# Master Equation and Control of an Open Quantum System with Leakage

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Given a multilevel system coupled to a bath, we use a Feshbach $P$, $Q$ partitioning technique to derive an exact trace-nonpreserving master equation for a subspace $\mathcal{S}_i$ of the system. The resultant equation properly treats the leakage effect from $\mathcal{S}_i$ into the remainder of the system space. Focusing on a second-order approximation, we show that a one-dimensional master equation is sufficient to study problems of quantum state storage and is a good approximation, or exact, for several analytical models. It allows a natural definition of a leakage function and its control and provides a general approach to study and control decoherence and leakage. Numerical calculations on an harmonic oscillator coupled to a room temperature harmonic bath show that the leakage can be suppressed by the pulse control technique without requiring ideal pulses.

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*Introduction.*—Control of quantum dynamics is of great interest for ‘‘quantum technology industries,’’ such as quantum computing. The control of closed quantum systems is well established and has been extensively studied in areas such as chemical physics [1,2]. Efforts to extend these studies to open systems, where the system interacts with an environment, are now underway [3]. Quantum information processing has already expended considerable effort on open systems. Hence, we anticipate that methods being developed in the latter area may well be useful in the former, and vice-versa [4].

A fundamentally difficult problem in quantum information processing is that of decoherence, i.e., the loss of quantum information in a system due to its interaction with its environment (or ‘‘bath’’) [5,6]. For multilevel systems, such as molecules, the interaction can also cause leakage (i.e., loss of population) from a system subspace of interest, denoted $\mathcal{S}_i$, into the system space outside of $\mathcal{S}_i$. Theoretical strategies for combating such deleterious environmental effects in the absence of natural decoherence free subspaces [6,7] invoke the dynamical control of system-environment interactions by external fields [8–14].

The aim of dynamical control in open systems is to suppress effects of the environment in order to control system processes at will. For example, ideal Bang-Bang (BB) control of decoherence, decay, and leakage [10,11] utilizes idealized zero-width pulses and the Trotter formula to achieve this goal, although higher-order finite-pulse widths have been considered in model cases [12].

Proposals to control decoherence by realistic (nonideal) pulses have often invoked Zwanzig's projection-operator techniques [13,14] resulting in a differential master equation for the density operator of the system up to second order in the system-bath coupling. This equation allows the tractable treatment of complicated quantum dynamical processes, eliminating the ideal zero-width pulses and Trotter formula assumptions and allowing natural dynamical evolution and dynamical control on an equal footing. For example, Ref. [13] provides a unified way to suppress the decoherence of two-level systems by arbitrary fields that control the system-bath interaction.

Below, we consider an $N$ dimensional system ($N$ can be infinite), spanned by the bases $\{|n\rangle\}$, coupled to a bath, and develop procedures to protect quantum information stored in a $d$-dimensional subspace $\mathcal{S}_i$ of the system. Specifically, we use a projection-operator approach to obtain an exact trace-*nonpreserving* master equation for the dynamics of the system subspace $\mathcal{S}_i$. We then introduce a characteristic leakage function which is used to consider dynamics and control of this subspace to second order in the system-bath interaction.

*Zwanzig's projection-operator approach.*—The most general Hamiltonian of the $N$ dimensional system plus the bath is

$$H = H_0 + H_I = H_S + H_B + H_I, \qquad (1)$$

where $H_S$ and $H_B$ are the system and bath Hamiltonians, respectively, and the system-bath interaction is $H_I = \sum_\alpha S_\alpha B_\alpha$, where $S_\alpha$ and $B_\alpha$ are Hermitian system and bath operators. During the dynamics, the information stored in the system subspace of interest $\mathcal{S}_i$ will distribute into the bath and *leak* into the system states outside of $\mathcal{S}_i$ if one does not protect the subspace. Therefore, in order to store the information in $\mathcal{S}_i$, we need to control the system through an interaction with an external field $H_c(t)$, protecting the information during the course of time $t$. The total system Hamiltonian including control is therefore $H_S(t) = H_S + H_c(t)$.

We first derive a *closed* master equation for $\mathcal{S}_i$. Since $\mathcal{S}_i$ opens both to bath and to other system states outside of $\mathcal{S}_i$, effects of leakage have to be considered in deriving a master equation. As usual, the derivation begins in the interaction representation with respect to $H_0$, in which the equation of motion is $\frac{\partial}{\partial t}\rho(t) = -i[H_I(t), \rho(t)] \equiv \mathcal{L}(t)\rho(t)$. Here, the system-bath interaction is $H_I(t)$ in the interaction representation, and the Liouville superoperator $\mathcal{L}(t)$ is defined by this equation [15]. The superprojection operation $\mathcal{P}$ that we seek defines the *relevant* part of the total density matrix [15] $\mathcal{P}\rho(t)$ for our new open system, i.e., the subspace $\mathcal{S}_i$ of the entire system. Specifically, the superprojection operation comprises two commuting parts: a trace over the bath components, and a projecting out of the $\mathcal{S}_i$ subspace from the full $N$-dimensional space of the system. The associated superprojector $\mathcal{P}$ is therefore defined as

$$\mathcal{P}\,\rho = \mathbf{P}\,\mathrm{tr}_B\{\rho\}\mathbf{P}\otimes\rho_B \equiv \eta\otimes\rho_B,$$

where $\mathbf{P} = \sum_{i=0}^{d-1}|i\rangle\langle i|$ denotes a projection onto $\mathcal{S}_i$ and $\eta$ is the relevant part of the total density matrix, which is projected from the total system density matrix $\rho_S = \mathrm{tr}_B\rho$ as $\eta \equiv \mathbf{P}\rho_S\mathbf{P}$. The matrix $\rho_B$ is chosen as the initial state of the bath. The trace of the matrix $\eta$ is not necessarily one, but $\eta$ satisfies a *closed* equation as does $\rho_S$, and plays the same rule as $\rho_S$, but in $\mathcal{S}_i$. That is, an arbitrary system observable acting only on $\mathcal{S}_i$ obeys the relation $O = \mathbf{P}O\mathbf{P}$. The expectation value of the operator $O$ when the system +bath is in state $\rho$ is $\mathrm{tr}\{\rho O\} = \mathrm{tr}\{\mathbf{P}\rho_S\mathbf{P}O\} = \mathrm{tr}\{\eta O\}$, which is the same as the expectation value of the operator $O$ in the state characterized only by $\eta$. Thus, the matrix $\eta$ provides a complete description of the physics in $\mathcal{S}_i$.

Applying the superprojector to the equation of motion for $\rho$ gives a time-local master equation

$$\frac{\partial}{\partial t}\mathcal{P}\rho(t) = \mathcal{K}(t)\mathcal{P}\rho(t), \tag{2}$$

where $\mathcal{K}(t)$ is the time-convolutionless generator [15]. Unlike the usual approach, our new system is a $d$-dimensional subspace of the $N$-dimensional space of the total system. Alternatively, Eq. (2) can be derived by applying the Feshbach projection-operator approach [16,17] to the traditional trace-preserving master equation for $\rho_S$.

Equation (2) is exact and holds for almost [15] all arbitrary systems and interactions, and for initial conditions $\rho(0) = \mathcal{P}\rho(0)$, i.e., where the quantum system is initially within $\mathcal{S}_i$. Since population can flow out of this subspace, the master equation is not trace-preserving. Unfortunately this equation is as difficult to solve as the original equation. Therefore, perturbation expansions are needed in order to apply the result to actual problems.

To second order in the coupling strength of the interaction, $\mathcal{K}(t) = \int_0^t ds\mathcal{P}\mathcal{L}(t)\mathcal{L}(s)\mathcal{P}$. Introducing the explicit expressions for the projection operator and the Liouville superoperator, we can obtain the second-order $d \times d$ dimensional master equation in the interaction representation,

$$\frac{\partial}{\partial t}\eta(t) = -\lambda^2\int_0^t ds\mathbf{P}\,\mathrm{tr}_B[H_I(t),[H_I(s),\eta(t)\otimes\rho_B]]\mathbf{P}. \tag{3}$$

Here, $\lambda H_I$ replaces $H_I$, with the small parameter $\lambda$ introduced to characterize the order of perturbation expansion. For the single component interaction $H_I(t) = S(t)B(t)$, Eq. (3) can be considerably simplified.

*One-dimensional dynamics and the principle of control.*—A primary example is the dynamics, control, and protection of one normalized state $|\phi\rangle$, in the interaction representation, within the $d$-dimensional subspace $\mathcal{S}_i$. [Spontaneous emission, for example, is a case where $|\phi\rangle$ is an energy eigenstate]. In general, $|\phi\rangle$ is a superposition of eigenstates, rather than a single eigenstate, and we can rearrange the bases of $\mathcal{S}_i$ so that $|\phi\rangle$ is one of the new orthonormal basis elements.

Suppose that the initial state $\eta(0) = |\phi\rangle\langle\phi|$. The subsystem evolves according to the closed Eq. (3) with $d = 1$ and, at time $t$, $\eta(t) = b(t)|\phi\rangle\langle\phi|$, where, in general, $b(t)$ is written as

$$b(t) = \exp[-L(t)], \tag{4}$$

with $b(t) \leq 1$ or $L(t) \geq 0$. Substituting $\eta(t)$ into the master Eq. (3) with $d = 1$ gives an analytic expression second order in $\lambda$,

$$L(t) = \lambda^2\int_0^t dsC(s), \tag{5}$$

where

$$C(s) = \int_0^s ds'\sum_{\alpha\beta}[\mathfrak{S}_{\alpha\beta}(s, s-s')\Phi_{\alpha\beta}(s') + \text{H.c.}], \tag{6}$$

and $\mathfrak{S}_{\alpha\beta}(s, s') = \overline{\Delta S_\alpha(s)\Delta S_\beta(s')}$. Here, $\Delta S_\alpha(t) = S_\alpha(t) - \overline{S_\alpha(t)}$ and $\overline{S_\alpha(t)} = \langle\phi|S_\alpha(t)|\phi\rangle$. $\Phi_{\alpha\beta}(t-s) = \mathrm{tr}_B[B_\alpha(t)B_\beta(s)\rho_B]$ is the bath correlation function for multiterm system-bath interactions. Note that $C(s)$ is a linear function of matrix elements $\Phi_{\alpha\beta}$. We term the time-dependent $L(t)$ a *leakage function*, by analogy with the decoherence function [15]. It describes the leakage from $\mathcal{S}_i$ due to the bath and into the space outside of $\mathcal{S}_i$. Higher than second-order effects in the leakage function are included in Eq. (2).

$L(t)$ is a functional of the initial state $|\phi\rangle$ and any added control $H_c$, the latter generally through incident external fields. Given a time $t$, the solution of the variational equation $\delta L(t) = 0$ with respect to the state $|\phi\rangle$ in the absence of $H_c$ yields, to second order, a self protected state. Alternatively, solution to this variational equation with respect to the incident electromagnetic fields for fixed $|\phi\rangle$ provides optimal control fields, to second order, to protect $|\phi\rangle$ against decoherence. Later in this Letter, we address optimizations with respect to the control fields for realistic molecular systems. Optimizations with respect to $|\phi\rangle$ are currently under study.

As in all approximation techniques, the utility of the second-order approximation [Eq. (3)] is examined by comparison with exact cases. We consider two examples.

*Example I: Pure leakage.*—Consider a pure leakage case, where the system is a one-dimensional Harmonic oscillator in which there is no system-bath interaction. The system is described by $H_S = \omega a^\dagger a$ and is polarized by the interaction $H_I = \lambda(a^\dagger + a) \otimes I_B$, where $I_B$ is the unit operator of a bath and $a^\dagger(a)$ is a bosonic creation (annihilation) operator for the harmonic oscillator. For the case of the ground state $|\phi\rangle = |0\rangle$, the exact solution is $b_{\rm ex}(t) = \langle 0|\rho_S(t)|0\rangle = \exp(-\frac{4\lambda^2}{\omega^2}\sin^2\frac{\omega t}{2})$. The second-order solution [Eqs. (4) and (5)] gives the same result. When $|\phi\rangle = |1\rangle$, the exact analytical solution $b_{\rm ex}(t) = (1 - \frac{4\lambda^2}{\omega^2}\sin^2\frac{\omega t}{2})^2 \exp(-\frac{4\lambda^2}{\omega^2}\sin^2\frac{\omega t}{2})$, while $b(t) = \exp(-\frac{12\lambda^2}{\omega^2}\sin^2\frac{\omega t}{2})$. In the superposition case of $|\phi\rangle = \frac{1}{\sqrt{2}} \times (|0\rangle + |1\rangle)$, $b(t) = \exp[-\frac{4\lambda^2}{\omega^2}(\sin^2\frac{\omega t}{2} + \sin^4\frac{\omega t}{2})]$ and the exact solution $b_{\rm ex}(t) = [1 - \frac{4\lambda^2}{\omega^2}(1 - \frac{\lambda^2}{\omega^2})\sin^4\frac{\omega t}{2}] \times \exp(-\frac{4\lambda^2}{\omega^2}\sin^2\frac{\omega t}{2})$. Their second-order expansions are all the same and are essentially equal to one another for $\frac{\lambda^2}{\omega^2} < 0.1$.

*Example II: Spin-bath model within the rotating wave approximation.*—Here, the system Hamiltonian reads $H_0 = \epsilon\sigma^z + \omega a^\dagger a$ and $H_I = \lambda(\sigma^+ a + \sigma^- a^\dagger)$, where $\sigma^\pm = (\sigma^x \pm i\sigma^y)/2$. Let $|\phi\rangle = |0\rangle$ and let the bath be in the state $a^\dagger|V\rangle$, where $|0\rangle$ is the spin-down state and $|V\rangle$ is the vacuum-bath state. Solving the problem exactly gives $b_{\rm ex}(t) = 1 - \sin^2[t\sqrt{(\frac{\epsilon-\omega}{2})^2 + \lambda^2}]\frac{\lambda^2}{\sqrt{(\frac{\epsilon-\omega}{2})^2+\lambda^2}}$. The second-order solution to the master equation is $b(t) = \exp(-\frac{16\lambda^2}{(\epsilon-\omega)^2}\sin^2\frac{(\epsilon-\omega)t}{2})$, which agrees with the exact result in second order. They can also be shown numerically to be in good agreement when $\frac{\lambda^2}{(\epsilon-\omega)^2} < 0.1$.

*Quantum control.*—Within the framework outlined here, the goal of quantum control becomes: given a time $t$ of interest, we computationally seek the solution of the variational equation $\delta L(t) = 0$ with respect to $H_c$, with the inclusion of any physical constraints of the control, The result is the control functional $H_c(s')$. Note that this approach does not just optimize $C(s)$ itself at time $s$, but rather includes the history of the time evolution of $C(s)$.

Although idealized BB control provides a possible mathematical solution, of $L(t) = 0$, it requires unrealistic (zero-width) pulses. Hence, our focus is to replace idealized control by an approximate variational or numerical solution that minimizes $L(t)$ under realistic pulse energy and pulse width constraints.

*Harmonic oscillator coupled to a harmonic bath.*—As an application of this framework, consider as the system the harmonic approximation (frequency $\Omega$) to a Morse oscillator [18,19] for, e.g., molecular iodine, which allows us to employ a simplifying symmetry. The total Hamiltonian, where the bath has $\ell$ oscillators, is $H = \Omega a^\dagger a + \sum_{j=1}^{\ell}\omega_j a_j^\dagger a_j + \lambda SB$, where $B = \sum\alpha(\omega_j)(a_j + a_j^\dagger)$. The interaction is separable with $S(t) = e^{-i\Omega t}a + e^{i\Omega t}a^\dagger$ and the bath correlation function is $\Phi(t) = \sum\alpha^2(\omega_j)[1 + n(\omega_j)e^{-i\omega_j t} + n(\omega_j)e^{i\omega_j t}]$ where $n_j = 1/[\exp(\beta\omega_j) - 1]$. At low energy, $I_2$ vibrational motion is harmonic, with $\Omega = 213.7\ \text{cm}^{-1}$ and $\alpha(\omega_j) = \sqrt{\frac{\omega_j\omega_d(1-e^{-5})}{40\pi\ell}}$ with $\omega_j = -\frac{\omega_d}{2}\ln(1 - \frac{j(1-e^{-5})}{\ell})$, where $\omega_d$ is the cutoff frequency at $j = \ell$.

As an example, consider a superposition of the eigenstates of the system harmonic oscillator as the state in need of protection in a bath at room temperature. Such states would be of interest, for example, in pump-dump coherent control scenarios [1,2] where this is the initially pumped state. In that case, one would be interested in maintaining this state over time scales of ≈ 700 fs, the system decoherence time [19].

Figure 1(a) shows $L(t)$ for the parameters shown in the figure caption. In some cases, $\mathfrak{S}(s, s - s')$ may be a function of $(s - s')$ only, in which case $L(t) \propto t$, shown as the dashed line in Fig. 1(a). This linear dependence on $t$ is similar to that of the usual decoherence function [15] in the long time limit. However, at shorter times, the dependence of $\mathfrak{S}(s, s - s')$ on $s$ is seen to contribute non-negligibly, leading to oscillatory $L(t)$. Below, we will numerically study the behavior of the leakage function in the short-

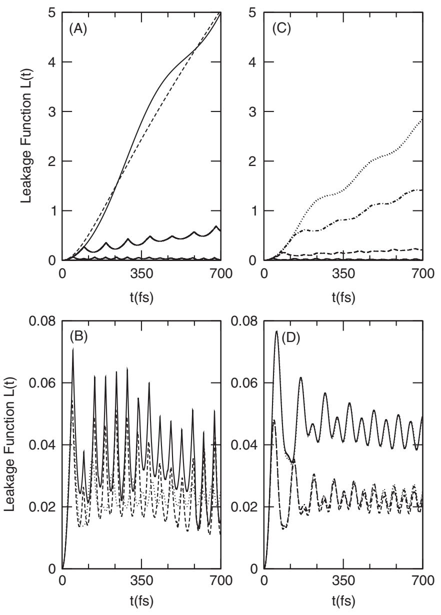

FIG. 1. (a) (upper solid curve) $L(t)$ in units of $\lambda^2$ for an Harmonic oscillator, in the initial state, $|\phi\rangle = \frac{1}{\sqrt{2}}(|0\rangle + |1\rangle)$ coupled to a bath of 1000 harmonic oscillators at $T = 300$ K. Here, $\omega_d = 0.01(fs)^{-1} \approx 1.5\ \Omega$. The dashed curve shows the dominant part, as discussed in the text. The three lower solid curves show $L(t)$ with $\Delta = 0$, $\phi_0 = \pi$ and $\tau = \frac{\pi}{20\Omega}, \frac{\pi}{10\Omega}$ and $\frac{\pi}{5\Omega}$, in the order from bottom to top. (b) $L(t)$ with $\tau = \frac{\pi}{10\Omega}$ and $\phi_0 = \pi$, but different values of $\Delta$. Solid curve $\Delta = \tau/5$; Dashed curve: $\Delta = \tau/2$; Dotted curve: $\Delta = \tau$. (c) $L(t)$ with $\tau = \frac{\pi}{10\Omega}$ and $\Delta = \frac{\tau}{2}$ but different pulse intensities, $\phi_0 = \frac{\pi}{10}, \frac{\pi}{5}, \frac{\pi}{2}$ from top to bottom. (d) two lower curves, $L(t)$ with $\phi_0 = \pi/100$, $\tau = \tau_0/100$ ($\tau_0 = \frac{\phi_0}{10\Omega}$) and $\phi_0 = \pi/10$ and $\tau = \tau_0/10$ all with $\Delta = \tau/2$. They have the same value of $\Omega_c$. The upper curves have the same values of $\phi_0$ and $\tau$ but different signs of $\phi_0$.

time region to determine the extent to which leakage can be controlled.

*Control.*—Physically, the origin of the control is that the frequency of the system (here, an harmonic oscillator) is periodically, dynamically, Stark shifted by the alternating field. To this end, we employ the realistic control Hamiltonian $H_S(t) = [\Omega + f(t)]a^\dagger a$, which results from strong laser pulses acting on electrons that induce an additional time-dependent nuclear potential. We model the control function as a periodic rectangular interaction: $f(t) = 0$ for regions other than $n\tau - \Delta < t < n\tau$, $n$ integer. Inside these regions, $f(t)$ is defined so that $\phi_0 = \int_{\tau-\Delta}^{\tau} f(t)dt$. That is, for nonzero $\Delta$, $f(t) = \phi_0/\Delta$ over the control interval, and for $\Delta = 0$, $f(t) = \delta(t - n\tau)$. The functional form contains three main control parameters: the time interval $\tau$, the pulse width $\Delta$, and the interaction intensity $\phi_0$. For comparison with realistic pulses, we show $L(t)$ with ideal impulsive phase modulation ($\Delta = 0$) in the three lower curves in Fig. 1(a). Clearly, the shorter the control interval, the better the control.

Figure 1(b) shows $L(t)$ with fixed $\tau$ and $\phi_0$, but with different pulse widths $\Delta$. The results show that the quality of the control is only weakly dependent on the pulse width. For example, the control is excellent even if the width of the pulse is equal to the control interval $\tau$. In this case, the control is equivalent to adding a constant frequency $\Omega_c = \phi_0/\tau$ to the harmonic oscillator frequency, i.e., shifting the system frequency by $\Delta/\tau = 1$ means shifting the system frequency to $(\Omega + \Omega_c)$. If $(\Omega + \Omega_c) > \omega_d$ (the cutoff frequency of the bath), which is the case in this figure, the function $\Im(s, s - s')$ oscillates faster than the rate of decay of the bath $\Phi(s)$. The integral, $C(s)$, of the product of the two functions generally oscillates around zero so that $L(t)$ is reduced at any time $t$. The way to achieve this goal (e.g., see Ref. [13]) is to increase the interaction of the pulse $\phi_0$ or decrease the pulse width $\tau$ in order to increase $\Omega_c = \phi_0/\tau$.

The dependence on the intensity is also of interest, as shown in Fig. 1(c). The quality of control is seen to decrease with decreasing intensity.

Finally, we consider the effects due to the different signs of $\phi_0$. The two lower curves in Fig. 1(d) correspond to results for different $\phi_0$ (or $\tau$) but the same $\Omega_c = \phi_0/\tau$. The upper curves have the same values but different signs. They show similar suppression, implying that the quality of suppression depends primarily on $\Omega_c$. Hence, the above discussions are also valid for the negative values of $\phi_0$ as shown in the two upper curves.

For the case of a diatomic molecule, we note that the ac Stark effect induced by an external laser field interacting with the electrons decreases (or increases) $\Omega$, an effect termed "bond softening (or hardening)." *Ab initio* calculations [20] show that, in the softening case, the frequency can be reduced by 10% for $H_2^+$ in a strong laser field. In the case of hardening, if the frequency can be enhanced by $p$ percent so that if $(1 + p)\Omega > \omega_d$, then leakage control will be effective. This is expected to be a considerable technical challenge.

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