Entropy exchange in laser cooling

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It is usually presumed that spontaneous emission is necessary to remove the entropy lost by laser-cooled atoms. Here we show that the changes in the laser beams themselves constitute a sufficiently large reservoir of N states accessible to the system that their entropy $S = k_B \ln(N)$ is sufficient to absorb the entropy lost by the atoms in the cooling process. Proper choice of laser parameters could possibly produce cooling of atoms or molecules over a wide range of temperatures without spontaneous emission.

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It is a long-standing and widely held tenet in the laser cooling community that spontaneous emission is required to carry away the entropy lost by a vapor of atoms being cooled. In this paper we show that spontaneous emission is not the only way of removing the entropy, and that the laser fields themselves are capable of absorbing it. We do this by comparing the entropy lost by the cooled atoms with the entropy capacity of the laser fields. This description requires that the light field be included as part of the system, and not just as an externally applied potential.

In the usual force-momentum description of laser cooling, the force is calculated from the momentum of absorbed light $h\nu/c \equiv \hbar k$ or $\mathrm{tr}(\rho \boldsymbol{\nabla}\mathcal{H})$. In the usual energy description, the exchange between atoms and light involves Doppler shifts or spatially dependent light shifts in inhomogeneous optical fields. In these usual $(\Delta E, \Delta p)$ views of laser cooling, the laser light is treated as a classical field with a fixed potential, and the entropy loss is usually dismissed with vague references to spontaneous emission without any proof.

When an atom absorbs light, its energy increase is compensated by an energy decrease of the light field. A popular way to depict this energy conservation process is the Jaynes-Cummings view, where the light field can be described as a number state. We see that energy conservation in atomic absorption requires that the light field be considered as part of the system, and not as an external fixed potential. If the light field is sufficiently intense that a coherent state description $|\alpha\rangle$ is called for, there is a quandary because the annihilation operator a does not change the state: $a|\alpha\rangle = \alpha|\alpha\rangle$. Energy conservation is at risk in such a description, and it will be considered below.

In the $(\Delta E, \Delta p)$ view, a sufficient condition for cooling a gas is the application of a velocity dependent force that either changes sign or becomes negligibly small over some finite velocity range. Thus if the force moves all the atoms in an extended region of velocity space to a narrower region because it nearly vanishes somewhere within the narrower region, then the velocity distribution is compressed. The one-dimensional case is particularly simple to envisage. As long as this compression does not come at the expense of comparable expansion in other dimensions of velocity space, we say the sample is cooled. If there is also no comparable expansion in configuration space, the volume of phase space is also reduced.

Such a description is incomplete because a velocity-dependent force does not conserve energy. Since such non-conservative forces cannot be described in terms of a potential, the energy change of a particle moving from point **A** to point **B** depends on its velocity. However, by including the light field as part of the system, the description becomes conservative because there is simply an energy exchange between the atoms and the light.

As an example, consider the case of Doppler cooling. The resonance condition, and hence the optical force, depends on the Doppler shift seen by moving atoms, $\omega_D \equiv -\vec{k}\cdot\vec{v}$, such that the light frequency $\omega_\ell = \omega_a - \omega_D$, where ω_a is the atomic frequency ($\vec{k}$ opposite to $\vec{v}$ gives a positive ω_D). The kinetic energy change on absorption is $\Delta E_{\mathrm{kin}} = Mv\Delta v = \hbar k v = -\hbar\omega_D$ (for $v \gg \Delta v = \hbar k/M$). Then energy conservation is satisfied with a velocity-dependent force with the light field included in the system. Analogous arguments apply to the velocity dependence of other cases of $(\Delta E, \Delta p)$ exchanges.

The most naive $(\Delta E, \Delta p)$ view of laser cooling violates the unitarity theorem. That is, atoms with initially different velocities have initially orthogonal wave functions because of their different deBroglie wavelengths. After cooling, their deBroglie wavelengths may be sufficiently similar that their wave packets are no longer orthogonal, thereby violating the theorem. Unitarity in laser cooling may be rescued by including the spontaneously emitted light from the excited atoms into the system. This fluorescence generally occupies orthogonal states of the radiation field thereby preserving the theorem. One of the purposes of this paper is to show that changes to the laser field itself are sufficient to preserve unitarity.

In this discussion of Doppler cooling, we have seen that including the optical field provides for satisfying three conditions: (1) energy conservation between the atomic internal energy and the field, (2) energy conservation between the atomic motion and the field, and (3) preservation of unitarity. Similar arguments hold for other laser cooling schemes. In each of these cases, the reduction of entropy that is concomitant with the reduction of atomic phase space volume is usually assumed, without proof, to be compensated by the increase of the entropy of the "required" spontaneously emitted light. Another purpose of this paper is to examine this assumption.

We now extend the discussion beyond the limits of the

usual $(\Delta E, \Delta p)$ view and treat the laser field as a dynamical variable. This notion is substantiated in the very eloquent statement from Ref. [1]:

> "Begin by deciding how much of the universe needs to be brought into the discussion. Decide what normal modes are needed for an adequate treatment of the problem under consideration."

Although this seems a bit superficial at first, in fact it is very profound. A complete description of the dynamics of any of these laser cooling processes requires that the entire light field be considered as part of the system. Only then can it absorb entropy and transport it out of the system. The atom-light interaction can then populate different field states by stimulated emission just as it does when spontaneous emission populates the otherwise empty field states. Spontaneous emission simply redistributes the light into some subset of a much larger set of accessible states, and stimulated emission can do likewise.

This different description of the light field has absolutely no effect on the $(\Delta E, \Delta p)$ discussion above, but only addresses the optical entropy capacity because now the total wave function includes the light field. Thus the exchange of entropy between the atoms and the light field does not violate the Liouville theorem, unitarity, or Ref. [2] because neither the total entropy of the system nor its phase space volume is reduced, but merely exchanged between its different parts.

Our one-dimensional (1D) comparison with the entropy capacity of the laser fields begins by first finding the entropy lost by atoms, ΔS_a. If the number of atoms is unchanged by the cooling process, the Sackur-Tetrode equation can be used to find $\Delta S_a = k_B \ln(V_{\text{final}}^{(\phi)} / V_{\text{init}}^{(\phi)})$, where $V^{(\phi)}$ is a phase-space volume. (For laser cooling, $\Delta S_a < 0$.) Moreover, the changes of the $V^{(\phi)}$'s are expressed by the product of the compression in velocity space $\Delta v_{\text{init}} / \Delta v_{\text{final}}$ with the expansion in configuration space $\Delta x_{\text{init}} / \Delta x_{\text{final}}$.

For the latter, we note that all laser cooling schemes have a characteristic cooling length and time found from the cooling force and the initial atomic kinetic energy spread, which is determined by the velocity capture range v_c of the force. We first make the natural choice for the initial spatial extent of the atoms as this cooling length Δx. The largest distance atoms can travel during slowing is another Δx, and only the fastest atoms that start near the extreme end of the sample will reach $2\Delta x$. Since this is a small fraction of all the atoms, for calculational convenience we choose the final spread to be $\Delta x_{\text{final}} \approx \sqrt{2}\Delta x$ for each direction, a total of $2\sqrt{2}\Delta x$.

For the width of the initial velocity distribution Δv_{init} we take the velocity capture range v_c and for Δv_{final} we take some measure of the cooling limit. Thus $\Delta S_a = k_B \ln(2\sqrt{2}\Delta v_{\text{final}}/v_c) < 0$, and is typically a few times $(-k_B)$ per atom.

It is straightforward to see how the entropy of the light field can be increased by its interaction with the atoms. The density matrix of the light field alone, ρ_ℓ, can be evaluated in any basis, and we suppose it starts in a pure state where $\rho_\ell = (\rho_\ell)^2$ so its entropy is zero [3].

After interaction with a two level atom having excited and ground states designated by $|e\rangle$ and $|g\rangle$, respectively, the total wave function of the (atom plus light) system is

$$|\Psi\rangle = c_e(t)|e\rangle|m\rangle + c_g(t)|g\rangle|n\rangle, \qquad (1)$$

where $|c_e(t)|^2 + |c_g(t)|^2 = 1$ and $|m\rangle$, $|n\rangle$ represent the different states of the light field when the atom is in $|e\rangle$ or $|g\rangle$ (in the simplest case, $|m\rangle$ and $|n\rangle$ are number states and $m = n-1$). The density operator for the (atom plus light) system is $|\Psi\rangle\langle\Psi|$ and we find the final density matrix for the light alone, ρ_ℓ', by tracing over the atomic states,

$$\rho_\ell' = \langle e|\Psi\rangle\langle\Psi|e\rangle + \langle g|\Psi\rangle\langle\Psi|g\rangle = |c_e|^2|m\rangle\langle m| + |c_g|^2|n\rangle\langle n|. \qquad (2)$$

Thus the matrix of ρ_ℓ' is diagonal with elements $|c_e|^2$ and $|c_g|^2$, and $\rho_\ell' \neq (\rho_\ell')^2$ unless one of them is zero. The original light field whose entropy was zero has been changed to one that is no longer a pure state so its entropy is greater than zero [3,4].

Although the natural choice for a description of the light beams might seem to be the familiar coherent states $|\alpha\rangle$, the usual description of $|\alpha\rangle$'s is not well suited to the exchange of light between beams caused by absorption-stimulated emission cycles of atoms. In particular, the transition term of the Jaynes-Cummings Hamiltonian is $\propto (ab^\dagger + a^\dagger b)$, and although $|\alpha\rangle$ is an eigenstate of a, $a^\dagger|\alpha\rangle$ is a complicated object [5–7]. Moreover, the $|\alpha\rangle$'s are not eigenstates of the Hamiltonian $a^\dagger a$ nor are they orthogonal. Still, they represent a suitable approximation as long as it is recognized that the annihilation operator a indeed *does* change the actual state of the field, even though in the exact (ideal) case, $|\alpha\rangle$ is an eigenstate of a [5–8].

A sample of atoms immersed in a light field is neither a closed system nor is it in thermal contact with a reservoir, so the ordinary thermodynamic entropy cannot be defined. Instead we use the information definition $S = k_B \ln(N)$ where N is the number of states accessible to the system.

If N includes all the empty modes of the radiation field that can be populated by spontaneous emission, its entropy capacity is huge [9,10]. Without spontaneous emission, however, we find the entropy capacity of the light beams ΔS_ℓ from the number of accessible states N that are sufficiently distinct (small overlap) after the stimulated emission processes have redistributed the light energy. Even though the coherent states $|\alpha\rangle$ and $|\alpha'\rangle$ are not very good approximations, we may choose the "distinct state" criterion from the overlap formula $e^{-|\alpha-\alpha'|}$ to be $1/e$. Then this overlap condition requires distinct states to have n values that differ by $\pm 2\sqrt{n}$, where $n \equiv |\alpha|^2$. Since Ref. [5] shows that the approximately coherent states are sub-Poissonnian, there are actually more accessible states than this lower limit estimate.

We need an estimate the field quantum number n, found from the amount of light that can interact with an atom in a large, continuous laser beam. We choose a cylinder of base area equal to the on-resonance atomic absorption cross section $\sigma = 3\lambda^2/2\pi$ and of length ct_{cool}, where t_{cool} is the cooling time mentioned above. For a beam of intensity $I \equiv sI_{\text{sat}}$ we find $n = I\sigma t_{\text{cool}}/h\nu = s\gamma t_{\text{cool}}/2$, where $I_{\text{sat}} \equiv \pi hc\gamma/3\lambda^3$ and $\tau \equiv 1/\gamma$ is the excited state lifetime. For Doppler cooling, $n \sim 10^3$.

TABLE I. A summary of the values for entry into Eq. (3). The velocity limit of Doppler cooling is $v_D \equiv \sqrt{\hbar\gamma/(2M)}$. See text for a discussion. For Sisyphus cooling $\delta \equiv \omega_\ell - \omega_a$ is the laser detuning. For the bichromatic case, 2δ is the frequency difference between the two fields, and typically $\delta \sim 20 - 40\gamma$.

	$\Delta v_{\text{final}}/v_D$	$\omega_r t_{\text{cool}}$	η
Doppler molasses	1	1	$1/\sqrt{s}$
Sisyphus cooling	$\sqrt{s\gamma/2\delta}$	γ/δ	$1/\sqrt{2}$
Bichromatic force	$\sqrt{\delta/2\gamma}$	$\pi/16$	$\sqrt{\approx 50(\gamma/\delta)}$

The maximum change of n required to stop an atom from $v = v_c$ is $\Delta n = Mv_c/\hbar k$ [11]. Since laser cooling requires compression of the velocity distribution from a wide range of velocities to a narrower one, there are many different values of Δn for the different atomic velocities in the sample, so many states of the light field can be populated. We find the number of independent states accessible to the laser field is $N = \Delta n/2\sqrt{n}$ so that

$$\Delta S_a + \Delta S_\ell = k_B \ln \underbrace{\left\{ \frac{2\Delta v_{\text{final}}}{v_r} \frac{1}{\sqrt{s\gamma t_{\text{cool}}}} \right\}}_{\equiv \eta}, \quad (3)$$

where $v_r \equiv \hbar k/M$ is the recoil velocity and $\omega_r \equiv \hbar k^2/2M$ is the recoil frequency. Moreover, there is a one-to-one correspondence between the states of the light field and the atomic motional states [12], just as in the families of velocity selective coherent population trapping [13,14], so their overlap is also small. As long as $\eta \sim 1$, meaning $\Delta S_\ell \sim |\Delta S_a|$, the light field has a large enough capacity to absorb the entropy lost by the atoms.

At this point further evaluation and discussion of Eq. (3) must be done case by case. The results are summarized in Table I. Curiously, there is neither any dependence on v_c in Eq. (3) nor buried in Δv_{final} or t_{cool}.

The first two entries for Doppler molasses are readily calculated from the well-known Doppler limit and velocity damping constant [15,16]. The cooling limit for Sisyphus cooling by polarization gradients, typically a few times v_r [16], is taken from Eq. (4.37) of Ref. [17] ($\delta \equiv \omega_\ell - \omega_a$), and experimentally confirmed in Ref. [18]. The second entry is readily calculated from the increased velocity damping coefficient of Sisyphus cooling [17,19].

The first entry for the bichromatic force, whose two frequencies are detuned by $\pm\delta$, comes from assuming that the nonadiabatic transitions leave the atoms distributed between two ground states at the end of the cooling process (it could be more) [20], and the second entry comes from dividing the velocity cooling range, estimated to be about $1/4$ of the velocity capture range $\pm\delta/4k$, by the (approximately constant) acceleration $2\hbar k\delta/\pi M$ [21]. The value $s = 3\delta^2/\gamma^2$ optimizes the bichromatic force.

Clearly $\eta \sim 1$ for all three cases so we can conclude that the laser beams themselves have sufficient capacity to absorb the entropy lost by the cooled atoms by redistribution of the laser light into a larger number of accessible states. Since the system is "open," the outgoing light beams carry away the entropy. Spontaneous emission is not required for this aspect of laser cooling. In some sense, this hypothesis may be related to cavity cooling (see Ref. [22], and references therein).

We see that the entropy lost by the atoms can be transferred to the light field, and it is not dissipated until the outgoing light hits the walls. Since the walls are *not* part of the system, this final destruction of the light field is indeed a dissipative, nonconservative, and irreversible process. The model here is different from those of previously studied cases [2,9,10] because it includes the applied light field as a dynamical variable rather than as a *c*-number parameter in the Hamiltonian.

Although it is now clear that spontaneous emission is not required to carry away the entropy removed from a vapor atoms during laser cooling, it is also true that spontaneous emission is indeed crucial for energy exchange in some forms of laser cooling. For example, the kinetic energy exchanged between an atom and a light field in a single excitation cannot exceed the larger of $\hbar k v$ or $\hbar \omega_r$.

Since excited atoms do not absorb resonantly, their return to the ground state without the spontaneous emission needed for further absorption can only occur by stimulated emission. If it occurs by the original exciting beam, there is no net energy exchange in the process. If stimulated emission is induced by another beam, for example, the counterpropagating beam in Doppler molasses or Sisyphus cooling, then the absorption-stimulation emission sequence in the opposite order is equally likely when spatially averaged over a wavelength, and on average there is no net energy transfer and no net force.

For Doppler molasses, only spontaneous emission between absorptions can exploit the Doppler shifts to allow energy loss because the average frequency of the emitted light is $\omega_a > \omega_\ell$. For Sisyphus cooling, where the force arises from the absorption-stimulated emission sequences in inhomogeneous fields (e.g., standing waves), spontaneous emission is needed for the transitions between the top of one hill to the bottom of another [17]. Without it, the energy exchange is limited to the light shift, and that is usually much smaller than atomic kinetic energies. Thus the indispensable role of spontaneous emission in many forms of laser cooling is not for entropy dissipation, but for energy transfer.

By contrast, the bichromatic force has no such limits [20,23]. With appropriate parameters and spatial offsets of the standing waves of the two different frequencies, there are positions where the light shifts and detuning just cancel, and exact crossings of the eigenstates can occur. Atoms passing through such positions can mediate the exchange of red-detuned light for blue, and vice versa, resulting in energy exchange between the atoms and the light beams [20]. This mechanism has no significant limit on the energy scale of interest here, and more important, has no counterpart in monochromatic light.

We have seen how the light beams have the capacity to absorb the entropy lost by laser-cooled atoms, and that spontaneous emission is not required for this task. We have also seen that it is indeed required to mediate the energy loss of

the atoms in some forms of laser cooling. The combination of these two results provides a completely different view of the role of spontaneous emission in laser cooling. Among the possible consequences may be the application of laser cooling to molecules, where the spontaneous emission to a plethora of ground state sublevels makes the desirable closed cycle schemes extremely difficult to attain.

[1] W. E. Lamb, Appl. Phys. B: Lasers Opt. **60**, 77 (1995).

[2] W. Ketterle and D. E. Pritchard, Phys. Rev. A **46**, 4051 (1992).

[3] E. Boukobza and D. J. Tannor, Phys. Rev. A **71**, 063821 (2005).

[4] D. Tannor and A. Bartana, J. Phys. Chem. A **103**, 10359 (1999).

[5] G. S. Agarwal and K. Tara, Phys. Rev. A **43**, 492 (1991).

[6] A. Zavatta, S. Viciani, and M. Bellini, Science **306**, 660 (2004).

[7] V. Parigi, A. Zavatta, M. Kim, and M. Bellini, Science **317**, 1890 (2007).

[8] B. R. Mollow, Phys. Rev. A **12**, 1919 (1975).

[9] M. E. Carrera-Patino and R. S. Berry, Phys. Rev. A **34**, 4728 (1986).

[10] S. J. van Enk and G. Nienhuis, Phys. Rev. A **46**, 1438 (1992).

[11] The momentum change for an absorption-stimulated emission cycle between counterpropagating beams is $2\hbar k$, but two beams are affected so the factors of 2 cancel.

[12] S. J. van Enk and H. J. Kimble, Quantum Inf. Comput. **2**, 1 (2002).

[13] A. Aspect, E. Arimondo, R. Kaiser, N. Vansteenkiste, and C. Cohen-Tannoudji, Phys. Rev. Lett. **61**, 826 (1988).

[14] J. Hack, L. Liu, M. Olshanii, and H. Metcalf, Phys. Rev. A **62**, 013405 (2000).

[15] D. J. Wineland and W. M. Itano, Phys. Rev. A **20**, 1521 (1979).

[16] H. Metcalf and P. vander Straten, *Laser Cooling and Trapping* (Springer-Verlag, New York, 1999).

[17] J. Dalibard and C. Cohen-Tannoudji, J. Opt. Soc. Am. B **6**, 2023 (1989).

[18] C. Salomon, J. Dalibard, W. Phillips, A. Clairon, and S. Guellati, Europhys. Lett. **12**, 683 (1990).

[19] Caution is needed since Eq. (4.3) of Ref. [17] defines the Rabi frequency to be twice as large as that used here. Our definition is $\hbar\Omega \equiv \vec{d}\cdot\vec{\mathcal{E}}$.

[20] L. Yatsenko and H. Metcalf, Phys. Rev. A **70**, 063402 (2004).

[21] In Refs. [24,25] the velocity range was measured to be $\delta/k(\pm\delta/2k)$, but in Ref. [20] it was shown that the part of the bichromatic force that arises from the mechanism described therein has a range $\delta/2k(\pm\delta/4k)$.

[22] K. Mürr, Phys. Rev. Lett. **96**, 253001 (2006).

[23] J. Söding, R. Grimm, Y. B. Ovchinnikov, P. Bouyer, and C. Salomon, Phys. Rev. Lett. **78**, 1420 (1997).

[24] M. T. Cashen and H. Metcalf, Phys. Rev. A **63**, 025406 (2001).

[25] M. Partlow, X. Miao, J. Bochmann, M. Cashen, and H. Metcalf, Phys. Rev. Lett. **93**, 213004 (2004).