Quantum control of dissipative systems

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Abstract

We study the effect of dissipation, i.e., uncontrollable interactions of a quantum system with the environment, on one's ability to control the system. In particular we show that dissipation, although often considered undesirable, opens up unique possibilities for quantum control by removing the constraint of unitary evolution, which restricts the set of reachable states and imposes bounds on the optimization of observables.

1 Density operators and mixed quantum states

In pure-state quantum mechanics the state of the system is usually represented by a (normalized) wavefunction $|\Psi\rangle$, which is a (unit) vector in a Hilbert space $\mathcal{H}$. For dissipative systems the state must be represented by a *density operator* ρ acting on the Hilbert space $\mathcal{H}$ instead. If the system is in the pure state $|\Psi\rangle$ then ρ is simply the projector onto this state, i.e., $\rho = |\Psi\rangle\langle\Psi|$. However, the density operator formalism enables us to deal with more general states of the system such as (statistical) ensembles of quantum states. Given a quantum system where a fraction w_1 of the system is in the state $|\Psi_1\rangle$, a fraction w_2 in the state $|\Psi_2\rangle$, etc., we can represent the state by a density operator $\rho = \sum_{n=1}^{N} w_n |\Psi_n\rangle\langle\Psi_n|$, where $0 \leq w_n \leq 1$ and $\sum_{n=1}^{N} w_n = 1$. The states of the ensemble must be orthonormal. Hence, $N \leq \dim \mathcal{H}$, but we may assume $N = \dim \mathcal{H}$ since we can enlarge a smaller set of independent quantum states to form a basis of the Hilbert space $\mathcal{H}$ by adding states with probability $w_n = 0$. A density operator with rank greater than one represents a non-trivial ensemble, called a mixed state, which cannot be expressed as a wavefunction.

Let E_n be the eigenvalues of the internal Hamiltonian H_0 of the system and $|n\rangle$ be the corresponding eigenstates such that $H_0|n\rangle = E_n|n\rangle$. We shall only consider quantum systems with a finite number of discrete energy levels here. The energy levels may be degenerate but we assume that the eigenstates $|n\rangle$ are chosen such that they form a complete orthonormal set for $\mathcal{H}$. We can expand the density operator ρ with respect to the eigenstates $|n\rangle$

$$\rho = \sum_{n=1}^{N} \rho_{nn} |n\rangle\langle n| + \sum_{n=1}^{N} \sum_{n'>n} (\rho_{nn'} |n\rangle\langle n'| + \rho^*_{nn'} |n'\rangle\langle n|). \tag{1.1}$$

The diagonal elements ρ_{nn} in this matrix representation of ρ determine the populations of the energy eigenstates $|n\rangle$, while the off-diagonal elements $\rho_{nn'}$ $(n \neq n')$ determine the coherences between the eigenstates. The latter distinguish coherent superpositions of energy eigenstates $|\Psi\rangle = \sum_{n=1}^{N} c_n |n\rangle$ from statistical ensembles of energy eigenstates $\rho = \sum_{n=1}^{N} w_n |n\rangle\langle n|$.

2 Dynamics of dissipative systems

For a non-dissipative system the evolution of the density matrix ρ is governed by $\rho(t) = U(t,t_0)\rho(t_0)U(t,t_0)^\dagger$, where the evolution operator $U(t,t_0)$ satisfies the Schrodinger equation $\mathrm{i}\hbar\frac{d}{dt}U(t,t_0) = H[\vec{f}(t)]U(t,t_0)$. ρ also satisfies the *quantum Liouville equation*

$$\mathrm{i}\hbar\dot{\rho} = [H[\vec{f}(t)],\rho] = H[\vec{f}(t)]\rho - \rho H[\vec{f}(t)] \quad \text{with} \quad H[\vec{f}(t)] = H_0 + \sum_{m=1}^{M} f_m(t)H_m, \tag{2.2}$$

where H_0 is the internal Hamiltonian, H_m $(1 \le m \le M)$ is the interaction Hamiltonian for the field f_m, and H is the total Hamiltonian of the (non-dissipative) control system.

When a quantum system interacts with the environment, two types of dissipation occur: phase relaxation and population relaxation. Phase relaxation or dephasing results when interactions of the system with the environment destroy the phase correlations between quantum states, which leads to a decay of the off-diagonal elements of the system's density matrix:

$$\mathrm{i}\hbar\dot{\rho}_{kn}(t) = ([H,\rho])_{kn} - \mathrm{i}\hbar\Gamma_{kn}\rho_{kn} \tag{2.3}$$

where Γ_{kn} (for $k \neq n$) is the dephasing rate between $|k\rangle$ and $|n\rangle$. Population relaxation occurs, for instance, when an excited state $|n\rangle$ with $E_n > E_1$ spontaneously emits a photon and decays to a less excited quantum state $|k\rangle$. It affects both the populations of the energy eigenstates $|n\rangle$, i.e., the diagonal elements of the density matrix ρ, and the coherences, i.e., the off-diagonal elements. The rate γ_{kn} of population relaxation from state $|n\rangle$ to $|k\rangle$ depends on the lifetime of the excited state, and in case of multiple decay pathways, the probability for the particular transition. Population relaxation forces us to modify the equations of motion for the diagonal elements of ρ:

$$\mathrm{i}\hbar\dot{\rho}_{nn}(t) = ([H,\rho])_{nn} - \mathrm{i}\hbar\sum_{k\neq n}\gamma_{kn}\rho_{nn} + \mathrm{i}\hbar\sum_{k\neq n}\gamma_{nk}\rho_{kk} \tag{2.4}$$

Since population relaxation induces phase relaxation, the off-diagonal elements of ρ must be modified as well (as discussed above) where the total dephasing rate Γ_{nk} is $\frac{1}{2}(\gamma_{nk}+\gamma_{kn})+\tilde{\Gamma}_{nk}$. $\tilde{\Gamma}_{kn}$ is the pure dephasing rate for the coherence ρ_{kn} and $\frac{1}{2}(\gamma_{nk}+\gamma_{kn})$ is the dephasing rate induced by population relaxation between the states $|n\rangle$ and $|k\rangle$.

Adding population relaxation and dephasing leads to the quantum Liouville equation for a driven, dissipative quantum system

$$\mathrm{i}\hbar\dot{\rho}(t) = [H_0,\rho] + \sum_{m=1}^{M} f_m(t)[H_m,\rho] - \mathrm{i}\hbar L_D(\rho), \tag{2.5}$$

where $\mathcal{L}_D(\rho)$ is the dissipation (super)operator.

3 Dissipation and entropy

One of the main consequences of dissipation is that interactions of the system with a bath (environment) can change the entropy of the system. The most useful measure of the entropy for our purposes is the Renyi entropy. Technically, there is a family of Renyi entropies $S_\alpha(\rho) = \frac{1}{1-\alpha}\,\mathrm{Tr}(\rho^\alpha)$, where $\alpha > 1$ is a real parameter. We choose $\alpha = 2$ and define $S(\rho) = S_2(\rho) + 1 = 1 - \mathrm{Tr}(\rho^2)$. Adding $+1$ ensures that the Renyi entropy of a pure-state system is zero and agrees with the von-Neumann or Shannon entropy.

For a coherently driven, non-dissipative quantum system, the constraint of unitary evolution partitions the set density operators into infinitely many kinematical equivalence classes of density operators with the same spectrum. The entropy of a non-dissipative, coherently driven quantum system is therefore conserved. For the Renyi entropy this follows directly from the fact that unitary transformations are trace-preserving. By applying coherent control fields, we can steer the system from one state ρ_0 to other states ρ_1 in the *same* kinematical equivalence class but we cannot reach states with different entropy [1].

By removing the constraint of unitary evolution, dissipation provides new opportunities for control by allowing us to reach states outside the kinematical equivalence class of states determined by the initial state, especially states whose entropy differs from the initial state. *Pure dephasing*, for instance, converts a coherent superposition state into an uncorrelated statistical mixture of energy eigenstates. Hence, dephasing enables us, in principle, to convert any given pure state into an arbitrary mixed state by creating a superposition state using coherent control, and letting the coherences decay. *Population relaxation* allows us, in principle, to convert a high entropy mixed state into a (zero entropy) pure state and vice versa. In the following sections, we shall present several applications of quantum control which rely on dissipation.

4 Conversion of a pure state into a mixed state

In absence of population relaxation, a coherent superposition of energy eigenstates $|\Psi\rangle = \sum_{n=1}^{N} c_n|n\rangle$ with $\sum_{n=1}^{N} c_n c_n^* = 1$ decays into a statistical mixture of the states $|n\rangle$ with discrete probability distribution $w_n = |c_n|^2$ for $1 \leq n \leq N$ as a result of pure dephasing. For dephasing times much greater than the control time we can design a control field that transforms the initial state into the required coherent superposition without worrying about the effect of dephasing, and then turn the field off to let dephasing transform this superposition state into the desired mixed state. However, if significant dephasing occurs during the coherent control phase, either due to rapid dephasing, or because the coherent control process takes too long, then this approach will fail.

For instance, consider a system with two non-degenerate energy levels. Suppose we wish to transform the initial pure state $|1\rangle$ into an equal and uncorrelated mixture of the states $|1\rangle$ and $|2\rangle$. Based on geometric control theory for non-dissipative systems, we might attempt

to apply a resonant Gaussian control pulse with effective pulse area $\frac{\pi}{2}$, which would create the superposition state $|\Psi\rangle = \frac{1}{\sqrt{2}}(|1\rangle + |2\rangle)$ in the non-dissipative case, and hope that this state will decohere into the desired mixed state due to dephasing. Unfortunately, our control calculations indicate that this scheme will definitely fail for dephasing rates of the order of the Rabi frequency of the control pulse. Straight-forward optimization with respect to the effective pulse area and length of the control pulse, however, indicate that the pulse length and pulse area can be chosen as to achieve the desired result. For instance, by increasing the effective pulse area of a Gaussian pulse lasting 50 vibrational periods from the predicted value of $\frac{\pi}{2}$ to 0.81π, we were able to create the desired maximum entropy state for a dephasing rate $\Gamma = 0.1$ in just over 50 vibrational periods.

5 Conversion of a mixed state into a pure state

An even more important application of controlled dissipative dynamics in quantum optics is optical pumping to drive a mixed-state system into a desired pure state using a combination of coherent control and population relaxation from an excited state. For instance, suppose we have a cloud of cold atoms whose electronic ground state is three-fold degenerate. If the system is not prepared in a particular pure state, it will usually be in a statistical mixture of the three degenerate substates, which we denote by $|1\rangle$, $|2\rangle$ and $|3\rangle$ for simplicity. For many applications, e.g., in quantum computing, it is crucial to prepare the system in a certain pure initial state. As we have seen, this is an aim impossible to realize by coherent control alone. To be able to take advantage of spontaneous emission to increase the purity of the system, we must couple the ground state to an excited electronic state with a finite lifetime. There are different ways of coupling the sublevels of the ground and excited states, depending on the polarization of the field. The trick is to select the right coupling.

For example, suppose the upper level is also three-fold degenerate and the coupling induced by the control field is as indicated in figure 1, i.e., the field couples states $|2\rangle$ and $|5\rangle$, as well as $|3\rangle$ and $|6\rangle$. The excited states can emit a photon and return to one of the ground states. Certain transitions are prohibited by atomic selection rules; the allowed decay modes are indicated in figure 1 (right). The simplest optical pumping schemes involve applying a constant amplitude field resonant with the transition frequency between the two levels and suitably polarized to couple only the levels indicated in figure 1. Without population relaxation due to spontaneous emission, the field merely leads to population cycling between states $|2\rangle$, $|5\rangle$, and $|3\rangle$, $|6\rangle$, respectively. Adding population relaxation changes the effect of the control field dramatically, leading to an accumulation of the population in state $|1\rangle$ as figure 2 shows. If the control field is applied for a sufficiently long time, all the population will eventually accumulate in state $|1\rangle$.

In the previous optical pumping scheme a simple constant amplitude resonant control field was sufficient to achieve the objective of driving the system into the desired pure state. However, this is not always the case. An application of optical pumping for dissipative

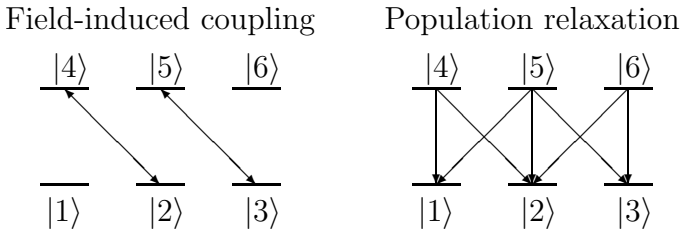

Figure 1: Optical pumping for a degenerate two-level system. Transition diagrams for the control field and population relaxation

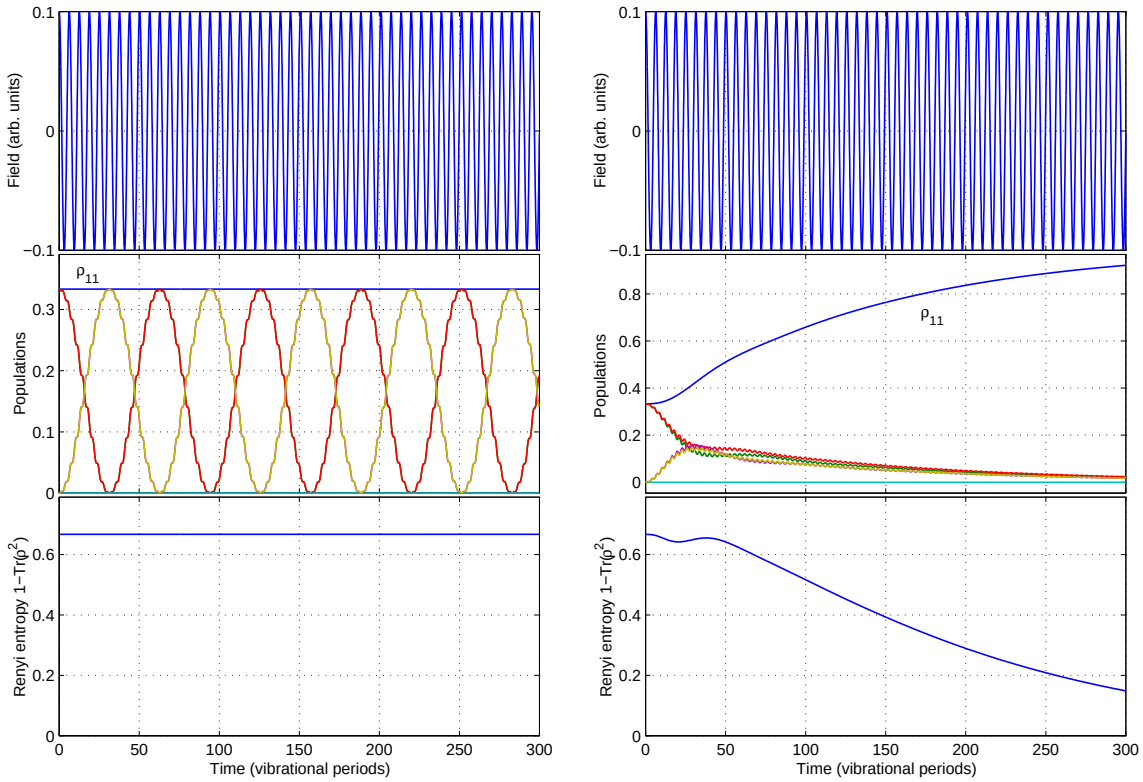

Figure 2: Optical pumping for a degenerate two-level system. Without population relaxation, the coherent control field induces population oscillations between states $|2\rangle$ and $|5\rangle$ as well as $|3\rangle$ and $|6\rangle$ (Rabi oscillations) and the entropy of the system remains constant (left). Population relaxation dramatically changes the effect of the control field, leading to an effective pumping of population into the lower left sublevel $|1\rangle$, and an entropy reduction as the system approaches a pure state (right).

systems, which relies on the interplay of carefully selected control pulses and dissipation, is laser cooling of internal molecular degrees of freedom. A molecular gas at room temperature consists of a statistical mixture of molecules in many different ro-vibrational states. Due to many closely spaced energy levels and lack of selection rules, there are many possible transitions with various transition probabilities that can be excited by applying a control field, and many different decay pathways. The situation is further complicated by the fact that the timescales for coherent control and population relaxation are often quite different. The problem thus appears to be nearly hopeless. Yet, it has been shown that this problem can be addressed successfully using optimal control for dissipative systems and creative control strategies [2, 3].

An approach that is especially promising for systems where the timescales for control and dissipation are quite different (as in our molecular cooling problem) involves breaking up the control problem into a sequence of excitation and relaxation steps. The goal in each step is to use control theory to design control fields to transfer the system from its initial state to a kinematically equivalent, dynamically reachable state, which has the same entropy but will (possibly after some time) decay into a state with lower entropy. In principle, the entropy of the system can be decreased until it is zero and the system is the desired pure state. The main difficulty of this approach is the choice of suitable target states for each optimization step, which requires a good understanding of the effects of population relaxation and dephasing on various kinematically equivalent states, in order to assure that the selected states will decay into a lower entropy state.

6 Conclusion

We have presented a general framework for coherent control of dissipative quantum systems, and have shown that dissipative effects open new possibilities for control by eliminating the kinematical constraint of unitary evolution. This allows us to change the entropy of the system and convert pure states into mixed states and vice versa, which is crucial for many applications in quantum optics, chemistry and computing. We hope that these applications will provide a motivation for further study of the control of dissipative systems.

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