Stochastic dynamics of open quantum systems: Derivation of the differential Chapman-Kolmogorov equation

Heinz-Peter Breuer and Francesco Petruccione

Fakultät für Physik, Albert-Ludwigs-Universität,

Hermann-Herder Straße 3, 79104 Freiburg im Breisgau, Federal Republic of Germany

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A formulation of quantum statistical ensembles in terms of probability distributions on a projective Hilbert space is developed. The combination of statistically independent systems and the reduction of a system to one of its subsystems are described by means of a tensor product of probability distributions and a reduction formula for the reduced probability distribution. Within this framework the dynamics of open quantum systems is investigated starting from a microscopic system-plus-reservoir model. Employing the Markov approximation of the classical theory of stochastic processes the short time behavior of the conditional transition probability is derived and shown to yield a differential Chapman-Kolmogorov equation. The latter has the form of a Liouville master equation for the reduced probability distribution of the open quantum system. Thus it is shown that the time-dependent wave function of an open quantum system represents a well-defined and unique stochastic process in the space of rays of the underlying Hilbert space. This stochastic process consists of a continuous time evolution generated by a nonlinear Schrödinger equation and a discontinuous jump process; the realizations of the processes correspond to those of the Monte Carlo wave function simulation method and to those of the jump-type evolution of the quantum trajectory method. The quantum master equation for the statistical operator is derived as the equation of motion for the two-point correlation function of the stochastic process.

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I. INTRODUCTION

The classical theory of probability and stochastic processes [1] provides powerful tools for the description of classical complex dynamical systems [2–6]. In this context, the variables of the system under study are considered as random variables, the time evolution of which is represented by a stochastic process. The latter is obtained either from phenomenological models or by applying various approximation schemes to the underlying microscopic theory.

In view of the great success of these stochastic methods it is natural to try a similar ansatz for the theoretical description of open quantum systems. In fact, there exist several approaches in the literature that exploit the methods of the theory of stochastic processes for the formulation of damping and fluctuations in open quantum systems [7–9]. Since the state of a quantum system containing all accessible information is represented by a wave function of the underlying Hilbert space, the most natural way to combine the methods of classical probability theory with the particular structure of quantum mechanics is to consider the wave function as a random vector in Hilbert space. The time evolution of the wave function is then represented as a stochastic process. This approach has been followed by several authors on phenomenological grounds. Basically, one may distinguish two classes of models. In the first class the stochastic wave function is described by means of stochastic Schrödinger-type equations. Particularly interesting in this context is the quantum state diffusion model first proposed by Gisin [10] and developed further by Gisin and Percival [11]. The dynamic equation of the quantum state diffusion model is a nonlinear stochastic Schrödinger-type equation which in turn is equivalent to a certain functional Fokker-Planck equation for the corresponding probability distribution on Hilbert space [12]. The second class of models considers stochastic jump processes that are defined in terms of algorithms by which realizations of the stochastic process are generated. This so-called Monte Carlo wave function simulation method has been proposed by Dalibard, Castin, and Mølmer [13]. A related method has been formulated a short time later by Dum, Zoller, and Ritsch [14] (see also Ref. [15]). Independently, the same method has been developed by Carmichael and co-workers [16,17] under the name "quantum trajectory" method. Recently, a different phenomenological jump process has been formulated in terms of a Liouville master equation for the corresponding probability distribution [18].

It is important to realize that introducing a random wave function in Hilbert space amounts to defining a probability distribution on Hilbert space. A mathematically rigorous formulation of probability measures on Hilbert space can be found in Ref. [19]. The introduction of a probability distribution on Hilbert space can be motivated by considering the ensemble definition of the statistical operator. Suppose we have given a quantum mechanical system S and an ensemble consisting of a large number N of identical copies of this system. According to quantum mechanics each member of the ensemble can be described by a normalized wave function $\psi^{(k)}(x)$ in the underlying Hilbert space, where $k \in \{1, 2, \ldots, N\}$ labels the different member systems. The statistical operator ρ

(in the position representation) can then be introduced as the mean value of the quantity $\psi(x)\psi^*(x')$, i.e., we may define (see, e.g., [20,21])

$$\rho(x, x') = \frac{1}{N} \sum_{k=1}^{N} \psi^{(k)}(x)\psi^{(k)*}(x') \quad . \tag{1}$$

On the basis of this definition one is immediately led to represent the above ensemble by a probability distribution $P[\psi]$ on Hilbert space, which enables one to interpret the right-hand side of Eq. (1) as the *statistical estimate of the expectation value* $\langle \psi(x)\psi^*(x')\rangle$, where the angular brackets denote an integral over Hilbert space, with the corresponding probability measure. In this way, the particular structure of quantum mechanics is combined with classical probability theory. Introducing a probability distribution on Hilbert space, the wave function ψ becomes a stochastic variable, that is, a random vector in Hilbert space, and the statistical operator is interpreted as the two-point correlation function. This interpretation is the basis of the aformentioned stochastic approaches that rely on the unraveling [17] of the quantum master equation for the reduced density operator [7,8] in terms of stochastic wave functions.

The central goals of the present paper can be described as follows. First, we are going to elaborate in detail the implications of the above ansatz and develop a general framework for the analysis of quantum ensembles in terms of probability distributions on projective Hilbert space. Second, we shall investigate within this framework the description of the dynamics of an open quantum system by means of a stochastic process in projective Hilbert space. The stochastic dynamics of the state vector is obtained from a microscopic system-plus-reservoir model by deriving within the Markov approximation the differential Chapman-Kolmogorov equation for the reduced probability distribution on the Hilbert space of the open system [22,23]. It is shown that the differential Chapman-Kolmogorov equation is a Liouville master equation. This means that the time-dependent wave function represents a stochastic process in projective Hilbert space that consists of a continuous time evolution according to the Liouville part and a discontinuous jump process defined by a gain-and-loss balance equation for the probability distribution.

It is important to note that the derivation of the stochastic process for the open system wave function relies on physical assumptions similar to those of the derivation of the quantum master equation for the reduced density operator; see, e.g., Refs. [7,8]. However, the derivation is performed completely within the framework of classical probability theory. Thus this derivation solely deals, right from the beginning, with ensembles of pure states (rays in projective Hilbert space) and therefore does not require the *a priori* knowledge of the quantum master equation for the reduced density operator.

The material of this paper is presented as follows. In Sec. II we formulate the general concepts for the description of quantum systems by means of probability distributions on Hilbert space. In Sec. II A we make more precise the idea of probability distributions on Hilbert

space. According to the principles of quantum mechanics, we only consider probability distributions that are concentrated on the unit sphere in Hilbert space and that do not depend upon the phase of the wave function. Thus these distributions can be regarded as probability distributions on the space of rays, that is, on projective Hilbert space. We then consider the dynamics of closed systems and derive the Liouville equation, which is a functional equation of motion for the probability distribution. Furthermore, we introduce the interaction representation of the probability distribution and deduce the corresponding Liouville equation. Section II B contains the derivation of two basic prescriptions: The first one defines a kind of tensor product of probability distributions, which enables us to determine the probability distribution of a quantum system that is composed of two statistically independent subsystems. The second prescription tells us how to obtain from a given probability distribution the reduced distribution of one of its subsystems.

The concepts developed in Sec. II enable us to study the dynamics of open quantum systems. Section III contains the derivation of the differential Chapman-Kolmogorov equation for the reduced system dynamics. To this end, the open system under study is coupled to an external heat bath. In order to simplify the presentation the coupling is assumed to be of the form $A \otimes B$, where A and B denote self-adjoint system and bath operators, respectively. In Sec. III A we give a detailed exposition of the procedure of the derivation. Furthermore, we precisely state the Markov approximation and the corresponding physical conditions. Section III B contains the derivation of the short time behavior of the conditional transition probability of the Markov process in second-order perturbation theory. This short time behavior immediately leads to the differential Chapman-Kolmogorov equation, which is the central result of this paper.

Since the material presented in Sec. III B is rather technical we have devoted Sec. IV to a detailed discussion of the physical meaning and the implications of the Liouville master equation. We demonstrate in Sec. IV that the Liouville master equation defines, in fact, a stochastic process on projective Hilbert space of the open system and we deduce the quantum master equation for the statistical operator as the equation of motion of the two-point correlation function of the stochastic process.

Finally, we summarize our results in Sec. V. In the Appendix we prove two equations, which are required in Sec. III B.

II. DESCRIPTION OF QUANTUM ENSEMBLES BY PROBABILITY DISTRIBUTIONS ON HILBERT SPACE: GENERAL CONCEPTS

In this section we develop the general framework for the description of quantum systems in terms of probability distributions on Hilbert space. Section II A is mainly devoted to the definition of distributions on projective Hilbert space and the derivation of the Liouville equation for closed systems in the Schrödinger and the interaction representation. In Sec. II B we formulate the combina-

tion of two systems and the reduction of a system to a subsystem in the language of probability distributions on Hilbert space.

A. Probability distributions on Hilbert space

In this subsection we study a closed quantum mechanical system S, the states of which are described by wave functions ψ from some Hilbert space $\mathcal{H}$. We write $\psi = \psi(x)$, where x represents a complete set of position coordinates and further quantum numbers for the internal degrees of freedom. Accordingly, the integral over x is understood to be an integral over the configuration space of the system and a sum over the quantum numbers referring to the internal degrees of freedom. The scalar product on $\mathcal{H}$ is written as

$$\langle \psi | \varphi \rangle \equiv \int dx \, \psi^*(x)\varphi(x) \tag{2}$$

and the corresponding norm is denoted by $||\psi|| \equiv \langle \psi | \psi \rangle^{1/2}$.

We consider an ensemble that consists of a large number of copies of the system S each member of which is described by its own wave function. This ensemble can be characterized by a probability distribution on Hilbert space $\mathcal{H}$ in the following way. The functional volume element on $\mathcal{H}$ is defined by

$$D\psi D\psi^* \equiv \prod_x d(\mathrm{Re}\psi(x))d(\mathrm{Im}\psi(x))$$
$$\equiv \prod_x \frac{i}{2} d\psi(x)d\psi^*(x) \quad , \tag{3}$$

where $\mathrm{Re}\psi(x)$ and $\mathrm{Im}\psi(x)$ are the real and the imaginary part of $\psi(x)$, respectively. The probability density $P[\psi]$ corresponding to the above ensemble can then be introduced by defining $P[\psi]D\psi D\psi^*$ to be the probability of finding the system in the volume element $D\psi D\psi^*$ around ψ. It should be clear that in the case of an infinite-dimensional Hilbert space $P[\psi]$ is a functional on Hilbert space and (3) is a functional measure. In this paper we leave aside all mathematical questions concerning the existence of such probability measures. Instead, we adopt a pragmatic point of view and always assume that the underlying Hilbert space is or can be approximated by a finite-dimensional subspace $\mathcal{H}_n$ of any dimension n.

It is important to note that the measure (3) is invariant with respect to linear unitary transformations $U : \mathcal{H} \longrightarrow \mathcal{H}$. This fact is expressed by the equation

$$\delta[U\psi] = \delta[\psi] \quad , \tag{4}$$

where

$$\delta[\psi(x)] \equiv \prod_x \delta(\mathrm{Re}\psi(x))\delta(\mathrm{Im}\psi(x)) \tag{5}$$

denotes the functional δ function on the Hilbert space $\mathcal{H}$ [$\delta(\ )$ is the ordinary delta function]. According to the general principles of quantum mechanics, the physical state of a system S is completely described by a normalized wave function and wave functions that differ by a

phase factor are equivalent. It is thus natural to impose the following restrictions on the probability distribution $P[\psi]$.

(i) P is normalized

$$\int D\psi D\psi^* P[\psi] = 1 \quad . \tag{6}$$

(ii) The probability distribution is concentrated on the unit sphere in Hilbert space defined by $\langle \psi | \psi \rangle \equiv ||\psi||^2 = 1$. This condition can be expressed by demanding that there exists a functional $Q[\psi]$ such that

$$P[\psi] = \delta(||\psi|| - 1)Q[\psi] \quad . \tag{7}$$

(iii) The probability distribution does not depend upon the phase of the wave function, i.e., we have for all $\chi \in \mathbb{R}$

$$P[e^{i\chi}\psi] = P[\psi] \quad . \tag{8}$$

It is important to note that conditions (7) and (8) imply that $P[\psi]$ can, in fact, be regarded as a probability density on the space of rays, that is, as a probability density on projective Hilbert space.

Having defined a probability distribution on Hilbert space, the expectation value of a functional $F[\psi]$ is given by

$$\langle F \rangle \equiv \int D\psi D\psi^* F[\psi]P[\psi] \quad . \tag{9}$$

In particular, the expectation value of a physical observable represented by a self-adjoint operator A is defined by the ensemble average of the quantum mechanical expectation value:

$$\langle A \rangle \equiv \langle\langle \psi | A | \psi \rangle\rangle$$
$$\equiv \int D\psi D\psi^* \int dx \, \psi^*(x)A\psi(x)P[\psi] \quad . \tag{10}$$

Equivalently, one can introduce the statistical operator as the two-point correlation function

$$\rho(x, x') \equiv \langle \psi(x)\psi^*(x') \rangle$$
$$= \int D\psi D\psi^* \psi(x)\psi^*(x')P[\psi] \tag{11}$$

and write Eq. (10) as

$$\langle A \rangle = \mathrm{tr}(A\rho) \quad , \tag{12}$$

in accordance with the quantum mechanical expression.

It should be clear at this stage that the space of probability distributions that fulfill properties (6)–(8) is much larger than the space of statistical operators, i.e., than the space of Hermitian and positive operators of trace one. We therefore need a prescription that provides the connection of the above formulation to the usual formulation of quantum ensembles in terms of statistical operators. To this end, we construct a correspondence between statistical operators and a certain type of probability distributions on Hilbert space. Assume that the ensemble has been prepared in a state that is described, according

to the rules of quantum mechanics, by means of a normalized statistical operator ρ. In the most general case, the spectral decomposition of ρ can be written

$$\rho = \sum_n \sum_{i=1}^{s_n} p_n |\varphi_{n,i}\rangle\langle\varphi_{n,i}| \quad , \tag{13}$$

where

$$p_n \geq 0 \ , \quad \sum_n \sum_{i=1}^{s_n} p_n = 1 \ , \quad \langle\varphi_{n,i}|\varphi_{m,j}\rangle = \delta_{nm}\delta_{ij} \quad . \tag{14}$$

Whereas the index n labels the different s_n-dimensional eigenspaces V_n of ρ, the index i labels the basis vectors that span these eigenspaces, i.e., we have $V_n = \text{span}\{\varphi_{n,1}, \varphi_{n,2}, \ldots, \varphi_{n,s_n}\}$. A natural choice for the probability distribution $P[\psi]$ on $\mathcal{H}$ is then given by [18]

$$P[\psi] = \sum_n p_n \frac{s_n}{|K_n|} \int_{K_n} d\omega_n(\lambda) \, \delta[\psi - \Phi_n(\lambda)] \quad . \tag{15}$$

Writing this equation we have introduced the $(2s_n - 1)$-dimensional unit spheres $K_n = \{\Phi_n(\lambda)\}$, where

$$\Phi_n(\lambda) = \sum_{i=1}^{s_n} \lambda_i \varphi_{n,i} \ , \quad \lambda_i \in \mathbb{C} \ , \quad \sum_{i=1}^{s_n} |\lambda_i|^2 = 1 \quad . \tag{16}$$

Furthermore, $d\omega_n(\lambda)$ denotes the volume element on K_n and $|K_n|$ is the total volume of K_n. Thus we see that the probability distribution $P[\psi]$ as given by Eq. (15) is concentrated on the unit spheres $K_n \subset V_n$ and that it is constant on each of these spheres. Note that the integration over the spheres K_n in Eq. (15) ensures that the probability distribution does not depend on the choice of basis functions within the degenerate subspaces V_n. Physically, the integral over the spheres K_n corresponds to a statistical mixture of all normalized states in V_n with equal weights [24]. In the special case of a nondegenerate statistical operator

$$\rho = \sum_n p_n |\varphi_n\rangle\langle\varphi_n| \quad , \tag{17}$$

Eq. (15) takes the form

$$P[\psi] = \sum_n p_n \int_0^{2\pi} \frac{d\chi}{2\pi} \delta[\psi - e^{i\chi}\varphi_n] \quad . \tag{18}$$

Obviously, the probability distribution (15) fulfills the basic properties (6)–(8). Moreover, for the distribution (15) the expectation value of any self-adjoint operator A is given by

$$\langle A\rangle = \int D\psi D\psi^* \int dx \, \psi^*(x) A\psi(x)$$
$$\times \sum_n p_n \frac{s_n}{|K_n|} \int_{K_n} d\omega_n(\lambda) \, \delta[\psi - \Phi_n(\lambda)] \quad . \tag{19}$$

Performing the integration over ψ yields

$$\langle A\rangle = \sum_n p_n \frac{s_n}{|K_n|} \int_{K_n} d\omega_n(\lambda)\langle\Phi_n(\lambda)|A|\Phi_n(\lambda)\rangle \quad . \tag{20}$$

Employing Eq. (16) this is seen to be equal to

$$\langle A\rangle = \sum_n \sum_{i=1}^{s_n} p_n\langle\varphi_{n,i}|A|\varphi_{n,i}\rangle = \text{tr}(A\rho) \quad . \tag{21}$$

We now turn to the description of the dynamics. We require that for the closed quantum system under consideration each member of the ensemble evolves according to the Schrödinger equation

$$i\frac{\partial\psi}{\partial t} = H\psi \quad , \tag{22}$$

where H is the Hamiltonian of the system, and choosing appropriate units we have assumed $\hbar = 1$. Introducing an initial probability distribution $P_0[\psi]$ that describes the initial state of the ensemble, ψ becomes a special kind of stochastic Markov process that is governed by a Liouville equation. Denoting the time-dependent probability distribution by $P = P[\psi, t]$ we write

$$P[\psi, t] = \int D\psi_0 D\psi_0^* \, \delta[e^{-iHt}\psi_0 - \psi]P_0[\psi_0] \quad . \tag{23}$$

This equation expresses that any initial ψ_0 drawn from the initial distribution P_0 evolves according to Schrödinger's equation. In other words, the conditional transition probability $\delta[e^{-iH(t_2-t_1)}\psi_1 - \psi_2]$ is non-zero if and only if ψ_2 evolves from ψ_1 according to the Schrödinger equation within the time interval from t_1 to t_2. Using (4) we obtain from Eq. (23) by integrating over ψ_0

$$P[\psi, t] = P_0[U^\dagger(t)\psi] \quad , \tag{24}$$

where $U(t) = \exp\{-iHt\}$ is the time-evolution operator. Differentiating (24) with respect to time and using the fact that H is self-adjoint yields the differential form of the Liouville equation, which is a first-order functional equation for the probability distribution:

$$\frac{\partial}{\partial t}P[\psi, t] = i \int dx \left\{ \frac{\delta P}{\delta\psi(x)}H\psi(x) - \psi^*(x)H\frac{\delta P}{\delta\psi^*(x)} \right\} , \tag{25}$$

where $\delta/\delta\psi(x)$ and $\delta/\delta\psi^*(x)$ are functional Wirtinger derivatives. Since the linear time-evolution operator $U(t)$ is unitary, it is easily seen that the Liouville equation (25) preserves the basic conditions (6)–(8), i.e., if $P_0[\psi]$ is a probability density on projective Hilbert space so is $P[\psi, t]$ for all $t > 0$. Moreover, the ensemble average (10) obeys the equation of motion

$$\frac{d}{dt}\langle A\rangle = i\langle[H, A]\rangle \quad , \tag{26}$$

which is identical to the corresponding equation in quantum mechanics. If the statistical operator is diagonal in the energy representation the probability distribution (15) is a stationary solution of the Liouville equation (25).

Another concept that will be important in the follow-

ing sections is the interaction representation of the time-dependent probability distribution on Hilbert space. In order to derive the equation of motion in the interaction picture we write

$$H = H_0 + H_I \quad , \tag{27}$$

where H_0 is the unperturbed part of the Hamiltonian and H_I denotes the interaction Hamiltonian. We define the probability distribution $\tilde{P}[\psi, t]$ in the interaction representation by

$$\tilde{P}[\psi, t] \equiv P[e^{-iH_0(t-t_0)}\psi, t] \quad . \tag{28}$$

At time $t = t_0$ the Schrödinger and the interaction representation coincide, that is, we have $\tilde{P}[\psi, t_0] = P[\psi, t_0]$. On differentiating Eq. (28) with respect to time and using the Liouville equation for $P[\psi, t]$ [Eq. (25)] one finds

$$\frac{\partial}{\partial t} \tilde{P}[\psi, t] = i \int dx \left\{ \frac{\delta \tilde{P}}{\delta \psi(x)} H_I(t)\psi(x) \right.$$
$$\left. - \psi^*(x)H_I(t)\frac{\delta \tilde{P}}{\delta \psi^*(x)} \right\}, \tag{29}$$

where

$$H_I(t) \equiv e^{iH_0(t-t_0)} H_I e^{-iH_0(t-t_0)} \tag{30}$$

is the interaction Hamiltonian in the interaction representation. The general solution of Eq. (29) takes the form

$$\tilde{P}[\psi, t] = \tilde{P}[\{U_I(t, t_0)\}^\dagger \psi, t_0] \quad , \tag{31}$$

where $U_I(t, t_0)$ is the time-evolution operator in the interaction picture.

Finally, we mention that if G is a transformation group acting on $\mathcal{H}$ by unitary transformations $U(g)$, $g \in G$, the probability distribution P transforms like a scalar,

$$P'[\psi] = P[U(g)\psi] \quad . \tag{32}$$

Invoking the invariance of the measure with respect to unitary transformations it follows that

$$\rho' = [U(g)]^\dagger \rho U(g) \quad , \tag{33}$$

as it must be according to the general principles of quantum mechanics.

B. Composition and reduction of systems

This subsection is devoted to a detailed discussion of two basic constructions that enable us to deal with the combination of statistically independent systems and with the reduced description in terms of a reduced probability distribution. The considerations presented in this section are needed for the derivation of the reduced system dynamics, which will be given in Sec. III.

The physical situation we have in mind is the following one. Suppose that we have two systems S_1 and S_2 with

corresponding Hilbert spaces $\mathcal{H}_1$ and $\mathcal{H}_2$. Wave functions in $\mathcal{H}_1$ are written as $\psi_1(x_1)$ and wave functions in $\mathcal{H}_2$ are denoted by $\psi_2(x_2)$. Furthermore, throughout the paper (unless stated otherwise) we use the convention that all quantities that refer to system S_i carry an index i, where $i = 1, 2$. For example, $\langle | \rangle_2$ denotes the scalar product on $\mathcal{H}_2$ and $|| \, ||_1$ is the norm in $\mathcal{H}_1$. According to the general principles of quantum mechanics, the Hilbert space $\mathcal{H}$ underlying the combined system $S = S_1 + S_2$ is given by the tensor product

$$\mathcal{H} = \mathcal{H}_1 \otimes \mathcal{H}_2 \quad . \tag{34}$$

Two questions then arise.

(i) Given two probability distributions $P_1[\psi_1]$ and $P_2[\psi_2]$ for the respective systems S_1 and S_2, what is the probability distribution for the combined system S on the Hilbert space $\mathcal{H}$ if the two systems are considered to be statistically independent?

(ii) Given a probability distribution $P[\psi]$ on the Hilbert space $\mathcal{H}$ for the combined system, what is the probability distribution $P_1[\psi_1]$ for the subsystem S_1 that enables us to determine expectation values for those observable that refer to S_1?

In order to answer the first question we consider the case that S_1 and S_2 are in pure states represented by wave functions ψ_1 and ψ_2, respectively. Quantum mechanics tells us that in this case S is also in a pure state represented by a wave function ψ, which is the product of the wave functions of the subsystems

$$\psi(x_1, x_2) = \psi_1(x_1)\psi_2(x_2) \quad . \tag{35}$$

According to our general prescriptions [see Eq. (18)] this pure state can be represented by a probability distribution on $\mathcal{H}$, which is given by (we ignore phase factors for a moment)

$$P[\psi] = \delta[\psi - \psi_1\psi_2] \quad . \tag{36}$$

The general case is obtained by averaging over ψ_1 and over ψ_2 with the corresponding probability distributions. Thus we define a tensor product

$$P = P_1 \otimes P_2 \tag{37}$$

of the probability distributions of the subsystems S_1 and S_2 by means of the equation

$$P[\psi] = (P_1 \otimes P_2)[\psi] \tag{38}$$
$$\equiv \int D\psi_1 D\psi_1^* \int D\psi_2 D\psi_2^* \, \delta[\psi - \psi_1\psi_2]$$
$$\times P_1[\psi_1]P_2[\psi_2].$$

It follows immediately from this expression that for any self-adjoint operator A on $\mathcal{H}$ of the form $A = A_1 \otimes A_2$ the following equation holds:

$$\langle A \rangle_{P_1 \otimes P_2} = \langle A_1 \rangle_{P_1} \langle A_2 \rangle_{P_2} \quad , \tag{39}$$

where the probability distributions by which the differ-

ent expectation values are defined have been indicated as indices of the angular brackets. Equation (39) means that for statistically independent systems S_1 and S_2 the expectation value in the combined system S of any product of operators is equal to the product of the expectation values of S_1 and S_2. In order to prove Eq. (39) we insert the definition (38) for the tensor product into the expression (10) for the expectation value of A to obtain

$$\langle A \rangle_{P_1 \otimes P_2} = \int D\psi D\psi^* \int D\psi_1 D\psi_1^* \int D\psi_2 D\psi_2^* \langle \psi | A | \psi \rangle$$
$$\times \delta[\psi - \psi_1 \psi_2] P_1[\psi_1] P_2[\psi_2] \quad . \tag{40}$$

On integrating over ψ we find

$$\langle A \rangle_{P_1 \otimes P_2} = \int D\psi_1 D\psi_1^* \int D\psi_2 D\psi_2^*$$
$$\times \langle \psi_1 \psi_2 | A_1 \otimes A_2 | \psi_1 \psi_2 \rangle P_1[\psi_1] P_2[\psi_2]$$
$$= \left(\int D\psi_1 D\psi_1^* \langle \psi_1 | A_1 | \psi_1 \rangle_1 P_1[\psi_1] \right)$$
$$\times \left(\int D\psi_2 D\psi_2^* \langle \psi_2 | A_2 | \psi_2 \rangle_2 P_2[\psi_2] \right) \quad ,$$

which obviously yields Eq. (39).

Let us check whether the basic conditions (6)–(8) are fulfilled for the tensor product. First, condition (6) follows from Eq. (39) by setting $A_1 = A_2 = \mathbf{1}$, where $\mathbf{1}$ denotes the identity operator on the respective Hilbert spaces. Condition (7) is obvious from the fact that $P[\psi]$ is nonzero only if ψ is the product of two normalized wave functions. Finally, also condition (8) is fulfilled since on using (4) for the unitary transformation given by the multiplication with the phase factor $\exp(i\chi)$ we obtain

$$P[e^{i\chi}\psi] = \int D\psi_1 D\psi_1^* \int D\psi_2 D\psi_2^*$$
$$\times \delta[\psi - e^{-i\chi}\psi_1 \psi_2] P_1[\psi_1] P_2[\psi_2]$$
$$= \int D\psi_1 D\psi_1^* \int D\psi_2 D\psi_2^*$$
$$\times \delta[\psi - \psi_1 \psi_2] P_1[e^{i\chi}\psi_1] P_2[\psi_2] \quad ,$$

and thus $P[\psi]$ is phase invariant if $P_1[\psi_1]$ is so.

Let us now turn to question (ii) above. In order to construct a probability distribution for the reduced subsystem on $\mathcal{H}_1$ from a distribution on $\mathcal{H}$ we first consider the case of a pure state $\psi \in \mathcal{H}$. According to quantum mechanics, the reduced statistical operator for the system S_1 is obtained by tracing over the states of the unobserved system S_2:

$$\rho_1 = \text{tr}_2 |\psi\rangle\langle\psi| = \sum_\alpha \langle \varphi_\alpha | \psi \rangle_2 \langle \psi | \varphi_\alpha \rangle_2$$
$$= \sum_\alpha w_\alpha \frac{\langle \varphi_\alpha | \psi \rangle_2}{w_\alpha^{1/2}} \frac{\langle \psi | \varphi_\alpha \rangle_2}{w_\alpha^{1/2}} \quad , \tag{41}$$

where $\{\varphi_\alpha\}$ is a complete orthonormal basis of $\mathcal{H}_2$ and we have defined

$$w_\alpha \equiv \int dx_1 |\langle \varphi_\alpha | \psi \rangle_2|^2 = ||\langle \varphi_\alpha | \psi \rangle_2||_1^2 \quad . \tag{42}$$

Note that the expression $w_\alpha^{-1/2} \langle \varphi_\alpha | \psi \rangle_2$ represents a normalized state in $\mathcal{H}_1$. The mixed state described by ρ_1 translates according to our general prescription [see Eq. (18)] into the probability distribution on $\mathcal{H}_1$ given by (again ignoring phase factors for a moment)

$$P_1[\psi_1] = \sum_\alpha w_\alpha \delta_1[w_\alpha^{-1/2} \langle \varphi_\alpha | \psi \rangle_2 - \psi_1] \quad , \tag{43}$$

where $\delta_1[\,]$ denotes the functional delta function on $\mathcal{H}_1$. Thus we have constructed P_1 for the case that S is in the pure state ψ. The general case is obtained by averaging the right-hand side of Eq. (43) over the probability distribution $P[\psi]$. Therefore, we define the reduced probability distribution on $\mathcal{H}_1$ by means of the equation

$$P_1[\psi_1] = \int D\psi D\psi^* \sum_\alpha w_\alpha \delta_1[w_\alpha^{-1/2} \langle \varphi_\alpha | \psi \rangle_2 - \psi_1] P[\psi] \quad . \tag{44}$$

Again, it is easy to check that the basic conditions (6)–(8) are fulfilled. Furthermore, we have for any operator A_1 on $\mathcal{H}_1$

$$\langle A_1 \rangle_{P_1} = \langle A_1 \otimes \mathbf{1} \rangle_P \quad . \tag{45}$$

To prove this relation one only has to employ the completeness (in $\mathcal{H}_2$) of the basis $\{\varphi_\alpha\}$.

We close this section by investigating the following consistency condition of our equations for the tensor product [Eq. (38)] of probability distributions and the reduced distribution [Eq. (44)]. Suppose that we have given two distributions P_1 and P_2 on their respective Hilbert spaces and that we form the tensor product $P = P_1 \otimes P_2$ according to Eq. (38). Applying then Eq. (44) we obtain a reduced probability distribution on $\mathcal{H}_1$, which is denoted by P_1'. The consistency condition then is that P_1' is equal to the original distribution P_1 on $\mathcal{H}_1$. In order to prove this we write P_1' in terms of P_1 and P_2 applying Eqs. (38) and (44):

$$P_1'[\psi_1] = \int D\psi D\psi^* \sum_\alpha w_\alpha \delta_1[w_\alpha^{-1/2} \langle \varphi_\alpha | \psi \rangle_2 - \psi_1]$$
$$\times \int D\tilde{\psi}_1 D\tilde{\psi}_1^* \int D\tilde{\psi}_1 D\tilde{\psi}_2^*$$
$$\times \delta[\psi - \tilde{\psi}_1 \tilde{\psi}_2] P_1[\tilde{\psi}_1] P_2[\tilde{\psi}_2] \quad .$$

On integrating over ψ we find

$$P_1'[\psi_1] = \int D\tilde{\psi}_1 D\tilde{\psi}_1^* \int D\tilde{\psi}_2 D\tilde{\psi}_2^*$$
$$\times \sum_\alpha w_\alpha \delta_1[w_\alpha^{-1/2} \langle \varphi_\alpha | \tilde{\psi}_1 \tilde{\psi}_2 \rangle_2 - \psi_1]$$
$$\times P_1[\tilde{\psi}_1] P_2[\tilde{\psi}_2], \tag{46}$$

where

$$w_\alpha \equiv \int dx_1 |\langle \varphi_\alpha | \tilde{\psi}_1 \tilde{\psi}_2 \rangle_2|^2$$
$$= ||\tilde{\psi}_1||_1^2 |\langle \varphi_\alpha | \tilde{\psi}_2 \rangle_2|^2 = |\langle \varphi_\alpha | \tilde{\psi}_2 \rangle_2|^2 \quad . \tag{47}$$

Thus we have

$$P_1'[\psi_1] = \int D\tilde{\psi}_1 D\tilde{\psi}_1^* \int D\tilde{\psi}_2 D\tilde{\psi}_2^* \sum_\alpha |\langle \varphi_\alpha | \tilde{\psi}_2 \rangle_2|^2$$

$$\times \delta_1 \left[\frac{\langle \varphi_\alpha | \tilde{\psi}_2 \rangle_2}{|\langle \varphi_\alpha | \tilde{\psi}_2 \rangle_2|} \tilde{\psi}_1 - \psi_1 \right] P_1[\tilde{\psi}_1] P_2[\tilde{\psi}_2]. \quad (48)$$

Since the factor in front of $\tilde{\psi}_1$ within the argument of the δ functional is a pure phase factor, we obtain, in view of the phase invariance of P_1,

$$P_1'[\psi_1] = \int D\tilde{\psi}_1 D\tilde{\psi}_1^* \int D\tilde{\psi}_2 D\tilde{\psi}_2^* \sum_\alpha |\langle \varphi_\alpha | \tilde{\psi}_2 \rangle_2|^2$$

$$\times \delta_1 [\tilde{\psi}_1 - \psi_1] P_1[\tilde{\psi}_1] P_2[\tilde{\psi}_2] \quad . \quad (49)$$

The sum over α in front of the δ functional is equal to 1 and hence we find, due to the normalization of P_2,

$$P_1'[\psi_1] = \int D\tilde{\psi}_1 D\tilde{\psi}_1^* \, \delta_1[\tilde{\psi}_1 - \psi_1] P_1[\tilde{\psi}_1] = P_1[\psi_1] \quad , \tag{50}$$

as required. Thus we have shown that on applying the reduction formula (44) to the tensor product (38) of two probability distributions the original distribution P_1 is recovered.

III. DERIVATION OF THE DIFFERENTIAL CHAPMAN-KOLMOGOROV EQUATION FOR THE REDUCED SYSTEM DYNAMICS

In the preceding section we have formulated the general concepts for the description of an ensemble of quantum mechanical systems in terms of probability distributions on the underlying Hilbert space. These concepts now enable us to derive an equation of motion for the reduced probability distribution pertaining to a system that is coupled to an external heat bath. The physical situation under consideration is similar to that considered in Sec. II B. The system S_1 (Hilbert space $\mathcal{H}_1$, wave functions ψ_1) is considered to be the system of interest (simply referred to as *the system*), whereas S_2 (Hilbert space $\mathcal{H}_2$, wave functions ψ_2) is the external heat bath. The Hamiltonian H of the combined system S acting on the Hilbert space $\mathcal{H} = \mathcal{H}_1 \otimes \mathcal{H}_2$ is written as

$$H = H_0 + H_I \quad , \tag{51}$$

where H_I is the interaction Hamiltonian and

$$H_0 = H_1 \otimes \mathbf{1} + \mathbf{1} \otimes H_2 \tag{52}$$

represents the free dynamics of the two subsystems S_1 (Hamiltonian H_1) and S_2 (Hamiltonian H_2). The interaction is assumed to be of the form

$$H_I = A \otimes B \quad , \tag{53}$$

where A and B are any self-adjoint operators acting on $\mathcal{H}_1$ and $\mathcal{H}_2$, respectively. We emphasize that the above form (53) for H_I has been chosen only for simplicity. The most general case will be the subject of a future paper [25].

It will be shown in this section that under suitable conditions to be specified below the equation governing the dynamics of the reduced probability distribution $P_1[\psi_1, t]$ for the system S_1 takes the form of a Liouville master equation [given by Eq. (101) below]. The following subsection A contains a precise formulation of the problem and gives an overview of the procedure. In subsection B we derive the short time behavior of the transition probability on Hilbert space within the Markov approximation and using second-order perturbation theory. The result enables us to derive the differential form of the Chapman-Kolmogorov equation for P_1, which turns out to be a Liouville master equation. A detailed physical discussion of the Liouville master equation is presented in Sec. IV.

A. The Markov approximation for the reduced system

We start by fixing an arbitrary time t_0 and an arbitrary time interval $\tau \equiv t - t_0$. We assume that (i) the system is so small compared to the bath that its influence upon the statistical properties of the bath can be neglected and (ii) the probability distribution of the bath is invariant under the time evolution of H_2, i.e., that P_2 is a stationary solution of the Liouville equation for the bath. Furthermore, we assume that at time t_0 the system and the bath are statistically independent. Under these assumptions we can write

$$P[\psi, t_0] = P_1[\psi_1, t_0] \otimes P_2[\psi_2] \quad . \tag{54}$$

The different steps in the derivation of the reduced system dynamics can be represented by the following scheme:

$$P[\psi, t_0] \xrightarrow{\;1\;} \tilde{P}[\psi, t] \xrightarrow{\;2\;} \tilde{P}_1[\psi_1, t] \xrightarrow{\;3\;} P_1[\psi_1, t] \quad , \tag{55}$$

where we have used (as in Sec. II) the convention that all distributions in the interaction representation carry a tilde. In the first step we transform to the interaction representation. To this end, we use the initial condition (54) and the definition of the tensor product (38) and determine the interaction picture probability distribution $\tilde{P}[\psi, t]$ of the total system at time $t = t_0 + \tau$. In the second step we employ our reduction formula (44) to obtain the reduced probability distribution $\tilde{P}_1[\psi_1, t]$ in the interaction representation. The Markov approximation is invoked in order to determine the short time behavior of $\tilde{P}_1[\psi_1, t]$. Finally, in the third step we transform back to the Schrödinger representation to obtain the reduced probability distribution $P_1[\psi_1, t]$ in the Schrödinger picture. The result immediately yields the differential Chapman-Kolmogorov equation. Let us now detail these steps indicated in the scheme (55).

Step 1: Transforming to the interaction representation

According to Eq. (28) the interaction picture probability distribution is given by

$$\tilde{P}[\psi, t] \equiv P[e^{-iH_0\tau}\psi, t] \quad . \tag{56}$$

Note that the interaction picture has been chosen in such a way that it coincides with the Schrödinger picture at $t = t_0$:

$$\tilde{P}[\psi, t_0] = P[\psi, t_0] \quad . \tag{57}$$

Thus we have in view of Eq. (54) and definition (38) for the tensor product

$$\tilde{P}[\psi, t_0] = \int D\tilde{\psi}_1 D\tilde{\psi}_1^* \int D\tilde{\psi}_2 D\tilde{\psi}_2^*$$
$$\times \delta[\psi - \tilde{\psi}_1\tilde{\psi}_2] P_1[\tilde{\psi}_1, t_0] P_2[\tilde{\psi}_2] \quad . \tag{58}$$

According to Eq. (31) the time evolution of this distribution in the interaction picture is given by

$$\tilde{P}[\psi, t] = \tilde{P}[U_I^\dagger(t, t_0)\psi, t_0]$$
$$= \int D\tilde{\psi}_1 D\tilde{\psi}_1^* \int D\tilde{\psi}_2 D\tilde{\psi}_2^* \, \delta[U_I^\dagger(t, t_0)\psi - \tilde{\psi}_1\tilde{\psi}_2]$$
$$\times P_1[\tilde{\psi}_1, t_0] P_2[\tilde{\psi}_2],$$

where

$$U_I(t, t_0) \equiv \mathcal{T} \exp\left\{-i \int_0^\tau ds H_I(s)\right\} \tag{59}$$

is the interaction picture time-evolution operator, $\mathcal{T}$ indicates time ordering, and

$$H_I(s) \equiv e^{iH_0 s} H_I e^{-iH_0 s} \tag{60}$$

is the interaction Hamiltonian in the interaction representation.

Step 2: Performing the Markov approximation

We now apply Eq. (44) to obtain the reduced probability distribution in the interaction representation:

$$\tilde{P}_1[\psi_1, t] = \int D\psi D\psi^* \int D\tilde{\psi}_1 D\tilde{\psi}_1^* \int D\tilde{\psi}_2 D\tilde{\psi}_2^*$$
$$\times \sum_\alpha w_\alpha \delta_1[w_\alpha^{-1/2}\langle\varphi_\alpha|\psi\rangle_2 - \psi_1]$$
$$\times \delta[U_I^\dagger(t, t_0)\psi - \tilde{\psi}_1\tilde{\psi}_2] P_1[\tilde{\psi}_1, t_0] P_2[\tilde{\psi}_2] \quad ,$$

where w_α is defined as in Eq. (42). Invoking Eq. (4) for the unitary operator $U_I(t, t_0)$ and integrating over ψ we obtain

$$\tilde{P}_1[\psi_1, t] = \int D\tilde{\psi}_1 D\tilde{\psi}_1^* \int D\tilde{\psi}_2 D\tilde{\psi}_2^*$$
$$\times \sum_\alpha w_\alpha \delta_1[w_\alpha^{-1/2}\langle\varphi_\alpha|U_I(t, t_0)\tilde{\psi}_1\tilde{\psi}_2\rangle_2 - \psi_1]$$
$$\times P_1[\tilde{\psi}_1, t_0] P_2[\tilde{\psi}_2],$$

where now

$$w_\alpha \equiv \int dx_1 |\langle\varphi_\alpha|U_I(t, t_0)\tilde{\psi}_1\tilde{\psi}_2\rangle_2|^2 \quad . \tag{61}$$

Thus we have

$$\tilde{P}_1[\psi_1, t] = \int D\tilde{\psi}_1 D\tilde{\psi}_1^* \, T[\psi_1, t|\tilde{\psi}_1, t_0] P_1[\tilde{\psi}_1, t_0] \quad , \tag{62}$$

where we have defined the functional kernel

$$T[\psi_1, t|\tilde{\psi}_1, t_0] \equiv \int D\tilde{\psi}_2 D\tilde{\psi}_2^*$$
$$\times \sum_\alpha w_\alpha \delta_1[w_\alpha^{-1/2}\langle\varphi_\alpha|U_I(t, t_0)\tilde{\psi}_1\tilde{\psi}_2\rangle_2$$
$$-\psi_1] P_2[\tilde{\psi}_2] \quad . \tag{63}$$

We now choose the following distribution to describe the external heat bath:

$$P_2[\psi_2] = \sum_\alpha p_\alpha \delta_2[\psi_2 - \varphi_\alpha] \quad , \tag{64}$$

where

$$\sum_\alpha p_\alpha = 1 \quad , \quad p_\alpha \geq 0 \quad , \tag{65}$$

and the complete orthonormal basis $\{\varphi_\alpha\}$ in $\mathcal{H}_2$ is chosen to be an eigenbasis of H_2,

$$H_2\varphi_\alpha = \varepsilon_\alpha\varphi_\alpha \quad . \tag{66}$$

Note that the distribution (64) is a stationary solution of the Liouville equation for the bath. Furthermore, we have ignored phase factors in Eq. (64) in order to simplify the notation. This is justified since P_2 only enters through the tensor product, which is already phase invariant if one of its factors is (see the proof of the phase invariance of the tensor product in Sec. II). Thus, if we make sure that P_1 is phase invariant we may omit the phase factors in the expression for P_2 (see also the discussion at the beginning of Sec. IV). On using Eq. (64) the functional kernel (63) can be written in the form

$$T[\psi_1, t|\tilde{\psi}_1, t_0] = \sum_{\alpha,\beta} w_{\alpha\beta} p_\beta \, \delta_1[w_{\alpha\beta}^{-1/2} L_{\alpha\beta}\tilde{\psi}_1 - \psi_1] \quad , \tag{67}$$

where for each pair (α, β) we have introduced the linear operator $L_{\alpha\beta} : \mathcal{H}_1 \longrightarrow \mathcal{H}_1$ defined by

$$L_{\alpha\beta}\tilde{\psi}_1 \equiv \langle\varphi_\alpha|U_I(t, t_0)\tilde{\psi}_1\varphi_\beta\rangle_2 \tag{68}$$

and

$$w_{\alpha\beta} \equiv \|L_{\alpha\beta}\tilde{\psi}_1\|_1^2 \quad . \tag{69}$$

Since

$$\sum_\alpha w_{\alpha\beta} = \sum_\beta w_{\alpha\beta} = 1 \qquad (70)$$

we have

$$\int D\psi_1 D\psi_1^* \, T[\psi_1, t|\tilde\psi_1, t_0] = 1 \quad . \qquad (71)$$

Furthermore, we have $U_I(t_0, t_0) = 1$ and thus

$$\lim_{\tau \to 0} w_{\alpha\beta} = \delta_{\alpha\beta} \quad . \qquad (72)$$

It follows from this equation that

$$\lim_{\tau \to 0} T[\psi_1, t|\tilde\psi_1, t_0] = \delta_1[\tilde\psi_1 - \psi_1] \quad . \qquad (73)$$

At this stage we are ready to state precisely the Markov assumption that allows us to derive a closed equation for the reduced system dynamics. Recall that any stochastic process is defined by specifying the infinite hierarchy of joint probability distributions (see, e.g., [2]). The Markov approximation then means that this hierarchy of joint probability distributions can be expressed completely in terms of the (one-time) probability distribution and the (two-time) conditional transition probability.

In the present case, the Markov assumption means that the time-dependent wave function $\psi_1(x_1, t)$ referring to the subsystem S_1 becomes a stochastic process (in the interaction representation) that is completely defined in terms of its probability distribution $\tilde P_1[\psi_1, t]$ and the conditional transition probability. In view of Eq. (62) it is precisely the functional kernel $T[\psi_1, t|\tilde\psi_1, t_0]$ that is to be interpreted as this conditional transition probability of the stochastic process (in the interaction picture). Thus $T[\psi_1, t|\tilde\psi_1, t_0]$, as given in Eq. (67), represents the conditional probability density for a transition from $\tilde\psi_1$ at time t_0 to ψ_1 at time $t = t_0 + \tau$ under the condition that at time t_0 the state $\tilde\psi_1$ is given. Equation (71) means that the total probability for a transition to any state is equal to 1 for all time intervals τ, whereas Eq. (73) expresses that for $\tau = 0$ the state $\tilde\psi_1$ is prescribed.

The transition probability of any stochastic Markov process obeys the Chapman-Kolmogorov equation. Under certain conditions regarding the short time behavior of the transition probability the Chapman-Kolmogorov equation can be written as a differential equation, which is the so-called differential Chapman-Kolmogorov equation [23]. We are now seeking for the differential Chapman-Kolmogorov equation for the stochastic process $\psi_1(x_1, t)$. This can be done directly by looking for a differential equation for the probability distribution itself since the transition probability is nothing but the corresponding fundamental solution. In order to find a differential equation for the reduced probability $\tilde P_1[\psi_1, t]$, we shall derive in subsection B the short time behavior of the transition probability $T[\psi_1, t|\tilde\psi_1, t_0]$. To be more precise we assume that there exists a time scale τ that obeys

$$\tau_B \ll \tau \ll \tau_S \quad , \qquad (74)$$

where τ_B is of the order of the bath correlation time and τ_S is of the order of the relaxation time of the system. As will be shown in Sec. III B we then obtain within second-order perturbation theory and to first order (on the time scale of the system dynamics) in the time interval τ

$$\Delta\tilde P_1 \equiv \tilde P_1[\psi_1, t_0 + \tau] - P_1[\psi_1, t_0] = \tau\mathcal{A}P_1[\psi_1, t_0] \quad , \quad (75)$$

where $\mathcal{A}$ is a (time-independent) linear functional operator which has the form of a Liouville master operator [see Eq. (99) in Sec. III B].

Step 3: Transforming to the Schrödinger representation

The final step consists of transforming Eq. (75) to the Schrödinger representation. To this end, we express the Schrödinger picture probability distribution P_1 as

$$P_1[\psi_1, t_0 + \tau] = \tilde P_1[e^{iH_1\tau}\psi_1, t_0 + \tau] \qquad (76)$$

from which we obtain, to first order in τ,

$$\begin{aligned}
\Delta P_1 &\equiv P_1[\psi_1, t_0 + \tau] - P_1[\psi_1, t_0] \\
&= \tau\frac{d}{ds}\tilde P_1[e^{iH_1 s}\psi_1, t_0]|_{s=0} + \tau\mathcal{A}P_1[\psi_1, t_0] \\
&= i\tau\int dx_1 \left\{\frac{\delta P_1}{\delta\psi_1}H_1\psi_1 - \psi_1^* H_1\frac{\delta P_1}{\delta\psi_1^*}\right\} \\
&\quad + \tau\mathcal{A}P_1[\psi_1, t_0] \quad .
\end{aligned} \qquad (77)$$

Dividing this equation by τ and performing the limit $\tau \longrightarrow 0$ immediately yields the differential Chapman-Kolmogorov equation for the reduced system dynamics. Thus it remains to derive the structure of the linear functional operator $\mathcal{A}$ from the short time behavior of the transition probability $T[\psi_1, t|\tilde\psi_1, t_0]$. This will be done in the next subsection, where also the final form of the Liouville master equation will be given [see Eq. (101)].

B. Short time behavior of the transition probability and derivation of the Liouville master equation

In this subsection we shall derive in second-order perturbation theory the short time behavior of the transition probability given by Eq. (67). We start by writing the time-evolution operator in the interaction picture in second order in H_I as

$$\begin{aligned}
U_I(t, t_0) = 1 &- i\int_0^\tau ds A(s)B(s) \\
&- \int_0^\tau ds \int_0^{\tau - s} ds' A(s + s')A(s)B(s + s')B(s),
\end{aligned} \qquad (78)$$

where we have defined the interaction picture operators

$$A(s) \equiv e^{iH_1 s}Ae^{-iH_1 s} \, , \quad B(s) \equiv e^{iH_2 s}Be^{-iH_2 s} \quad . \quad (79)$$

For the sake of technical simplicity we assume that the time dependence of $A(s)$ can be neglected over time scales of the order of τ (this assumption ensures that in the quantum master equation, which is derived in Sec. IV, only one Lindblad operator is present). The most general case will be treated in a future paper [25]. Introducing the quantities

$$f_{\alpha\beta}(\tau) \equiv -i \int_0^\tau ds \langle \varphi_\alpha | B(s) | \varphi_\beta \rangle_2, \qquad (80)$$

$$g_{\alpha\beta}(\tau) \equiv -\int_0^\tau ds \int_0^{\tau-s} ds' \langle \varphi_\alpha | B(s+s')B(s) | \varphi_\beta \rangle_2, \qquad (81)$$

we have

$$L_{\alpha\beta}\tilde{\psi}_1 = \left\{ \delta_{\alpha\beta} + f_{\alpha\beta}A + g_{\alpha\beta}A^2 \right\} \tilde{\psi}_1 \quad . \qquad (82)$$

We insert this expression into Eq. (69) and collect the terms of zeroth, first, and second order. This yields

$$w_{\alpha\beta} = \delta_{\alpha\beta} \left\{ 1 - \sum_\gamma |f_{\gamma\beta}|^2 \, ||A\tilde{\psi}_1||_1^2 \right\} + |f_{\alpha\beta}|^2 ||A\tilde{\psi}_1||_1^2, \qquad (83)$$

where we have ensured that the normalization (70) is valid. On using Eq. (83) we find, to leading order in the interaction,

$$w_{\alpha\beta}^{-1/2} L_{\alpha\beta}\tilde{\psi}_1 = \frac{f_{\alpha\beta}}{|f_{\alpha\beta}|} \frac{A\tilde{\psi}_1}{||A\tilde{\psi}_1||_1} \quad \text{for } \alpha \neq \beta \quad , \qquad (84)$$

$$w_{\alpha\alpha}^{-1/2} L_{\alpha\alpha}\tilde{\psi}_1 = \left\{ 1 + \frac{1}{2}||A\tilde{\psi}_1||_1^2 \sum_\gamma |f_{\gamma\alpha}|^2 + g_{\alpha\alpha}A^2 \right\} \tilde{\psi}_1, \qquad (85)$$

where we have assumed for simplicity that $f_{\alpha\alpha} = 0$.

The results (83), (84), and (85) enable us to determine the short time behavior of the transition probability. To this end, we decompose the transition probability T as

$$T = T_{\text{nondiag}} + T_{\text{diag}} \quad , \qquad (86)$$

where

$$T_{\text{nondiag}} \equiv \sum_{\alpha\neq\beta} w_{\alpha\beta}p_\beta\delta_1[w_{\alpha\beta}^{-1/2}L_{\alpha\beta}\tilde{\psi}_1 - \psi_1] \quad , \qquad (87)$$

$$T_{\text{diag}} \equiv \sum_\alpha w_{\alpha\alpha}p_\alpha\delta_1[w_{\alpha\alpha}^{-1/2}L_{\alpha\alpha}\tilde{\psi}_1 - \psi_1] \quad . \qquad (88)$$

Inserting Eqs. (83) and (84) into Eq. (87) we obtain

$$T_{\text{nondiag}} = \sum_{\alpha,\beta} |f_{\alpha\beta}|^2 \, ||A\tilde{\psi}_1||_1^2 \, p_\beta$$
$$\times \delta_1 \left[\frac{f_{\alpha\beta}}{|f_{\alpha\beta}|} \frac{A\tilde{\psi}_1}{||A\tilde{\psi}_1||_1} - \psi_1 \right]. \qquad (89)$$

The argument of the δ functional contains the pure phase factor $f_{\alpha\beta}/|f_{\alpha\beta}|$. Recall that the functional kernel T acts

only upon phase-invariant functionals. In this case the above phase factor has no effect and can therefore be omitted. Thus the argument of the δ functional does not depend upon α and β and the sum in front of the δ functional is just a multiplicative factor. We show in the Appendix that this sum is given approximately by

$$\sum_{\alpha,\beta} |f_{\alpha\beta}|^2 p_\beta \approx \tau\gamma \quad , \qquad (90)$$

where

$$\gamma \equiv \int_{-\infty}^{+\infty} dt \sum_\alpha p_\alpha \langle \varphi_\alpha | B(t)B | \varphi_\alpha \rangle \qquad (91)$$

is the damping time of the system. Therefore, we can write the nondiagonal part of the transition probability as

$$T_{\text{nondiag}} \approx \tau\gamma ||A\tilde{\psi}_1||_1^2 \, \delta_1 \left[\frac{A\tilde{\psi}_1}{||A\tilde{\psi}_1||_1} - \psi_1 \right] \quad . \qquad (92)$$

Equation (88) for the diagonal part of the transition probability can be interpreted in the following way. For each α the transition $\tilde{\psi}_1 \longrightarrow \psi_1 = w_{\alpha\alpha}^{-1/2} L_{\alpha\alpha}\tilde{\psi}_1$ takes place with probability $w_{\alpha\alpha}p_\alpha$. In view of Eq. (85) the size of these transitions is small for small τ [in contrast to the case $\alpha \neq \beta$; see Eq. (84)]. It is thus justified to replace for small τ these transitions by a single transition, which is given by the sum of the transitions weighted with the probabilities p_α,

$$\tilde{\psi}_1 \longrightarrow \psi_1 = \sum_\alpha p_\alpha w_{\alpha\alpha}^{-1/2} L_{\alpha\alpha}\tilde{\psi}_1 \qquad (93)$$

and which happens with probability

$$\sum_\alpha w_{\alpha\alpha}p_\alpha = 1 - \tau\gamma ||A\tilde{\psi}_1||_1^2 \quad . \qquad (94)$$

It is shown in the Appendix that

$$\sum_\alpha g_{\alpha\alpha}p_\alpha \approx -\frac{1}{2}\tau(\gamma + i\gamma') \quad , \qquad (95)$$

where γ is defined in Eq. (91) and γ' is a constant real number. Thus we obtain from Eq. (85)

$$\sum_\alpha p_\alpha w_{\alpha\alpha}^{-1/2} L_{\alpha\alpha}\tilde{\psi}_1$$

$$= \left\{ 1 + \frac{1}{2}\tau\gamma ||A\tilde{\psi}_1||_1^2 - \frac{1}{2}\tau(\gamma + i\gamma')A^2 \right\} \tilde{\psi}_1 \quad . \qquad (96)$$

Inserting these results into Eq. (88) yields

$$T_{\text{diag}} \approx \left(1 - \tau\gamma ||A\tilde{\psi}_1||_1^2 \right)$$
$$\times \delta_1 \left[\left\{ 1 + \frac{1}{2}\tau\gamma ||A\tilde{\psi}_1||_1^2 \right. \right.$$
$$\left. \left. - \frac{1}{2}\tau(\gamma + i\gamma')A^2 \right\} \tilde{\psi}_1 - \psi_1 \right] \quad . \qquad (97)$$

Adding Eqs. (92) and (97) we finally obtain

$$T[\psi_1, t|\tilde{\psi}_1, t_0] \approx \tau\gamma||A\tilde{\psi}_1||_1^2 \, \delta_1 \left[\frac{A\tilde{\psi}_1}{||A\tilde{\psi}_1||_1} - \psi_1\right]$$

$$+ \left(1 - \tau\gamma||A\tilde{\psi}_1||_1^2\right) \delta_1 \left[-\left\{1 + \frac{1}{2}\tau\gamma||A\tilde{\psi}_1||_1^2 - \frac{1}{2}\tau(\gamma + i\gamma')A^2\right\}\tilde{\psi}_1 - \psi_1\right]. \tag{98}$$

This equation represents the short time behavior of the transition probability. It is important to note that Eqs. (71) and (73) remain true for the approximate expression (98).

By virtue of Eq. (98) it is now easy to demonstrate that the infinitesimal generator $\mathcal{A}$ introduced in Eq. (75) is in fact a Liouville master operator. Inserting Eq. (98) into Eq. (62) we obtain to first order in τ

$$\Delta\tilde{P}_1 \equiv \tilde{P}_1[\psi_1, t_0 + \tau] - P_1[\psi_1, t_0] = \tau\mathcal{A}P_1[\psi_1, t_0] = -\frac{\tau}{2} \int dx_1 \left\{\frac{\delta}{\delta\psi_1} \left(\gamma||A\psi_1||_1^2 - (\gamma + i\gamma')A^2\right) \psi_1 P_1[\psi_1, t_0] + \text{c.c.}\right\}$$

$$+ \tau \int D\tilde{\psi}_1 D\tilde{\psi}_1^* \left\{W[\psi_1|\tilde{\psi}_1]P_1[\tilde{\psi}_1, t_0] - W[\tilde{\psi}_1|\psi_1]P_1[\psi_1, t_0]\right\} \quad, \tag{99}$$

where c.c. means complex conjugated and we have introduced the transition functional

$$W[\psi_1|\tilde{\psi}_1] \equiv \gamma||A\tilde{\psi}_1||_1^2 \, \delta_1 \left[\frac{A\tilde{\psi}_1}{||A\tilde{\psi}_1||_1} - \psi_1\right] \quad. \tag{100}$$

Thus we see from Eq. (99) that the linear functional operator $\mathcal{A}$ is a Liouville master operator (see the discussion in Sec. IV).

Finally, we insert Eq. (99) into Eq. (77), divide the whole equation by τ, and perform the limit $\tau \longrightarrow 0$. The result is the Liouville master equation for the probability distribution P_1 of the reduced subsystem S_1. Since any reference to the bath variables has been eliminated, we omit in the following the index 1 from all quantities except from the probability distribution P_1 (in order to remind the reader that it is the *reduced* probability distribution). Furthermore, we write t instead of t_0. The Liouville master equation for the reduced probability distribution P_1 then takes the form

$$\frac{\partial}{\partial t}P_1[\psi, t] = i \int dx \left\{\frac{\delta}{\delta\psi(x)}G(\psi)(x)\right.$$

$$\left. - \frac{\delta}{\delta\psi^*(x)}[G(\psi)]^*(x)\right\}P_1[\psi, t]$$

$$+ \int D\tilde{\psi}D\tilde{\psi}^* \left\{W[\psi|\tilde{\psi}]P_1[\tilde{\psi}, t]\right.$$

$$\left. - W[\tilde{\psi}|\psi]P_1[\psi, t]\right\} \quad. \tag{101}$$

Here the transition functional is defined by

$$W[\psi|\tilde{\psi}] \equiv \gamma||A\tilde{\psi}||^2 \, \delta \left[\frac{A\tilde{\psi}}{||A\tilde{\psi}||} - \psi\right] \tag{102}$$

and we have introduced the nonlinear and non-Hermitian operator $G(\psi)$ given by

$$G(\psi) \equiv H_{\text{eff}}\psi + \frac{i}{2}\gamma||A\psi||^2\psi \quad, \tag{103}$$

where

$$H_{\text{eff}} \equiv H + \frac{\gamma'}{2}A^2 - \frac{i\gamma}{2}A^2 \tag{104}$$

is a non-Hermitian linear operator. Note that according to the above convention H denotes the system Hamiltonian. The Liouville master equation (101) constitutes the central result of this paper.

IV. DISCUSSION

This section is devoted to a detailed discussion of the physical meaning and implications of the Liouville master equation (101) governing the dynamics of the reduced probability distribution $P_1[\psi, t]$ in the Markov approximation. We start by investigating the structure of the Liouville master equation (101). The Liouville part of this equation, which is given by the first line in (101), describes the rate of change of P_1 induced by the flow corresponding to the nonlinear (deterministic) Schrödinger-type equation

$$\frac{\partial\psi}{\partial t} = -iG(\psi) = -iH_{\text{eff}}\psi + \frac{\gamma}{2}||A\psi||^2\psi \quad. \tag{105}$$

It is easily verified that the solution of Eq. (105) corresponding to the initial condition $\psi(0) = \psi_0$ ($||\psi_0|| = 1$) reads

$$\psi(t) = \frac{e^{-iH_{\text{eff}}t}\psi_0}{||e^{-iH_{\text{eff}}t}\psi_0||} \quad. \tag{106}$$

Thus the time evolution of ψ is generated by the non-Hermitian Hamiltonian H_{eff} and the nonlinearity of Eq. (105) has the effect of confining ψ to the unit sphere in Hilbert space. The linear operator H_{eff} [see Eq. (104)] is the sum of the Hermitian Hamiltonian H of the system, the non-Hermitian part $-i\gamma A^2/2$, which causes damping due to the coupling to the heat bath, and an additional Hermitian part $\gamma'A^2/2$, which induces a shift of the energy levels of the system. This shift is referred to as the Lamb shift (see, e.g., [7]).

The master part of our Liouville master equation given

by the second line of Eq. (101) describes the rate of change of P_1, which is due to the discontinuous jumps

$$\tilde{\psi} \longrightarrow \psi = \frac{A\tilde{\psi}}{||A\tilde{\psi}||} \qquad (107)$$

occurring with probability per unit time given by

$$\{\text{rate of transition } \tilde{\psi} \longrightarrow \psi\} = \gamma ||A\tilde{\psi}||^2 \quad . \qquad (108)$$

Obviously, the master part in (101) has the typical structure of a gain-and-loss equation for the probability density P_1 of each state ψ: The quantity $\int D\tilde{\psi} D\tilde{\psi}^* W[\psi|\tilde{\psi}] P_1[\psi, t]$ describes the probability density per unit time for a transition from the state ψ into any other state, whereas $\int D\tilde{\psi} D\tilde{\psi}^* W[\psi|\tilde{\psi}] P_1[\tilde{\psi}, t]$ represents the probability density per unit time for a transition from any state into the state ψ. We emphasize that Eq. (101) is a Liouville master equation in the sense of classical probability theory and *not* an equation for the density matrix. Furthermore, Eq. (101) should be carefully distinguished from the Pauli master equation [7,8], which is an equation for the diagonal part of the relevant statistical operator (in the sense of projection operator techniques; see, e.g., [26,27]).

Summarizing, a typical trajectory $\psi(t)$ of the stochastic process defined by our Liouville master equation can be described as follows: The deterministic time evolution of $\psi(t)$ according to the nonlinear Schrödinger equation (105) is interrupted by discontinuous jumps (107), which happen with probability per unit time given by Eq. (108).

Let us briefly check whether the Liouville master equation preserves the three basic conditions formulated in Sec. II. Of course, the normalization of P_1 is conserved as it is for any differential Chapman-Kolmogorov equation. Also condition (7) holds for all times. This is obvious from the fact that both the deterministic part of the time evolution according to the nonlinear Schrödinger equation (105) and the transitions (107) conserve the norm of the wave function and therefore the dynamics is confined to the unit sphere in Hilbert space. Finally, the phase invariance expressed by condition (8) is preserved under the time evolution. This follows from the fact that $P_1'[\psi, t] \equiv P_1[e^{i\chi}\psi, t]$ is a solution of the Liouville master equation if $P_1[\psi, t]$ is a solution of this equation.

Thus it has been shown that the reduced dynamics of an open quantum system yields, employing the basic postulates formulated in Sec. II and the Markov approximation, a well-defined and unique stochastic jump process in the space of rays of the underlying Hilbert space. It is important to note that the realizations of the stochastic process defined by our Liouville master equation are very similar to those generated by the piecewise deterministic quantum jump methods [13–17]. Obviously, the transitions (107) and the corresponding transition rates are the same at least for the case of only one Lindblad operator, which has been studied here. As far as the continuous part of the evolution is concerned, the above mentioned stochastic methods work with a linear time evolution generated by the non-Hermitian Hamiltonian

H_{eff}. Since we have confined the dynamics to the unit sphere in Hilbert space our continuous time evolution is of course nonlinear. The difference between both representations is thus merely a different normalization of the wave function [see Eq. (106)].

The dynamic equation of the quantum state diffusion model [10,11] is a nonlinear stochastic Schrödinger-type equation, which in turn is equivalent to a certain functional Fokker-Planck equation for the corresponding probability distribution on Hilbert space [12]. The connection of the Liouville master equation derived above to this representation in terms of a stochastic Schrödinger-type equation can be established by performing the diffusion-noise approximation [2] of the Liouville master equation. An example for such a diffusion approximation is given in Ref. [18].

As is well known from the general theory of stochastic processes, the formulation of a stochastic Markov process by means of the corresponding differential Chapman-Kolmogorov equation immediately leads to the whole set of equations of motion for the n-point correlation functions of the stochastic variables. Since the Liouville master equation (101) is nonlinear it is to be expected that, in general, the dynamic equations for the correlation functions of the stochastic process $\psi(x, t)$ are not closed and form a coupled hierarchy of equations (as it is the case, e.g., for a master equation formulation of turbulence [28]). However, as will be shown next the stochastic process defined by our Liouville master equation has the interesting property that the equation of motion for its two-point correlation function

$$
\begin{aligned}
\rho_t(x, x') &\equiv \langle \psi(x)\psi^*(x') \rangle \\
&= \int D\psi D\psi^* \psi(x)\psi^*(x') P_1[\psi, t] \qquad (109)
\end{aligned}
$$

is closed and identical to the general Lindblad form [29,30] for the equation of motion of the statistical operator. In order to prove this property we differentiate Eq. (109) with respect to time and invoke the Liouville master equation to express the time derivative of P_1. The rate of change of ρ_t is then found to be the sum of two parts. The first part describes the rate of change that is due to the smooth motion according to the nonlinear Schrödinger equation (indicated by an index $\mathcal{L}$) and can be written as

$$
\left. \frac{\partial}{\partial t} \right|_{\mathcal{L}} \langle \psi(x)\psi^*(x') \rangle
$$

$$
\begin{aligned}
&= -i[H_{\text{eff}}\langle \psi(x)\psi^*(x') \rangle - \langle \psi(x)\psi^*(x') \rangle H_{\text{eff}}^\dagger] \\
&\quad + \gamma \langle ||A\psi||^2 \psi(x)\psi^*(x') \rangle.
\end{aligned}
$$

The second part represents the rate of change of (109) that is induced by the discrete jumps (indicated by an index $\mathcal{M}$). This part takes the form

$$
\left. \frac{\partial}{\partial t} \right|_{\mathcal{M}} \langle \psi(x)\psi^*(x') \rangle
$$

$$
= \gamma A \langle \psi(x)\psi^*(x') \rangle A - \gamma \langle ||A\psi||^2 \psi(x)\psi^*(x') \rangle.
$$

Adding both parts we see that the four-point correlation functions on the right-hand sides cancel and we obtain a closed equation of motion for the two-point correlation function:

$$\frac{\partial}{\partial t}\langle\psi(x)\psi^*(x')\rangle$$

$$= -i[H_{\text{eff}}\langle\psi(x)\psi^*(x')\rangle - \langle\psi(x)\psi^*(x')\rangle H_{\text{eff}}^\dagger]$$
$$+\gamma A\langle\psi(x)\psi^*(x')\rangle A \quad .$$

In view of definition (109) this equation is identical to

$$\frac{\partial}{\partial t}\rho_t = -i\left[H + \frac{\gamma'}{2}A^2, \rho_t\right]$$
$$+\gamma\left(A\rho_t A - \frac{1}{2}A^2\rho_t - \frac{1}{2}\rho_t A^2\right) \quad . \qquad (110)$$

This equation, which is written in the general Lindblad form, is identical to the so-called quantum master equation [8,31] for the statistical operator for the open system under study (see, e.g., [7]). Thus we have demonstrated that the quantum master equation for the reduced system dynamics of an open quantum system can be derived as the equation of motion for the two-point correlation function of the stochastic process defined by our Liouville master equation. The fact that only one Lindblad operator A is present in Eq. (110) is due to our special ansatz (53). It is, however, easy to generalize the derivation presented in Sec. III to include the general case of any number of Lindblad operators. This more technical derivation will be given in a future paper [25].

V. SUMMARY

Let us summarize the reasoning and the main results of this paper. The ensemble definition (1) for the statistical operator naturally leads to the introduction of a probability distribution $P[\psi]$ on Hilbert space. Since according to quantum mechanics a state is described by a ray in Hilbert space, we made more precise this idea by constructing probability distributions on the space of rays, that is, on projective Hilbert space. Within this formulation the wave function ψ becomes a random variable and the statistical operator appears as the two-point correlation function of ψ. For closed systems the different members of the ensemble evolve according to Schrödinger's equation and consequently the dynamics of the ensemble is formulated in terms of a Liouville equation, which is a first-order functional equation for the time-dependent probability distribution $P[\psi, t]$.

When dealing with an open system that is considered to be a part of some larger system two important constructions have to be introduced. First, it is important to define precisely the probability distribution for a combined system S that is made up of two statistically independent subsystems S_1 and S_2, i.e., to define the probability distribution on the tensor product of the Hilbert spaces of the subsystems. Second, given a probability distribution on the total Hilbert space it is to be clarified

what is meant by the reduced probability distribution on the Hilbert space of one of the subsystems. The corresponding equations for these constructions have been worked out in Sec. II.

On the basis of the general formal setting developed in Sec. II it was possible in Sec. III to derive the differential Chapman-Kolmogorov equation, which governs the dynamics of open quantum systems within the Markov approximation. This was done by analyzing, in second-order perturbation theory, the short time behavior of the transition probability, i.e., the conditional probability density for the transitions among the rays in Hilbert space. It turns out that the differential Chapman-Kolmogorov equation has in fact the form of a Liouville master equation. The Liouville part of this equation describes the continuous flow of a nonlinear, dissipative, and norm-conserving Schrödinger equation. The master part defines a discontinuous stochastic jump process. Finally, we have shown that the equation of motion for the two-point correlation function of the stochastic process defined by the Liouville master equation is closed and exactly of the form of the quantum master equation for the reduced density operator.

Employing a formulation of ensembles of quantum systems in terms of probability distributions on the underlying projective Hilbert space, we have shown that the dynamics of open quantum systems can be described by a unique classical stochastic process on projective Hilbert space. This has been achieved by starting from a general microscopic description of the interaction between system and reservoir. Since the realizations of the stochastic process correspond to the trajectories generated by the Monte Carlo wave function simulation method and by the quantum trajectory method with jump-type evolution, the present derivation confirms the validity of these approaches and shows how to formulate them within the mathematical formalism of probability theory.

APPENDIX: PROOF OF EQS. (90) AND (95)

In order to prove Eq. (90) we use Eq. (80) to find

$$\sum_{\alpha,\beta}|f_{\alpha\beta}|^2 p_\beta = \int_0^\tau ds \int_0^\tau ds' \langle B(s)B(s')\rangle \quad , \qquad (A1)$$

where

$$\langle B(s)B(s')\rangle \equiv \sum_\alpha p_\alpha\langle\varphi_\alpha|B(s)B(s')|\varphi_\alpha\rangle \qquad (A2)$$

denotes the correlation function of B obtained by tracing over the bath variables. By virtue of the invariance of the bath under the time evolution generated by H_2 we have

$$\langle B(s)B(s')\rangle = \langle B(s-s')B\rangle \qquad (A3)$$

and therefore

$$\sum_{\alpha,\beta} |f_{\alpha\beta}|^2 p_\beta = \int_0^\tau ds \int_0^\tau ds' \langle B(s-s')B \rangle$$

$$= \int_0^\tau dt' \int_{-t'}^{\tau-t'} dt \langle B(t)B \rangle \quad , \tag{A4}$$

where we have transformed to new time variables $t = s - s'$, $t' = s'$. Since $\tau \gg \tau_B$ the correlation function vanishes very fast outside a small strip along the t' axis. Thus we can approximate

$$\sum_{\alpha,\beta} |f_{\alpha\beta}|^2 p_\beta \approx \int_0^\tau dt' \int_{-\infty}^{+\infty} dt \langle B(t)B \rangle = \tau\gamma \quad , \tag{A5}$$

and Eq. (90) is proved. Similarly, we find

$$\sum_\alpha g_{\alpha\alpha} p_\alpha = - \int_0^\tau dt' \int_0^{\tau-t'} dt \langle B(t)B \rangle \quad . \tag{A6}$$

On using the same argument as above we obtain

$$\sum_\alpha g_{\alpha\alpha} p_\alpha \approx - \int_0^\tau dt' \int_0^{+\infty} dt \langle B(t)B \rangle$$

$$= -\tau \int_0^{+\infty} dt \langle B(t)B \rangle \quad . \tag{A7}$$

We have

$$\int_0^{+\infty} dt \langle B(t)B \rangle = \frac{1}{2}(\gamma + i\gamma') \quad , \tag{A8}$$

where $\gamma'/2$ denotes the imaginary part of this integral. Thus we finally obtain

$$\sum_\alpha g_{\alpha\alpha} p_\alpha \approx -\frac{1}{2}\tau(\gamma + i\gamma') \quad , \tag{A9}$$

which is Eq. (95).

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