivity:

$$
\langle\varphi_0|\mathcal{L}_{t_0}\rho_0|\varphi_0\rangle < 0. \tag{10}
$$

Consider now an ensemble representation of ρ_0 that is generated through the SSE (4): $\rho_0 = \sum_i p_i|\psi_i\rangle\langle\psi_i|$ with $\langle\psi_i|\psi_i\rangle = 1$, $p_i > 0$ and $\sum_i p_i = 1$. We then have

$$
\langle\varphi_0|\rho_0|\varphi_0\rangle = \sum_i p_i|\langle\varphi_0|\psi_i\rangle|^2 = 0.
$$

It follows that $|\varphi_0\rangle$ is orthogonal to all members of the ensemble, *i.e.*, $\langle\varphi_0|\psi_i\rangle = 0$. Evaluating condition (10), one therefore finds

$$
\langle\varphi_0|\mathcal{L}_{t_0}\rho_0|\varphi_0\rangle = \sum_{m,i} p_i\Delta_m(t_0)|\langle\varphi_0|C_m|\psi_i\rangle|^2 < 0.
$$

Hence, there must exist indices m and i such that $\Delta_m(t_0) < 0$ and $\langle\varphi_0|C_m|\psi_i\rangle \neq 0$. It follows that $C_m|\psi_i\rangle$ has a nonzero component in the direction of $|\varphi_0\rangle$ and that the state vector $|\psi\rangle = C_m|\psi_i\rangle/\|C_m|\psi_i\rangle\|$ does not belong to the ensemble $\{|\psi_i\rangle\}$. In other words, $P[|\psi\rangle, t_0] = 0$. We conclude that the point of violation of positivity implies the breakdown of the SSE (4) because there exists an open channel with negative rate while the probability of being in the source state of the corresponding jump vanishes.

Of course, the formal mathematical solution of the master equation (1) does not halt at the point of time when the positivity is lost: The dynamics continues to reduce the occupation probability of a given state beyond the zero-point. However, the evolution given by the SSE (4) stops at the zero-point since the number of realizations in a given state cannot, by construction, have negative values. Thus, the stochastic process developed here identifies the point of time where the master equation loses the positivity, preventing excursions to unphysical solutions. While the stochastic unravelling of the master equation is in general not unique, we expect that the connection between a breakdown of the positivity and a singularity of the SSE holds for all stochastic representation of the form constructed here.

In conclusion, we have derived a piecewise deterministic process which describes the dynamics of non-Markovian systems. The stochastic Schrödinger equation constructed reveals the fundamental mathematical and physical difference between time-local master equations which appear with positive and with negative rates. The corresponding Poisson increments have a distinct structure and the negative rate process clearly shows how non-Markovian effects are manifested.

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[2]It is assumed here that $\dot{\lambda}(t_0)$ does not vanish, which is obviously the generic case.

Markovian and non-Markovian processes are widely used for the modelling of dynamical systems in many areas of physics, chemistry and biophysics. Our results indicate how to treat master equations with negative rates and memory effects also for classical systems. In fact, the standard simulation algorithm for a classical Markovian master equation corresponding to the stochastic wave function method is known as Gillespie algorithm [25]. The method proposed here could therefore lead to the development of an efficient non-Markovian generalization of the Gillespie algorithm and thus opens the way to many further studies in the dynamics of complex system.

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