

Is the dynamics of open quantum systems always linear?

Karen M. Fonseca Romero,* Peter Talkner, and Peter Hänggi

Institut für Physik, Universität Augsburg, Universitätsstrasse 1, D 86315 Augsburg, Germany

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We study the influence of the preparation of an open quantum system on its reduced time evolution. In contrast to the frequently considered case of an initial preparation where the total density matrix factorizes into a product of a system density matrix and a bath density matrix the time evolution generally is no longer governed by a linear map nor is this map affine. Put differently, the evolution is truly nonlinear and cannot be cast into the form of a linear map plus a term that is independent of the initial density matrix of the open quantum system. As a consequence, the inhomogeneity that emerges in formally exact generalized master equations is in fact a nonlinear term that vanishes for a factorizing initial state. The general results are elucidated with the example of two interacting spins prepared at thermal equilibrium with one spin subjected to an external field. The second spin represents the environment. The field allows the preparation of mixed density matrices of the first spin that can be represented as a convex combination of two limiting pure states, i.e., the preparable reduced density matrices make up a convex set. Moreover, the map from these reduced density matrices onto the corresponding density matrices of the total system is affine only for vanishing coupling between the spins. In general, the set of the accessible total density matrices is nonconvex.

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I. INTRODUCTION

Closed systems are known to be an idealization. In general, real systems interact with their environment and exhibit properties that cannot be observed in finite closed systems, such as irreversibility of the time evolution and, related, the relaxation of observables toward stationary values and dephasing, or decoherence. Various techniques have been developed to treat the dynamics of open systems without explicitly considering the full Hamiltonian dynamics [1]. For example, effective equations for the reduced density matrix of the considered open system, known as master equations, have been proposed long ago [2–4] and still are of considerable interest because of their conceptual simplicity and potential usefulness [5,6]. New challenges in this field of fundamental physics have come from nanotechnology [7] and quantum computing [8].

Any equation determining the time evolution of a density matrix has to obey several general properties which guarantee that the density matrix stays self-adjoint, positive, and normalized in the course of time. These general properties still leave much freedom and, in order to further restrict possible dynamical laws, additional requirements for the dynamics have been postulated [9]. One seemingly natural property that often is assumed without even being mentioned is the *linearity* of the time evolution, which generally is understood as a consequence of the linearity of the Schrödinger and the Liouville–von Neumann equation for closed systems. This argument, however, only works by analogy and no proof of the necessity of this requirement is available. Just on the contrary Pechukas [10,11] has recently shown that linearity may only hold if the initial state of the total system factorizes into a product of a density matrix for the open system and

another one for the environment, and if a sufficient number of pure states can be prepared. The assumption of linearity is a prerequisite of a Markovian dynamics and of complete positivity [12,13]. These properties then lead to the mathematically well characterized class of Lindblad master equations. From the physical point of view, however, these equations suffer from certain deficiencies [10]. They are restricted to the regime of weak coupling between the considered system and its environment. In particular, the weak-coupling assumption will fail at sufficiently low temperatures [14,15]. Moreover, there are various general statistical-mechanical properties that are incompatible with the assumption of a Markovian dynamics [16].

A few microscopic models of systems interacting with their environment can be exactly reduced to Lindblad master equations with time-dependent coefficients [17,18], thereby describing the single-time non-Markovian reduced dynamics.

The dynamics of an open system is determined by both, the full dynamics of the considered system interacting with its environment and the initial state of the complete system. The significance of the initial state was emphasized in several works [19–27]. In an experiment this initial state is imposed by a preparation procedure. Here we will be concerned only with equilibrium preparations that lead to a thermal equilibrium of the total system in the presence of external fields that only act on the system and that are switched off finally. In this way, the initial state is described by a canonical density matrix:

$$\rho^{\mathbf{F}} = Z^{-1} \exp \left\{ -\beta \left(H - \sum_j F_j X_j \right) \right\}, \quad (1)$$

where β is the inverse temperature, H the Hamiltonian governing the dynamics of the total system, $\mathbf{F} = (F_j)$ are external, i.e., classical, fields, X_j the corresponding conjugate operators of the open system, and $Z = \text{Tr} \exp \{ -\beta (H + \sum_j F_j X_j) \}$ is

*On leave from Departamento de Física, Universidad Nacional, Bogotá, Colombia.

the partition function of the total system. Here, Tr denotes the trace over the total system. In this way, initial states of the total system are reproducibly prepared. The system is in thermal equilibrium with the environment at a given temperature and is additionally constrained by the external fields F_j . The set of density matrices that can be obtained upon variation of the fields forms the equilibrium preparation class. The reduced states belonging to this preparation class are determined by the trace over the environment, which is denoted by Tr_B :

$$\rho_S^F = \text{Tr}_B \rho^F. \quad (2)$$

The calculation of this trace is nontrivial in most cases and in general does not lead to the canonical distribution of the uncoupled system, $Z_S^{-1} \exp\{-\beta H_S\}$ [14,28–30], where H_S is the Hamiltonian of the system in the presence of the external fields and Z_S the respective partition function. This particular form is obtained only in the limit of weak coupling between the system and the environment [3,29,31]. In this limit, the equilibrium density matrix of the total system factorizes into a product of a system and an environment density matrix. This is an example of the factorizing preparation which leads to a product of a particular density matrix of the environment ρ_B and an arbitrary density matrix ρ_S of the system:

$$\rho^{\text{fac}} = \rho_S \rho_B. \quad (3)$$

The factorizing preparation is assumed in most theoretical investigations though it is often difficult, if not impossible, to realize it experimentally.

A more general class of preparations has been suggested in the context of the path-integral approach to open systems [22]:

$$\rho^O = \sum_j O_j \rho^F O_j', \quad (4)$$

where ρ^F is defined as in the equilibrium preparation, Eq. (1), and O_j and O_j' are operators that only act on the system's Hilbert space. For applications of this preparation class we refer the reader to Ref. [22].

The state of the open system after the preparation results from the density matrix of the full system at that time by means of the trace over the environment, i.e.,

$$\rho_S(t) = \text{Tr}_B U(t) \rho U^\dagger(t), \quad (5)$$

where

$$U(t) = \exp\left\{-\frac{i}{\hbar} H t\right\} \quad (6)$$

is the unitary time evolution operator of the full system and $\rho(0) = \rho$ the density matrix resulting from the preparation.

According to Eq. (5), the reduced density matrix at time t is a linear image of the initial *full* density matrix ρ under the successive action of the unitary time evolution of the full system and the operation of the trace. In order to obtain a map from the initial *reduced* density matrix ρ_S to its value at a later time t , we introduce the *blow-up* map $R(\rho_S)$ that as-

signs to each reduced initial density matrix ρ_S one that belongs to the total system:

$$\rho = R(\rho_S). \quad (7)$$

The particular form of the blow-up map depends on the initial preparation of the system. Expressing the initial total density matrix ρ by the reduced initial state with the help of the blow-up map one obtains the reduced time evolution of the system:

$$\rho_S(t) = \text{Tr}_B U(t) R(\rho_S) U^\dagger(t). \quad (8)$$

The reduced time evolution is linear only if the blow-up map is linear.

Taking into account the structure of generalized master equations that may contain an inhomogeneous term one may ask whether an affine time evolution could possibly provide a relevant class of time evolution laws. Any affine time evolution of the reduced density matrix $\rho_S(t)$ can be written as the sum of a term linear in ρ_S and another one independent of ρ_S , i.e.,

$$\rho_S(t) = T(t) \rho_S + I(t), \quad (9)$$

where $T(t)$ is a linear operator and $I(t)$ is independent of ρ_S . So affine time evolutions are apparently more general than linear ones. An affine time evolution as described in Eq. (9) will result only from an affine blow-up map. Hence, one may ask under which conditions the blow-up map is affine.

An equivalent characterization of affine blow-up maps can be given in the following way. Because a density matrix is a positive normalized operator, we require that the blow-up map R acts on a convex set of reduced density matrices, i.e., a set that contains with each pair ρ_{S1} and ρ_{S2} all convex combinations $\lambda \rho_{S1} + (1-\lambda) \rho_{S2}$ for all $0 < \lambda < 1$. We assume next that the considered preparation provides such a convex set of reduced density matrices. For any particular preparation one has to check this property. We then further may ask under which conditions the blow-up map R preserves the convexity condition. Put differently, under which conditions a full density matrix that corresponds to a convex combination of reduced density matrices again is given by a convex combination. Then the preparation class also forms a convex set. For such a convex blow-up map one obtains

$$R[\lambda \rho_{S,1} + (1-\lambda) \rho_{S,2}] = \lambda R(\rho_{S,1}) + (1-\lambda) R(\rho_{S,2}). \quad (10)$$

This then implies that the blow-up map is affine:

$$R(\rho_S) = L \rho_S + \varphi, \quad (11)$$

where L is a linear operator that maps reduced density matrices onto full density matrices and φ is an operator of the full system. A proof of this statement is given in Appendix A.

We note that the mixing parameter λ has to be identical on the left- and on the right-hand side of Eq. (10). This is a consequence of the fact that the trace of $R(\rho_S)$ over the Hilbert space of the environment must coincide with the system density matrix ρ_S :

$$\text{Tr}_B R(\rho_S) = \rho_S. \quad (12)$$

For the factorizing preparation (3) the blow-up map R is always linear. It simply acts as the multiplication by the reference environment density operator ρ_B . In the case of a classical system dynamics the role of density matrices is taken over by probability densities defined on the respective phase space. Then, any preparation can be characterized by a conditional probability density $\rho(\mathbf{x}|\mathbf{x}_S)$ for the state \mathbf{x} of the total system given the state \mathbf{x}_S of the system [19–21]. The corresponding blow-up map R is then given by the multiplication with this conditional probability and, hence, is always linear. No such simple construction scheme is available in quantum mechanics and Pechukas [10] showed that the factorizing preparation is the only one for which the blow-up map is linear, provided that a sufficient number of pure states of the reduced system are contained in the preparation class. For the convenience of the reader, the precise formulation of the theorem and a proof is given in Appendix B.

In the present work, we will consider the influence of the preparation of an open quantum system on its reduced time evolution by means of the simple example of two interacting spins. One of those is considered as the system, the other one plays the role of the environment. The second spin is only a very crude caricature of a true environment which clearly fails to cause dissipation or dephasing in the system because of its finiteness. Nevertheless, it suffices to illustrate the influence of the preparation on the subsequent dynamics of the system.

We assume that the total system starts from an equilibrium preparation, i.e., its initial state is described by a density matrix of the form of Eq. (1). It will be shown that in general this preparation renders the time evolution of the reduced system *nonlinear*.

II. TWO SPINS

Both interacting spins $\boldsymbol{\sigma}_1 = (\sigma_1^x, \sigma_1^y, \sigma_1^z)$ and $\boldsymbol{\sigma}_2 = (\sigma_2^x, \sigma_2^y, \sigma_2^z)$ with Pauli spin matrices σ_α^i , $\alpha=1,2$, $i=x,y,z$, are of total length $s=1/2$. The first spin $\boldsymbol{\sigma}_1$ is considered as the system and the second one $\boldsymbol{\sigma}_2$ takes over the role of the environment. Every density matrix of the total system then assumes the form

$$\rho = \frac{1}{4}(1 + \mathbf{S}_1 \cdot \boldsymbol{\sigma}_1 + \mathbf{S}_2 \cdot \boldsymbol{\sigma}_2 + \mathbf{C} \cdot \boldsymbol{\sigma}_2), \quad (13)$$

where

$$\mathbf{S}_\alpha = \langle \boldsymbol{\sigma}_\alpha \rangle \quad (14)$$

denotes the Bloch vector of the spin $\alpha=1,2$ and the matrix

$$\mathbf{C} = \langle \boldsymbol{\sigma}_1 \boldsymbol{\sigma}_2 \rangle \quad (15)$$

denotes the correlation matrix of the two spins. The dot product denotes the scalar product in three dimensions, e.g., $\mathbf{S}_1 \cdot \boldsymbol{\sigma}_1 = S_1^x \sigma_1^x + S_1^y \sigma_1^y + S_1^z \sigma_1^z$. The reduced density matrix of the system is given by the trace over the environment (i.e., the second spin), and hence becomes

$$\rho_S = \text{Tr}_2 \rho = \frac{1}{2}(1 + \mathbf{S}_1 \cdot \boldsymbol{\sigma}_1). \quad (16)$$

Here we want to study the opposite direction, that is, to go from ρ_S to ρ . In particular, we look for conditions under

which the respective blow-up map $R(\rho_S) = \rho$ is convex. We recall that the blow-up map is determined by the preparation process of the system. In the process of a preparation the state of the system is controlled by external fields \mathbf{F} that ideally act only on the system, as given in Eq. (1) for the equilibrium preparation. For the considered spin one can think of static magnetic fields. For interacting spins, the two Bloch vectors and the correlation matrix will depend on the applied magnetic field.

We next investigate the requirement of the convexity of the blow-up map R . A necessary condition for this property to hold is the convexity of the domain of definition of R . This implies that for any pair of reduced density matrices ρ_S^k , $k=1,2$ that result from two different values of the field, all convex linear combinations can be prepared by means of at least one other value \mathbf{F}_3 of the field:

$$\rho_S^3 = \lambda \rho_S^1 + (1-\lambda) \rho_S^2, \quad (17)$$

with \mathbf{F}_3 depending on the fields \mathbf{F}_1 , \mathbf{F}_2 , and λ . Using the general form of the reduced density matrix in Eq. (16) one finds a respective equation for the Bloch vectors, reading

$$\mathbf{S}_1(\mathbf{F}_3) = \lambda \mathbf{S}_1(\mathbf{F}_1) + (1-\lambda) \mathbf{S}_1(\mathbf{F}_2). \quad (18)$$

Note that for the equilibrium preparation $\mathbf{S}_1(\mathbf{F})$ is a uniquely defined function of the field \mathbf{F} . This is the case because the derivatives of the Bloch vector components with respect to the field components form the susceptibility matrix, which is an invertible matrix in thermal equilibrium. Hence, $\mathbf{S}_1(\mathbf{F})$ has a uniquely defined inverse $\mathbf{F}(\mathbf{S}_1)$.

We study the further consequences of the convexity condition (10) of the blow-up map for the spin system in the equilibrium preparation. Using Eqs. (17) and (13) for the total density matrix of the full system in Eq. (10) one finds the following relations for both the Bloch vector of the second spin and for the correlation matrix:

$$\mathbf{S}_2(\mathbf{F}_3) = \lambda \mathbf{S}_2(\mathbf{F}_1) + (1-\lambda) \mathbf{S}_2(\mathbf{F}_2),$$

$$\mathbf{C}(\mathbf{F}_3) = \lambda \mathbf{C}(\mathbf{F}_1) + (1-\lambda) \mathbf{C}(\mathbf{F}_2). \quad (19)$$

Here \mathbf{F}_3 is defined by Eq. (18). The nontrivial conditions (19) in general will not be satisfied. Expressing next the fields \mathbf{F}_i by those Bloch vectors $\mathbf{S}_1^i = \mathbf{S}_1(\mathbf{F}_i)$ that result for the respective fields we find, by use of Eq. (18), the following relations for the Bloch vectors of the second spin and the correlation matrix:

$$\mathbf{S}_2[\lambda \mathbf{S}_1^1 + (1-\lambda) \mathbf{S}_1^2] = \lambda \mathbf{S}_2[\mathbf{S}_1^1] + (1-\lambda) \mathbf{S}_2[\mathbf{S}_1^2],$$

$$\mathbf{C}[\lambda \mathbf{S}_1^1 + (1-\lambda) \mathbf{S}_1^2] = \lambda \mathbf{C}[\mathbf{S}_1^1] + (1-\lambda) \mathbf{C}[\mathbf{S}_1^2]. \quad (20)$$

Hereby we introduced the notation $\mathbf{S}_2[\mathbf{S}_1] = \mathbf{S}_2(\mathbf{F})$ and $\mathbf{C}[\mathbf{S}_1] = \mathbf{C}(\mathbf{F})$ where the dependence on the external field \mathbf{F} is expressed by the corresponding, equivalent value of the Bloch vector $\mathbf{S}_1 = \mathbf{S}_1(\mathbf{F})$. From these equations it follows that both $\mathbf{S}_2[\mathbf{S}_1]$ and $\mathbf{C}[\mathbf{S}_1]$ are affine functions, see Appendix A. Therefore they can be represented as

$$\mathbf{S}_2[\mathbf{S}_1] = \mathbf{A} \cdot \mathbf{S}_1 + \mathbf{B},$$

$$\mathbf{C}[\mathbf{S}_1] = \mathbf{D} \cdot \mathbf{S}_1 + \mathbf{E}, \quad (21)$$

where \mathbf{B} is a constant vector, \mathbf{A} and \mathbf{E} are constant, second-order tensors, and \mathbf{D} is a constant third-order tensor. Hence, these quantities must neither depend on the applied field \mathbf{F} nor on the Bloch vector \mathbf{S}_1 .

III. AN EXPLICIT ILLUSTRATION

We consider the equilibrium preparation for the following simple two-spin Hamiltonian as an example,

$$H = -F_z \sigma_1^z + e \sigma_2^z + g \sigma_1^x \sigma_2^x. \quad (22)$$

Here we allow for a field only in the z direction. The two spins interact by their x components. We study the equilibrium preparation class that results if the field F_z assumes all possible values, $-\infty < F_z < \infty$ at the fixed inverse temperature β :

$$\rho^{F_z} = Z^{-1} \exp\{-\beta H\}. \quad (23)$$

Because the Hamiltonian H commutes with the operator $\sigma_1^z \sigma_2^z$ one can diagonalize H in the eigenspaces of $\sigma_1^z \sigma_2^z$. This diagonalization then yields the four eigenvalues \mathcal{E}_i ,

$$\begin{aligned} \mathcal{E}_1 &= -\sqrt{(F_z - e)^2 + g^2}, & \mathcal{E}_2 &= \sqrt{(F_z - e)^2 + g^2}, \\ \mathcal{E}_3 &= -\sqrt{(F_z + e)^2 + g^2}, & \mathcal{E}_4 &= \sqrt{(F_z + e)^2 + g^2}, \end{aligned} \quad (24)$$

and the corresponding eigenprojection operators P_i ,

$$P_i = \frac{1}{4} \left(1 + \sigma_1^z \sigma_2^z - \frac{F_z - e}{\mathcal{E}_i} (\sigma_1^z + \sigma_2^z) + \frac{g}{\mathcal{E}_i} (\sigma_1^x \sigma_2^x - \sigma_1^y \sigma_2^y) \right), \quad (25)$$

$i = 1, 2,$

$$P_i = \frac{1}{4} \left(1 - \sigma_1^z \sigma_2^z - \frac{F_z + e}{\mathcal{E}_i} (\sigma_1^z - \sigma_2^z) + \frac{g}{\mathcal{E}_i} (\sigma_1^x \sigma_2^x + \sigma_1^y \sigma_2^y) \right), \quad (26)$$

$i = 3, 4,$

such that $H = \sum_i \mathcal{E}_i P_i$ holds. The canonical density matrix ρ^{F_z} at the inverse temperature β is a mixture of the pure states P_i with the Boltzmann weights $p_i = \exp\{-\beta \mathcal{E}_i\} / [2\{\cosh(\beta \mathcal{E}_1) + \cosh(\beta \mathcal{E}_3)\}]$, i.e.,

$$\begin{aligned} \rho^{F_z} &= \sum_{i=1}^4 p_i P_i = \frac{1}{4} (1 + S_{1z} \sigma_1^z + S_{2z} \sigma_2^z + C_{xx} \sigma_1^x \sigma_2^x + C_{yy} \sigma_1^y \sigma_2^y \\ &\quad + C_{zz} \sigma_1^z \sigma_2^z). \end{aligned} \quad (27)$$

The x and y components of the two Bloch vectors vanish. The nonvanishing z components read

$$\begin{aligned} S_{1z} &= \beta [F_z \mathcal{F}_+(\beta \mathcal{E}_1, \beta \mathcal{E}_3) - e \mathcal{F}_-(\beta \mathcal{E}_1, \beta \mathcal{E}_3)], \\ S_{2z} &= \beta [F_z \mathcal{F}_-(\beta \mathcal{E}_1, \beta \mathcal{E}_3) - e \mathcal{F}_+(\beta \mathcal{E}_1, \beta \mathcal{E}_3)], \end{aligned} \quad (28)$$

where the auxiliary functions $\mathcal{F}_\pm(x, y)$ are defined by

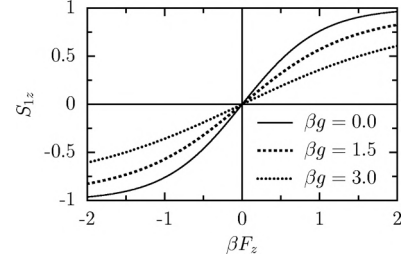


FIG. 1. The Bloch vector component S_{z1} as a function of the field βF_z resulting from Eq. (28) for $\beta e = 1$. With increasing coupling parameter $\beta g = 0.5, 1, 1.5$ the slope at $F_z = 0$ decreases. Apparently, the functions $S_{z1}(F)$ are monotonic and hence possess a unique inverse.

$$\mathcal{F}_\pm(x, y) = \frac{y \sinh(x) \pm x \sinh(y)}{xy [\cosh(x) + \cosh(y)]}. \quad (29)$$

Finally, the nonvanishing elements of the correlation matrix \mathbf{C} are given by

$$\begin{aligned} C_{xx} &= -\beta g \mathcal{F}_+(\beta \mathcal{E}_1, \beta \mathcal{E}_3), \\ C_{yy} &= \beta g \mathcal{F}_-(\beta \mathcal{E}_1, \beta \mathcal{E}_3), \\ C_{zz} &= \frac{\cosh(\beta \mathcal{E}_1) - \cosh(\beta \mathcal{E}_3)}{\cosh(\beta \mathcal{E}_1) + \cosh(\beta \mathcal{E}_3)}. \end{aligned} \quad (30)$$

As already stated above, in the present case of the equilibrium preparation the z component of the Bloch vector of the first spin is a uniquely invertible function of the magnetic field F_z . This can be shown by inspection from Eq. (29), see also Fig. 1. We note that the equilibrium preparation contains the pure system states $\rho_S = \frac{1}{2}(1 \pm \sigma_z)$ asymptotically in the infinite field limit $F_z \rightarrow \pm \infty$. If one also took into account fields that couple to the other spin components σ_x, σ_y , the eigenstates of these components could also be prepared asymptotically. In the case studied here, however, no other pure states than the eigenstates of σ_z can be prepared. Thus, for this particular case one of the conditions of the Pechukas theorem to hold is not met.

A. Testing convexity of the blow-up map

We now come to the discussion of Eqs. (21) which are necessary conditions for the blow-up map to be convex. Because in the considered preparation class only the z component of the field is varied and because of the symmetries of the considered Hamiltonian, these equations need only be checked for the Bloch vector component S_{z2} and the diagonal elements of the correlation matrix, respectively as functions of S_{z1} :

$$S_{z2}[S_{z1}] = A S_{z1} + B,$$

$$C_{xx}[S_{z1}] = D_{xx} S_{z1} + E_{xx},$$

$$C_{yy}[S_{z1}] = D_{yy} S_{z1} + E_{yy},$$

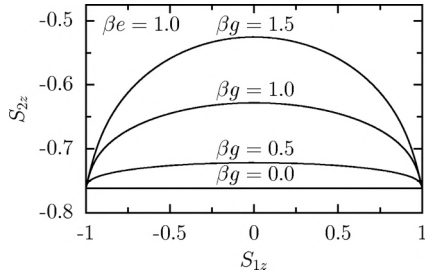


FIG. 2. The Bloch vector component S_{22} as a function of S_{z1} resulting from Eq. (28) for $\beta e=1$ and different values $\beta g = 0, 0.5, 1, 1.5$. Note that a strictly linear dependence results only for the case of vanishing coupling $g=0$. An approximate linear regime exists in a neighborhood of $S_{1z}=0$.

$$C_{zz}[S_{z1}] = D_{zz}S_{z1} + E_{zz}. \quad (31)$$

If the blow-up map were convex these equations would have to result from Eqs. (28) and (30) by eliminating the external field F_z . There is no need to perform the lengthy calculation to see that the above equations hold only if the system-environment interaction is absent, i.e., if the coupling constant g in the Hamiltonian vanishes. We note that with $\mathcal{E}_1(-F_z) = \mathcal{E}_3(F_z)$ and the symmetries of the auxiliary functions $\mathcal{F}_\pm(x, y) = \pm \mathcal{F}_\pm(y, x)$, S_{1z} becomes an odd function whereas C_{xx} is an even function of F_z . If Eq. (31) was to hold, D_{xx} would have to vanish and C_{xx} would have to be a constant. This is true if and only if $g=0$. Figures 2 and 3 depict the dependences of S_{22} and of the correlation functions, respectively, on S_{z1} for finite coupling strengths. In all cases, except for $g=0$, and for the correlation C_{zz} , the deviations from linearity are strikingly obvious. We note, however, that for small values of the Bloch vector component S_{1z} the component S_{22} and correlation C_{xx} are almost constant and the other correlations C_{zz} and C_{yy} are linear. This behavior is in accordance with a convex blow-up map. Below we will come back to the blow-up in the linear-response regime when the external fields are small.

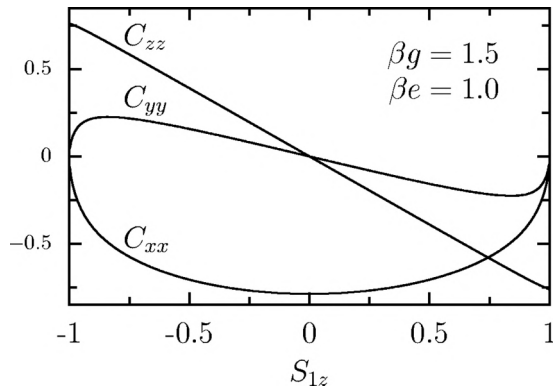


FIG. 3. The nonvanishing correlation functions C_{xx}, C_{yy}, C_{zz} as a function of the Bloch vector component S_z for $\beta e=1$ and $\beta g=1.5$. The correlation functions C_{xx} and C_{yy} clearly deviate from straight lines following from a convex blow-up map, see Eq. (31). For the correlation functions also an approximately linear regime exists in a neighborhood of $S_{1z}=0$.

IV. DISCUSSION, IMPLICATIONS, AND CONCLUSIONS

We have illustrated Pechukas' verdict on the linear time evolution of open quantum systems [10] by a simple example. Moreover, we have demonstrated that the theorem holds for affine time evolutions: If two complete sets of pure system states can be prepared this more general class of evolution implies a factorizing preparation of the total initial density matrix where the environment density matrix must be independent of the system density matrix. Actually, Pechukas' original proof [10] applies as well in the affine case. Nowhere in the proof he made explicit use of the homogeneity condition, i.e., $R(\lambda\rho_S) = \lambda R(\rho_S)$, λ real, that would render an affine R a linear map.

At first, this may only seem a modest generalization of the original conclusion but it sheds some light on the crucial role of the inhomogeneity term of (generalized) master equations which appears when the initial density matrix does not factorize. The present analysis excludes that this term is *independent* of the initial reduced density matrix. This term, actually is not merely an inhomogeneity of the otherwise linear master equation but must depend on the reduced density matrix in a *nonlinear way*. In the cases, in which a Markovian dynamics is approached for long times this nonlinear term must vanish for sufficiently large times. It will do so, however, in a characteristic manner that depends on the particular initial reduced density matrix.

In the present example only one set of pure system states, the eigenstates of σ_z , are preparable and yet the affinity of the blow-up map implies a factorizing preparation. We note that in general the condition on the number of preparable pure states of the Pechukas theorem cannot be relaxed. An example is provided by the work of Karleim and Grabert [27] who considered a harmonic oscillator coupled to a bath of harmonic oscillators with a nonfactorizing thermal preparation. This preparation allows for pure position states and still leads to a linear master equation.

Another example of a nonfactorizing preparation that leads to a linear time evolution results from the following preparation procedure: (i) start with a factorizing density matrix $\rho_{S0}\rho_{B0}$ at a time $t=-t_0$, $t_0 > 0$, (ii) turn on the interaction between the system and the bath, and (iii) use the density matrix that has evolved at $t=0$ as a result of the preparation. We term this preparation the *factorize-and-wait* preparation. The blow-up map $R(\rho_S)$ then assumes the form

$$R(\rho_S) = e^{-(i\hbar)Ht_0} G_{t_0}^{-1}(\rho_S) \rho_{B0} e^{(i\hbar)Ht_0}, \quad (32)$$

where G_t is the linear propagator of the reduced density matrix for the factorizing preparation, i.e.,

$$\rho_S(t) = G_t(\rho_S(0)) = \text{Tr}_B e^{-(i\hbar)Ht} \rho_S(0) \rho_{B0} e^{(i\hbar)Ht}. \quad (33)$$

Here, the inverse of the propagator G_t is needed in order to infer the proper system part of the factorizing density at $t=-t_0$ from the density matrix ρ_S that is to be prepared at $t=0$. In view of possible fast relaxation processes and the buildup of system-bath correlations [23,24,26] after the interaction has been switched on, this inverse propagator will not be defined on the total set of possible density matrices [32]. Still, it is a linear operator and thus the blow-up map (32)

also is linear. It is obvious, however, that no pure states of the system can be prepared at time $t=0$ in this way, i.e., the factorize-and-wait preparation does not provide pure states. Therefore, the conditions for the Pechukas theorem are not met and the theorem is thus not in conflict with the linearity of the blow-up map of the entangled factorize-and-wait preparation.

Finally, we discuss the equilibrium preparation in the limit of weak external forces. In the region close to thermal equilibrium, Mori's generalized quantum Langevin equations [33] provide a proper description of the time evolution of the set of system operators that couple to the external fields. In this case, the preparable density matrices of the total system follow from Eq. (1) in linear approximation in the external fields F_j , and hence assume the linear-response form [34]

$$\rho^F = \rho^0 + \beta \sum_i \int_0^1 dx (\rho^0)^{1-x} (X_i - \langle X_i \rangle_0) (\rho^0)^x F_i, \quad (34)$$

where ρ^0 denotes the equilibrium density matrix of the total system in the absence of external fields and $\langle X \rangle_0$ the average of the operator X with respect to the density matrix ρ^0 . The corresponding density matrix of the reduced system $\rho_S^F = \text{Tr}_B \rho^F$ then becomes

$$\rho_S^F = \rho_S^0 + \beta \sum_i \int_0^1 dx \text{Tr}_B (\rho^0)^{1-x} (X_i - \langle X_i \rangle_0) (\rho^0)^x F_i. \quad (35)$$

We recall that the external fields act on the system, i.e., the conjugate operators X_i are system operators. The expectation values of these operators with respect to the density matrix ρ_S^F are linear functions of the external fields by construction and can be written as

$$\langle X_j - \langle X_j \rangle_0 \rangle \equiv \text{Tr}_S (X_j - \langle X_j \rangle_0) \rho_S^F = \sum_i \chi_{ji} F_i, \quad (36)$$

where the response matrix χ_{ij} is obtained by inserting Eq. (34) into the middle term of Eq. (36):

$$\chi_{ij} = \beta \int_0^1 dx \text{Tr} (X_i - \langle X_i \rangle_0) (\rho^0)^{1-x} (X_j - \langle X_j \rangle_0) (\rho^0)^x. \quad (37)$$

Because χ_{ij} is an invertible matrix we may solve Eq. (36) for the external fields F_i ,

$$F_i = \sum_j \chi^{-1}_{ij} \langle X_j - \langle X_j \rangle_0 \rangle. \quad (38)$$

In this way the external fields are expressed in terms of the linear functional $\langle X_j - \langle X_j \rangle_0 \rangle$ of the system density matrix ρ_S^F . Using this relation in Eq. (34) we find for the blow-up map the affine form $R(\rho_S^F) = L\rho_S^F + \varphi$, where

$$L\rho_S^F = \beta \sum_{i,j} \int_0^1 dx (\rho^0)^{1-x} (X_i - \langle X_i \rangle_0) (\rho^0)^x \chi^{-1}_{ij} \text{Tr}_S (X_j - \langle X_j \rangle_0) \rho_S^F,$$

$$\varphi = \rho^0. \quad (39)$$

With Eq. (5) one obtains for the time evolution of the reduced density matrix,

$$\rho_S^F(t) = \text{Tr}_B U(t) L(\rho_S^F) U^\dagger(t) + \text{Tr}_B \rho^0, \quad (40)$$

where we used that the total density matrix ρ^0 is invariant under the full time evolution. Actually this is a linear equation in the reduced density matrix ρ_S . In order to see this one puts $t=0$ in Eq. (40), to express the trace of ρ^0 over the environment in terms of the reduced density matrix ρ_S : $\text{Tr}_B \rho^0 = \rho_S - \text{Tr}_B L(\rho_S)$. Hence, for the Mori preparation the time evolution of the reduced density matrix is linear. This is also in agreement with the findings for the above discussed model, see Figs. 2 and 3.

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APPENDIX A: CONVEX MAPS ARE AFFINE

We prove that a differentiable convex map M from a Banach space B_1 into a Banach space B_2 is also affine. The derivative of $M(x)$ at $x \in B_1$ is defined as the linear map $DM(x): B_1 \rightarrow B_2$ that is tangential to M at x . For further mathematical details see, e.g., Ref. [35]:

$$\lim_{\|h\|_1 \rightarrow 0} \frac{\|M(x+h) - M(x) - DM(x)h\|_2}{\|h\|_1} = 0, \quad (A1)$$

where $\|\cdot\|_i$, $i=1,2$ denotes the norm in the respective Banach space. The convexity of M then requires that its domain of definition $\mathcal{D}(M)$ is convex, i.e., with each pair x,y of elements of $\mathcal{D}(M)$ also all convex linear combinations, $\lambda x + (1-\lambda)y$, with $0 < \lambda < 1$, belong to $\mathcal{D}(M)$. Moreover, this property is conserved under the convex map M :

$$M(\lambda x + (1-\lambda)y) = \lambda M(x) + (1-\lambda)M(y). \quad (A2)$$

Taking the derivative with respect to λ one finds from Eq. (A2),

$$DM(\lambda x + (1-\lambda)y)(x-y) = M(x) - M(y). \quad (A3)$$

For $\lambda=0$ one obtains

$$M(x) = D M(y)x + M(y) - D M(y)y. \quad (A4)$$

The first term on the right-hand side is linear with respect to x and the second and the third term are independent of x and hence M is affine.

APPENDIX B: PECHUKAS' THEOREM

Pechukas proved in Ref. [10] that a preparation is factorizing if the following conditions are satisfied: (i) the corresponding blow-up map of the preparation is convex; (ii) two

different complete sets of pure system states can be prepared in the considered preparation class.

The proof consists of two steps. The first step is a consequence of the positivity of the blow-up map $R(\rho_S)$ and makes use of the possibility to prepare pure states of the system. Assume $|\psi\rangle$ is a pure state of the system that can be prepared. In the first step of the proof it is shown that the density matrix ρ of the full system has the form

$$\rho = R(|\psi\rangle\langle\psi|) = |\psi\rangle\langle\psi|\chi, \quad (\text{B1})$$

where the environment density matrix χ in general depends on the system state $|\psi\rangle$.

Following Pechukas [10], we consider the diagonal matrix element $\langle\phi, e|\rho|\phi, e\rangle$ of the density matrix ρ with a product state $|\phi, e\rangle$, where ϕ is an element of the system Hilbert space that is orthogonal on the prepared pure state ψ , $\langle\phi, \psi\rangle=0$, and $|e\rangle$ is an arbitrary normalized element of the environment Hilbert space. The positivity of ρ implies $\langle\phi, e|\rho|\phi, e\rangle \geq 0$. On the other hand, $\langle\phi, e|\rho|\phi, e\rangle \leq \langle\phi|\text{Tr}_B\rho|\phi\rangle = |\langle\phi|\psi\rangle|^2 = 0$. Therefore

$$\rho|\phi, e\rangle = 0 \quad (\text{B2})$$

must hold for all environment states $|e\rangle$ and all system states $|\phi\rangle \perp |\psi\rangle$. This implies (B1) with an environment reference density matrix

$$\chi = \text{Tr}_S\rho = \langle\psi|\rho|\psi\rangle. \quad (\text{B3})$$

Up to this point we have not made use of the convexity of the blow-up map and therefore the reduced environment density matrix χ may still depend on the system state $|\psi\rangle$.

In the second step of the proof we first take two pairs of orthonormal system states, $|\psi_1\rangle, |\psi_2\rangle$ and $|\varphi_1\rangle, |\varphi_2\rangle$, that span the same two-dimensional subspace:

$$|\psi_1\rangle\langle\psi_1| + |\psi_2\rangle\langle\psi_2| = |\varphi_1\rangle\langle\varphi_1| + |\varphi_2\rangle\langle\varphi_2| \equiv P, \quad (\text{B4})$$

where P is the projection operator on this subspace. The absolute values of the mutual scalar products then are determined by an angle α :

$$\begin{aligned} |(\psi_1, \varphi_1)|^2 &= \cos^2 \alpha, & |(\psi_1, \varphi_2)|^2 &= \sin^2 \alpha, \\ |(\psi_2, \varphi_1)|^2 &= \sin^2 \alpha, & |(\psi_2, \varphi_2)|^2 &= \cos^2 \alpha. \end{aligned} \quad (\text{B5})$$

We assume that the two pairs of states ψ_i and φ_i can be prepared. According to the first step of the proof, the full density matrix that corresponds to either of the pure states is

a product of the pure state and a density matrix of the environment:

$$\begin{aligned} R(|\psi_i\rangle\langle\psi_i|) &= |\psi_i\rangle\langle\psi_i|\chi(\psi_i), & R(|\varphi_i\rangle\langle\varphi_i|) &= |\varphi_i\rangle\langle\varphi_i|\chi(\varphi_i), \\ & & i &= 1, 2, \end{aligned} \quad (\text{B6})$$

where $\chi(\psi_i)$ and $\chi(\varphi_i)$ are bath density matrices. We shall show that they all are the same. For this purpose we make use of the convexity of the blow-up map and consider its action on the density matrix that is proportional to the projection P onto the two-dimensional subspace, $\rho_S = \frac{1}{2}P$. The action of the blow-up map can be represented in either of two ways by convex combinations of pure states, see Eq. (B4):

$$\begin{aligned} |\psi_1\rangle\langle\psi_1|\chi(\psi_1) + |\psi_2\rangle\langle\psi_2|\chi(\psi_2) \\ = |\varphi_1\rangle\langle\varphi_1|\chi(\varphi_1) + |\varphi_2\rangle\langle\varphi_2|\chi(\varphi_2). \end{aligned} \quad (\text{B7})$$

Calculating the matrix elements with the four pure states ψ_i, φ_i , one obtains the following equations relating the reference density matrices $\chi(\psi_i)$ and $\chi(\varphi_i)$:

$$\begin{pmatrix} \chi(\psi_1) \\ \chi(\psi_2) \end{pmatrix} = \begin{pmatrix} |\cos \alpha|^2 & |\sin \alpha|^2 \\ |\sin \alpha|^2 & |\cos \alpha|^2 \end{pmatrix} \begin{pmatrix} \chi(\varphi_1) \\ \chi(\varphi_2) \end{pmatrix} \quad (\text{B8})$$

and

$$\begin{pmatrix} \chi(\varphi_1) \\ \chi(\varphi_2) \end{pmatrix} = \begin{pmatrix} |\cos \alpha|^2 & |\sin \alpha|^2 \\ |\sin \alpha|^2 & |\cos \alpha|^2 \end{pmatrix} \begin{pmatrix} \chi(\psi_1) \\ \chi(\psi_2) \end{pmatrix}. \quad (\text{B9})$$

We may exclude the trivial cases when the pairs ψ_i and φ_i coincide and either $\cos^2 \alpha=1$ or $\sin^2 \alpha=1$. In all other cases, the four equations only have a solution with $\chi(\psi_1) = \chi(\psi_2) = \chi(\varphi_1) = \chi(\varphi_2)$. Hence, the environment density matrix is identical for all preparable system density matrices in the considered subspace such that a factorizable state of the total system results on this subspace.

In order to apply this argument to higher-dimensional system Hilbert spaces it must be possible to prepare sufficiently many pure system states. Starting as above with a two-dimensional subspace that can be spanned by two pairs of preparable bases $\{\psi_{ij}\}, \{\varphi_{ij}\}$, $i=1,2$ one next considers the subspace spanned by either $|\psi_1\rangle$ and $|\psi_3\rangle$ or by $|\varphi_1\rangle$ and $|\varphi_3\rangle$ and finds $\chi(\psi_1) = \chi(\psi_3) = \chi(\varphi_1) = \chi(\varphi_3)$ for the reference bath operators in the density matrix of the full system. That means that two different sets of pure states spanning the system's Hilbert space must be preparable.

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- [1] U. Weiss, *Quantum Dissipative Systems* (World Scientific, Singapore, 1999).
 [2] W. Pauli, *Probleme der Modernen Physik*, edited by P. Debye (Hirzel Verlag, Leipzig, 1928), p. 30.
 [3] L. van Hove, *Physica* (Amsterdam) **23**, 441 (1957).
 [4] F. Haake, in *Springer Tracts in Modern Physics*, Vol. 66 (Springer, Berlin 1974), p. 98.
 [5] K. Blum, *Density Matrix Theory and Applications*, 2nd ed.

(Plenum, New York, 1996).

- [6] V. May and O. Kühn, *Charge and Energy Transfer Dynamics in Molecular Systems* (Wiley-VCH Verlag, Berlin, 2003).
 [7] *Processes in Molecular Wires*, Chem. Phys. **281**, 111 (2002), edited by P. Hänggi, M. Ratner, and S. Yalriki.
 [8] M. A. Nielsen and I. L. Chuang, *Quantum Computation and Quantum Information* (Cambridge University Press, Cambridge, 2000).

- [9] E. C. G. Sudarshan, P. M. Mathews, and J. Rau, *Phys. Rev.* **121**, 920 (1961).
- [10] P. Pechukas, *Phys. Rev. Lett.* **73**, 1060 (1994).
- [11] P. Pechukas, *Phys. Rev. Lett.* **75**, 3021 (1995).
- [12] V. Gorini, A. Kossakowski, and E. C. G. Sudarshan, *J. Math. Phys.* **17**, 821 (1976).
- [13] G. Lindblad, *Rep. Math. Phys.* **10**, 393 (1976).
- [14] H. Grabert, U. Weiss, and P. Talkner, *Z. Phys. B: Condens. Matter* **55**, 87 (1984).
- [15] P. S. Riseborough, P. Hänggi, and U. Weiss, *Phys. Rev. A* **31**, 471 (1985).
- [16] P. Talkner, *Ann. Phys. (San Diego)* **167**, 390 (1986).
- [17] K. M. Fonseca Romero and M. C. Nemes, *Phys. Lett. A* **235**, 432 (1997).
- [18] K. M. Fonseca Romero and M. C. Nemes, *Physica A* **325**, 333 (2003).
- [19] H. Grabert, P. Talkner, and P. Hänggi, *Z. Phys. B* **26**, 389 (1977).
- [20] H. Grabert, P. Talkner, P. Hänggi, and H. Thomas, *Z. Phys. B* **29**, 273 (1978).
- [21] H. Grabert, P. Hänggi, and P. Talkner, *J. Stat. Phys.* **22**, 537 (1980).
- [22] H. Grabert, P. Shramm, and G.-L. Ingold, *Phys. Rep.* **168**, 115 (1988).
- [23] W. Bez, *Z. Phys. B: Condens. Matter* **39**, 319 (1980).
- [24] A. Suarez, R. Silbey, and I. Oppenheim, *J. Chem. Phys.* **97**, 5101 (1992).
- [25] J. S. Canizares and F. Sols, *Physica A* **212**, 181 (1994).
- [26] S. Gnutzmann and F. Haake, *Z. Phys. B: Condens. Matter* **101**, 263 (1996).
- [27] R. Karrlein and H. Grabert, *Phys. Rev. E* **55**, 153 (1997).
- [28] U. Zürcher and P. Talkner, *Phys. Rev. A* **42**, 3267 (1990).
- [29] S. Kohler, T. Dittrich, and P. Hänggi, *Phys. Rev. E* **55**, 300 (1997).
- [30] G.-L. Ingold, in *Coherent Evolution in Noisy Environments*, edited by A. Buchleitner and K. Hornberger, *Lecture Notes in Physics* Vol. 611 (Springer, Berlin, 2002), pp. 1–53.
- [31] H. Spohn, *Rev. Mod. Phys.* **52**, 569 (1980).
- [32] P. Hänggi, H. Thomas, H. Grabert, and P. Talkner, *J. Stat. Phys.* **18**, 155 (1978).
- [33] H. Mori, *Prog. Theor. Phys.* **33**, 423 (1965).
- [34] R. Kubo, *Rep. Prog. Phys.* **29**, 255 (1966).
- [35] R. Abraham, J. E. Marsden, and T. Ratiu, *Manifolds, Tensor Analysis, and Applications* (Addison-Wesley, London, 1983).