The typical quantum Markovian processes do not support the ensemble interpretation

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Abstract For some typical open quantum system Markovian processes we show the absence of the ensemble ”unraveling” of the dynamics. The unraveling we are concerned with is not directly linked with the so-called piece-wise-deterministic processes unraveling. The general Markovian scenario is briefly considered. Comparison of our results with the similar results is made.

1. Introduction

Physical interpretation of mixed quantum states (statistical operators, i.e. density matrices) is a deep problem intimately linked with the foundations and interpretations of quantum theory as well as with the diverse applications in quantum optics, quantum metrology, complex quantum systems, quantum thermodynamics and the emerging quantum technology [1–3] (and the references therein). The problem remains even if the subtle conceptual mathematical problems on interpretation of probability [4, 5] are ignored.

Traditionally, mixed quantum states for an isolated system are called ”proper mixtures” if they arise from the subjective ignorance of the state of individual elements of the statistical ensemble [6]. On the other hand, quantum states of systems quantum correlated with other quantum systems (or fields) are referred to as ”improper mixtures” due to impossibility to define quantum state of such systems [6]. In this sense, the improper mixtures step aside of the ensemble conceptualization thus presenting an aspect of the quantum measurement problem as well as of the problem of the ”transition from quantum to classical” [4,5,9].

Modern approaches to the issue not only extend the list of the statistical ensembles [1] but the proposals appear to remedy the conceptual conundrum by withdrawing from the concept of statistical ensemble [5]. Both approaches target the single quantum systems. The former aims at theoretical description of the experiments with the single atomic systems [1], while the latter describes the single systems by the mixed states [5]. Those novel approaches...
to quantum statistics are based and endowed by some methods developed in
the open systems \[1,10\], and the quantum information \[3\] theories.

In this paper we investigate the ensemble aspect of the mixed states for
certain well-known and well-studied homogeneous Markovian processes \[1,10\]. We slightly extend and use a condition \[11\] for the pure-state-dynamics
for the Markovian processes. While the condition is known for a relatively
long time and has been applied in the phenomenological modelling of the
damped harmonic oscillator \[12\], its use in the present context is, to the
best of our knowledge, here presented for the first time. Our findings are
somewhat unexpected: for the considered models, there appear inevitable,
typically \textit{a priori} arguments against the ensemble picture of the open system’s
dynamics. We briefly extend our analysis to the general Markovian s
enario and compare with some other approaches \[1,5\] to the topic of interpretation
of the mixed states of open quantum systems.

2. The pure-state conditions for the Markovian processes

The unitary dynamics admits an ensemble presentation. If the mixed
state is (nonuniquely) initially decomposed as
\[
\rho = \sum_k p_k |\psi_k\rangle \langle \psi_k|,
\]
then linearity of the unitary evolution "sustains" the ensemble decomposition,
\[
\rho(t) = \sum_k p_k U(t) |\psi_k\rangle \langle \psi_k| U^\dagger(t) = \sum_k p_k |\psi_k(t)\rangle \langle \psi_k(t)|.
\]
For the linear non-unitary dynamical maps, one might expect an analogous possibility. How-
ever, as we show below, this is not the case already for the open-system Markovian processes.

By Markovian process we assume a differentiable and CP-divisible dy-
namical map \[10\] that admits the Lindblad form of the master equation of the
general form (in the Schrödinger picture):

\[
\frac{d\rho(t)}{dt} = \mathcal{L}[\rho(t)] = -\frac{i}{\hbar} [H, \rho(t)] + \sum_i \gamma_i \left( L_i \rho(t) L_i^\dagger - \frac{1}{2} \{ L_i^\dagger L_i, \rho(t) \} \right). \tag{1}
\]

In general, the damping factors \(\gamma_i \geq 0\) and all the Lindblad operators may
carry time dependence, while the time dependence of the Hamiltonian \(H\)
may be allowed in the case of the week external field(s) \[10\].

For a sufficiently smooth dynamics, arbitrary decomposition \(\rho(t) = \sum_k p_k(t) \langle \psi_k(t)| \langle \psi_k(t)|, \sum_k p_k(t) = 1, \forall t, \) in general, implies:

\[
\frac{d\rho(t)}{dt} = \sum_k \left( \frac{dp_k(t)}{dt} |\psi_k(t)\rangle \langle \psi_k(t)| + p_k(t) \frac{d}{dt} (|\psi_k(t)\rangle \langle \psi_k(t)|) \right). \tag{2}
\]
The knowledge of the (not necessarily orthogonal but normalized) $|\psi_k\rangle$s determines also the "populations" $p_k$ thus presenting eq.(2) as a mathematically well defined problem. Placing $p_k = 1$ and $p_{k'} = 0, \forall k' \neq k$ in eq.(2) gives rise to the pure-state dynamics, $d\rho/dt = d(|\psi_k\rangle\langle\psi_k|)/dt$, which admits for eq.(1) the following form for a pure state in every instant of time [11]:

$$\frac{d}{dt}(|\psi\rangle\langle\psi|) = -i(\mathcal{H}|\psi\rangle\langle\psi| - |\psi\rangle\langle\psi|\mathcal{H}^\dagger),$$

(3)

where, for simplicity, we dropped the index $k$, and:

$$\mathcal{H} = H + i \sum_i \gamma_i \left( \langle L_i\rangle L_i - \frac{1}{2} \langle L_i^\dagger L_i \rangle - \frac{1}{2} L_i^\dagger L_i \right),$$

(4)

while $\langle * \rangle \equiv \langle \psi | * | \psi \rangle$ and we assume $\hbar = 1$. A derivation of equations (3) and (4) for the semigroup maps can be found in [11]; the main steps in the derivation, also applying for the general Markovian case, are presented in Appendix. Solutions to eq.(3)–if such exist–determine also the "populations" $p_k$ in eq.(2) as presented in Appendix.

Now, $p_k$ and $|\psi_k\rangle$ may be regarded to "unravel" the imagined (although non-unique) ensemble composition of the mixed state $\rho$, which is subject of the dynamics eq.(1). That is the idea we follow in this paper: to investigate dynamics of certain Markovian processes so as to see if any ensemble "structure", i.e. the set $\{p_k(t), |\psi_k(t)\rangle\}$, of the state $\rho$ could in principle appear. In Discussion section we carefully emphasize that our task does not have much in common with the so-called piece-wise-deterministic processes (PDP) unraveling [1,13].

For the pure states applies $\rho^2 = \rho$, which is equivalent with $tr\rho^2 = 1$. Hence the equality $dtr(\rho^2)/dt = 0$, i.e. $tr(pdp/dt) = 0$. We are interested in the Markovian processes, which are the completely positive and trace preserving processes, i.e. leave invariant (represent contractions on) the Banach space of the statistical operators [10]. Therefore, with the use of eq.(1), we obtain the following condition for the pure-state dynamics for the Markovian processes:

$$tr \left( \rho \frac{d\rho}{dt} \right) = tr(\rho L[\rho]) = \sum_i \gamma_i \left( tr(\rho L_i^\dagger L_i) - tr(\rho^2 L_i^\dagger L_i) \right) = 0,$$

(5)

where we used the commutation under the tracing out operation.

The equality (5) is well known for the dynamical semigroups, e.g., [11,12] (and the references therein) and here is extended to every Markovian process.
when the time dependence may appear for both $\gamma$s as well as for the Lindblad operators in eq.(1). The equality is often written in the form of:

$$\sum_i \gamma_i \langle \psi | L_i | \psi \rangle \langle L_i^\dagger | \psi \rangle = \sum_i \gamma_i \langle \psi | L_i^\dagger L_i | \psi \rangle,$$

(6)

and applies for any instant in time, $t \geq 0$, as well as for every Markovian process [11, 12]. That is, the equality (6) is necessary in order to have the pure-state continuous-in-time dynamics, $\rho^2(t) = \rho(t)$, for eq.(1) in every instant of time.

Then unraveling of a mixed state, which satisfies eq.(1), assumes continuous-in-time dynamics for a set of pure states $|\psi_k(t)\rangle$, which satisfy eq.(3) and eq.(6), with the statistical weights $p_k(t)$ that appear in eq.(2), for $k \geq 2$.

It is worth emphasizing: despite the formal similarity, the equality (6) cannot generally follow from the Cauchy-Schwarz inequality. To see this, let us introduce an instantaneous normalized state $|\chi\rangle$ and define the following two vectors: $|u\rangle = |\chi\rangle$ and $|v\rangle = L_i |\chi\rangle$. Then the Cauchy-Schwarz (CS) inequality, $|\langle u|v \rangle|^2 \leq \langle u|u \rangle \langle v|v \rangle$, implies:

$$\langle \chi | L_i | \chi \rangle \langle L_i^\dagger | \chi \rangle \leq \langle \chi | L_i^\dagger L_i | \chi \rangle.$$

(7)

Due to the CS inequality, the equality sign in eq.(7) may appear iff the two vectors are collinear, i.e. iff $L_i |\chi\rangle = l_i |\chi\rangle$. But summation over the index $i$ in eq.(7) could lead to eq.(6) iff the state $|\chi\rangle$ is a common eigenstate of all $L_i$s. Bearing in mind that the typical Markovian processes employ the non-commutative Lindblad operators, in general, the purity condition eq.(6) cannot be derived from neither it can be interpreted due to the Cauchy-Schwarz inequality.

### 3. Analysis of some Markovian processes

Consider some typical examples of the homogeneous Markovian processes in the context of the pure-state dynamics of Section 2.

#### 3.1 The two-level system

Choose the system’s basis states denoted $|\pm\rangle$ and construct the observable $\sigma_\pm |\pm\rangle = \pm |\pm\rangle$. Introduce also the nonhermitian operators $\sigma_- = |\rangle \langle -| = \sigma_-^\dagger$.

The homogeneous Markovian master equation (in the interaction picture) for such system is given in the general form [11, 10]:

$$\frac{d\rho}{dt} = \gamma_1 \left( \sigma_+ \rho \sigma_- - \frac{1}{2} \{\sigma_+ \sigma_- , \rho\} \right) + \gamma_2 \left( \sigma_- \rho \sigma_+ - \frac{1}{2} \{\sigma_- \sigma_+ , \rho\} \right)$$

(8)
where the curly brackets denote the anticommutator. Physically, for the case of $\gamma_1 = 0$, we have the case of a single-qubit amplitude damping process \[3\], or the model of the two-level atomic system in contact with the environment on the absolute zero temperature [1][10]. For the atomic system,

$$\gamma_1 = \gamma_0 N, \quad \gamma_2 = \gamma_0 (N + 1),$$

(9)

where

$$N = (e^{\beta \omega_0} - 1)^{-1} \geq 0,$$

(10)

is the mean number of quanta for the environmental mode of frequency $\omega_0$ on the inverse temperature $\beta$, while the real $\gamma_0 > 0$.

Let us separately consider the two cases: when $\gamma_1 = 0$, and when $\gamma_i \neq 0, i = 1, 2$.

The case $\gamma_1 = 0$. For this case of the absolute zero, $T = 0$ (and $N = 0$), the master equation (8) reduces to the second term, $\gamma_2 = \gamma_0$, with the only one Lindblad operator, $L = \sigma_-$. Then eq.(6) easily gives the pure-state dynamics condition to read:

$$p_+ (1 - p_+) = p_+,$$

(11)

where $p_+ \equiv |(|\psi\rangle|^2$. That is, eq.(11) reveals the only one possible solution: $p_+ = 0$ and therefore the only one pure state allowed for every instant of time—the ground state $|\rangle$. The same conclusion is reached by employing the Cauchy-Schwarz inequality: due to the only one Lindblad operator, the state satisfying the equality in eq.(7) must be an eigenstate of the Lindblad operator $\sigma_-$—that is the ground state $|\rangle$.

The case $\gamma_i \neq 0, i = 1, 2$. Now consider the full master equation (8) with both nonzero damping factors $\gamma_i$. In this case there are the two Lindblad operators, $\sigma_-$ and $\sigma_+$. A straightforward application of eq.(6) with the definitions eq.(9) and (10) gives the pure-state dynamics condition:

$$(2N + 1)p_+^2 - 2Np_+ + N = 0,$$

(12)

which leads to the solutions for the $p_+$:

$$p_{+1,2} = \frac{N \pm \sqrt{N^2 - N}}{2N + 1}. $$

(13)

Again, from eq.(13) follows the only one solution: $N = 0$, which implies $p_+ = 0, \forall t$. Needless to say, $N = 0$ is equivalent with $T = 0$—which is analysed above—thus emphasizing the absence of even a single pure state satisfying eq.(6) for the finite temperature.
On the other hand, eq.(8), for \( \gamma_i \neq 0, i = 1, 2 \), is known to have a well-defined stationary (steady) mixed state—the thermal-equilibrium Gibbs canonical state—which is the asymptotic \((t \to \infty)\) limit for the dynamics \([1][10]\). According to eq.(13), there does not exist any representation of \( \rho \) of the form of \( \rho(t) = \sum_k p_k(t) |\psi_k(t)\rangle \langle \psi_k(t)| \) such that the pure ensembles' \(|\psi_k(t)\rangle\)'s dynamics is governed by eq.(3), i.e. that the condition (6) is fulfilled, while providing the mixed canonical state as the asymptotic limit \((\lim_{t \to \infty} \rho(t))\).

3.2 The damped linear harmonic oscillator

The one-dimensional (i.e. one-mode) harmonic oscillator damped by the bosonic heat bath is described by the widely used master equation (in the interaction picture) \([1][10]\):

\[
\frac{d\rho}{dt} = \gamma \circ (N + 1) \left( apa^\dagger - \frac{1}{2} \{a^\dagger a, \rho\} \right) + \gamma_o N \left( a^\dagger pa - \frac{1}{2} \{aa^\dagger, \rho\} \right),
\]

where appear the standard bosonic operators, \([a, a^\dagger] = I\), with the \(N\) defined by eq.(10), while the real \(\gamma_o > 0\).

Again, for the bath on the absolute zero, the second term on the rhs of eq.(14) disappears.

The case \(T = 0\). Then \(N = 0\) and hence the master equation reduces to the first term with only one Lindblad operator, \(L = a\). For this case the condition eq.(6) reads:

\[
\langle \psi | a | \psi \rangle \langle \psi | a^\dagger | \psi \rangle = \langle \psi | a^\dagger a | \psi \rangle.
\]

The solution to eq.(15) is obvious, also following from the Schwarz inequality: every eigenstate of the Lindblad operator \(a\) is the solution to eq.(15). That is, every "coherent state" \(|\alpha\rangle\) satisfying the eigen-problem \(a|\alpha\rangle = \alpha|\alpha\rangle\) also satisfies the condition eq.(15) for the pure-state dynamics. In the context of the ensemble unraveling, this means that the only possibility is to have a continuous transition between the "coherent states", of the form \(|\alpha(t)\rangle\), such that \(a|\alpha(t)\rangle = \alpha(t)|\alpha(t)\rangle\), \(\forall t\). However, this is not possible (for \(T = 0\)). If in any instant of time \(t_o \geq 0\), the state is a coherent state, it remains unchanged for the rest of the evolution \(t \geq t_o\) \([1]\).

The case \(T > 0\). Then both terms in the master equation are nonzero \((N > 0)\) giving rise to the two Lindblad operators, \(a\) and \(a^\dagger\). It is easy to show that the pure-dynamics condition (6) now gives:

\[
\langle a^\dagger a \rangle + \frac{N}{2(N + 1)} = \langle a \rangle \langle a^\dagger \rangle.
\]
According to the Cauchy-Schwarz inequality, \( \langle a^\dagger a \rangle \nless \langle a \rangle \langle a^\dagger \rangle \), so we conclude that it is necessary to have the condition \( N = 0 \) in contradiction with the assumption of nonzero temperature of the thermal bath. That is, for the finite temperature, there is not a single pure state that could fulfill the condition for the pure-state dynamics of the damped harmonic oscillator. It is rather obvious that placing \( N = 0 \) in eq.(16) returns eq.(15).

This finding is even more striking than in the previous case in Section 3.1. While existence of the asymptotic and stationary, the [mixed] Gibbs canonical state for the damped harmonic oscillator is well known [1, 10], there is not even a single pure state which could unravel the mixed state dynamics.

3.3 Comments

The models of sections 3.1 and 3.2 are central and paradigmatic for the whole field of the open quantum systems. It is thus not surprise that the similar results are found for some other Markovian models that can be found in the literature, as presented in Supplementary Material. Therefore we find the strong and a priori arguments against the unraveling (Section 2) of the considered open-systems dynamics.

Extension of our considerations to the general Markovian processes does not go far yet. For the semigroup dynamics (which includes the cases emphasized above), the assumption of the weak coupling with the environment distinguishes the Lindblad operators \( L_i \) as the eigenoperators of the open system’s free Hamiltonian \( H_S \) to fulfill the commutator relations of the form:

\[
[H_S, L_i] = \omega_i L_i,
\]

with the so-called Bohr frequencies \( \omega_i \) of \( H_S \) [1,10]. Then the Lindblad operators generate the dynamical Lie algebra for the system [13]. Introduce the commutators \([L_i, L_j^\dagger]\) = \( \theta_i L^{(i)} \) for the algebra, where all \( \theta_i \) are real and the Hermitian \( L^{(i)} \). Assume that for every index \( i \), both the \( L_i \) and \( L_i^\dagger \) appear in the master equation; of course, this need not be the case as illustrated by the above zero-temperature cases. Then the condition eq.(6) leads to the equality:

\[
\sum_i (\gamma_i + \gamma'_i)((L_i L_i^\dagger) - \langle L_i \rangle \langle L_i^\dagger \rangle) + \sum_i \gamma'_i \langle L^{(i)} \rangle = 0,
\]

where the primed coefficients \( \gamma'_i \) regard the adjoint Lindblad operators \( L_i^\dagger \).

For Hermitian \( L_i \)s, \( \theta_i = 0, \forall i \), thus reducing eq.(18) to an “extended” quantum uncertainty relation (whose counterparts are extensively investigated for some models [11,12] (and the references therein)):
\[ \sum_i (\gamma_i + \gamma_i') (\Delta L_i)^2 = 0. \] (19)

Non-negativity of the gamma-factors implies \( \Delta L_i = 0, \forall i \). Needless to say, this condition can be fulfilled iff the state is a common eigenstate of the \( L_i \)s. Therefore continuous-variable (CV) systems are typically beyond the reach of the equality (19).

If relaxed the condition (19):

\[ \sum_i (\gamma_i + \gamma_i') (\Delta L_i)^2 \ll 1, \] (20)

a slight impurity of the state is allowed while eq.(20) may apply even for the CV systems. Then a search for the proper solutions (if allowed by the, typically temperature-dependent, damping factors \( \gamma_i \)) may (just like in the models of Sections 3.1 and 3.2) coincide with the search for the ”preferred” (approximate stationary) states for the dynamics [14]. Again, this does not necessarily (if at all) provide the desired solution—a continuous ”set” of states that could support the time-continuous pure-state dynamics of the open system. For example, the fixed values for the standard deviations \( \Delta L_i \) may lead to a single solution, i.e. to unique pure state [15], as it is already observed in Sections 3.1 and 3.2.

Without further ado, we conclude that the search ensemble-presentation of some Markovian processes may be regarded typically to fail.

4. Discussion

While our task of ”unraveling” the Markovian master equations may sound like the program of the master equations ”unraveling” in the context of the so-called piece-wise-deterministic processes (PDP) models [1] (and the references therein), the similarity is only in the narrative [1, 2, 13]. Actually, the PDP unraveling [1] regards the completely different physical situation, which extends the continuous-in-time dynamics eq.(1) by the occasional instantaneous state collapses. While it is provided that averaging over the state collapse events returns the continuous dynamics eq.(1), the collapse mechanism is essential for the physical situation and is recognized in the context of the so-called indirect (”continuous”) quantum measurement scheme [1] (and the references therein). This is physically a tripartite and hybrid model: the open system interacts with its environment, which, in turn, is monitored by a classical apparatus, whose actions on the environment (the ”probe” of the measurement scheme) induce the collapse to the open system’s dynamics [1]. In contrast to this picture, we are concerned with the standard bipartite
setup of the fully quantum system “open system+environment” that is subject of the unitary-only dynamics (i.e. the environment does not induce any state collapse events). The formal side of this discrepancy is also interesting.

Our considerations regard every Markovian (not necessarily “homogeneous”) processes so that the pure-state condition eq.(6) is satisfied along with the dynamics eq.(3), which is equivalent with the Schrödinger-like dynamics ($\hbar = 1$):

$$\frac{d|\psi(t)\rangle}{dt} = -i\mathcal{H}|\psi(t)\rangle,$$

and $\mathcal{H}$ is defined by eq.(4). On the other hand, the PDP unraveling process [1] is also defined by eq.(21) but with the different ”hamiltonian”, which reads:

$$\mathcal{H}' = H - \frac{i}{2} \sum_i \gamma_i |L_i\rangle\langle L_i| + \frac{i}{2} \sum_i \gamma_i \langle L_i\rangle L_i.$$

Compared to eq.(4), eq.(22) is missing the term $i \sum_i \langle L_i\rangle L_i$ while having the opposite sign for the last term.

Therefore we may say that the possibility to ”unravel” the master equations for the PDP processes [1] does not have much in common with our considerations based on eq.(3) and eq.(6). That is, our conclusions are not limited by the PDP method or by the usefulness of its formalism [1, 2, 13].

Nonexistence of the solutions for eq.(3) and/or eq.(6) establishes nonexistence of the ensemble unraveling of the related master equation (1). In such cases, that include the models considered in Section 3 (as well as in Supplementary Material), for the most of the time instants, the state is mixed without the pure-state unraveling.

Therefore our findings emphasize inadequacy, or at least limitations, of the concept of statistical ensemble for the open quantum system dynamics. Bearing in mind the ”improper mixtures” from Introduction, this may be not surprise. However, the concept of ”improper mixtures” [6] is non-transparent itself and its meaning and the role in the foundations of quantum theory is interpretation-dependent [16–19]. On the other hand, the new approaches targeting the single quantum system [1, 5] as well as our considerations are closer to the standard quantum mechanical formalism and thus less dependent on the quantum interpretations. Concretely, they all go towards the hard problem of (re)interpreting the concept of probability and statistical randomness [4]. To this end, for the Markovian processes, we can detect the two possibly important directions whose elaboration will be presented elsewhere. The first one regards the status of the concept of ”quantum system” [9, 20, 22] as a basis for the ensemble conceptualization—with the obsta-
cles imposed by the quantum nonseparability \cite{6} and nonlocality \cite{23}. Another direction regards dynamics of single quantum systems \cite{1,2} as well as the concept of mixed states for the individual (single) quantum systems \cite{5}—while bearing in mind that in this case the quantum measurement problem seems to fade out \cite{24}.

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Appendix A

From a decomposition \( \rho = \sum_k p_k |\psi_k\rangle \langle \psi_k| \) follows:

\[
\rho_{ii} = \sum_k p_k |\langle i| \psi_k\rangle|^2,
\]

(23)

where \( \rho_{ii} = \langle i|\rho|i\rangle \) for an orthonormalized basis \( |i\rangle \) of the system’s Hilbert state space; \( i = 1, 2, 3, ..., n \). Since existence of the \( \rho_{ii}s \) is guaranteed by eq.(1), and the states \( |\psi_k\rangle, k = 1, 2, 3, ..., m \leq n \), are solutions to eq.(3) (assuming that such exist), the equation (23) is well-defined algebraic equation for the unknown ”populations” \( p_k \) for every instant in time. This establishes consistency of the mathematical task posed by eq.(2), without a need to refer to the derivatives of \( p_k \) appearing in eq.(2).

For completeness, we present the main steps leading to eqs.(3) and (4) as it can be found in \cite{11}.

Assume the pure state \( \rho(t) = |\varphi(t)\rangle \langle \varphi(t)|, \forall t \). Introduce arbitrary pure state (a time-independent vector) \( |\theta\rangle \). Then \( \rho|\theta\rangle = \langle \varphi|\theta\rangle|\varphi\rangle \) so it is straightforward to obtain:

\[
\frac{d}{dt} \rho|\theta\rangle = \mathcal{L}[\rho]|\theta\rangle + \rho \mathcal{L}[\rho]|\theta\rangle.
\]

(24)

Now the use of \( \rho * \rho = (\text{tr} * \rho)\rho \) and the, possibly time dependent, Liouvillian \( \mathcal{L} \) from eq.(1) easily lead to the Schrödinger-like equation:

\[
\frac{d|\varphi\rangle}{dt} = -\frac{i}{\hbar} \mathcal{H}|\varphi\rangle,
\]

(25)

which is equivalent with eq.(3) and eq.(4).
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Following the analysis performed in Section 3 of the main text, we analyse some standard Markovian models of the open quantum systems theory. The findings justify the findings of sections 3.1 and 3.2 in the main text.

A. A three-level atom

A three-level atom with the (non-degenerate) energies $E_1 < E_2 < E_3$ is endowed by the dipole transitions which exclude the transitions between

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A three-level atom with the (non-degenerate) energies $E_1 < E_2 < E_3$ is endowed by the dipole transitions which exclude the transitions between
the two lower levels. Defining the operators \( \sigma_{ij} \equiv |i\rangle\langle j|, i \neq j = 1, 2, 3 \), the master equation (in the interaction picture) follows from the quantum-optical master equation [1, 2]:

\[
\dot{\rho} = \gamma_1(N_1 + 1) \left( \sigma_{13} \rho \sigma_{31} - \frac{1}{2} \{ \sigma_{31} \sigma_{13}, \rho \} \right) + \gamma_1 N_1 \left( \sigma_{31} \rho \sigma_{13} - \frac{1}{2} \{ \sigma_{13} \sigma_{31}, \rho \} \right) + \\
\gamma_2(N_2 + 1) \left( \sigma_{23} \rho \sigma_{32} - \frac{1}{2} \{ \sigma_{32} \sigma_{23}, \rho \} \right) + \gamma_2 N_2 \left( \sigma_{32} \rho \sigma_{23} - \frac{1}{2} \{ \sigma_{23} \sigma_{32}, \rho \} \right).
\]

(S.1)

In eq.(S.1): \( N_i \equiv N(\omega_i) = (e^{\omega_i/k_BT} - 1)^{-1} \), with the transition frequencies \( \omega_i, i = 1, 2 \); we assume \( (h = 1) \).

Therefore the four Lindblad operators (for the finite temperature): \( \sigma_{13}, \sigma_{31}, \sigma_{23}, \sigma_{32} \); generalization to the degenerate case is straightforward. Then the pure-state condition eq.(6) of the main text gives the equality:

\[
\gamma_1 N_1 (p_1 + p_3 - 2p_1 p_3) + \gamma_1 (p_3 - p_1 p_3) + \gamma_2 N_2 (p_2 + p_3 - 2p_2 p_3) + \gamma_2 (p_3 - p_2 p_3) = 0,
\]

(S.2)

where \( p_i \equiv |\langle i | \psi \rangle|^2 \).

The case \( T = 0 \). For \( T = 0 \), \( N_1 = 0 = N_2 \) and eq.(S.2) requires \( p_3 = 0 \), with arbitrary \( p_i, i = 1, 2 \). That is, every pure state \( |\psi\rangle = c_1|1\rangle + c_2|2\rangle \) is allowed. However, since \( \sigma_{i3} |\psi\rangle = 0, \forall i = 1, 2, 3 \), placing \( N_1 = 0 = N_2 \) into eq.(S.1) reveals that every pure state \( |\psi\rangle \) (i.e. for arbitrary \( c_i, i = 1, 2 \)) is a stationary state. That is, such states do not evolve in time and hence there is not even a single pure state that could be used to unravel the mixed state dynamics eq.(S.1).

The case \( T > 0 \). Then \( N_1 \neq 0 \neq N_2 \) and the pure-state condition reads:

\[
\gamma_1(N_1 + 1)(p_3 - p_1 p_3) + \gamma_1 N_1 (p_1 - p_1 p_3) + \gamma_2(N_2 + 1)(p_3 - p_2 p_3) + \gamma_2 N_2 (p_2 - p_2 p_3) = 0,
\]

(S.3)

with the constraint \( p_1 + p_2 + p_3 = 1 \). Without loss of generality, choose (like for the models of the atomic dark states and induced transparency [2,3]) \( \gamma_1 = 1 = 100 \gamma_2 \) and \( N_1 = 0.4 = 1000 N_2 \). By inspection it can be seen that eq.(S.3) does not return any relevant solutions. For example, eliminate \( p_3 \) and solve eq.(S.3) for \( p_1 \). It is obtained that the minimum value for \( p_2 \) that returns a real \( p_1 \) is \( p_2 \approx 0.83 \), while then the minimum value of \( p_1 \approx 0.32 \). Elimination of the other terms (\( p_2 \), or \( p_3 \)) does not offer any solution for the unknown population in the interval \([0, 1]\). Therefore in this case even a single pure state cannot be found to fulfill the pure-state condition.
B. Multimode system

Consider a linear system of \( n \) harmonic oscillators or more generally modes in contact with the thermal bath on the temperature \( T \). In any case this system can be transformed into a set of mutually uncoupled normal coordinates, i.e. of uncoupled modes, here presented by the commuting Bose annihilation operators \( a_i, i = 1, 2, ..., n; \ [a_i, a_j^\dagger] = \delta_{ij} \). Then a generalization of eq.(14) of the main text is straightforward:

\[
\dot{\rho} = \sum_{i=1}^{n} \gamma_i \left( (N(\omega_i) + 1) \left( a_i \rho a_i^\dagger - \frac{1}{2} \{a_i^\dagger a_i, \rho\} \right) + N(\omega_i) \left( a_i^\dagger \rho a_i - \frac{1}{2} \{a_i a_i^\dagger, \rho\} \right) \right).
\]

(S.4)

The Lindblad operators are all the Bose operators, \( a_i \) and \( a_i^\dagger \). Then eq.(4) of the main text gives the condition for the pure pure-state dynamics for eq.(S.4):

\[
\sum_{i=1}^{n} \gamma_i \left( (N(\omega_i) + 1) \left( \langle a_i^\dagger a_i \rangle - \langle a_i \rangle \langle a_i^\dagger \rangle \right) + N(\omega_i) \left( \langle a_i a_i^\dagger \rangle - \langle a_i^\dagger \rangle \langle a_i \rangle \right) \right) = 0.
\]

(S.5)

Due to the Cauchy-Schwarz inequality (cf. the main text), as well as to \( \gamma_i, N(\omega_i) \geq 0, \forall i \), all the terms in eq.(S.5) are non-negative. Therefore the only possibility to fulfill the equality in (S.5) is already recognized in Section 3.2 of the main text:

\[
N(\omega_i) = 0, \quad \langle a_i^\dagger a_i \rangle - \langle a_i \rangle \langle a_i^\dagger \rangle = 0,
\]

(S.6)

for every mode \( i \). Mutual independence of the modes implies the conclusion drawn in Section 3.2 for every mode \( i \) as well as for the solution, \( \otimes_i |\alpha_i\rangle \), of eq.(S.5) (where \( \alpha_i \) states for the \( i \)th "coherent state" for the \( i \)th mode).

C. The phase damped harmonic oscillator

This model [2] is comprised of a single harmonic oscillator in a contact with the thermal bath such that the bosonic number operator, \( N = a^\dagger a \), is coupled with the bath’s variable(s). The effective master equation (in the Schrödinger picture) is (\( \hbar = 1 \)):

\[
\dot{\rho} = -i\omega_0 [N, \rho] + \gamma \left( N \rho N - \frac{1}{2} \{N^2, \rho\} \right).
\]

(S.7)

The only Lindblad operator \( N \) commutes with the system Hamiltonian and hence the pure-state condition:
\[ \Delta N = 0, \quad (S.8) \]
determines its eigenstates \( |n\rangle \) as the solutions. However, those states are the exact "pointer basis" states \[4\] that do not evolve in time. Thus there is not a single pure state whose dynamics could describe the decoherence-dynamics eq.(S.7). This conclusion applies to all the similar Markovian decoherence-models for a qubit or a continuous-variable (CV) systems \[1, 4, 5\].

**D. One-qubit depolarizing channel**

The so-called generalized one-qubit depolarizing channel is modelled by the following master equation (in the interaction picture) \[6\]:

\[ \dot{\rho} = \sum_{i=x}^{z} \gamma_i (\sigma_i \rho \sigma_i - \rho), \quad (S.9) \]

where appear the standard Pauli operators \( \sigma_i, i = x, y, z \).

Therefore there are three Lindblad operators, \( L_i = \sigma_i \), which give rise to the pure-state condition:

\[ \sum_i \gamma_i (1 - \langle \sigma_i \rangle^2) = 0. \quad (S.10) \]

It is obvious that eq.(S.10) implies \( \langle \sigma_i \rangle = 1, \forall i \), which cannot be fulfilled for any pure state. This result may be expected due to the fact that this is a unital channel, i.e., dynamics preserving the fully mixed state, \( I/2 \), where \( I \) is the identity operator. This is a situation also for all the unital maps for which the Lindblad operators do not have even a single common eigenstate \[1\].

**E. Decay of a two-level atom into a squeezed field vacuum**

This is another standard model in quantum optics for a two-level atom that is described by the following master equation \[1, 2\]:

\[ \dot{\rho} = \gamma_o \left( C \rho C^\dagger - \frac{1}{2} [C^\dagger C, \rho] \right), \quad (S.11) \]

where \( C = \cosh(r) \sigma_- + e^{i\theta} \sinh(r) \sigma_+ \), and the environmental squeeze parameters \( r \) and \( \theta \), while \( \sigma_- = |g\rangle \langle e| \) for the excited \( (e) \) and the ground \( (g) \) atomic states.

Since there is only one Lindblad operator, \( L = C \), the pure-state condition gives:

\[ \cosh^2(r)p_e + \sinh^2(r)(1 - p_e) - 2p_e(1 - p_e) \sinh(r) \cosh(r) \cos(\omega) = 0; \quad (S.12) \]
in eq. (S.12): \( p_e = |\langle e|\psi\rangle|^2 \), \( \omega = \theta + 2\delta \), with the arbitrary phase \( \delta \).

Solutions to eq. (S.12) are readily found to read: 
\[
(1 + e^{i\omega}\coth(r))^{-1} \quad \text{and} \quad 1 - (1 + e^{i\omega}\tanh(r))^{-1}.
\]
The presence of the complex term can be eliminated for either \( \omega = 0 \) or \( \omega = \pi \). For the latter the negative values for \( p_e \) are obtained. Therefore the only possibility is \( \omega = 0 \) (i.e. \( \delta = -\theta/2 \)) when the two solutions become equal. Hence the unique solution for \( p_e = (1 + \coth(r))^{-1} \), which is independent of \( \theta \).

Thus, for the environmental state with the fixed \( r \) and \( \theta \) parameters, the unique state of the two-level system is found to read:

\[
|\psi\rangle = (1 + e^{i\theta}\coth(r))^{-1/2}|e\rangle + e^{-i\theta/2}(1 - (1 + e^{i\theta}\coth(r))^{-1})^{1/2}|g\rangle. \quad (S.13)
\]

Certainly, the single pure state eq. (S.13) cannot unravel the mixed-state dynamics, which is known (for \( \theta = 0 \)) \([1]\) to have the unique asymptotic state, \( \rho_\infty = (I + \sigma_z/(2\sinh^2(r) + 1))/2 \). Similar results are found for the related model of the environment in the initial squeezed-thermal state \([1]\).

F. A nonadiabatic Markovian model

For an externally driven damped harmonic oscillator, the following Markovian master equation applies (in the interaction picture) \([4]\):

\[
\dot{\rho} = |\xi(t)|^2\gamma(t)\left(F_+\rho F_- - \frac{1}{2}\{F_-F_+, \rho\} + e^{-i\alpha(t)/\kappa_B T}(F_-\rho F_+ - \frac{1}{2}\{F_-F_+, \rho\})\right),
\]

where \( F_+ = Ax + Bp = F_+^\dagger \) and \( A = (1 + i\mu/\kappa)/2, B = i/m\omega(0)\kappa \), with all the positive parameters, \( m, \omega(0), \kappa > 0 \) as well as \( \gamma(t), \alpha(t) \geq 0, \forall t \).

From (S.14), the two Lindblad operators are found: \( L_1 = F_+ \) and \( L_2 = e^{-i\alpha(t)/2\kappa_B T}F_- \). Then applying eq. (6) from the main text gives:

\[
(\langle F_-F_+ \rangle - \langle F_-\rangle \langle F_+ \rangle)(1 + e^{-i\alpha(t)/\kappa_B T}) + e^{-i\alpha(t)/\kappa_B T}\langle [F_+, F_-] \rangle = 0. \quad (S.15)
\]

With the use of the commutator, \([F_-, F_+] = \hbar/m\omega(0)\kappa\), eq. (S.15) becomes a sum of the non-negative terms:

\[
(\langle F_-F_+ \rangle - \langle F_-\rangle \langle F_+ \rangle)(1 + e^{-i\alpha(t)/\kappa_B T}) + \frac{\hbar e^{-i\alpha(t)/\kappa_B T}}{m\omega(0)\kappa} = 0. \quad (S.16)
\]

Since the second term on the rhs of (S.16) cannot equal zero, we conclude that there does not exist even a single pure state for the master equation (S.14) unraveling.
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