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On the Interplay of Control Fields and Spontaneous Emission in Laser Cooling

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Optimal control theory (OCT) is applied to the problem of cooling molecular rotations. The optimal field gives rise to a striking behavior, in which there is no noticeable increase in the lowest rotational state population until the last percent or so of the control interval, at which point the population jumps to 1. Further analysis of the intermediate time interval reveals that cooling is taking place all along, in the sense that the *purity* of the system, as measured by $\mathrm{Tr}(\rho^2)$, is increasing monotonically in time. Once the system becomes almost completely pure, the external control field can transfer the amplitude to the lowest rotational state by a completely Hamiltonian manipulation. This mechanism is interesting because it suggests a possible way of accelerating cooling, by exploiting the cooling induced by spontaneous emission to *all* the ground electronic state levels, not just the lowest rotational level. However, it also raises a major paradox: it may be shown that external control fields, no matter how complicated, cannot change the value of $\mathrm{Tr}(\rho^2)$; changing this quantity requires spontaneous emission which is inherently uncontrollable. What place is there then for control, let alone optimal control, using external fields? We discuss the resolution to this paradox with a detailed analysis of cooling in a two-level system.

I. Introduction

In previous work we have described the application of optimal control theory (OCT) to laser cooling of molecular vibrations.[1] It was shown that OCT generated a pulse sequence that caused the population in the lowest vibrational level to grow monotonically. The underlying mechanism of the optimal pulse is an analogue of velocity selective coherent population trapping (VSCPT) in atoms, in which any population which emits by chance to the desired final state becomes decoupled from the field and no longer reabsorbs, while the population which emits to any other levels is continually repumped, to give it additional chances to emit to the desired state.[2] Here we report on the application of OCT to the cooling of molecular rotations, where a completely different behavior is observed. In contrast with vibrations, in rotations the optimal pulse sequence generates no noticeable increase in population in the ground state level until the last percent or so of the control interval. However, during the intermediate time the system is increasing monotonically in purity, as measured by $\mathrm{Tr}(\rho^2)$; when the system becomes almost completely pure, the external field transfers the amplitude to the lowest rotational state using a fully coherent manipulation. This second mechanism apparently takes over when the optimization procedure is given only a short time interval to act; as a result this mechanism may be a promising approach to accelerating the rate of cooling by exploiting all the spontaneous emission for purification of the system, not only the emission to the ground state.

The rotational cooling mechanism gives a new perspective on laser cooling—it is the attempt to use external controls to increase the purity of a state. This perspective has far-reaching consequences for the design of cooling strategies but raises a number of profound paradoxes. First, it is widely appreciated that external fields, no matter how complicated, cannot change the purity of the system, as measured by $\mathrm{Tr}(\rho^2)$;[3] changes in the system's purity can be achieved only through the inclusion of spontaneous emission (or any other "bath" which can serve to dump the system's entropy). Yet, spontaneous emission is inherently uncontrollable, so what place is there for control, let alone optimal control? But the external fields *must* play a role: with only spontaneous emission and no external field for optical excitation again, there will be no cooling since there will be no excited state populations (neglecting the much slower process of IR or microwave spontaneous emission). Furthermore, external fields applied randomly would tend to lead to heating rather than cooling! What then is the interplay between absorption, stimulated emission, and spontaneous emission necessary to achieve cooling? Can this interplay be quantified?

The brief answer is that the purity-preserving transformation that can be performed by external fields, while they do not themselves change the purity of the system, do affect the subsequent sign and rate of change of purity due to spontaneous emission. This is consistent with the observation that $\mathrm{Tr}(\dot{\rho}^2)$ is independent of the external field, while $\mathrm{Tr}(\ddot{\rho}^2)$ is not; loosely speaking, the second derivative affects the observable after two time steps, via a two-step sequential process: first, a purity-preserving transformation induced by the external field, followed by purity-changing transformations induced by the spontaneous emission. The optimization of the external field is the optimization of the purity-preserving transformation so that the purity will increase maximally in the subsequent spontaneous emission.

Although the interplay of absorption, stimulated emission, and spontaneous emission plays a central role in the extant theories of laser cooling, those theories focus on the conditions favorable to reduction of the *energy* content of the system. The rotational cooling mechanism described above highlights the fact that key to cooling is the increase in the system's *purity*, or a decrease in the system's entropy; once this is achieved, any state, including the ground state, is easily populated. Thus, the present work is an important step toward a much-needed thermodynamic analysis of laser cooling.

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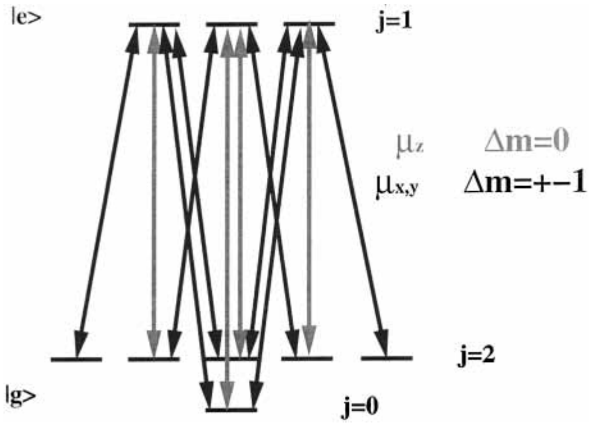

Figure 1. The level structure in the rotational model.

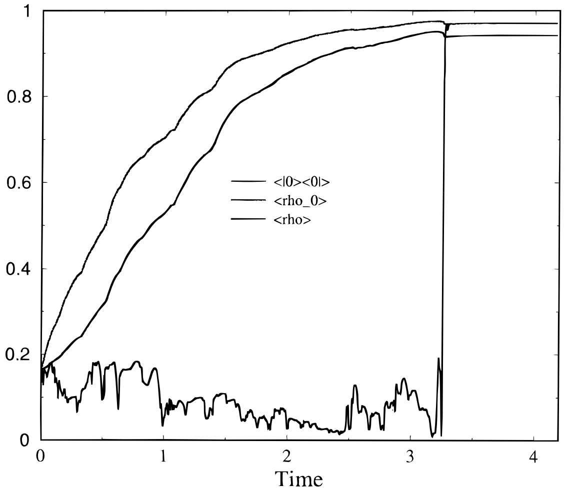

Figure 2. The projection on the target state <<equation:contr34>> is shown (solid line) with Renyi entropy Tr(ρ^2) (dashed line). Time is measured in rotational periods.

II. Rotational Cooling

The Model. The Hamiltonian we used to study rotational cooling is

$$\hat{\mathbf{H}} = \begin{pmatrix} \hat{\mathbf{H}}_{\mathbf{e}} & -\epsilon_x(t)\hat{\mu}_x - \epsilon_y(t)\hat{\mu}_y - \epsilon_z(t)\hat{\mu}_z \\ -\epsilon_x(t)^*\hat{\mu}_x - \epsilon_y(t)^*\hat{\mu}_y - \epsilon_z(t)^*\hat{\mu}_z & \hat{\mathbf{H}}_{\mathbf{g}} \end{pmatrix} \tag{2.1}$$

where $\hat{\mathbf{H}}_{\mathbf{e/g}} = B_{e/g}l(l + 1)$. For simplicity, we have included only three l levels, $l = 0$, 2 on the ground electronic state and $l = 1$ on the excited electronic state (Figure 1). The choice of these states is motivated by the selection rule $\Delta l = \pm1$. We include all $2l + 1$ of the m states for each l state. Furthermore, we assume that all three polarizations of light are possible, e.g. by having a coherent laser beam which is split and then allowed to impinge on the sample from two directions. This allows transitions $\Delta m = 0, \pm1$. Initially the system is assumed to be in the ground electronic state, with equal populations in each of the rotational states (i.e. one-sixth in $l = 0$ and one-sixth in each of the five $l = 2$ states.

The OCT iterative algorithm described elsewhere in our primary was applied to this truncated rotational system, with the objective of maximizing Tr($\rho|0\rangle\langle0|$) at the final time. The results are shown in Figure 2: the quantity Tr($\rho|0\rangle\langle0|$) is almost constant and in fact decreases during most of the time interval, jumping to 1 in the last 1% of the pulse sequence. However, the purity of the system, as measured by Tr(ρ^2) (shown in the same figure) increases monotonically during the control pulse. This drives home the point discussed in the Introduction: that true cooling should be measured by an increase in purity; if the system can be obtained in any pure state, then coherent manipulations can move it virtually completely to the ground state. Focusing on overall purity rather than population in the lowest rotational state allows the OCT pulse to exploit *all* the spontaneous emission for purification, and thus cooling is not limited to the rate at which chance emission to the lowest rotational state occurs. Yet, as mentioned above, this leads to a paradox: if spontaneous emission is inherently uncontrollable, how can the purity be increasing so systematically during the action of the external field? This resolution to this paradox is discussed in the next two sections.

III. Purity Increasing Transformations

Using Tr(ρ^2) as a measure of purity, the object of cooling is then to take Tr(ρ^2) from its initial value, which is less than 1, to a final value of 1. Now for the bad news: external control fields, no matter how complicated, cannot change the value of Tr(ρ^2). This is easily seen by considering the quantity d(Tr(ρ^2))/dt:

$$\frac{\mathrm{d}}{\mathrm{d}t}\mathrm{Tr}(\rho^2) = 2\mathrm{Tr}(\rho\dot{\rho}) \tag{3.1}$$

$$= \frac{2}{i\hbar}\mathrm{Tr}(\rho[H, \rho]) \tag{3.2}$$

$$= \frac{2}{i\hbar}\mathrm{Tr}(\rho(H\rho - \rho H)) \tag{3.3}$$

$$= 0 \tag{3.4}$$

where in the last step we have used the cyclic invariance of the trace. In eq 3.4 we have assumed completely Hamiltonian evolution:

$$\dot{\rho} = -\frac{i}{\hbar}[H,\rho] \tag{3.5}$$

Since the external field enters only through H, and since eq 3.4 holds for any H, this shows that the external field cannot change the purity of the state. Thus, we are faced with the following paradox: external control fields (absorption and stimulated emission) cannot change the purity of the state and, hence, cannot produce true cooling. To achieve true cooling, we need to add spontaneous emission to the model, but spontaneous emission is intrinsically uncontrollable! What then is the role of the external field in producing cooling? We may answer this question by first asking another question: spontaneous emission can lead at times to cooling and at times to heating. Can we formulate general rules or delineate regions in parameter space of the density matrix which determine when cooling occurs and when heating occurs? To give a quick preview of the answer, the answer will be yes, we can delineate regions in the parameter space of ρ corresponding to heating and corresponding to cooling. This in turn explains the function of the external control field: to manipulate the elements of the density matrix via purity-preserving transformations in such a way that maximizes the rate of increase in purity. To give a colorful, if somewhat macabre, analogy, the situation may be compared to having a person on a roof who is considering jumping (spontaneous emission). We cannot push the person off the roof (i.e., we cannot induce spontaneous emission), but we can affect the outcome by moving around nets on the ground below (we can change the effect that the spontaneous emission has on the purity of the system by manipulating the phases of the coherences in the levels into which emission takes place). It is worth mentioning that the spontaneous emission rate can also be affected by changing both the populations and the coherences in the excited electronic state.

To put these ideas on a more quantitative basis, we consider the explicit form for $\dot{\rho}$, including spontaneous emission:[5]

$$\dot{\rho} = -\frac{i}{\hbar}[H, \rho] + L\rho \tag{3.6}$$

$$= -\frac{i}{\hbar}[H,\rho] + F\,\rho F^{\ddagger} - \frac{1}{2}F^{\dagger}\,F\,\rho - \frac{1}{2}\rho F^{\dagger}\,F \tag{3.7}$$

where

$$H = \begin{pmatrix} E_g & -E(t)\mu \\ -E^*(t)\mu & E_e \end{pmatrix} F = \begin{pmatrix} 0 & 1 \\ 0 & 0 \end{pmatrix} F^{\dagger} = \begin{pmatrix} 0 & 0 \\ 1 & 0 \end{pmatrix} \tag{3.8}$$

The term $F\rho F^{\dagger}$ controls the changes in populations and coherences in the downstairs manifold, while the terms $-1/2F^{\dagger}F\,\rho$ and $-1/2\rho F^{\dagger}\,F$ control the changes in populations and coherences in the upstairs manifold; all three terms together control the changes in the optical coherences connecting the two manifolds. It is easily verified that

$$\mathrm{Tr}(\dot{\rho}) = 0 \tag{3.9}$$

consistent with the fact that $\mathrm{Tr}(\rho) = 1$.

Writing

$$\rho = \begin{pmatrix} \rho_{\mathrm{g}} & \rho_{\mathrm{c}} \\ \rho_{\mathrm{c}}^{\dagger} & \rho_{\mathrm{e}} \end{pmatrix} \tag{3.10}$$

we find

$$\mathrm{Tr}(\dot{\rho^2}) = 2\mathrm{Tr}(\rho\dot{\rho}) \tag{3.11}$$

$$= 2\mathrm{Tr}[\rho(-i/\hbar)[H,\rho] + L\rho] \tag{3.12}$$

$$= 2\mathrm{Tr}[\rho L\rho] \tag{3.13}$$

$$= 2\mathrm{Tr}(\rho_{\mathrm{e}}(\rho_{\mathrm{e}} - \rho_{\mathrm{g}}) - \rho_{\mathrm{c}}^{\ddagger}\rho_{\mathrm{c}}) \tag{3.14}$$

A first glimmer of the resolution to our paradox of how control fields can control purity content is obtained by noting that the second derivative, $\mathrm{Tr}(\ddot{\rho^2})$, does depend on the external field. Loosely speaking, we may interpret the independence of the first derivative and dependence of the second derivative on the control field as saying that the control of spontaneous emission is achieved only by a two-stage process: preparation of the initial state by the control field, followed by spontaneous emission into that recipient state. To put this two-stage interpretation on a more rigorous basis, in the next section we explore in some detail how this two-stage mechanism operates in a two-level system.

IV. Illustrative Example: Cooling in the Two-Level System

Consider a 2 × 2 density matrix, ρ. The general form is

$$\rho = \begin{pmatrix} a & b \\ \mathrm{c} & d \end{pmatrix} \tag{4.1}$$

where a and d are real and $c = b^*$ are complex.

For a two-level system, the constraints $\mathrm{Tr}(\rho) = 1$ and $\mathrm{Tr}(\rho^2) \le 1$ take the following forms:

$$\mathrm{Tr}(\rho) = a + d = 1 \tag{4.2}$$

$$\mathrm{Tr}(\rho^2) = a^2 + 2|b|^2 + d^2 \le 1 \tag{4.3}$$

The quantity $|b|^2$ is a measure of the coherence of the system; its maximum allowed value is $|b^2| = ad$, in which case

$$\mathrm{Tr}(\rho^2) = (a + d)^2 = 1 \tag{4.4}$$

corresponding to a pure state. It is convenient to define a parameter γ, $0 \le \gamma \le 1$, such that

$$|b|^2 = \gamma ad \tag{4.5}$$

With this definition we can characterize the density matrix by two parameters which range between 0 and 1: d, which is a measure of the population in the excited state, and γ, which is a measure of the coherence of the system. Technically, these two parameters do not give a complete characterization of the 2 × 2 density matrix—there is a phase of b which is still unspecified. But in all the expressions we examine below only the product $|b|^2$ enters, and hence d and γ give a full parametrization.

Substituting eqs 4.2 and 4.5 into 4.3, we obtain

$$\mathrm{Tr}(\rho^2) = 2(1 - \gamma)d^2 - 2(1 - \gamma)d + 1 \tag{4.6}$$

Now consider $\mathrm{Tr}(\dot{\rho^2})$. For the two-level system we have

$$F = \begin{pmatrix} 0 & 1 \\ 0 & 0 \end{pmatrix} \qquad F^{\ddagger} = \begin{pmatrix} 0 & 0 \\ 1 & 0 \end{pmatrix} \tag{4.7}$$

Now

$$\dot{\rho} = -\frac{i}{\hbar}(H\rho - \rho H) + L\rho \tag{4.8}$$

$$= -\frac{i}{\hbar}(H\rho - \rho H) + \begin{pmatrix} d & -b/2 \\ -c/2 & -d \end{pmatrix} \tag{4.9}$$

This implies that

$$\mathrm{Tr}(\dot{\rho^2}) = 2\mathrm{Tr}(\rho\dot{\rho}) \tag{4.10}$$

$$= 2\mathrm{Tr}[\rho(-i/\hbar)[H,\rho] + L\rho] \tag{4.11}$$

$$= 2\mathrm{Tr}\left[\begin{pmatrix} \mathrm{a} & \mathrm{b} \\ \mathrm{c} & \mathrm{d} \end{pmatrix}\begin{pmatrix} d & -b/2 \\ -c/2 & -d \end{pmatrix}\right] \tag{4.12}$$

$$= 2\mathrm{Tr}\begin{pmatrix} ad - (bc/2) & -(ab/2) - bd \\ cd - (cd/2) & -(bc/2) - d^2 \end{pmatrix} \tag{4.13}$$

$$= 2(ad - bc - d^2) \tag{4.14}$$

$$= 2(d(1 - \gamma) - d^2(2 - \gamma)) \tag{4.15}$$

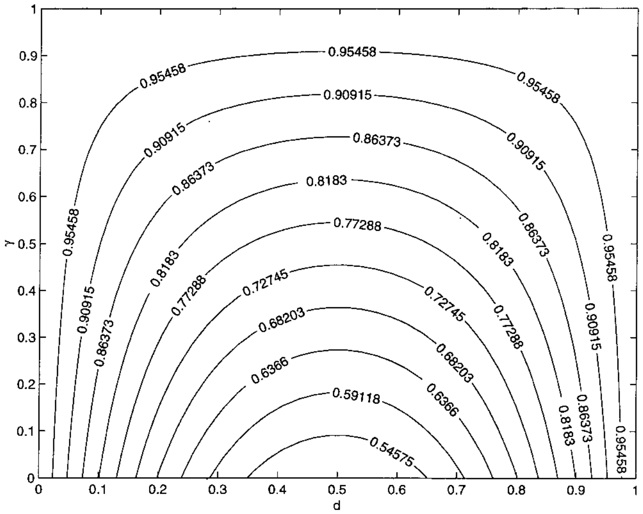

Figure 3. Isopurity, or isocoherence contours (contours of fixed Tr(ρ^2)) as a function of the parameters d and γ (eq 4.6). The contour takes its maximum value of 1, corresponding to a pure state, along the three contiguous lines $d = 0$, $d = 1$, and $\gamma = 1$, while the function takes its minimum value of 1/2, representing the most impure state, at $d = 1/2$, $\gamma = 0$. Note the symmetry with respect to the line $d = 1/2$, reflecting the fact that the purity is unchanged if the role of the ground and excited state are interchanged. Control fields can perform only those transformations on d and γ that keep the system on the same purity contour.

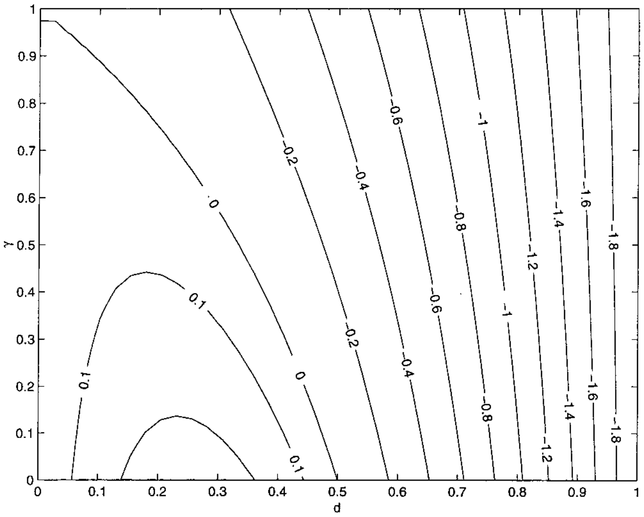

Figure 4. Contour map of Tr($\dot{\rho^2}$) as a function of the parameters d and γ (eq 4.15). The region of high d is negative, leading to heating, while the region of low d is positive, leading to cooling. For fixed d, the maximum occurs at the boundary $\gamma = 0$. The global maximum is at $d = 1/2$, $\gamma = 0$. The goal of the control field, therefore, is to move the system along an isopurity contour to the point of maximum positive Tr($\dot{\rho^2}$).

A full perspective on the effect of the external field may be obtained by inspection of equipotential contours of Tr(ρ^2) and Tr($\dot{\rho^2}$) as a function of d and γ (Figure 3). The manipulations allowed by the external field are those that move the system along a contour of constant value of Tr(ρ^2)—an isocoherence contour; it is clear from Figure 4 that the location on this contour has a profound affect on Tr($\dot{\rho^2}$). This gives a second perspective on how the external field cannot directly change Tr(ρ^2) but can still affect the rate of change of Tr(ρ^2). If we imagine that at every instant in time the external field moves the system along the instantaneous isocoherence contour until it intersects the curve of maximum Tr($\dot{\rho^2}$), *that* would provide an optimal cooling strategy. This last observation is the crux of our cooling theory and puts into sharp perspective the role played by the external field: while the external field cannot itself change the purity of the system, it can perform purity-preserving transformations which subsequently affect the rate of change of purity.

To express this idea mathematically, we seek to maximize Tr($\dot{\rho^2}$) with respect to the parameters d and γ, subject to the constraint Tr(ρ^2) = C. In principle Tr($\dot{\rho^2}$)$^{\mathrm{opt}}$ will be obtained as a function of d and γ. These values of d and γ in turn fix Tr(ρ^2). Combining these equations gives Tr($\dot{\rho^2}$) as a function of Tr(ρ^2), which is a differential equation for the trajectory Tr(ρ^2)(t). If the trajectory agrees with the results of the OCT simulation, and if, moreover, the values of d and γ as a function of time agree with the OCT simulation, we will have confirmed that we understand the underlying mechanism of the OCT.

Note that the differential equation obtained from this approach will never agree perfectly with the results of a simulation. The above formulation is essentially an adiabatic formulation of the process: the spontaneous emission is considered to be slow compared with the time scale for the purity-preserving transformations generated by the external field, which is what allows us to assume in the theory that the external field manipulation along the isocoherence contour is instantaneous. If the external field is sufficiently intense, the population transfer may become nearly instantaneous relative to the spontaneous emission, and the adiabatic approximation will be excellent.

For the two-level system the optimal cooling strategy is somewhat trivial; it can be seen graphically from Figures 3 and 4. For any value of d, Tr($\dot{\rho^2}$) is maximal for $\gamma = 0$. Thus, the optimal path follows the line $\gamma = 0$ from time 0 until the final time t. Thus, at the very first time step the system should move along the isocoherence contour to the value ($d \leq {}^1/_2$, $\gamma = 0$) and then proceed along the line $\gamma = 0$ until reaching ($d = 0$, $\gamma = 0$). Note that for all values of ($d < {}^1/_2$, $\gamma = 0$) we have Tr($\dot{\rho^2}$) > 0 and the system will cool. At the isolated point ($d = {}^1/_2$, $\gamma = 0$) the cooling rate Tr($\dot{\rho^2}$) = 0; this is a fixed point: instantaneously, the system will not cool nor are there any other points on the isocoherence contour that can be accessed by an external field. However, to second order in time the system's purity will increase, and the point will evolve to values of $d < {}^1/_2$.

We now derive the differential equation for cooling. Taking $\gamma = 0$, we have

$$\mathrm{Tr}(\rho^2) = 2d^2 - 2d + 1 \tag{4.16}$$

$$\mathrm{Tr}(\dot{\rho^2}) = 2(d - d^2) \tag{4.17}$$

Solving eq 4.16 for d gives

$$d = \frac{1 \pm [1 - 2(1 - \mathrm{Tr}(\rho^2))]^{1/2}}{2} \tag{4.18}$$

Substituting this expression into eq 4.17 gives

$$\mathrm{Tr}(\dot{\rho^2}) = 1 - 2\mathrm{Tr}(\rho^2) \mp [2\mathrm{Tr}(\rho^2) - 1]^{1/2} \tag{4.19}$$

We choose the positive sign since the negative sign leads to depurification of the system, or heating. The solution to this differential equation is

$$\mathrm{Tr}(\rho^2) = (1 - (1 - \sqrt{\mathrm{Tr}(\rho^2)_0})\mathrm{e}^{-t/2})^2 \tag{4.20}$$

At long times we have

$$\mathrm{Tr}(\rho^2) = 1 - 2(1 - \sqrt{\mathrm{Tr}(\rho^2)_0})\mathrm{e}^{-t/2}) \tag{4.21}$$

which shows an exponentially fast approach to a pure state.

It is interesting to consider the regions where $\mathrm{Tr}(\dot{\rho^2}) < 0$ as well. We conjecture that these regions correspond to regions where lasing occurs. The conjecture is based on the following considerations:

(1) Note that for $\gamma = 0$, d takes on the values $d > {}^1/_2$ in this region. This corresponds to the conventional criterion for lasing: that population in the excited state be larger than in the ground state.

(2) The fact that in this region the system coherence is decreasing leaves open the possibility that coherence elsewhere can increase. In particular, excitation with incoherent light can lead to emission of coherent light. This is precisely the reverse situation as with laser cooling, where coherent light is transformed to incoherent light (spontaneous emission), increasing the level of coherence of the system.

(3) The regions with $\mathrm{Tr}(\dot{\rho^2}) < 0$ and $d < {}^1/_2$ necessarily imply $\gamma > 0$, i.e. coherences between the ground and excited state. This may correspond to lasing without population inversion, an effect which has attracted a great deal of attention in recent years, and is made possible by coherences between the ground and excited states.

V. Discussion/Conclusions

We have shown numerical results for optimal control of cooling of molecular rotations. The population in the lowest rotational state is almost constant during the interval in which the control field acts, but jumps to 1 at the end of the interval. However, a more general definition of cooling, based on the purity of the state as measured by $\mathrm{Tr}(\rho^2)$, showed that cooling was taking place all along. We then noted that control fields cannot change the quantity $\mathrm{Tr}(\rho^2)$, which can be changed only via spontaneous emission which is inherently uncontrollable! What is the mechanism, then, which the control fields exploit to ensure than the purity will increase monotonically? We proceeded to show that control fields can affect the rate of change of purity via what may loosely be called a two-step process. In the first step, the control field generates a purity preserving transformation on the density matrix; in the second step spontaneous emission changes the purity of the density matrix. The key is to realize that the magnitude and sign of the change in purity induced by the spontaneous emission is sensitive to the prior transformation induced by the control fields. We showed in detail how this works in the simple case of cooling a two-level system. In a separate publication we will present a similar analysis for multilevel systems. In particular, the growth of $\mathrm{Tr}(\rho^2)$ at intermediate times indicates that rotational coherences are forming. It should be possible to show analytically what the optimal phases of these coherences should be and how this choice of phase accelerates the increase in $\mathrm{Tr}(\rho^2)$. If the magnitude and phases of the optimal coherences agree, at least qualitatively, with the results that emerge from an OCT calculation on the corresponding multilevel system, we can say with some confidence that we understand the cooling mechanism which OCT is exploiting.

The formulation presented here is quite general and we believe should also include existing laser cooling schemes for atoms within its framework. Future work will involve reexamining the laser cooling schemes for atoms, e.g. Doppler cooling, Sisyphus cooling, sideband cooling, Raman cooling, and velocity selective coherent population trapping,[6] to see how $\mathrm{Tr}(\rho^2)$ is behaving as a function of time, what the role of the control field is in manipulating the system to a region of favorable $\mathrm{Tr}(\dot{\rho^2})$, and whether any of these schemes can be further optimized on the basis of such an analysis.

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