Biexponential decay and ultralong coherence of a qubit

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Abstract – A quantum two-state system, weakly coupled to a heat bath, is traditionally studied in the Born-Markov regime under the secular approximation with completely positive linear master equations. Despite its success, this microscopic approach exclusively predicts exponential decays and Lorentzian susceptibility profiles, in disagreement with a number of experimental findings. On the contrary, in the absence of the secular approximation they can be explained but with the risk of jeopardizing the positivity of the density matrix. To avoid these drawbacks, we use a physically motivated nonlinear master equation being both thermodynamically and statistically consistent. We find that, beyond a temperature-dependent threshold, a bifurcation in the decoherence time $T_2$ takes place; it gives rise to a biexponential decay and a susceptibility profile being neither Gaussian nor Lorentzian. This implies that, for suitable initial states, a major prolongation of the coherence can be obtained in agreement with recent experiments. Moreover, $T_2$ is no longer limited by the energy relaxation time $T_1$ offering novel perspectives to elaborate devices for quantum information processing.

Introduction. – The duration of energy relaxation and decoherence is of significance for a wide scope of quantum nanodevices. Preserving their coherence is a particularly challenging task in the presence of noisy environments [1]. The archetypical example is a qubit whose coherence time must be longer than the duration of a logic-gate operation to adequately carry out a quantum computation [2]. It also plays a central role in long-distance quantum communication [3], environment-assisted transport [4], long-lived coherence of photosynthetic complexes [5,6], quantum chaos [7,8] and others [9–11]. Theoretically, the energy relaxation and decoherence lifetimes, respectively denoted by $T_1$ and $T_2$, are often computed in the context of open quantum systems from the celebrated Lindblad-Davies master equation (LDME) [12–14]. This equation is obtained in the weak-coupling limit (WCL), where the coupling constant of the system-bath interaction is taken towards zero after a time rescaling. Although the LDME is related to an underlying Hamiltonian description only in this scaling limit, its linear and robust thermodynamic character [15] makes it an appealing tool to compute lifetimes at small but finite values of the coupling constant. However, due to its structural properties the LDME exclusively predicts polarization decays of exponential kind, corresponding to Lorentzian profiles for the susceptibility. Precisely such features are violated in a plethora of experimental findings ranging from electron/nuclear spins [16–20] or nitrogen-vacancy (NV) centers [21,22] to chromophoric molecules [23,24] which display a biexponential decay of the polarizations leading to two $T_2$ times, a short and a long one. This decay process was clearly associated to homogeneous non-Lorentzian susceptibility profiles in quantum dots (QDs) [25–31] and NV centers [32,33]. In addition, using materials doped with rare-earth ions, the decoherence can be slowed down by one order of magnitude for an initial Bloch vector being properly sized and oriented [34,35]. On the other hand, the complete positivity of the LDME implies that $T_2$ is at most twice as large as $T_1$, while equality is reached only in the absence of pure dephasing [36,37]. Although no experimental evidence has yet broken the theoretical bound $T_2 \leq 2T_1$, it has been highly disputed, see [38–48]. Indeed, the LDME is just one pos-
sible phenomenological Markovian linear master equation for finite couplings whose complete positivity is by far too restrictive [49–55]. To leave this strict paradigm, one can terminate the microscopic derivation before applying the secular approximation (SA) giving the famous Bloch-Redfield master equation (BRME). Unfortunately, it can violate the positivity of the density matrix and the second law of thermodynamics [56]. Alternatively, one could resort to stochastic models [57,58], non-Markovianity [59,60] or nonlinear dynamics for the reduced (open) quantum system [47,61–72].

In this letter we describe the emergence of two $T_2$ decoherence times, namely a short and a long one, for a qubit undergoing a physically sound nonlinear Markovian dynamics beyond the WCL\(^1\). Thereby, the polarizations follow a biexponential decay coming along with a non-Lorentzian susceptibility profile. In this context, we explain how an appropriate choice of the initial state can be exploited to overcome the well-known too restrictive [49–55]. To leave this strict paradigm, one can terminate the microscopic derivation before applying the secular approximation (SA) giving the famous Bloch-Redfield master equation (BRME). Unfortunately, it can violate the positivity of the density matrix and the second law of thermodynamics [56]. Alternatively, one could resort to stochastic models [57,58], non-Markovianity [59,60] or nonlinear dynamics for the reduced (open) quantum system [47,61–72].

Below, all relevant quantities are expressed in terms of the absorption rate $a(\omega) = h(-\omega)$ for $\omega > 0$. For the sake of simplicity, we use for the qubit a parametrization in terms of the Pauli matrices\(^2\) $\sigma_x$, $\sigma_y$ and $\sigma_z$ yielding $H_0 = -i(\Delta/2)\sigma_z$ with $\Delta = E_2 - E_1$ the energy gap. The operator $Q$, dimensionless so that the units of energy are fully assigned to $h(\omega)$, carries the real elements $Q_{11}$ and $Q_{12}$ on the diagonal while the off-diagonal elements read $Q_{12} = e^{i\theta}Q_{21}^*$ with $\theta \in [0, 2\pi)$.

**Thermodynamic formulation.** – To go beyond the standard linear master equation (1), exclusively producing exponential decays, we make use of the nonlinear thermodynamic master equation (NTME)\(^{[74]}\)

\[
\dot{\rho} = -i[H_s, \rho] + \frac{1}{2} \sum_{\omega} h(\omega) \left( A_{\omega} \rho A_{\omega}^* - A_{\omega}^* A_{\omega} \rho + h.c. \right) + \frac{1}{2} \sum_{\omega, \omega'} \sqrt{h(\omega)h(\omega')} \lambda \sum_{\lambda} \frac{1}{2} \left[ A_{\lambda}^* \rho A_{\lambda} - A_{\lambda}^* A_{\lambda} \rho + h.c. \right].
\]

where $S(\rho) = -\ln \rho$ is the von Neumann entropy operator. This equation, inspired by a derivation\([64]\) as well as by thermodynamical\([70,71]\) and statistical\([75]\) arguments, generates a modular dynamical semigroup ensuring the preservation of the hermiticity, the trace and the positivity of $\rho$ as expected from a physical master equation\(^3\). Moreover, it converges to the Gibbs state and gives rise to a positive entropy production. Besides, the NTME (3) gives back the LDME (1) in the WCL, asymptotically describing the exact Hamiltonian dynamics in the long time limit\([76]\), by applying the time-averaging procedure mentioned in\([75]\). The NTME (3) carries a sum over two sets of Bohr frequencies like the BRME (2), due to the absence of the SA, but without jeopardizing the positivity of $\rho$.

Pechukas\([51]\) and Romero\([69]\) noted that beyond the WCL nothing forbids to have a nonlinear equation with respect to the state for the reduced system. For example, the nonlinearity of a reduced system can arise by eliminating the irrelevant degrees of freedom of the total system’s density matrix using generalized Nakajima-Zwanzig methods in the absence and in the presence of system-environment correlations\([61,62,64,68]\). Of course, the full system evolves under a linear von Neumann equation. In this respect, the present nonlinear Markovian reduced dynamics is pertinent from a physical perspective.

**Linear response regime.** – Given the nonlinear nature of the NTME, the various lifetimes can only be accessed through the first-order susceptibility. As shown in\([77]\) the latter is obtained by means of the fluctuation-dissipaton theorem (FDT) in the time-domain through the expression $\chi_{ab}(t) = -\beta \partial_t \text{tr}(A_a^C \kappa_b \rho)$ relative to the self-adjoint observables $A$ and $B$. It requires the lineariza-

\(^{[74]}\)It is shown in\([74]\) that the environment can be modelled as a quantum or classical object; here, we use a quantum heat bath.

\(^1\)Small but non-vanishing coupling strength contrary to the WCL.
tion $\dot{\rho} = \mathbf{L}\rho$ of (3) near equilibrium obtained from
\begin{equation}
\rho^\dagger[\mathbf{A}_2, S(\rho - \beta H_0)]\rho^{\lambda - 1} = \pi^\lambda[\mathbf{A}_2, \mathbf{K}_2^\lambda - \rho]\pi^{1 - \lambda}.
\end{equation}
Above, $\mathbf{K}_2^\lambda - \rho = \int_0^\infty d\tau \pi^\lambda(\sqrt{\mathbf{A}}\tau A^\dagger\pi^{1 - \lambda})$ known as the equilibrium Kubo-Mori superoperator [78]. Note that, contrary to the thermodynamically robust NTME, its linearized version does not preserve positivity far away from equilibrium and one should limit its use to the linear response regime where the lifetimes/decay rates are defined.4

To perform the calculations, it is advantageous to switch to the Liouville space (see, e.g., Chap. 3 of [9]) highlighted hereafter with a bold notation. Choosing the vector representation $\mathbf{\rho} = (\rho_{11}, \rho_{12}, \rho_{21}, \rho_{22})^T$ for the density matrix coefficients in the Hamiltonian basis, the linearized NTME (3), (4) reads
\begin{equation}
\mathbf{L} = \begin{pmatrix}
-x & 0 & 0 & xe^{\beta i} \\
0 & -y + i\Delta & z e^{2\beta i} & 0 \\
z e^{-2\beta i} & -y - i\Delta & 0 & 0 \\
x & 0 & 0 & -xe^{\beta i}
\end{pmatrix}
\end{equation}
depending on three real positive variables
\begin{equation}
x = a(\Delta), \\
y = \frac{1 + e^{\beta i}}{\beta} a(\Delta)/2 - \Gamma_2^z, \\
z = (\beta/2) e^{\beta i/2} \coth(\beta/2) a(\Delta)
\end{equation}
for the NTME. For the LDME we get $\mathbf{L}$ with $z = 0$ whereas for the BRME we obtain it with $z = (1 + e^{\beta i}) a(\Delta)/2$.

**Spectral analysis.** – The four eigenvalues of the generator (5) are $0, -\Gamma_1$ and $-\Lambda_\pm$ given by $\Gamma_1 = (1 + e^{\beta i}) x$ and $\Lambda_\pm = y \pm \Omega$ with $\Omega = \sqrt{\Delta^2 - \Delta^2}$ being real or imaginary. From $\mathbf{L}$ and its eigenvalues it naturally follows that all decay rates are relative either to energy relaxation or decoherence appearing in the model. The transition from one to two decay rates happens for $z > \Delta$, or equivalently, past the absorption rate $a(\Delta)$ thresholds
\begin{equation}
a_{\text{NTME}}^{\text{abs}} = \Delta e^{-\beta/2} \tanh(\beta/2)/\Delta, \\
a_{\text{BRME}}^{\text{abs}} = 2\Delta/(1 + e^{\beta}),
\end{equation}
whereas for the BRME we obtain it with $\beta \gg 1$. The microscopically origin of the threshold can be tracked back to the non-secular terms of the BRME (2), i.e. with $\omega \neq \omega$, yielding the element $\mathbf{L}_{23} = z e^{2\beta i} = \mathbf{L}_{32}$. Thus, the Liouville matrix (5) for $z \neq 0$ offers a straightforward modeling tool to describe the bifurcation in the decoherence time. It is remarkable that such a temperature-dependent transition from one to two decoherence times, which are associated to a biexponential decay of the coherence, was reported in low-temperature experiments of nuclear spins’ impurities in silicon crystals [19,20] or InGaAs QDs [27].

**Susceptibility analysis.** – We note that the bifurcation phenomenon has simple and striking consequences on the susceptibility $\chi_{\omega}(t)$ associated to the dipole operator $D = \mu e^{i\psi} \sigma_x + e^{-i\psi} \sigma_y$ with $\sigma_\pm = \sigma_x \pm i\sigma_y$ and $\mu > 0$. Switching to the frequency domain the aforementioned FDT becomes $\chi_{\omega}(\omega) = -\beta \cdot \mathbf{A} \cdot \mathbf{L} (\mathbf{L} - i\nu)^{-1} \mathbf{K}_\pi \mathbf{B}$, where the scalar product is defined as $\mathbf{X} \cdot \mathbf{Y} = \sum_\mu x^\mu y^\mu \cdot \Lambda_\pm = \nu$ and $\mathbf{B}$ are again self-adjoint observables now expressed as $4 \times 4$ vectors whereas $\mathbf{K}_\pi$ is a $4 \times 4$ diagonal Kubo matrix evaluated at equilibrium whose non-zero elements read $\{ (1 + e^{-\beta})^{-1}; \beta^{-1}\tanh(\beta/2); \beta^{-1}\tanh(\beta/2); (1 + e^{\beta})^{-1} \}$ and $\mathbf{I}$ is a $4 \times 4$ identity matrix. The dipolar susceptibility arising from the FDT then reads
\begin{equation}
\chi_{\omega}(\omega) = (\mu^2/\Delta) \tanh(\beta/2) \times \sum_\pm \left(1 \pm \frac{z}{\Omega} \cos(2(\theta - \psi)) \right) \frac{\Lambda_\pm}{\Lambda_\pm + i\nu},
\end{equation}
Prior to the bifurcation, the absorption $\text{Im}(\chi_{\omega}(\nu))$ can essentially be approximated by a superposition of two Lorentzians, centered at $\text{Im}(\Lambda_\pm)$, with a unique linewidth equal to $\Gamma_\pm$ for frequencies $\nu \approx \text{Im}(\Lambda_\pm)$, i.e. close to the resonances. After the bifurcation, however, the two resonances are both located at the frequency $\nu = 0$, each of which is characterized by its own linewidth $\Gamma_\pm$, being neither of Lorentzian nor Gaussian type. Indeed, we have a superposition of two non-Lorentzian functions $\text{Im}(\chi_{\omega}(\nu)) = \sum_\pm c_\pm \nu/(\Lambda_\pm^2 + \nu^2)$ with suitable real coefficients $c_\pm$ (see footnote 6). Notably, non-Lorentzian lineshapes were measured for nuclear spins [16], a wide range of QDs [25–29], NV centers [32,33] and for a quantum well [79]. Moreover, if one of the two contributions

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4Decay times have an experimental significance for long times near equilibrium (42). The decay rates obtained by fitting the polarizations far away from equilibrium are not equal to the one defined in the linear response regime; this is even more true with the NTME which is nonlinear (i.e. distinct initial conditions can yield different decay rates; match is only recovered close to equilibrium).

5For large pure dephasing, $\langle Q_{11} - Q_{22} \rangle$ is negligible in regard of $\langle Q_{11} - Q_{22} \rangle^2$ producing the zero entries in $\mathbf{L}$.

6The approximation $\nu = 0$ at the numerator cannot be used to restore a combination of Lorentzian in case $\Lambda_\pm = 0$ so that the profile is truly non-Lorentzian.
The NTME has been designed to be Markovian and we checked this fact with a non-Markovianity measure [83].

Note that the full decay pattern is much richer than what the NTME predicts in the linear response regime, e.g., see fig. 14 on p. 93 of [81] for an overview of decays at short, intermediate and long times.

Look, for example, at Laird’s eqs. (A1)–(A4) in the appendix only makes sense for linear dynamics, it does not need to be a physical requirement and CP [53]. Moreover, the standard notion of CP only makes sense for linear dynamics [49,52,90] and, as mentioned by Pechukas [50] as well as by Shahjai and Sudarshan [54], it does not need to be a physical requirement in spite of its mathematical attractiveness.

Conclusion and perspective. – The secular LDME is over-restricted by CP and does not allow to interpret
numerous experimental results, for example the problem of Cooper-pair pumping [91]. On the contrary, the non-
secular terms lead to a bifurcation phenomenon beyond a temperature-dependent threshold, associated with a biex-
ponential decay of the polarizations and non-Lorentzian susceptibility profiles. Unfortunately, the BRME does
not in general preserve the positivity of $\rho$ and leads to a negative entropy production [56]. The phenomenolog-
ical NTME predicts the same unorthodox effects as the BRME but it is free of its drawbacks. The biexponential
decay can be used to drastically prolong the coherence for optimal orientation of the initial Bloch vectors. More-
over, according to our analysis nothing forbids to overcome the bound $T_2 \leq 2T_1$ although its experimental realization
would clearly be challenging. Such conclusions could be made directly from the Liouville matrix (5) and its asso-
ciated Bloch equation which can be obtained from various other models supporting the idea that the presented bifur-
cation phenomenon is of ubiquitous nature. For example, it can be observed for Laird and co-workers’ equation [38]
or the dynamically time coarse-grained master equations proposed in [74].

Remarkably, ultralong coherence is not limited to a single qubit and could be “scaled-up” since biexponential de-
cays were measured in epitaxial QD arrays [92]. It could allow to delay entanglement sudden-death and enhance
revival [93–95]. Away from the linear response regime, the decay pattern becomes richer. One then finds initial
optimal states either by screening strategies or applying search/reinforcement learning algorithms. Moreover, at
intermediate pure dephasing, where the populations and coherences are interwoven, the NTME predicts population
beating and coherence revival which have been observed in photosynthetic complexes [5,6], quantum kicked
rotors [7,8] as well as anions solvated in water [96]. Taking all this into account, we hope that the present findings will
provide some leads to develop ground-breaking nanoscale devices in the near future.

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