Dynamical cooling of a single-reservoir open quantum system via optimal control

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(Dated: October 6, 2010)

Based on an exact description of open quantum systems in terms of stochastic Liouville-von Neumann equations optimal control is investigated without rotating-wave or Markovian approximations. Within this scheme we generalize Krotov's iterative algorithm, preserving its monotonic convergence. This formalism is applied to the problem of controlling a particle in a harmonic potential whose thermal bath is characterized by an ohmic spectrum. Interestingly, optimal control can modify the quantum dissipative dynamics to the point where its entropy change turns negative. We provide an example where translational motion is cooled without any involvement of internal degrees of freedom.

PACS numbers: 02.30.Yy, 03.67.-a, 05.70.Ln

Introduction. The control of quantum dynamics or the accurate preparation of a prescribed quantum state by a tailored time-dependent field is a task of key importance in quantum physics and related disciplines. For instance, the creation of ultrashort light pulses of well-defined shape has led to the optical control of ultrafast processes in quantum chemistry [1]. In the last decade, for the successful realization of schemes for quantum information processing (QIP), optimal control theory (OCT) has become a key ingredient in strategies to tame the destructive effect of decoherence and other imperfections or to speed up operations. Accordingly, OCT has been used to improve the performance of simple quantum gates [2–6] or to directly construct more complex operations [7]. In several implementations it is also necessary to transport computational states between different operational sites. This usually requires the optimal control not only of the quantum bits themselves but also of auxiliary degrees of freedom, typically translational motion.

An important issue for all types of implementations in QIP is the interaction of the relevant quantum system with a fluctuating environment. Atomic systems such as a neutral atom or ion in an electromagnetic trap, are exposed to fluctuations of the (comparatively hot) trapping chip surface [8]. In solid state devices diffusing charges and electromagnetic fluctuations affect the fidelity quite substantially [9]. Despite the ubiquitous nature of such interactions, however, optimal control schemes have treated them mostly by heuristic or approximate methods so far. A simple strategy has been followed in [10], where environmental influences were taken into account by assuming an initial thermal state for translational motion while neglecting the effect of the environment completely during its dynamics. Fully dynamical approaches based on standard Markovian Master equations, however, become inconsistent for strong control fields unless additional field-dependent memory terms in the dissipator are introduced [11, 12]. The rotating wave approximation (RWA), usually invoked to ensure complete positivity in quantum master equations, is also known to be unreliable in driven systems [11]. This observation raises questions on the fundamental properties of driven quantum dynamics since the current understanding of quantum statistical aspects such as entropy production seems to depend largely on the RWA [13].

In this Letter we describe quantum dissipation through non-Markovian stochastic Liouville-von Neumann (SLN) equations [14] instead of Master equations. This representation of quantum dissipation, closely related to the non-Markovian Schrödinger equation of Diósi et al. [15], is completely equivalent to influence functionals in the path integral approach [16]. It is independent of the free dynamics of the undamped system and, in particular, remains unchanged when external driving is introduced.

The non-Markovian nature of the environment, however, does not lead to explicit time-retarded self-interactions, but it appears only through correlations of complex-valued noise forces. Its use has been demonstrated for both discrete quantum systems [14, 17] and problems involving translational motion [18]. The resulting equations of motion are formally local in time, as required by most standard optimal control algorithms. SLN equations thus provide the basis for an approach to optimal control which is both realistic and practical even in the presence of low-temperature, non-Markovian quantum noise. We outline the general mathematical and algorithmic structure and address as a specific example the control of a harmonic degree of freedom such as the motional state of an atom or ion under electromagnetic confinement or the low energy dynamics of the phase in a Josephson junction. Moreover, the entropy production induced by the control field is analyzed and shown to deviate strongly from predictions based on the standard RWA-Lindblad master equation.

Open-system dynamics and control algorithm. We consider a quantum system characterized by a Hamiltonian $\hat{H}_{\rm s}$ representing a potential model, augmented by a term $\hat{H}_{\rm c}(t)$ describing the influence of time-dependent control

fields. Additionally, we assume a dissipative environment, which can be characterized by the power spectrum of its fluctuations. The SLN equation for the controlled dissipative dynamics

$$\begin{aligned}\dot{\hat{\varrho}}_{\xi,\nu}(t) &= -\frac{i}{\hbar}[\hat{H}_{\rm s},\hat{\varrho}_{\xi,\nu}(t)] - \frac{i}{\hbar}[\hat{H}_{\rm c},\hat{\varrho}_{\xi,\nu}(t)] \\ &\quad + \frac{i}{\hbar}\xi(t)[\hat{q},\hat{\varrho}_{\xi,\nu}(t)] + \frac{i}{2}\nu(t)\{\hat{q},\hat{\varrho}_{\xi,\nu}\} \quad (1)\end{aligned}$$

contains the Gaussian, complex-valued stochastic variables $\xi(t)$ and $\nu(t)$ with the properties (i) the autocorrelation of ξ matches the quantum noise of the reservoir, (ii) the cross-correlations of ξ and ν match the dynamical response of the environment and (iii) the autocorrelation function $\langle\nu(t)\nu(t')\rangle$ vanishes identically [14].

In the case of an ohmic environment, to be discussed below, a partial average over ν simplifies Eq. (1) to [19]

$$\begin{aligned}\dot{\hat{\varrho}}_{\xi}(t) &= \hat{\mathfrak{L}}\hat{\varrho}_{\xi}(t) := -\frac{i}{\hbar}[\hat{H}_{\rm s},\hat{\varrho}_{\xi}(t)] - \frac{i}{\hbar}[\hat{H}_{\rm c},\hat{\varrho}_{\xi}(t)] \\ &\quad - \frac{i}{2\hbar}\gamma_0[\hat{q},\{\hat{p},\hat{\varrho}_{\xi}(t)\}] + \frac{i}{\hbar}\xi(t)[\hat{q},\hat{\varrho}_{\xi}(t)]\,, \quad (2)\end{aligned}$$

where γ_0 is the damping rate of a Brownian particle in the environment. The physical density matrix is a stochastic average of the form $\hat{\varrho}(t) = \mathbb{E}[\hat{\varrho}_{\xi}(t)]$.

At the price of introducing an explicit noise variable $\xi(t)$, Eq. (2) represents the *exact non-Markovian* dynamics in terms of a stochastic ensemble with *time-local* equations of motion. All memory effects are contained in the temporal correlations of $\xi(t)$. This consideration of intrinsic non-Markovian features of the reservoir differs from the approach of Rebentrost et al. [20], where non-Markovian fluctuations result from the characteristic timescales of a small additional subsystem mediating energy exchange between the system of interest and a Markovian environment. Equation (2) becomes Markovian in the high-temperature limit, where it reduces to the master equation of Caldeira and Leggett [21].

We now generalize the monotonically convergent algorithm of Krotov [22, 23] to optimization objectives defined through averages of the stochastic variable $\hat{\varrho}_{\xi}(t)$. The objective associated with a final-state observable $\hat{M}$ has the form of a quantum statistical expectation value at the end time T, $\mathrm{tr}\{\hat{M}\hat{\varrho}(T)\}$. Assuming the control Hamiltonian $\hat{H}_{\rm c}(t)$ to be a function of a vector of control fields $\mathbf{u}(t) = (u_1(t),\ldots,u_N(t))$, the form of the (terminal) objective functional

$$\mathsf{F}[\mathbf{u}(t),\{\hat{\varrho}_{\xi}(t)\}] = \mathbb{E}[\mathrm{tr}\{\hat{M}\hat{\varrho}_{\xi}(T)\}] \quad (3)$$

depends on the control fields only implicitly through the equation of motion (2).

The objective functional F may be replaced by the expectation value of an extended functional L_{ξ} which coincides with F for admissible processes, i.e., pairs $(\mathbf{u}(t),\{\hat{\varrho}_{\xi}(t)\})$ which obey the equation of motion (2). Following Ref. [23], we choose

$$\begin{aligned}\mathsf{L}_{\xi}[\mathbf{u}(t),\hat{\varrho}_{\xi}(t);\phi_{\xi}] &:= \mathsf{G}_{\xi}(\hat{\varrho}_{\xi}(T)) - \int_0^T \mathrm{d}t\mathsf{R}_{\xi}(t,\mathbf{u}(t),\hat{\varrho}_{\xi}(t)) \\ &\quad - \phi_{\xi}(0,\hat{\varrho}_{\xi}(0)), \quad (4)\end{aligned}$$

where $\phi_{\xi}(t,\hat{\varrho}_{\xi}(t))$ is a real-valued function of the time t, the noise realization $\xi(t)$, and the state sample $\hat{\varrho}_{\xi}(t)$, and

$$\mathsf{G}_{\xi}(\hat{\varrho}_{\xi}(T)) := \mathrm{tr}\{\hat{M}\hat{\varrho}_{\xi}(T)\} + \phi_{\xi}(T,\hat{\varrho}_{\xi}(T)), \quad (5)$$

$$\mathsf{K}_{\xi}(t,\mathbf{u}(t),\hat{\varrho}_{\xi}(t),\,\cdot\,) := \mathrm{tr}\{\,\cdot\,\hat{\mathfrak{L}}\hat{\varrho}_{\xi}(t)\}, \quad (6)$$

$$\begin{aligned}\mathsf{R}_{\xi}(t,\mathbf{u}(t),\hat{\varrho}_{\xi}(t)) &:= \mathsf{K}_{\xi}(t,\mathbf{u}(t),\hat{\varrho}_{\xi}(t),\frac{\partial}{\partial\hat{\varrho}_{\xi}}\phi_{\xi}(t,\hat{\varrho}_{\xi}(t))) \\ &\quad + \frac{\partial}{\partial t}\phi_{\xi}(t,\hat{\varrho}_{\xi}(t)). \quad (7)\end{aligned}$$

For admissible processes, L_{ξ} does not depend on the choice of ϕ_{ξ}, which may be adjusted as the iterative algorithm progresses. The following property of ϕ_{ξ} must be chosen for Krotov's algorithm: For any admissible process, L_{ξ} is at a maximum with respect to $\hat{\varrho}_{\xi}$ for any function $\mathbf{u}(t)$. This amounts to the two conditions

$$\mathsf{G}_{\xi}(\hat{\varrho}_{\xi}^{(j)}(T)) = \max_{\hat{\varrho}_{\xi}}\{\mathsf{G}_{\xi}(\hat{\varrho}_{\xi}(T))\}, \quad (8)$$

$$\mathsf{R}_{\xi}(t,\mathbf{u}^{(j)}(t),\hat{\varrho}_{\xi}^{(j)}(t)) = \min_{\hat{\varrho}_{\xi}}\{\mathsf{R}_{\xi}(t,\mathbf{u}^{(j)}(t),\hat{\varrho}_{\xi}(t))\}. \quad (9)$$

Each iteration cycle j proceeds in two steps: First a time-local update law (a map $\mathbf{u}^{(j)}(t) \mapsto \mathbf{u}^{(j+1)}(t)$) is chosen which improves the objective for any ensemble of states $\{\hat{\varrho}_{\xi}\}$, including the (yet unknown) updated ensemble. Consistently propagating the states using Eq. (2) and the updated control then yields a new admissible process. Finally, the function ϕ_{ξ} is adapted to the new process $(\mathbf{u},\{\hat{\varrho}_{\xi}\})$.

For the linear dynamics of Eq. (2), it turns out that the function $\phi_{\xi}(t,\{\hat{\varrho}_{\xi}\})$ appears in the conditions (8) and (9) only in form of its derivative with respect to the state, i.e., the co-state $\hat{\Lambda}_{\xi} = \partial\phi_{\xi}/\partial\hat{\varrho}_{\xi}$. After promoting $\hat{\Lambda}_{\xi}$ to the state of a dynamical variable, the extremal conditions lead to the co-state equation of motion

$$\begin{aligned}\dot{\hat{\Lambda}}_{\xi}(t) &= -\hat{\mathfrak{L}}^{\dagger}\hat{\Lambda}_{\xi}(t) = -\frac{i}{\hbar}[\hat{H}_{\rm s},\hat{\Lambda}_{\xi}(t)] - \frac{i}{\hbar}[\hat{H}_{\rm c},\hat{\Lambda}_{\xi}(t)] \\ &\quad - \frac{i}{2\hbar}\gamma_0\{\hat{p},[\hat{q},\hat{\Lambda}_{\xi}(t)]\} + \frac{i}{\hbar}\xi(t)[\hat{q},\hat{\Lambda}_{\xi}(t)] \quad (10)\end{aligned}$$

accompanied by $\hat{\Lambda}_{\xi}(T) + \hat{M} = 0$ resulting from (8).

Now, following [23] one determines that

$$u_k^{(j+1)}(t) = u_k^{(j)}(t) + \frac{1}{\lambda_k(t)}\mathbb{E}\left[\mathrm{tr}\left\{\hat{\Lambda}_{\xi}^{(j)}(t)\frac{\partial\hat{\mathfrak{L}}}{\partial u_k}\hat{\varrho}_{\xi}^{(j+1)}(t)\right\}\right] \quad (11)$$

is a suitable update law for $k = 1,\ldots,N$. Here $\lambda_k(t)$ may be adjusted to tune convergence properties. This update

always reduces the objective, $\mathbb{E}[\mathsf{F}_\xi[\mathbf{u}^{(j+1)}(t),\hat{\varrho}_\xi^{(j+1)}(t)]] \leq \mathbb{E}[\mathsf{F}_\xi[\mathbf{u}^{(j)}(t),\hat{\varrho}_\xi^{(j)}(t)]]$ which can be seen as follows:

$$\begin{aligned}\mathbb{E}[\mathsf{L}_\xi[\mathbf{u}^{(j)}(t),\hat{\varrho}_\xi^{(j)}(t),\phi_\xi]] - \mathbb{E}[\mathsf{L}_\xi[\mathbf{u}^{(j+1)}(t),\hat{\varrho}_\xi^{(j+1)}(t),\phi_\xi]] \\ = \Delta_1+\Delta_2+\Delta_3, \qquad (12)\end{aligned}$$

where

$$\begin{aligned}\Delta_k \;=\; \int_0^T \mathrm{d}t\mathbb{E}[\mathsf{R}(t,\mathbf{u}^{(j+k-1)}(t),\hat{\varrho}_\xi^{(j+1)}(t)) \\ -\mathsf{R}(t,\mathbf{u}^{(j)}(t),\hat{\varrho}_\xi^{(j+k-1)}(t))] \qquad (13)\end{aligned}$$

for $k=1,2$ and $\Delta_3 = \mathbb{E}[\mathsf{G}(\hat{\varrho}_\xi^{(j)}(T)) - \mathsf{G}(\hat{\varrho}_\xi^{(j+1)}(T))]$. Now $\Delta_1 \geq 0$ and $\Delta_3 \geq 0$ follow from Eq. (9) and Eq. (8), respectively, whereas $\Delta_2 \geq 0$ is a property of the update law [23]. We have thus demonstrated monotonicity for the modified algorithm, which ensures convergence for an arbitrary initial guess.

Application. As a generic model we consider a harmonic oscillator, i.e., $\hat{H}_\mathrm{s} = \frac{\hat{p}^2}{2\,m} + \frac{m\omega^2}{2}\hat{q}^2$, which is subject to an additional quadratic potential with two independent control fields, $\hat{H}_\mathrm{c}(t) = -u_1(t)\hat{q} + \frac{u_2(t)}{2}\hat{q}^2$. This situation is not only a model for typical realizations as e.g. trapped atoms or ions or low energy dynamics of Josephson junctions, but also a non-trivial application of the non-Markovian control formalism due to the quadratic coupling of the system coordinate to the field $u_2(t)$. We emphasize that Eq. (2) is completely general and not restricted to quadratic potentials.

As initial states we consider Gaussian wavepackets, conveniently parameterized by the first and second cumulants (means and variances) $\langle\hat{q}\rangle_c$, $\langle\hat{p}\rangle_c$, $\langle\hat{q}^2\rangle_c$, $\langle\hat{p}^2\rangle_c$ and by $\langle\frac{1}{2}(\hat{q}\hat{p}+\hat{p}\hat{q})\rangle_c$. Under the equation of motion (2), the samples $\hat{\varrho}_\xi$ remain Gaussian, which allows us to re-phrase the equation of motion (2) as a system of five ordinary differential equations. A similar consideration holds for the co-state dynamics (10) if a maximal overlap with a Gaussian target state is chosen as optimization objective, i.e., $\hat{M} = 1-\hat{A}$, where $\hat{A} = |\alpha\rangle\langle\alpha|$ projects onto a coherent state. We thus obtain closed equations of motion for the first two cumulants for the propagation of both the state $\hat{\varrho}_\xi(t)$ and the co-state $\hat{\Lambda}_\xi(t)$. While the effect of the linear control $u_1(t)$ alone is obviously given by linear response theory, the dynamical squeezing through a time-dependent $u_2(t)$ leads to non-trivial dynamics, as does the combined action of both controls. We have explored these effects numerically, computing the expectation values in Eq. (11) explicitly through a large number of samples (typically 10^4). This has the advantage of being justified from first principles, without resorting to approximations of the dynamics. It reconciles the intrinsically non-Markovian character [16] of quantum dissipation with the requirement of time-local equations of motion for both dynamical state and co-state in optimal control theory.

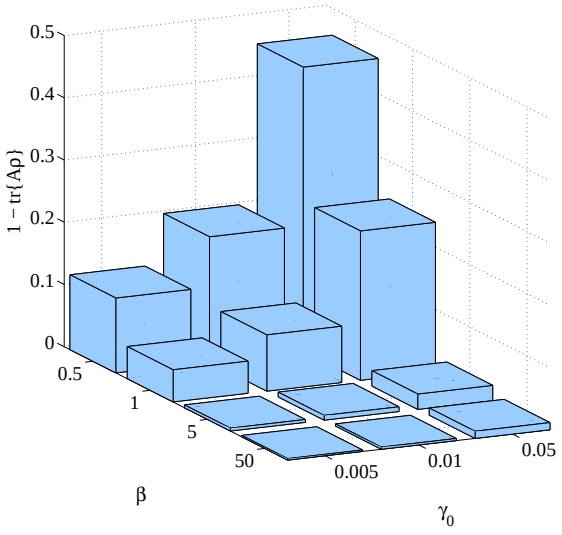

Figure 1: (color online) Control of an open quantum system with linear and parametric driving: Remaining error $1-\mathrm{tr}\{\hat{A}\hat{\varrho}(T)\}$ for different damping rates γ_0 and different inverse thermal energies of the reservoir β.

In the following, we use natural units ($\hbar\omega$ for energies, $\sqrt{\hbar/(m\omega)}$ for lengths, $\sqrt{\hbar m\omega}$ for momenta) and choose a minimal-uncertainty wavepacket centered around $q=1$ and $p=0$ as both initial and target state. Values of the temperature and the damping constant are chosen in the range typical of superconducting solid-state devices [9]. The propagation time $T=20$ is about the same order of magnitude as the relaxation time in the examples to be discussed.

Figure 1 shows how optimal control drives the dissipative dynamics towards the objective functional for varying environmental parameters. For thermal energies at or above the level spacing, the optimized final state is still visibly distinct from the target state even for fairly weak damping. At low temperatures, this disparity virtually vanishes, as evidenced by a numerical value of $1-\mathrm{tr}\{\hat{A}\hat{\varrho}(T)\} \approx 0.0023$ for the shortest column. The algorithmic property of monotonic convergence is confirmed by our numerical results.

Quantum dissipation invariably creates mixed states in the subsystem of interest. But can optimal control prevent an increase of entropy? To investigate this question, we choose the oscillator ground state as target and prepare both system and environment as thermal states with equal inverse temperature $\beta=1$. Since the effect of the force $u_1(t)$ amounts to a unitary transform of the final state, in the following we consider only the control field $u_2(t)$. With the expression

$$\begin{aligned}S(\hat{\varrho}) \;&=\; f\left(\sqrt{\langle q^2\rangle_c\langle p^2\rangle_c - \langle pq+qp\rangle_c^2/4}\right)\,, \qquad (14)\\ f(x) \;&=\; \left(x+\tfrac{1}{2}\right)\log\left(x+\tfrac{1}{2}\right) - \left(x-\tfrac{1}{2}\right)\log\left(x-\tfrac{1}{2}\right)\end{aligned}$$

for the von Neumann entropy of a Gaussian state [24], we

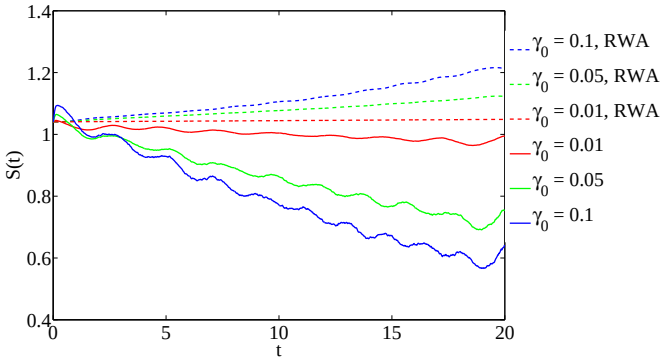

Figure 2: (color online) An open quantum system initially equilibrated with its surroundings loses entropy under an optimized control field (solid). This result is not reproduced within an RWA approximation (dashed).

obtain the remarkable result that a time-dependent control field can modify dissipative dynamics to the point where its entropy change turns negative (Fig. 2). Likewise, the subsystem energy of the final state decreases below its original thermal value. This strongly indicates the interpretation of our result as a *dynamical cooling effect* (with the caveat that the final state may be not considered a thermal state with a uniquely defined temperature). The simplistic approach of adding $H_c(t)$ to the system Hamiltonian *after* performing the RWA fails in this scenario (Fig. 2, dashed lines). Moreover, the described phenomenon intimately depends on the interplay of non-Markovian fluctuations and control field and cannot be reproduced within a RWA-Markov-type of approximation. In contrast to recent proposals for quantum refrigerators [25, 26], which rely on intricate band or level structures, we have chosen a model with a very basic structure. The cooling effect found here seems to arise from general properties of driven dynamics, not from specific features of the system. We also note that no internal degree of freedom is needed for the effect to occur.

Conclusions. Based on the dissipative dynamics in terms of exact SLN equations, we have introduced a version of Krotov's optimal control algorithm adapted to non-Markovian environmental fluctuations. First implementations on a harmonic degree of freedom subject to two control fields illustrate the performance of the algorithm. Efficient computations are feasible for environmental couplings from small values up to damping corresponding to a quality factor as low as $Q \approx 10$. This allows applications to solid state devices such as superconducting circuits with Josephson junctions and condensed-matter phenomena such as reactive dynamics of small molecules in a solvent or on a surface. Extensions to systems with anharmonic potentials and more complex spectral bath densities are currently investigated, relevant e.g. to the emerging field of ultracold quantum chemistry. Optimal control of a dissipative quantum system can extract entropy from a system initially at the same temperature as its environment. This cooling effect in a generic system without special structural features should be of importance for mesoscopic quantum refrigerators.

Acknowledgements. We gratefully acknowledge helpful conversations with S. Montangero and M. Murphy. Financial support was provided by the Postgraduate Scholarship Program of the State of Baden-Württemberg (R.S.), DFG (SFB569 – J.T.S., J.A., SFB/TRR21 – A.N., T.C.), EU (Marie Curie FP7-IEF – A.N., IP-AQUTE, PICC – T.C.) and Ulm University and UUG (A.N.).

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