

Sharpening accepted thermodynamic wisdom via quantum control: or cooling to an internal temperature of zero by external coherent control fields without spontaneous emission

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Abstract. Cooling of internal atomic and molecular states via optical pumping and laser cooling of the atomic velocity distribution, rely on spontaneous emission. The outstanding success of such examples, taken together with general arguments, has led to the widely held notion that radiative cooling requires spontaneous emission. We here show by specific examples and direct calculation, based primarily on breaking emission–absorption symmetry as in lasing without inversion, that cooling of internal states by external coherent control fields is possible. We also show that such coherent schemes allow us to practically reach absolute zero in a finite number of steps, in contrast to some statements of the third law of thermodynamics.

1. Introduction

The laws of thermodynamics have aptly been described as expressions of human frustration. The first law implies that there are no perpetual motion machines which would provide useful work or heat periodically. The second law teaches us that it is only possible to extract all of the thermal energy of a body as useful work by coupling to a reservoir at T = 0. However, the third law seems to have the consequence, that it is impossible to reach T = 0, where the entropy vanishes. As such, the fundamental laws of thermodynamics are very useful in telling us what is not possible.

Moreover, thermo-statistical wisdom is being supplemented and refined at a rapid pace. For example, the advent of new technology such as the laser has led to new thermodynamical and/or statistical mechanical concepts such as negative

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temperature [1] and phase transitions far from equilibrium [2] on the one hand, and new techniques like laser cooling [3] and breaking of emission-absorption symmetry via quantum control, on the other. Thus it behooves us to carefully reconsider accepted wisdom (or 'dogma') in the light of such advances.

In the present paper we show that the recent theoretical and experimental studies concerning lasing without inversion [4] point the way to extending thermodynamical foundations by cooling to absolute zero (in a physical sense of this notion) via external control fields without spontaneous emission. In the following paragraphs we present our main results by citing three specific examples of common statistical wisdom[†] and indicating our conclusions in juxtaposition. More detailed discussion and comparison with other work will be given in the main body of the paper.

(1) We find that *radiative cooling does not require spontaneous emission*. This is in contrast to some aspects of current thinking, which is aptly summarized as per the following statement.

It may be shown that external control fields, no matter how complicated, cannot lead to cooling; this requires spontaneous emission which is inherently uncontrollable.

To be sure, radiative cooling typically involves spontaneous emission. For example, optical pumping [5] is governed by spontaneous emission. Likewise, cooling of centre of mass (COM) degrees of freedom [3] is achieved as the moving atoms go through cycles of absorption and spontaneous emission of radiation. In fact, the effective temperature for a laser cooled gas is typically (but not always [6]) governed by the spontaneous emission rate into the lowest levels. The outstanding success of radiative cooling schemes has elevated the notion of cooling via spontaneous emission to the level of a kind of accepted opinion such that spontaneous emission is held to be an essential ingredient in radiative cooling. In the present paper we show that radiative cooling can be achieved by external coherent control fields without spontaneous emission [7], in particular, although other dissipation mechanisms often play a similar role; and in certain cases without irreversible dynamics of any kind, i.e. via complete coherent control.

(2) We also show that breaking emission-absorption symmetry via external control fields does not violate unitarity. One frequently hears the opposite statement, namely that unitarity forbids cooling via external coherent control fields. This—incorrect—argument runs as follows.

Consider an atom with upper state $|a\rangle$ and lower state $|b\rangle$ the U matrix description of emission is $U|a\rangle = |b\rangle$ but if absorption is cancelled, then $U|b\rangle = |b\rangle$. And if we multiply by U^{\dagger} , then we have $|a\rangle = U^{\dagger}|b\rangle$ (emission) and $|b\rangle = U^{\dagger}|b\rangle$ (no absorption). Thus we have a contradiction to $|a\rangle \neq |b\rangle$, and so unitarity forbids breaking emission-absorption symmetry by coherent control fields.

[†]The quotes given in the following three examples are from papers/textbooks/talks of outstanding workers in the field. However, we prefer not to attribute the quotes to specific individuals since they represent generally held opinions and we do not wish to criticize colleagues unfairly. That is, we hope to offend everyone, not just a few. Following the logic of LWI we show that cooling via external control fields is possible, whereby the interaction Hamiltonian describing the process is non-Hermitian. That is, a cooling process can be realized by breaking emission–absorption symmetry via coherent control. We give several examples and indicate how they can be, and are being, profitably applied to other problems in thermodynamics.

(3) Conventional wisdom teaches that there is no way leading to absolute zero temperature, to wit:

It is impossible by any procedure, no matter how idealized, to reduce the temperature of any system to absolute zero in a finite number of operations.

This is a correct statement under the usual macroscopic schemes due to the fact that heat capacity vanishes as we approach absolute zero.

However, we may envision coherent cooling schemes which are not so limited and we present and analyse such a scheme. The proposed quantum cooler is not limited by the usual entropy arguments. That is, the third law need not apply to the process of cooling by coherent control. Thus, we find that the third law of thermodynamics—as it regards the availability of the absolute zero temperature—is not an entirely general law. This means that there is in fact a possibility to cool to absolute zero, with a physical understanding of this notion, in a rather small number of steps.

In the next section we present the LWI based cooling scheme and in section 3 we present a cooling protocol based on the 'state selection maser' such as that of the original NH_3 maser of Gordon, Zeiger and Townes and/or the H maser of Kleppner and Ramsey and conclusions are given in section 4.

2. Cooling by breaking emission-absorption detailed balance

2.1. Cooling via adiabatic 'counter intuitive' pulsing

Proceeding with the analysis of cooling via external control fields, consider the situation in figure 1. In the usual optical pumping approach to cooling, atoms are promoted from c to a and decay via spontaneous emission on the direct $a \rightarrow b$ transition and the cooling proceeds on the time scale of γ_b^{-1} . In the present case γ_b is equal to zero, i.e. there is no spontaneous emission on the direct transition and cooling via optical pumping is not possible.

However, we can use coherent control to transfer the atomic population to b as in figure 1. There we see a short high power π pulse that promotes all atoms from bto a. After several times γ_c^{-1} we apply the counter intuitive pulse sequence, namely Ω_b 'dresses' the empty b state first, followed by Ω_c , which transfers all population from c to b.

The Hamiltonian for this procedure is

$$H(t) = \hbar \Omega_b(t) [|a\rangle \langle b| + |b\rangle \langle a|] + \hbar \Omega_c(t) [|a\rangle \langle c| + |c\rangle \langle a|], \tag{1}$$

where the notation is explained in figure 1 (b). By applying the pulses in the socalled counter-intuitive sequence the atom evolves from the initial dark state at t = 0



Figure 1. (a) Optical pumping from $|c\rangle$ to $|b\rangle$ via a pulse of Rabi frequency Ω_c due to spontaneous emission coupling levels $|a\rangle$ and $|b\rangle$. (b) Coherent control protocol demonstrating cooling without direct spontaneous emission on the $|a\rangle$, $|b\rangle$ transition. This is accomplished by first applying a π pulse promoting all of $|b\rangle$ atoms to $|a\rangle$. After several radiative decay times the atoms are in state $|c\rangle$ and the counterintuitive pulse sequence is applied transferring the population to the ground state $|b\rangle$. Note that spontaneous emission between $|a\rangle$ and $|b\rangle$ is essential for cooling by optical pumping, but not for cooling via coherent control.

$$|\Psi_{\text{dark}}(0)\rangle = \frac{\Omega_b(0)|c\rangle - \Omega_c(0)|b\rangle}{\left[|\Omega_b(0)|^2 + |\Omega_c(0)|^2\right]^{1/2}} \bigg|_{\Omega_b(0) \gg \Omega_c(0)} \Longrightarrow |c\rangle, \tag{2}$$

which is 'dark' in the sense that $H(0)|\Psi_{dark}(0)\rangle = 0$; to the final dark state $|b\rangle$ at time τ

$$\Psi_{\text{dark}}(\tau)\rangle = \frac{\Omega_b(\tau)|c\rangle - \Omega_c(\tau)|b\rangle}{\left[\left|\Omega_c(\tau)\right|^2 + \left|\Omega_b(\tau)\right|^2\right]^{1/2}} \bigg|_{\Omega_c(\tau) \gg \Omega_b(\tau)} \Longrightarrow - \left|b\right\rangle \tag{3}$$

for which $H(\tau)|\Psi_{\text{dark}}(\tau)\rangle = 0$. The transition from $|c\rangle$ to $|b\rangle$ proceeds, of course, by adiabatically turning the fields $\Omega_b(t)$ and $\Omega_c(t)$ on and off [8]. We note that the adiabatic transfer process proceeds on a time scale governed by the Rabi frequency and is independent of the decay rate.

Please note the following key points: (a) we have shown that cooling is possible even without decay on the direct transition; (b) coherent control provides a potentially useful tool for more effective cooling when $\gamma_b \ll \gamma_c$.

2.2. Cooling via Fano interference and recycling

The next example is based on the experimental reality [9] of lasers which operate without population inversion (LWI). In such devices the absorption– emission symmetry present in most radiating systems is broken. That is, LWI works by arranging the individual radiators (e.g. atoms) to emit but not to absorb radiation, an operation that can be used to yield cooling of internal atomic states, which is an essential feature of the present paper.

We focus on the LWI of Harris–Fano type [10, 11] which illustrates the nonreciprocity between absorption and emission in systems displaying Fano-type interference profiles. In figure 2 an arch-type Fano system is indicated in which an excited state doublet is coupled to a tunnelling [12] Fano continuum.

For the tunnel coupled states [13] of figure 2 we have

$$\dot{\alpha}_1 = -i\varDelta\alpha_2, \quad \dot{\alpha}_2 = -i\varDelta\alpha_1 - 2\gamma\alpha_2,$$
(4)

where Δ is the tunnelling rate and γ is the removal rate. Now we include a laser field of frequency ν which resonantly couples the ground state $|b\rangle$ with the excited $|\alpha_2\rangle$, and the Rabi frequency is denoted by Ω_l .

The tunnelling interaction is diagonalized in the usual way by introducing $2^{1/2}|a_{2,1}\rangle = |\alpha_2\rangle \pm |\alpha_1\rangle$ yielding

$$\dot{a}_2 = -\mathrm{i}\varDelta a_2 - \gamma a_2 - \gamma a_1, \quad \dot{a}_1 = \mathrm{i}\varDelta a_1 - \gamma a_1 - \gamma a_2. \tag{5}$$

Please note, the damping terms now couple a_1 and a_2 , which is the essence of Fano interference.

Writing the energies of states $a_{1,2}$ related to b as $\nu \pm \Delta$, where ν is the centrally tuned laser frequency, we arrive at

$$\dot{\Psi} = -\Gamma \Psi - \frac{\mathrm{i}}{\hbar} (H_0 + V) \Psi, \qquad (6)$$

where



Figure 2. (a) Dressed state picture in which tunnelling is incorporated into states a_1 and a_2 ; recycling of the electron is depicted by dotted lines. (Inset shows tunnelling between levels α_1 and α_2 at rate Δ). (b) Initial thermal state is cooled by (multiple) LWI interactions with coherent radiation cooling sub-bands in a double quantum well. The upper part of (b) exhibits the thermal and the lower part of the dark state.

and $\Delta = \omega_2 - \nu$, $\Omega_{1,2} = \wp_{1,2} E_0/\hbar$ (= $\Omega_l/2^{1/2}$ in our case), and $\wp_{1,2}$ are the dipole matrix elements between b and $a_{1,2}$. In addition, H_0 is the diagonal part of the given matrix (with elements $\pm \Delta$ and 0).

It is useful to rewrite equation (6) in a dressed state picture in which Γ is diagonal ($\tilde{\Gamma}_{11}/\gamma = 1 + x$, $\tilde{\Gamma}_{22}/\gamma = 1 - x$, where $x = (1 - \tilde{\Delta}^2)^{1/2}$ with $\tilde{\Delta} = \Delta/\gamma$; and zero for other components). In such a basis we find [14] that the transformed interaction matrix $2^{1/2}\tilde{V}$ is given by

$$\begin{bmatrix} 0 & 0 & [\Omega_2(x+i\tilde{\Delta})+\Omega_1]/x \\ 0 & 0 & [\Omega_2(x-i\tilde{\Delta})-\Omega_1]/x \\ [\Omega_2+\Omega_1(x-i\tilde{\Delta})] & [\Omega_2-\Omega_1(x+i\tilde{\Delta})] & 0 \end{bmatrix}.$$
 (7)

Note that $(\tilde{V})_{ij} \neq (\tilde{V})_{ji}^*$, so the transformed interaction Hamiltonian is non-Hermitian. We emphasize that it is this non-Hermitian nature of \tilde{V} that accounts for the non-unitary dynamics, thus breaking emission–absorption symmetry. It is not difficult to show, see [14] for details, that this implies the LWI result,

LWI absorption rate :
$$\dot{\rho}_{bb} = -\kappa (\Omega_1 - \Omega_2)^2 \rho_{bb},$$
 (8)

where $\kappa = 2/\Delta^2$.

To appreciate the significance of (8) recall that for a detuned two-level system (TLS) with the upper state decaying into another state $|d\rangle$ at a rate γ we find

TLS absorption rate : $\dot{\rho}_{bb} = -L\Omega^2 \rho_{bb},$ (9)

TLS emission rate :
$$\dot{\rho}_{aa} = -L\Omega^2 \rho_{aa} - \gamma \rho_{aa},$$
 (10)

where $L = \gamma / [\Delta^2 + (\gamma/2)^2]$.

Thus, for the TLS, the emission rate from $|a\rangle$ to $|b\rangle$ due to the coherent drive is the same as the rate of absorption from $|b\rangle$ to $|a\rangle$. But in the LWI case of equation (8) things are very different, e.g. when $\Omega_1 = \Omega_2$ absorption vanishes. Further when the system is prepared in the state $|a_1\rangle$ the rate of emission is essentially the same as that given by equation (10), that is the LWI emission rate is essentially the same as that given by the usual TLS gain analysis. This is the basis of the Harris–Fano lasing without inversion.

The preceding shows how cooling can be achieved without spontaneous emission: LWI amplification of the external field serves to cool the internal atomic degrees of freedom. We emphasize that the non-unitary dynamics which allows breaking of emission-absorption symmetry follows from the non-Hermitian nature of \tilde{V} as given by equation (10).

How one uses such a LWI scheme to cool is clarified by introducing the atomic analogue of figure 2. For example, it may be thought that the removal from $|a_1\rangle$ and $|a_2\rangle$ is simply a form of evaporative cooling and may be obtained equally well in the simple TLS associated with equation (10). But this would miss the key point, namely, the electrons in the appropriate ('dark state') combination of $|b\rangle$, $|a_1\rangle$, and $|a_2\rangle$, as discussed in figures 2 and 3, are 'locked in'. And only electrons which are not in this locked-in configuration leave the right-hand quantum well. Furthermore, these electrons can be recycled and eventually induced into the dark state, see figure 2 (*a*).

In order to further clarify this important point, we consider atomic LWI following Imamoglu and Harris [15] as in figure 3(a). In such a case Fano



Figure 3. Atomic version of cooling by transferring atoms from dressed states to the dark states.

interference between $|a_1\rangle$ and $|a_2\rangle$ is achieved by allowing both levels to decay via spontaneous emission. We note, however, that the spontaneous decay is *not* between $|a_1\rangle$, $|a_2\rangle$ and $|b\rangle$. We assume that the spontaneous emission between levels being cooled (i.e. levels $|a\rangle$ and $|b\rangle$ of figures 3 (b) and (c)) is so slow that it can be neglected. Such an upper level doublet can be established via the three-level Λ arrangement of figure 3 (b) by introducing the dressed states $2^{1/2}|a_2\rangle = |a\rangle + |c\rangle$, $2^{1/2}|a_1\rangle = |a\rangle - |c\rangle$. In the tunnelling example the removal was due to the extraction of electrons from the right-hand well, whereas in the atomic example the removal is due to spontaneous emission from $|a\rangle$ to $|c\rangle$. Furthermore, in this Λ system we do not lose atomic population because the decay is within the atomic levels of interest, i.e. the decay from $|a\rangle$ to $|c\rangle$ serves as the recycle process.

Next, consider the cooling cycle of figure 3(b). There the original thermal distribution is cooled by the two-step process of first applying the resonant coherent control fields having Rabi frequencies Ω_b and Ω_c . The cooling process may be understood by noting that the dark state $\Omega|D\rangle = \Omega_c|b\rangle - \Omega_b|c\rangle$ (where $\Omega^2 = \Omega_b^2 + \Omega_c^2$) does not couple to the radiation fields while the bright state $\Omega|B\rangle = \Omega_b|b\rangle + \Omega_c|c\rangle$ does. Thus the bright state will be promoted to $|a\rangle$ by the action of radiation and the decay as in the previous example. In the tunnelling example the removal was due to the extraction of electrons from the right-hand well, whereas in the atomic example the removal is due to spontaneous emission.



Figure 4. Time evolution of the populations $|a_i|^2$ and $|b(t)|^2$ with the initial conditions (a) b(0) = 1 and (b) $a_{2,1}(0) = \pm 1/2^{1/2}$. Note that after a short time population concentrates in b, i.e. the ground state population does not change, and the absorption ceases.

In particular, we see that the atom decays from the upper level $|a\rangle$ to level $|c\rangle$. The decay process is equivalent to the recycling as is illustrated in figure 3 (c). Hence, after a few cycle times, the population will be in the dark state. The density matrix goes from the initial thermal distribution $\rho(0) = Z^{-1} \sum_{\alpha=}^{a,b,c} \exp(-\beta \varepsilon_{\alpha}) |\alpha\rangle \langle \alpha|$, with $Z = \sum_{\alpha=}^{a,b,c} \exp(-\beta \varepsilon_{\alpha})$ to the final dark state $\rho(T) = |D\rangle \langle D| \Longrightarrow |b\rangle \langle b|$ when $\Omega_c \gg \Omega_b$. This is completely at variance from the case in which spontaneous emission couples *a* and *b*. In the LWI case Ω_b controls the cooling rate. In the usual optical pumping case the cooling rate is governed by $\gamma_{a\to b}$. The detailed time evolution for absorption and emission is shown in figure 4.

Finally we make connection with the previous tunnelling Fano interference of figure 2 by noting that the dark state may be written by using the dressed state representation for state $2^{1/2}|c\rangle = (|a_2\rangle - |a_1\rangle)$ yielding

$$|D\rangle = \frac{\Omega_c}{\Omega} |b\rangle - \frac{\Omega_b}{2^{1/2}\Omega} \left(|a_2\rangle - |a_1\rangle \right). \tag{11}$$

Thus we see that the dark state in the case of tunnelling corresponds to the the electron primarily residing in the sub-band b but having small probabilities of residing in bands a_1 and a_2 . In this way it is clear that the electron in the $|D\rangle$ state is indeed trapped, i.e. does not exit the junction.

A similar method of cooling, utilizing emission-absorption asymmetry, has been discussed in [16]. The scheme uses electromagnetically induced transparency and thus is more efficient than conventional cooling methods. However, it is different in many ways when compared to the present scheme. It needs continuous presence of the cooling and driving lasers, and most importantly, the decay on the cooling transition is still present and affects the cooling rate in a considerable manner.

3. Cooling via the state selection maser

The previous LWI based example of cooling via coherent control is simple enough to allow a rather complete analytical solution. However the underlying physics is not widely known. Therefore, we next present a simple, if somewhat idealized, example containing and illustrating many of the previous points. This example involves a simple gedanken experiment based on an extension of a Stern-Gerlach apparatus (SGA), as illustrated in figure 5.

To illustrate, we have a beam of atoms with internal energy-level structure (e.g. hyperfine sublevels) of states a, b and e. The starting thermal mixture corresponds to arbitrary occupations of levels a and b. Level e is so far up that it is practically unoccupied. The beam is then passed through a SGA to separate the a and bcomponents. The crucial step is cooling of *a* atoms to *b* which is achieved through a process similar to the counter-intuitive pulse sequence of section 2.1. The difference is the level structure needed and absence of any kind of decay processes in the present case. A pulse sequence as shown in figure 5 accomplishes transfer of all a atoms to b through an auxiliary upper level e much on the same lines as population transfer from level c to level b through a in figure 1 as discussed in section 2.1. The point to be noted is that cooling is achieved in both cases on a - btransition. The current scheme allows a one-shot cooling mechanism with complete success. Just to note, there is a similar scheme [17] which achieves cooling on a two-level transition through an upper third level. However, the mechanism of cooling is totally different and the presence of heat baths restricts their scheme to conventional macroscopic thermodynamics and maximum efficiency achievable to the Carnot efficiency.

The behaviour of the atomic beam as it passes through the apparatus can be studied through the density matrix formalism. The density matrix representing the atomic system before entering the SGA is



Figure 5. The beam of atoms with internal structure is split by the Stern–Gerlach device, and the population of level a is transferred to level b through adiabatic counter-intuitive pulsing via level e. All these atoms with the lowest level b occupied are collected in Box 2, thus corresponding to a system with internal (spin) temperature zero.

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$$\rho_{0} = \left[\frac{\exp\left(-\beta\epsilon_{a}\right)}{Z}|a\rangle\langle a| + \frac{\exp\left(-\beta\epsilon_{b}\right)}{Z}|b\rangle\langle b|\right]$$
$$\otimes |\psi_{0}(y)\rangle\langle\psi_{0}(y)|\sum_{P}\frac{\exp\left(-\beta\epsilon_{P}\right)}{Z_{P}}|\psi_{P}(z)\rangle\langle\psi_{P}(z)|, \qquad (12)$$

where $Z = \exp(-\beta\epsilon_a) + \exp(-\beta\epsilon_b)$ and $Z_P = \sum_P \exp(-\beta\epsilon_P)$. Tracing over the COM degrees of freedom yields the initial density matrix for the internal atomic states, $\rho_0(\text{internal}) = [\exp(-\beta\epsilon_a)|a\rangle\langle a| + \exp(-\beta\epsilon_b)|b\rangle\langle b|]/Z$.

After the passage through the SGA and transferring the population from level $|a\rangle$ to level $|b\rangle$, which is achieved through the adiabatic counter-intuitive pulsing sequence (see section 2.1, figures 1 and 5), the density matrix (12) becomes

$$\rho_{1} = |b\rangle\langle b|$$

$$\otimes \frac{1}{Z} [\exp(-\beta\epsilon_{a})|\psi_{a}(y)\rangle\langle\psi_{a}(y)| + \exp(-\beta\epsilon_{b})|\psi_{b}(y)\rangle\langle\psi_{b}(y)|]$$

$$\otimes \sum_{P} \frac{\exp(-\beta\epsilon_{P})}{Z_{P}}|\psi_{P}(z)\rangle\langle\psi_{P}(z)|, \qquad (13)$$

and the reduced spin density matrix is now given by $\rho_1(\text{internal}) = |b\rangle\langle b|$, so that $\text{Tr} [\rho_1(\text{internal})^2]^2 = 1$, which signifies that the internal temperature of the system is zero.

We note that the wave packet describing the COM motion of the incident atoms corresponds to a pure case density matrix. Since the incident COM state has a single velocity in the z direction, the time of flight is the same for every atom, and we can choose this time such that population transfer from $|a\rangle$ to $|b\rangle$ is ensured upon passage through the cavity, which contains two fields with Rabi frequencies Ω_b and Ω_c . In effect the atoms are then all prepared in the lowest state $|b\rangle$ and the Boltzmann weighting factors go from being associated with the internal degrees of freedom to the COM degree of freedom. Hence, it is as if the COM motion served as a kind of reservoir into which the internal thermal noise is transferred.

An important point to be noted here is that, even though the incoming atomic beam might have a spread of velocities, the field inside the cavities can be arranged in such a way that cooling by counter-intuitive pulsing (see figure 5) takes place with unit probability. Thus a single step is sufficient to arrive at the internal (spin temperature of zero, in contrast to the third law statement given in the introduction).

4. Conclusions

We have demonstrated that it is possible to affect radiative cooling without spontaneous emission on the direct transition in example 1 (section 2.1) and via a controllable dissipative mechanism which replaces indirect spontaneous emission in example 2 (section 2.2). Finally in example 3 (section 3) we have used a Stern-Gerlach apparatus together with coherent π pulse spin flipping to generate radiative cooling via coherent control fields alone.

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