DISSIPATIVE QUANTUM CONTROL

ALLAN I. SOLOMON$^{1,2}$ AND SONIA G. SCHIRMER$^3$

$^1$ Department of Physics and Astronomy, The Open University, Milton Keynes, MK7 6AA, United Kingdom
$^2$ LPTL, University of Paris VI, France
$^3$ Dept of Applied Maths & Theoretical Physics, University of Cambridge, Wilberforce Road, Cambridge, CB3 0WA, United Kingdom
E-mail: a.i.Solomon@open.ac.uk, sgs29@cam.ac.uk

Nature, in the form of dissipation, inevitably intervenes in our efforts to control a quantum system. In this talk we show that although we cannot, in general, compensate for dissipation by coherent control of the system, such effects are not always counterproductive; for example, the transformation from a thermal (mixed) state to a cold condensed (pure state) can only be achieved by non-unitary effects such as population and phase relaxation.

1 Representation of quantum states for dissipative systems

In closed-system, pure-state quantum mechanics the state of the system is usually represented by a wavefunction $|\Psi\rangle$, which is an element of the Hilbert space $\mathcal{H}$. For open quantum systems, however, a quantum statistical mechanics formulation is necessary since dissipative effects due to the interaction of the system with its environment convert pure states into statistical ensembles and vice versa. The state of the system must therefore be represented by a density operator $\hat{\rho}$, i.e., a positive trace-one operator acting on $\mathcal{H}$. It is convenient to expand this density operator in terms of a complete orthonormal set of energy eigenstates $\{|n\rangle: 1, 2, \ldots, N = \dim \mathcal{H}\}$ of the system:

$$\hat{\rho} = \sum_{n=1}^{N} \rho_{nn}|n\rangle\langle n| + \sum_{m>n} (\rho_{nm}|n\rangle\langle m| + \rho_{mn}^*|m\rangle\langle n|)$$

such that the diagonal elements $\rho_{nn}$ in this expansion determine the populations of the energy eigenstates $|n\rangle$, while the off-diagonal elements $\rho_{nm}$ for $n \neq m$ determine the coherences between energy eigenstates. The latter distinguish coherent superposition states $|\Psi\rangle = \sum_{n=1}^{N} c_n|n\rangle$ from statistical ensembles of energy eigenstates (i.e., mixed states) $\hat{\rho} = \sum_{n=1}^{N} w_n|n\rangle\langle n|$. To see the difference between the two, consider $\hat{\rho}_1 = \begin{bmatrix} 1/2 & 0 \\ 0 & 1/2 \end{bmatrix}$ and $\hat{\rho}_2 = \begin{bmatrix} 1 & 1 \\ 1 & 1 \end{bmatrix}$.

$\hat{\rho}_1$ is diagonal and represents a two-level system in an incoherent mixed state with equal populations in states $|1\rangle$ and $|2\rangle$ but no correlation between both states. Note that such a state cannot be represented by a wavefunction. $\hat{\rho}_2$ has off-diagonal elements and diagonalization shows that it rep-
represents the coherent superposition state $|\Psi\rangle = \frac{1}{\sqrt{2}}(|1\rangle + |2\rangle)$ since we have
\[
\hat{\rho} = |\Psi\rangle\langle\Psi| = \frac{1}{\sqrt{2}}[1, 1]^T \times \frac{1}{\sqrt{2}}[1, 1].
\]

2 Dynamics of dissipative quantum control systems

For Hamiltonian systems the evolution of the state $\hat{\rho}(t)$ with $\hat{\rho}(t_0) = \hat{\rho}_0$ is governed by
\[
\hat{\rho}(t) = \hat{U}(t)\hat{\rho}_0\hat{U}(t)^\dagger, \tag{2}
\]
where $\hat{U}(t)$ is the evolution operator satisfying the Schrödinger equation
\[
\frac{i\hbar}{dt}\hat{U}(t) = \hat{H}(t)\hat{U}(t), \quad \hat{U}(0) = \hat{I}, \tag{3}
\]
and $\hat{I}$ is the identity. $\hat{\rho}(t)$ also satisfies the quantum Liouville equation
\[
\frac{i\hbar}{dt}\hat{\rho}(t) = [\hat{H}(t), \hat{\rho}(t)] = \hat{H}(t)\hat{\rho}(t) - \hat{\rho}(t)\hat{H}(t). \tag{4}
\]
$\hat{H}(t)$ is the total system Hamiltonian, which depends on the control fields $f_m$:
\[
\hat{H}(t) = \hat{H}_0 + \sum_{m=1}^{M} f_m(t)\hat{H}_m, \tag{5}
\]
where $\hat{H}_0$ is the internal Hamiltonian and $\hat{H}_m$ is the interaction Hamiltonian for the field $f_m$ for $1 \leq m \leq M$.

The advantage of the Liouville equation \(\text{(4)}\) over the unitary evolution equation \(\text{(2)}\) is that it can easily be adapted for dissipative systems by adding a dissipation (super)operator $L_D[\hat{\rho}(t)]$:
\[
\frac{i\hbar}{dt}\hat{\rho}(t) = [\hat{H}_0, \hat{\rho}(t)] + \sum_{m=1}^{M} f_m(t)[\hat{H}_m, \hat{\rho}(t)] + i\hbar L_D[\hat{\rho}(t)]. \tag{6}
\]
Under certain assumptions (semi-group dynamics, norm continuity and conservation of probability), it can be shown that the dissipation operator has the form
\[
L_D[\hat{\rho}(t)] = \frac{1}{2} \sum_{s} \left( [\hat{V}_s\hat{\rho}(t), \hat{V}_s^\dagger] + [\hat{V}_s, \hat{\rho}(t)\hat{V}_s^\dagger]\right), \tag{7}
\]
where the $\hat{V}_s$ are arbitrary $N \times N$ matrices, i.e., bounded operators on the Hilbert space $\mathcal{H}$.
3 Population relaxation and phase decoherence

In general, uncontrollable interactions of the system with its environment lead to two types of dissipation: population relaxation (decay) and phase decoherence (or dephasing).

The former occurs, for instance, when a quantum particle in state $|n\rangle$ spontaneously emits a photon and decays to another quantum state $|k\rangle$, which changes the populations according to

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

where $\gamma_{kn}\rho_{nn}$ is the population loss for level $|n\rangle$ due to transitions $|n\rangle \rightarrow |k\rangle$, and $\gamma_{nk}\rho_{kk}$ is the population gain caused by transitions $|k\rangle \rightarrow |n\rangle$. The total population relaxation rate $\gamma_{kn}$ is determined by the lifetime of the state $|n\rangle$, and for multiple decay pathways, the relative probability for the transition $|n\rangle \rightarrow |k\rangle$.

The latter occurs when the interaction with the environment destroys the phase correlations between states, which leads to a decay of the off-diagonal elements of the density matrix:

\[
\dot{\rho}_{kn}(t) = -\frac{i}{\hbar}([\hat{H}(f),\hat{\rho}(t)])_{kn} - \Gamma_{kn}\rho_{kn}(t)
\]

where $\Gamma_{kn}$ (for $k \neq n$) is the dephasing rate between $|k\rangle$ and $|n\rangle$. Note that population relaxation always induces dephasing since decay destroys the phase correlations between states. However, there may be other sources that contribute to the loss of coherence of the system.

The effects of dephasing and population relaxation can be accounted for by adding a dissipation super-operator defined by $(LD)_{lm} = (LD)_{nk}\rho_{nk}(t)$ to the Liouville equation. The latter can be represented by an $N^2 \times N^2$ matrix whose non-zero elements are

\[
(L_D)_{kn,kn} = -\Gamma_{kn}, \quad (L_D)_{nn,kk} = +\gamma_{nk}, \quad k \neq n
\]

\[
(L_D)_{nn,nn} = -\sum_{n\neq k} \gamma_{kn}.
\]

Population decay and dephasing allow us to overcome kinematical constraints such as unitary evolution to create statistical ensembles from pure states, and pure states from statistical ensembles, which is important for many applications such as optical pumping. However, there are instances when this is not desirable such as in quantum computing, where these effects destroy quantum information. Hence, there are situations when we would like to prevent decay and dephasing. A cursory glance at the quantum Liouville equation for coherently driven, dissipative systems suggests that it might be possible to prevent population and phase relaxation by applying suitable
control fields such that
\[
\sum_{m=1}^{M} f_m(t)[\hat{H}_m, \hat{\rho}(t)] + i\hbar L_D[\hat{\rho}(t)] = 0.
\] (11)

Unfortunately, however, a more careful analysis reveals that this is not possible, in general, as we shall now show explicitly for a two-level system, or qubit in quantum computing parlance.

4 Dynamics of a dissipative, coherently driven 2-level system

The Hamiltonian for a driven two-level system with energy levels \( E_1 < E_2 \) is
\[
\hat{H}[f(t)] = \hat{H}_0 + f_1(t)\hat{H}_1 + f_2(t)\hat{H}_2
\] (12)
where \( \hat{H}_0 \) is the internal Hamiltonian and \( \hat{H}_1 \) and \( \hat{H}_2 \) are Hamiltonians describing the interaction with independent (real-valued) control fields \( f_1(t) \) and \( f_2(t) \). For a spin system, for example, the two control fields might be two orthogonal polarizations of an electromagnetic field affecting rotations about two orthogonal axes. In general, we can assume without loss of generality that the internal and interaction Hamiltonians have the form:
\[
\hat{H}_0 = \begin{bmatrix} E_1 & 0 \\ 0 & E_2 \end{bmatrix}, \quad \hat{H}_1 = d_1 \begin{bmatrix} 0 & 1 \\ 1 & 0 \end{bmatrix}, \quad \hat{H}_2 = d_2 \begin{bmatrix} 0 & -1 \\ 1 & 0 \end{bmatrix}.
\]

where \( d_1, d_2 \) are the (real-valued) dipole moments for the transition and \( \omega = (E_2 - E_1)/\hbar \) is the transition frequency.

We can re-write the Liouville equation in matrix form in a higher dimensional space (Liouville space). Straightforward computation shows
\[
\frac{d}{dt}|\rho(t)\rangle = -i[L_0 + f_1(t)L_1 + f_2(t)L_2 + iL_D]|\rho(t)\rangle
\] (13)
where \( |\rho(t)\rangle = [\rho_{11}(t), \rho_{12}(t), \rho_{21}(t), \rho_{22}(t)]^T \) and the free dissipative evolution is given by
\[
L_0 = \omega \begin{bmatrix} 0 & 0 & 0 & 0 \\ 0 & -1 & 0 & 0 \\ 0 & 0 & +1 & 0 \\ 0 & 0 & 0 & 0 \end{bmatrix}, \quad L_D = \begin{bmatrix} -\gamma_{21} & 0 & 0 & \gamma_{12} \\ 0 & -\Gamma & 0 & 0 \\ 0 & 0 & -\Gamma & 0 \\ \gamma_{21} & 0 & 0 & -\gamma_{12} \end{bmatrix}
\] (14)
with \( \gamma_{12} \) being the rate of population relaxation from \( |2\rangle \) to \( |1\rangle \), \( \gamma_{21} \) the rate of population relaxation from \( |1\rangle \) to \( |2\rangle \), and \( \Gamma \) the dephasing rate. The control action is given by
\[
L_1 = \frac{d_1}{\hbar} \begin{bmatrix} 0 & -1 & +1 & 0 \\ -1 & 0 & 0 & +1 \\ +1 & 0 & 0 & -1 \\ 0 & +1 & -1 & 0 \end{bmatrix}, \quad L_2 = \frac{d_2}{\hbar} \begin{bmatrix} 0 & -i & -i & 0 \\ +i & 0 & 0 & -i \\ +i & 0 & 0 & -i \\ 0 & +i & +i & 0 \end{bmatrix}
\] (15)
Observe that the matrix elements of the control Liouville operators $L_1$ and $L_2$ are zero where the matrix elements of the dissipation operator $L_D$ are non-zero, and vice versa. Thus, no matter how we choose the control fields, we cannot cancel the effect of the dissipative terms without introducing additional terms such as measurements and feedback or the ability to control the coupling of the system to the reservoir.

Note that the dissipation operator as defined in (14) is equivalent to the Lindblad form since inserting

$$
\hat{V}_1 = \begin{pmatrix} 0 & 0 \\ \sqrt{\gamma_{12}} & 0 \end{pmatrix}, \quad \hat{V}_2 = \begin{pmatrix} 0 & \sqrt{\gamma_{12}} \\ 0 & 0 \end{pmatrix}, \quad \hat{V}_3 = \begin{pmatrix} \sqrt{2\Gamma} & 0 \\ 0 & 0 \end{pmatrix}
$$

with $\hat{\Gamma} = \Gamma - \frac{1}{2}(\gamma_{12} + \gamma_{21})$ into (7) gives

$$
L_D[\hat{\rho}(t)] = \frac{1}{2} \sum_{s=1}^{3} \left( [\hat{V}_s, \hat{\rho}(t)], [\hat{V}_s, \hat{\rho}(t)]^\dagger \right) + [\hat{V}_s, \hat{\rho}(t) \hat{V}_s^\dagger]
$$

$$
= \begin{pmatrix}
-\gamma_{21}\rho_{11} & -\frac{1}{2}\gamma_{21}\rho_{12} \\
-\frac{1}{2}\gamma_{21}\rho_{21} & \gamma_{21}\rho_{22} \\
\gamma_{12}\rho_{12} & -\frac{1}{2}\gamma_{12}\rho_{11}
\end{pmatrix} + \begin{pmatrix}
\gamma_{12}\rho_{22} & -\frac{1}{2}\gamma_{12}\rho_{12} \\
-\frac{1}{2}\gamma_{12}\rho_{21} & -\gamma_{12}\rho_{22} \\
\hat{\rho}_2 & 0
\end{pmatrix} = \begin{pmatrix}
-\gamma_{21}\rho_{11} + \gamma_{12}\rho_{22} & -\gamma_{12}\rho_{21} \\
-\gamma_{21}\rho_{12} & -\frac{1}{2}\gamma_{21}\rho_{11} - \gamma_{12}\rho_{22}
\end{pmatrix}
$$

which is equivalent to $L_D[\rho(t)]$ with $L_D$ as in (7).

5 Dissipation and entropy

One of the main consequences of dissipation is that interactions of the system with a bath (environment) can change the entropy and purity of the system. A useful measure of the entropy and purity for our purposes is $1 - \text{Tr}[\rho(t)^2]$, which essentially determines the Renyi entropy of the system, although the latter is often defined to be $-\log \text{Tr}[\rho(t)^2]$. For a non-dissipative, coherently driven quantum system the entropy is conserved because the evolution must remain unitary. Dissipation provides new opportunities for control by enabling us to reach states outside the orbit of the initial state under the unitary group, especially states whose entropy differs from the initial state. Dephasing, for instance, converts coherent superposition states into uncorrelated statistical mixtures of energy eigenstates and hence enables us, in principle, to convert a 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 to convert a high entropy mixed state into a (zero entropy) pure state and vice versa.

5.1 Conversion of a pure state into a mixed state

When only pure dephasing occurs 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 mix-
ture of the states $|n\rangle$ with a discrete probability distribution $w_n = |c_n|^2$ for $1 \leq n \leq N$. 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 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 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 pure state $|1\rangle$ into a statistical mixture of the states $|1\rangle$ and $|2\rangle$. Based on geometric control for non-dissipative systems, we might try 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 decoherence will convert this state into the desired mixed state. Our control calculations indicate, however, that this scheme will fail for dephasing rates of the order of the Rabi frequency of the control pulse. However, straight-forward optimization with respect to the effective pulse area and length of the control pulse indicates that the pulse length and pulse area can be chosen such 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 $\frac{\pi}{2}$ for a closed system to $0.81\pi$, we were able to create the desired maximum entropy state for a dephasing rate $\Gamma = 0.1$ in just over 50 vibrational periods.

5.2 Conversion of a mixed state into a pure state

A perhaps 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 mixture of the three degenerate substates, which we shall 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 generally 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. The sublevels of the ground and excited states can be coupled in various ways 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. Some applications of optical pumping such as laser cooling of internal molecular degrees of freedom rely on the interplay of carefully selected control pulses and dissipation. For example, a molecular vapor 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.

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 the problem up 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 is likely to 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 shown that although coherent control of the system dynamics in absence of feedback or the ability to alter the interaction of the system with its
Field-induced coupling

\[
\begin{align*}
|4\rangle & \\
|5\rangle & \\
|6\rangle & \\
|1\rangle & \\
|2\rangle & \\
|3\rangle &
\end{align*}
\]

Population relaxation

\[
\begin{align*}
|4\rangle & \\
|5\rangle & \\
|6\rangle & \\
|1\rangle & \\
|2\rangle & \\
|3\rangle &
\end{align*}
\]

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

Environment cannot compensate for dissipative effects such as dephasing and population relaxation in open systems, such effects need not be detrimental to control, but are in fact crucial for many interesting applications such as ensemble preparation or system purification by laser cooling.
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