Recovery of Purity in Dissipative Tunneling Dynamics

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Abstract

The time evolution of purity for an initially localized state of a symmetric two-level system coupled to a dissipative bath is investigated using numerically exact real-time path integral methods. With strong system-bath coupling and high temperature, the purity decays monotonically to its fully mixed value, with a short-time Gaussian behavior which is subsequently followed by exponential evolution. However, under low-temperature and weak coupling conditions, a substantial recovery of purity is observed. A simple theoretical analysis reveals three contributions that correspond to completely incoherent, eigenstate population difference and rate terms. The last two of these terms can counter the early drop of purity and are responsible for its rebound. These findings caution against using purity as a measure of decoherence in the dynamics of quantum dissipative systems.

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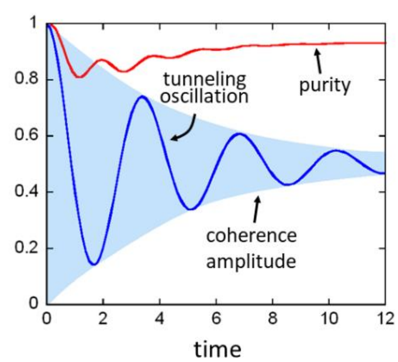

Quantum mechanical coherence is responsible for some of the most intriguing phenomena encountered in the microscopic world. The interference pattern of Young's double slit experiment, the persisting tunneling oscillations in a two-level system and the survival probability recurrences of a displaced wavepacket in an anharmonic potential are typical manifestations of quantum coherence in small, isolated systems. These effects arise from delicate quantum phase relations and are easily destroyed by the noisy effects of fluctuating polyatomic environments (or a generic "bath"). Quantum coherence and its destruction are of interest not only for achieving a deeper understanding of countless physical and biological phenomena, but also in the design for nanoscale devices with a targeted function, e.g. solar energy storage and transport or quantum computers.

Much effort has been spent on understanding the characteristics of decoherence in quantum systems in contact with dissipative environments. The main paradigm in most studies has been the dissipative two-level system[1] (TLS), which represents the simplest case of the more general system-dissipative bath Hamiltonian.[2] The coupling to a bath manifests itself as noise, which gradually destroys the delicate quantum mechanical phase relationships, quenching the coherent tunneling oscillations of the bare system. At low temperature and with weak system-bath coupling the oscillation amplitude of a symmetric TLS decreases slowly as time progresses. The oscillatory dynamics is damped more rapidly with increasing temperature and/or system-bath coupling strength. Strongly dissipative environments fully quench the tunneling oscillations, typically leading to monotonic population decay and rate dynamics.

The idempotence or "purity" of a system's reduced density matrix (RDM) $\tilde{\rho}(t)$, defined as

$$Q(t) \equiv \mathrm{Tr}\,\tilde{\rho}^2(t), \tag{1}$$

which is also related to the linear entropy $S_{\mathrm{lin}} = 1 - Q$, has often been used as a well-defined, basis-independent measure of decoherence. The density matrix of an isolated quantum system in a pure state corresponds to $Q = 1$. If the system and its environment are initially uncorrelated and the system is placed in a pure state, $\mathrm{Tr}\,\tilde{\rho}(0)^2 = 1$ and the purity is equal to unity. Over time, interactions with the environment introduce entanglement between system and bath states, destroying the purity of the RDM and leading to $Q(t) < 1$. In that case the system, in the presence of its environment, is an incoherent mixture of two or more quantum states. The purity is also intimately connected to the behavior of the off-diagonal elements of the RDM (the "coherences"), and decoherence of an initially pure quantum system when placed in a dissipative environment has been associated with a decrease in purity.

Previous work to determine the dynamics of purity in quantum dissipative systems has been largely confined to approximate analytical treatments. Quadratic expansion in time has shown that the early dynamics of a quantum system's linear entropy closely resembles the classical result.[3] Perturbative expansions about $t = 0$ predict a Gaussian decay with a characteristic coefficient given by the inverse of the decoherence time.[4-7] The quantitative behavior of purity in system-bath problems remains unexplored, owing to the computational challenges of full quantum mechanical treatments in condensed phase environments.

In this work we use numerically exact real-time path integral methods to follow the time evolution of purity in the tunneling dynamics of a TLS coupled to a harmonic bath across a variety of parameter regimes. Our results reveal a substantial recovery of purity following its initial drop, which can be nearly quantitative in the low-temperature, weak coupling regime, and a partial recovery under different conditions. This effect may at first seem counterintuitive. However, the equilibrium density matrix will be dominated by the ground state at sufficiently low temperatures in the regime of weak dissipation. Not surprisingly, the long-time RDM of the TLS closely resembles an almost pure state under such conditions.

We focus on the simple case of a symmetric two-level quantum system bilinearly coupled to a bath of harmonic oscillators, which clearly displays the interplay between the important, fundamentally quantum mechanical phenomenon of tunneling and the decohering effects from an environment. In the local (or site) basis, the symmetric TLS Hamiltonian has the standard form

$$\hat{H}_{\rm TLS} = -\hbar\Omega\left(|{\rm R}\rangle\langle{\rm L}| + |{\rm L}\rangle\langle{\rm R}|\right) = -\hbar\Omega\hat{\sigma}_x \tag{2}$$

where $2\hbar\Omega > 0$ is the tunneling splitting, σ_x is the Pauli spin operator, and $|{\rm R}\rangle$ and $|{\rm L}\rangle$ are the 'right' and 'left' localized states, which are related to the TLS eigenstates Φ_0, Φ_1 (with energy eigenvalues $E_0 = -\hbar\Omega,\ E_1 = \hbar\Omega$) through the usual sum and difference combinations,

$$|\Phi_0\rangle = \frac{1}{\sqrt{2}}\left(|{\rm R}\rangle + |{\rm L}\rangle\right), \quad |\Phi_1\rangle = \frac{1}{\sqrt{2}}\left(|{\rm R}\rangle - |{\rm L}\rangle\right) . \tag{3}$$

The TLS is coupled to a bath of harmonic oscillators through the additional term

$$\hat{H}_{\rm bath} = \sum_j \frac{\hat{p}_j^2}{2m_j} + \frac{1}{2} m_j \omega_j^2 \left(\hat{q}_j - \frac{c_j \hat{\sigma}_z}{m_j \omega_j^2} \right)^2 , \tag{4}$$

where $\hat{\sigma}_z = |{\rm R}\rangle\langle{\rm R}| - |{\rm L}\rangle\langle{\rm L}|$. As is well known, all collective parameters of the bath are contained in the spectral density function

$$J(\omega) = \tfrac{1}{2}\pi \sum_j \frac{c_j^2}{m_j \omega_j} \delta\left(\omega - \omega_j\right) . \tag{5}$$

In this work we employ the commonly used Ohmic spectral density with an exponential cutoff, $J(\omega) = \tfrac{1}{2}\pi\hbar\xi\omega e^{-\omega/\omega_{\rm c}}$, which is known[1] to produce rich dynamical behaviors. In this, the TLS-bath coupling is quantified by the dimensionless Kondo parameter ξ and $\omega_{\rm c}$ is the cutoff frequency. All dynamical properties of the system can be obtained from the RDM,

$$\tilde{\rho}_{\alpha''\alpha'}(t) = {\rm Tr}_{\rm bath} \langle \alpha'' | e^{-i\hat{H}t/\hbar} \hat{\rho}(0) e^{i\hat{H}t/\hbar} | \alpha' \rangle , \tag{6}$$

where $\alpha', \alpha'' = {\rm R}, {\rm L}$ or 0, 1.

For the symmetric TLS, it is easy to show that the purity is bounded by the inequality $\tfrac{1}{2} \le Q \le 1$. The minimum value $Q = \tfrac{1}{2}$ is associated with the maximally mixed state $\tilde{\rho} = \tfrac{1}{2}\left(|{\rm R}\rangle\langle{\rm R}| + |{\rm L}\rangle\langle{\rm L}|\right)$, while the highest value $Q = 1$ is attained when the RDM corresponds to a pure state. While the density matrix can be prepared in a pure state at $t=0$, evolution in contact with a dissipative environment leads to mixing. After a long time, the process reaches equilibrium, thus $\lim_{t\to\infty} \tilde{\rho}_{\alpha'\alpha''}(t) = \mathrm{Tr}_{\mathrm{bath}} \langle \alpha' | e^{-\beta \hat{H}} | \alpha'' \rangle / \mathrm{Tr}\, e^{-\beta \hat{H}}$, where $\beta = 1/k_{\mathrm{B}}T$ is the inverse temperature.

The evolution of the RDM is calculated using the numerically exact quasi-adiabatic propagator path integral (QuAPI) methodology,[8,9] and (for parameters associated with long memory) the small matrix disentanglement of the path integral[10,11] (SMatPI) with the blip decomposition[12] of the relevant matrices. Once converged to the desired accuracy with respect to time step and included memory length, these methods produce exact results and allow propagation to long times with effort that scales linearly with the number of path integral time steps. In particular, the SMatPI decomposition eliminates the need for storage and manipulation of large QuAPI matrices, facilitating calculations with long memory and multisite systems.

As is customary, we use a factorized initial condition, $\hat{\rho}(0) = \hat{\rho}_{\mathrm{sys}}(0)\hat{\rho}_{\mathrm{bath}}(0)$ where $\hat{\rho}_{\mathrm{bath}}(0)$ is the Boltzmann operator for the free bath Hamiltonian, given by Eq. (4) with $c_j = 0$. In all calculations presented in the next section the system was initially prepared in the R state, i.e., $\hat{\rho}_{\mathrm{sys}}(0) = |\mathrm{R}\rangle\langle\mathrm{R}|$, and the purity was followed until equilibrium was reached. The behavior of purity dynamics during relaxation from the excited TLS eigenstate is investigated in separate work.

In Figure 1 we show the purity dynamics of this symmetric TLS for four parameter sets: (i) $\xi = 2,\ \omega_{\mathrm{c}} = \Omega,\ \hbar\Omega\beta = 0.125$, (ii) $\xi = 0.3,\ \omega_{\mathrm{c}} = 5\Omega,\ \hbar\Omega\beta = 1$, (iii) $\xi = 0.3,\ \omega_{\mathrm{c}} = 7.5\Omega,\ \hbar\Omega\beta = 5$ and (iv) $\xi = 0.1,\ \omega_{\mathrm{c}} = 7.5\Omega,\ \hbar\Omega\beta = 5$. These are chosen to display a variety of TLS population behaviors, ranging from monotonic decay toward equilibrium to weakly damped oscillatory dynamics. With parameter set (i) the TLS is strongly coupled to a slow, high-temperature bath. The purity is seen to drop monotonically to its equilibrium value $Q_{\mathrm{eq}} \simeq 0.51$, which practically corresponds to a completely mixed state. The small value of the bath cutoff frequency induces long-lasting memory effects which lead to Gaussian purity evolution that (as seen from the logarithmic plot shown in the inset of Fig. 1) persists only up to $\Omega t \simeq 0.25$ and is subsequently replaced by exponential decay. The transition to exponential dynamics occurs even earlier with larger values of ω_{c}. In parameter set (ii) the TLS interacts with a moderately dissipative environment at an intermediate temperature. In this case the equilibrium RDM is a mixed state with $Q_{\mathrm{eq}} \simeq 0.67$. The purity decays to this value nearly monotonically, although it is seen to drop to the slightly lower value $Q_{\min} \simeq 0.63$ before plateauing.

With parameter set (iii) the TLS population exhibits quenched oscillations. The coupling to the bath is again of intermediate strength, but the low temperature prevents a monotonic decay of the population. In this case the purity initially falls to the value $Q_{\min} \simeq 0.70$, but subsequently rises to the equilibrium value $Q_{\mathrm{eq}} \simeq 0.81$. Last, parameter set (iv) shows the purity dynamics in the case of weakly dissipative conditions at a low temperature. Following an initial drop to about $Q_{\min} \simeq 0.81$, the purity subsequently recovers, eventually rising to the value $Q_{\mathrm{eq}} \simeq 0.94$, which represents an almost pure state. At this low temperature the equilibrium distribution is dominated by the ground state, and since the TLS-bath coupling is weak in this case, the projection of the equilibrium density on the TLS subspace results in a RDM that closely resembles the TLS ground state. The nearly complete recovery of purity observed in this case follows an oscillatory pattern.

While the RDM purity decays monotonically to 0.5 in the high-temperature, strongly dissipative regime, its dynamics is not monotonic in the other situations. At lower temperatures and with weaker system-bath coupling, the lowest value the purity reaches is seen to be considerably larger than 0.5. Most interestingly, the initial drop is followed by a rebound, as the RDM evolves toward its long-time equilibrium value. As seen in Fig. 1, the minimum value reached can be considerably larger than 0.5, and the subsequent recovery can be substantial and nearly complete.

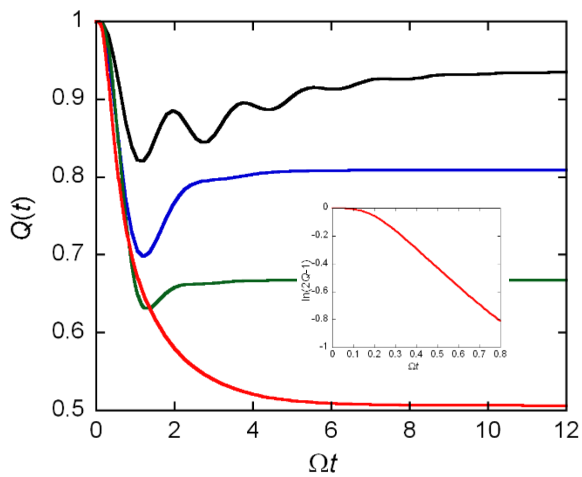

Fig 1. Purity of symmetric TLS coupled to a dissipative bath for four sets of parameters. Red: parameter set (i), $\xi = 2,\ \omega_c = \Omega,\ \hbar\Omega\beta = 0.125$. Green: parameter set (ii), $\xi = 0.3,\ \omega_c = 5\Omega,\ \hbar\Omega\beta = 1$. Blue: parameter set (iii), $\xi = 0.3,\ \omega_c = 7.5\Omega,\ \hbar\Omega\beta = 5$. Black: parameter set (iv), $\xi = 0.1$, $\omega_c = 7.5\Omega$, $\hbar\Omega\beta = 5$. The inset shows a logarithmic plot for parameter set (i).

To understand these behaviors, we express Eq. (1) in terms of the TLS RDM elements,

$$Q(t) = \tilde{\rho}_{\rm RR}(t)^2 + \tilde{\rho}_{\rm LL}(t)^2 + 2\left(\operatorname{Re}\tilde{\rho}_{\rm RL}\right)^2 + 2\left(\operatorname{Im}\tilde{\rho}_{\rm RL}\right)^2 . \tag{7}$$

Next, we rewrite this expression in terms of quantities that have a clear physical meaning. It is easy to see that the real part of the off-diagonal element is related to the difference between the TLS eigenstate populations, while the imaginary part is proportional to the time derivative of the site population,[13] i.e.,

$$\operatorname{Re}\tilde{\rho}_{\rm RL}(t) = \tfrac{1}{2}\left(\tilde{\rho}_{00}(t) - \tilde{\rho}_{11}(t)\right), \quad \operatorname{Im}\tilde{\rho}_{\rm RL}(t) = \tfrac{1}{2}\Omega^{-1}\dot{\tilde{\rho}}_{\rm RR}(t) . \tag{8}$$

Using these relations, the purity becomes the sum of three terms,

$$Q(t) = Q_{\rm incoh}(t) + Q_{\text{pop-dif}}(t) + Q_{\text{t-der}}(t) \tag{9}$$

where

$$
\begin{aligned}
&Q_{\text{incoh}}(t) \equiv \tilde{\rho}_{\text{RR}}(t)^2 + \tilde{\rho}_{\text{LL}}(t)^2 \\
&Q_{\text{pop-dif}}(t) \equiv \tfrac{1}{2}\left(\tilde{\rho}_{00}(t) - \tilde{\rho}_{11}(t)\right)^2 \\
&Q_{\text{t-der}}(t) \equiv \tfrac{1}{2}\Omega^{-2}\dot{\tilde{\rho}}_{\text{RR}}(t)^2
\end{aligned}
\tag{10}
$$

The first contribution to Eq. (9), Q_{incoh}, is the sum of the squared diagonal elements. This is a completely incoherent contribution which is dominant in the strong-dissipation, high-temperature regime. For a localized initial state, $\tilde{\rho}_{\text{RR}}(0)=1$, Q_{incoh} is initially equal to unity and begins to decrease as soon as the site populations change in value. Eventually, the site populations reach the value $\frac{1}{2}$, thus the sum of the squared site populations approaches the maximally mixed value $\frac{1}{2}$. The second term $Q_{\text{pop-dif}}$, the eigenstate population difference, vanishes at $t=0$ as $\tilde{\rho}_{00}(0)=\tilde{\rho}_{11}(0)=\frac{1}{2}$. As time progresses, population begins to transfer from the excited to the ground state, until the two populations attain the Boltzmann relation (projected onto the TLS subspace), thus the population difference term rises gradually.

The behavior of the third term $Q_{\text{t-der}}$, the square of the instantaneous rate, depends strongly on the parameter regime. This term starts at zero and rises slightly in the overdamped regime characterized by monotonic population decay. However, the oscillatory population evolution typical of low-temperature, weakly dissipative conditions causes rapid, large-amplitude oscillations in the rate, which can counter the contribution of the first term in Eq. (9) early on, stopping the decay of purity. The contributions of the three components given in Eq. (9) to the purity dynamics for the four parameter sets discussed earlier are seen in Figure 2.

In the high-temperature, overdamped regime of parameter set (i), the eigenstate population difference remains very small because of the Boltzmann factor. Once the site populations have entered the exponential decay regime $\tilde{\rho}_{\text{RR}}(t)=\frac{1}{2}+\frac{1}{2}e^{-kt}$, $\tilde{\rho}_{\text{RR}}(t)^2+\tilde{\rho}_{\text{LL}}(t)^2=\frac{1}{2}+\frac{1}{2}e^{-2kt}$ and $\dot{\tilde{\rho}}_{\text{RR}}(t)=-\frac{1}{2}ke^{-kt}$. (These forms do not apply to very short times, at which the evolution is Gaussian.) From these expressions we obtain $Q(t)\simeq\frac{1}{2}+\frac{1}{2}(1+\frac{1}{8}k^2\Omega^{-2})e^{-2kt}$, i.e. the purity decays exponentially to the thermodynamic limit $\frac{1}{2}$. As seen in Fig. 2, the purity is dominated by the sum of squared populations at all times in this regime, as the rate contribution is also very small in this case.

In the moderate-dissipation, intermediate-to-low temperature regime of parameter sets (ii) and (iii), the site populations exhibit rapidly damped oscillatory dynamics. As a result, the sum of the squared populations displays a small hump after falling to its minimum value $\frac{1}{2}$, subsequently stabilizing at its thermodynamic value. The contribution of the eigenstate population difference is significant at intermediate temperatures and even larger at low temperature, gradually increasing the value of purity. The rate term, while not particularly large in this regime, plays an important role at early times by shifting the local minimum of the RDM purity to a higher value.

The low-temperature, weak coupling regime of parameter set (iv) shows the complex interplay of all three contributions. In this regime the site populations exhibit damped oscillations, which can be described approximately by the form $\tilde{\rho}_{\text{RR}}(t)=\frac{1}{2}+\frac{1}{2}\cos at\, e^{-kt}$, $\tilde{\rho}_{\text{LL}}(t)=\frac{1}{2}-\frac{1}{2}\cos at\, e^{-kt}$, where a is the renormalized tunneling frequency.[1] Thus $Q_{\text{incoh}}(t)=\frac{1}{2}+\frac{1}{2}\cos^2 at\, e^{-2kt}$, i.e. the sum of the squared site populations shows persistent oscillations, periodically falling to the fully mixed value $\frac{1}{2}$. The contribution of the eigenstate population difference increases steadily over time and is responsible for the nearly complete recovery of purity at long times. The time derivative of the site population has the form $\dot{\tilde{\rho}}_{\text{RR}}(t)=-\frac{1}{2}\left(k\cos at+a\sin at\right)e^{-kt}$. Since the eigenstate population difference is small at early times, the large, highly oscillatory $Q_{\text{t-der}}$ term is responsible for preventing the fall of purity below the relatively high minimum value observed in this regime. Overall, we find that the purity in the low-temperature, weakly dissipative regime satisfies the inequality $Q(t) > \frac{1}{2} + \frac{1}{2}\cos^2 at\, e^{-2kt} + \frac{1}{16}\Omega^{-2}\left(k\cos at + a\sin at\right)^2 e^{-2kt}$, which provides a lower bound.

The three contributions to the time evolution of purity in each of the regimes discussed above are clearly identified in the numerical results presented in Fig. 2.

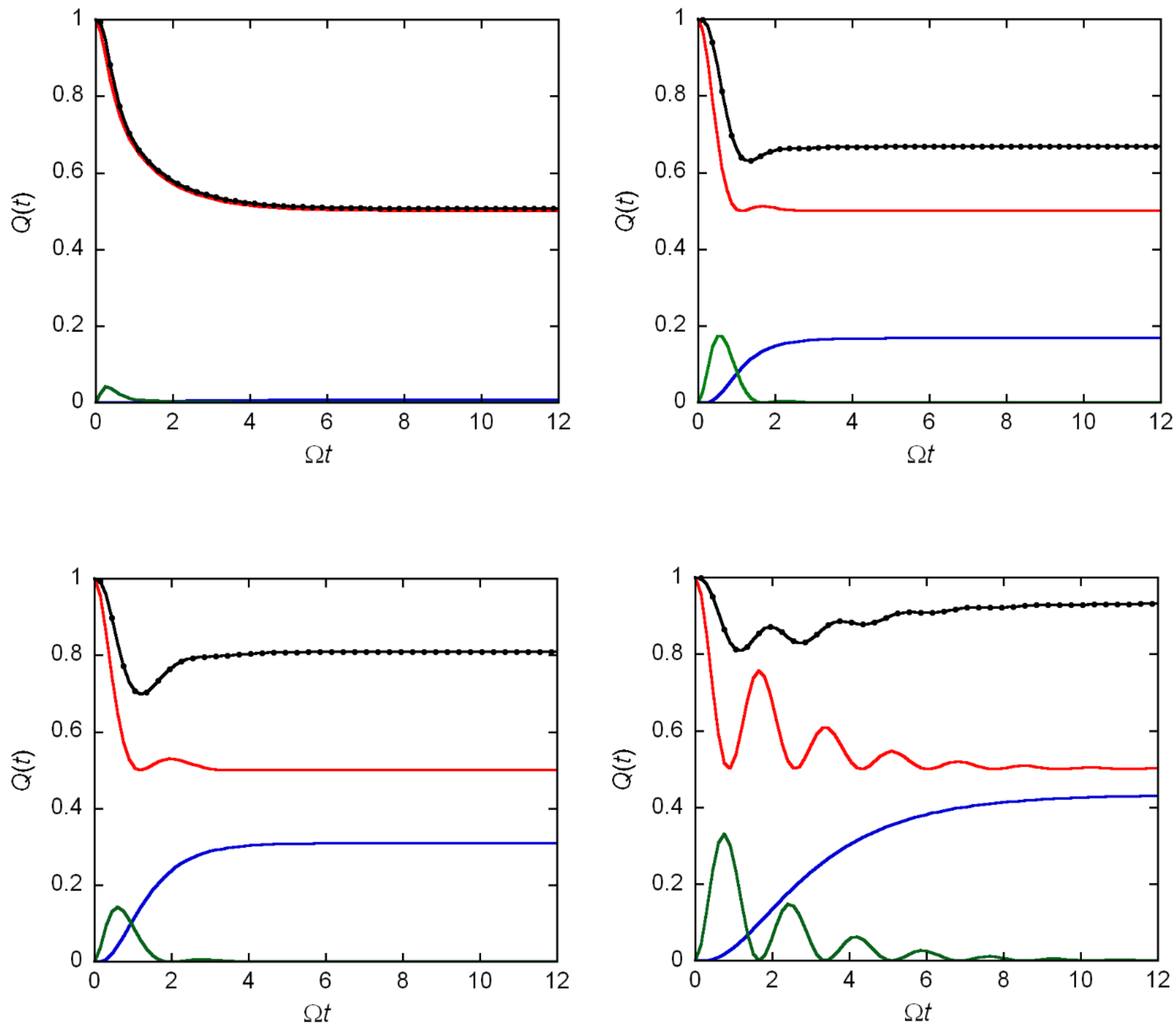

Fig. 2. The three contributions to purity for a two-level system. Red: incoherent term, Q_{incoh}. Blue: term related to eigenstate population difference, $Q_{\text{pop-dif}}$. Green: term related to the time derivative of the site population, $Q_{\text{t-der}}$. The black markers show the sum of these three contributions, which is identical to the calculated purity shown as a black line. Top left: $\xi = 2,\ \omega_c = \Omega,\ \hbar\Omega\beta = 0.125$. Top right: $\xi = 0.3,\ \omega_c = 5\Omega,\ \hbar\Omega\beta = 1$. Bottom left: $\xi = 0.3,\ \omega_c = 7.5\Omega,\ \hbar\Omega\beta = 5$. Bottom right: $\xi = 0.1,\ \omega_c = 7.5\Omega,\ \hbar\Omega\beta = 5$.

These behaviors have important implications for the use of purity as a measure of decoherence. When the system has reached the equilibrium distribution in the presence of its environment, all recurrences in its dynamics have ended, and one would say that quantum coherence has been completely quenched. However, the loss of coherence at equilibrium does not necessarily imply that the purity has decayed to its fully mixed value. In particular, under low-temperature, weak coupling conditions, the equilibrium purity can be almost equal to unity. Similarly, the transient short-time decrease of purity may not even approximately characterize the time length of persisting oscillatory dynamics over which the system remains partially coherent. Thus, these findings caution against the use of purity as a measure of quantum coherence and its destruction.

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