Reabsorption of Light by Trapped Atoms

Y. Castin,¹ J. I. Cirac,² and M. Lewenstein³

¹*Laboratoire Kastler Brossel, Ecole Normale Supérieure, 24, Rue Lhomond, F-75231 Paris Cedex 05, France*

²*Institut f ür Theoretische Physik, Universität Innsbruck, Technikerstrasse 25, A-6020 Innsbruck, Austria* ³*CEA*y*DRECAM*y*SPAM, Centre d'Etudes de Saclay, 91191 Gif-sur-Yvette Cedex, France*

(Received 19 December 1997)

We investigate the effect of reabsorption of photons in laser cooling of trapped atoms, in view of a possible achievement of Bose-Einstein condensation by purely optical means. Reabsorption can be strongly suppressed by using strongly deformed traps and by reducing the fluorescence rate to a value smaller than the trap frequencies. [S0031-9007(98)06386-8]

PACS numbers: 32.80.Pj, 03.75.Fi, 32.70.Jz, 42.50.Fx

Light reabsorption is believed to be the major obstacle in the way of achieving low temperature, high density, laser cooled, trapped atoms [1–4]. This is one of the main reasons why in order to obtain Bose-Einstein condensation (BEC) of dilute atomic gases [5] additional cooling methods, such as evaporative and sympathetic cooling, have to be used [5,6]. Nevertheless, the achievement of Bose-Einstein condensation via all-optical means remains as one of the challenges of atomic physics, and several experimental groups are pursuing this goal [7–9].

There are several laser cooling schemes that allow us to reach low temperatures, even below the photon recoil limit [10,11]. These schemes exploit single atom "dark states," i.e., states which cannot be excited by the cooling laser, but can be populated via spontaneous emission. However, it is not clear whether these schemes operate at the high densities required for BEC. The dark states are not dark with respect to the photons spontaneously emitted by other atoms. Thus, at sufficiently high densities, dark state cooling may cease to work, since each reabsorption can take one atom out of the dark state and can add, on average, one recoil energy to the atoms.

Usually, one estimates the probability of absorption of a fluorescence photon in terms of the absorption cross section σ . In free space and for an atom at rest (i.e., neglecting Doppler effect) σ is of the order of $1/k_0^2$, where k_0 is the optical wave vector. This result holds even if the fluorescence photon is far from resonance with the bare atom [2]: The atoms are actually dressed by the laser and their fluorescence frequency matches the dressed states' energy difference. According to the simple model of Ref. [2], such a high value of σ excludes the possibility of reaching Bose-Einstein condensation with pure laser cooling in a trap of frequency ω smaller than the recoil frequency, $\omega_R = \hbar k_0^2 / m$, where *m* is the atomic mass.

Several strategies have been proposed to circumvent this difficulty. One can use strongly anisotropic traps, either cigar or disk shaped, so that only the photons emitted along the axis of slow harmonic motion can be reabsorbed. No detailed quantitative analysis of this idea has been given so far. One can also use a trap with a frequency $\omega \approx \omega_R$; a two atom calculation [12] has shown indeed that the relative role of reabsorption is then significantly reduced. It is, however, unclear how this result scales for a large number of atoms, as the atomic density becomes rapidly high in such a strongly confining trap. A third possibility is the *festina lente* scenario, where the fluorescence rate Γ is much smaller than ω [13]. The motion of the atom during the reabsorption process can then no longer be neglected. As is shown in Ref. [13], the reabsorption processes in which the atoms change energy are suppressed, which diminishes the heating effects for $\Gamma \ll \omega$. In actual Raman cooling experiments, Γ is the pumping rate and can be adjusted at will. If, however, Γ becomes too small, the cooling process becomes so slow that trap losses and ground state collisions become problematic. Therefore, in order to evaluate to what extent the *festina lente* regime can lead to high phase space densities, a quantitative analysis of the heating effects as a function of Γ is crucial.

In this Letter we derive a formula for the energy $\Delta E^{(2)}$ reabsorbed by the trapped atoms which is valid for all values of Γ/ω and arbitrary trap geometries. As we will show, only when $\Gamma \gg \omega$, ΔE factorizes into a geometrical form factor and a cross section, and therefore the geometrical and dynamical effects can be separately understood. Our formula is correct in the temperature regime where bosonic effects can be neglected and $k_B T \gg$ $h\omega$. Furthermore, the formula is derived in the low density limit where the mean spacing between the atoms is much larger than $1/k₀$. These regimes correspond to the situation of all present experiments in laser cooling.

In the limit of optically thin samples, we can restrict ourselves to a study of reabsorption between two atoms only. We expect that the results can be directly extended to the optically thick case by summing the scattering events between pairs of atoms as in the derivation of Beer's law [14]. Therefore we consider two distinguishable atoms, *A* and *B*, confined in a harmonic trap. In the simplest model of Raman cooling each atom has two internal ground state levels: $|g\rangle$ and $|e\rangle$ (Fig. 1). The atoms are transferred from $|g\rangle$ to $|e\rangle$ in a velocity selective nondissipative way, and repumped from $|e\rangle$ to $|g\rangle$ through a spontaneous Raman transition. We concentrate here on the latter process, and consider the situation in which initially the atom *A* is in

 $|g\rangle_A$, the atom *B* is in $|e\rangle_B$, and their motional degrees of freedom are described by a Boltzmann distribution with temperature *T*. As shown, for instance, in Ref. [15], the dynamics of the density operator is described by the master equation

$$
\dot{\rho} = \mathcal{L}_0 \rho + \mathcal{L}_1 \rho = (\mathcal{L}_0^A + \mathcal{L}_0^B) \rho + (S_1 + J_1) \rho \,.
$$
\n(1)

The single atom terms are given by $(s = A, B)$

$$
\mathcal{L}_{0}^{s} \rho = \frac{1}{i\hbar} \left[H_{\text{ho}}^{s}, \rho \right]
$$

$$
- \frac{\Gamma}{2} (|e\rangle_{s} \langle e|\rho + \rho |e\rangle_{s} \langle e|) + J_{0}^{s} \rho , (2)
$$

and the harmonic oscillator Hamiltonian is

$$
H_{\rm ho} = \frac{\vec{p}^2}{2m} + \frac{1}{2} m(\omega_x^2 x^2 + \omega_y^2 y^2 + \omega_z^2 z^2), \quad (3)
$$

with \vec{p} and $\vec{r} = (x, y, z)$ being the momentum and position operators. The superoperator

$$
J_0^s \rho = \Gamma \int \frac{d\Omega}{4\pi} e^{-i\vec{k}_0(\Omega)\cdot \vec{r}_s} \sigma_s^- \rho \sigma_s^+ e^{i\vec{k}_0(\Omega)\cdot \vec{r}_s} \quad (4)
$$

describes spontaneous emission of a photon of wave vector $\vec{k}_0(\Omega)$, with $|\vec{k}_0(\Omega)| = k_0 \equiv \omega_0/c$. Here, σ_s^+ $|e\rangle_s\langle g| = (\sigma_s^{-})^{\dagger}$. The interactions terms in Eq. (1) are given by

$$
S_1 \rho = -i \frac{\Gamma}{2} G(\vec{r}_{AB}) (\sigma_A^+ \sigma_B^- + \sigma_A^- \sigma_B^+) \rho + \text{H.c.},
$$

$$
J_1 \rho = \Gamma \int \frac{d\Omega}{4\pi} e^{-i\vec{k}_0(\Omega) \cdot \vec{r}_A} \sigma_A^- \rho \sigma_B^+ e^{i\vec{k}_0(\Omega) \cdot \vec{r}_B} + \text{H.c.},
$$

where $\vec{r}_{AB} = \vec{r}_A - \vec{r}_B$, and $G(\vec{r}) = -\exp(ik_0 r)/k_0 r =$ $G_R(\vec{r}) + iG_I(\vec{r})$ has the Fourier representation:

$$
G_R(\vec{r}) = -\frac{1}{\pi} \int d\lambda \, \mathcal{P} \, \frac{\lambda}{\lambda - 1} \int \frac{d\Omega}{4\pi} \, e^{-i\lambda \vec{k}_0(\Omega) \cdot \vec{r}}, \tag{5a}
$$

$$
G_I(\vec{r}) = -\int \frac{d\Omega}{4\pi} e^{-i\vec{k}_0(\Omega)\cdot\vec{r}}.
$$
 (5b)

 G_R is the dipole-dipole interaction potential and G_I is a modification to the fluorescence rate. We have used a scalar model for the electromagnetic field, which gives the correct far zone dipole-dipole interaction but fails at short range $(r \leq 1/k_0)$. We refer to [16] for a discussion of the short range effects.

FIG. 1. Diagrammatic representation of the considered situation: (a) Two atoms *A* and *B* in a harmonic trap; (b) atomic internal structure.

Our aim is to solve the master equation (1) for $\Gamma t \gg 1$, where both atoms are in the internal state *g*, and then calculate the increase of motional energy ΔE with respect to the initial state. In the considered regime, the thermal energy $k_B T$ is of the order of the recoil energy and is much larger than the $\hbar\omega_i$'s; in this case $k_0\Delta r \gg 1$, where Δr is the typical distance between the atoms. As the matrix elements of \mathcal{L}_1 for two atoms separated by \vec{r}_{AB} scale as $1/k_0r_{AB}$ [since S_1 and J_1 are proportional to $G(\vec{r}_{AB})$ and $G_I(\vec{r}_{AB})$, respectively], we treat this interaction term perturbatively which amounts to expanding ρ in a series: $\rho(t) = \rho^{(0)}(t) + \rho^{(1)}(t) + \rho^{(2)}(t) + \dots$ We find $\rho^{(1)}(t = +\infty) = 0$ and a nonvanishing expression for $\rho^{(2)}(t = +\infty)$. The energy increase $\Delta E = \text{Tr}\{H[\rho(\infty) - \rho^{(2)}(t = +\infty)]\}$ $\rho(0)$] is expanded as $\Delta E^{(0)} + \Delta E^{(2)} + \dots$. We find $\Delta E^{(0)} = \hbar \omega_R/2 = \hbar^2 k_0^2/(2m) = E_R$, where E_R is the recoil energy. The first correction is

$$
\Delta E^{(2)} = -\frac{\hbar \Gamma}{2} \operatorname{Im} \langle (\bar{G}_R - G_R)G \rangle + E_R \operatorname{Im} \langle \bar{G}_I G \rangle, \tag{6}
$$

where the expectation value $\langle \ldots \rangle$ is taken with respect to the Boltzmann distributions for both atoms. We have denoted $G = G(\vec{r}_{AB})$ and defined

$$
\bar{G}_{R,I} = \Gamma \int_0^\infty d\tau \, e^{-\Gamma \tau} G_{R,I}[\vec{r}_{AB}(\tau)],\tag{7}
$$

and $\vec{r}_{AB}(\tau) = e^{i(H_{\text{ho}}^A + H_{\text{ho}}^B)\tau} \vec{r}_{AB} e^{-i(H_{\text{ho}}^A + H_{\text{ho}}^B)\tau}$. In the considered limit $k_0 \Delta r \gg 1$, Eq. (6) can be further simplified and we get

$$
\Delta E \simeq E_R(1 + d + p), \tag{8a}
$$

$$
d = -(\Gamma/\omega_R) \operatorname{Im}\langle \bar{G}_R G_R \rangle, \tag{8b}
$$

$$
p = \text{Re}\langle \bar{G}_I G_I \rangle. \tag{8c}
$$

Here *d* is the energy change due to the dipole-dipole interaction potential G_R and p is the probability of reabsorption of the fluorescence photon.

We perform the averages using Eqs. $(5a)$ and $(5b)$:

$$
p = \Gamma \int_0^\infty dt \, e^{-\Gamma t} \int \frac{d\Omega}{4\pi} \frac{d\Omega'}{4\pi} \cos\left(\sum_{i=x,y,z} \frac{\omega_R}{\omega_i} n_i n_i' \sin \omega_i t\right) \times \exp\left[-\frac{1}{2} \sum_{i=x,y,z} k_0^2 \Delta r_i^2 [n_i^2 + n_i'^2 - 2n_i n_i' \cos(\omega_i t)]\right],
$$
\n(9)

and a similar expression holds for *d*. We have introduced unit vectors $\vec{n} = \vec{k}_0(\Omega)/k_0$, $\vec{n}' = \vec{k}_0(\Omega')/k_0$. The quantities $\Delta r_i = \sqrt{2k_B T/m\omega_i^2}$, $i = x, y, z$ are the rms coordinates for the relative motion between the two atoms. We now discuss two different regimes depending on the ratio of Γ to the trap frequencies.

(*i*) *Free space limit.*—In the limit $\Gamma \gg \omega_i$, $i = x, y, z$, the atoms do not have time to oscillate in the trap. The integral over solid angles and time in Eq. (9) can be performed using the Laplace method. Only the saddle points with $t = 0$ contribute, as Γ is large, and we get

$$
\Delta E^{(2)} = E_R \frac{\sigma_E}{4\pi} \left\langle \frac{1}{r_{AB}^2} \right\rangle, \tag{10}
$$

where σ_E is the cross section for energy reabsorption:

$$
\sigma_E = \frac{4\pi}{k_0^2} \text{Re}\left[\frac{i\sqrt{\pi} \Gamma \zeta^*}{2\omega_R} \exp(\zeta^2) \text{erfc}(\zeta)\right]. \qquad (11)
$$

In this expression, erfc (\cdot) denotes the complementary In this expression, erfc(\cdot) denotes the complementary
error function, and $\zeta = (\Gamma + i\omega_R)/\sqrt{2} \omega_D$, where $\omega_D =$ $k_0\omega_j\Delta r_j = k_0\Delta v$ is a *j*-independent Doppler width. In Eq. (10) the dynamical effects inherent to σ_E are nicely separated from the geometrical effects contained in the $\langle 1/r_{AB}^2 \rangle$ factor.

Let us discuss the dynamical part first. In the limit $\Gamma \gg \omega_D$, the atomic motion is negligible and σ_E reduces to the resonant absorption cross section $4\pi/k_0^2$. As Γ becomes much smaller than the Doppler width ω_D the emitted photon can be reabsorbed only when the atom *A* is in a narrow velocity class of width Γ/k_0 . In effect, the cross section decreases linearly with Γ :

$$
\sigma_E \sim \frac{4\pi}{k_0^2} \frac{\sqrt{\pi} \Gamma}{2\sqrt{2} \omega_D} e^{-\omega_R^2/2\omega_D^2}.
$$
 (12)

In this limit we note that $\Delta E^{(2)} = E_R p$ is dominated by the reabsorption process [i.e., $d = O(\Gamma^2)$]. The full dependence of σ_E with Γ is plotted in Fig. 2.

We now turn to the influence of the trap geometry. Restricting to cylindrically symmetric traps around the *z* axis, we deform the trap keeping its mean oscillation frequency $(\omega_x \omega_y \omega_z)^{1/3}$ constant; the ratio of $\Delta E^{(2)}$ for the deformed trap to the $\Delta E^{(2)}$ for the isotropic trap is a function of ω_z/ω_x that can be calculated analytically.

FIG. 2. Cross section σ_E as a function of Γ for $\omega_D^2 = 10\omega_R^2$. Short-dashed (long-dashed) line represents the limiting cases $\Gamma \gg \omega_D$ ($\Gamma \ll \omega_D$) discussed in the text.

Figure 3 shows that by deforming the trap the reabsorption probability decreases. Note, however, that deformations of the order $\omega_z/\omega_x = 20$ (disk shape) and $\omega_z/\omega_x = 1/30$ (cigar shape) are needed to obtain a modest 50% decrease.

(*ii*) *Festina lente.*—This limit is defined by $\Gamma \leq \omega_i$ and is more difficult to analyze because atoms have time to oscillate before undergoing spontaneous emission. In the case of a *spherically symmetric trap,* we include in the Laplace method the saddle points $t = 0, \pi/\omega, \ldots$; they are such that $n'_i = \pm n_i$, $i = x, y, z$, where \vec{n} can take any value on the sphere. We find that the form of Eq. (10) is preserved with a cross section now depending on the trap frequency ω . As in Eq. (12), only the contribution of p to $\Delta E^{(2)}$ is relevant:

$$
\sigma_E = \frac{4\pi}{k_0^2} \operatorname{Re} \left\{ \frac{\sqrt{\pi} \Gamma}{2\sqrt{2} \omega_D} \exp(\zeta^2) \times \left[\operatorname{erfc}(\zeta) + \frac{2}{e^{\pi \Gamma/\omega} - 1} \right] \right\}.
$$
 (13)

A similar formula applies to the case of the *asymmetric trap with "commensurable" frequencies, with* $\omega_i = \tilde{\omega} q_i$ *,* where the integers $q_i \leq k_0 \Delta r_i$ have no (trivial) common factors. The reabsorption probability is then dominated by contributions of saddle points with $t = 0, \pi/\tilde{\omega}, \ldots$, and $n'_i = \pm n_i$, \vec{n} taking any value on the sphere. The corresponding formula is obtained from (13) by replacing ω by $\tilde{\omega}$ in the last term in the square brackets. In the limit of a vanishing Γ , σ _E tends to a finite value $(4\pi\tilde{\omega}/\sqrt{2\pi}\omega_D k_0^2)$ exp $(-\omega_R^2/2\omega_D^2)$. This is illustrated in Fig. 4, where the two upper curves represent the reabsorption probability calculated numerically from Eq. (9) (solid line) and using the saddle point method (diamonds) for $\omega_x = 0.06\omega_R$, $\omega_y = 0.08\omega_R$, and $\omega_z = 0.1\omega_R$.

The lower curve in Fig. 4 corresponds to the case of *"incommensurable" frequencies,* which is the situation in a real experiment. Here, \sim 2% random variations of the frequencies have been done, so that $\omega_x = 0.061793 \omega_R$,

FIG. 3. Ratio of $\Delta E^{(2)}$ for a cylindrically symmetric trap to $\Delta E^{(2)}$ for an isotropic trap as a function of ω_z/ω_x , for a fixed mean oscillation frequency $(\omega_x \omega_y \omega_z)^{1/3}$.

FIG. 4. Reabsorption probability as a function of Γ for $\omega_D^2 =$ $10\omega_R^2$ in an asymmetric trap with commensurable (upper solid line) and incommensurable (lower solid line) frequencies (see text). Diamonds represent the result of the analytic formula, Eq. (13).

 $\omega_y = 0.079272\omega_R$, and $\omega_z = 0.097989\omega_R$. Even such a small frequency variation changes the results appreciably and in a favorable way for small Γ . For the incommensurable case we have obtained an analytical result in the limit $\Gamma \ll \omega_i$. The dominant contribution then comes from $\vec{n} \approx \vec{n}'$ close to an eigenaxis of the trap:

$$
p \sim \sqrt{\frac{8}{\pi^5}} \frac{(\sum_i \Delta r_i)}{k_0^5 (\prod_i \Delta r_i)^2} \ln^2 \left[k_0 \left(\prod_i \Delta r_i \right)^{1/3} \right] e^{-\omega_R^2/2\omega_D^2}.
$$
\n(14)

As $k_0 \Delta r_i \gg 1$ this expression is much smaller than the limit of *p* for $\Gamma \rightarrow 0$ in the commensurable case. This can be understood from Eq. (9): Expanding the time dependentification in Eq. (2). Expanding the time dependent integrand in a Fourier series with frequencies $\sum_i s_i \omega_i$, where s_i are integers, only the components with a vanishing frequency have a nonzero contribution when $\Gamma \rightarrow 0$. When the trap frequencies are incommensurable, only the term with $s_x = s_y = s_z = 0$ survives and Eq. (14) is obtained. For commensurable frequencies an infinite number of terms, all positive, survive.

Let us finally show the implications of our results for the achievement of BEC with all-optical means. In a harmonic trap, the number of atoms *N* required to reach BEC is $N \ge N_c \simeq 1.2(k_BT)^3/\hbar^3\omega_x\omega_y\omega_z$. To satisfy this condition with a small probability of reabsorption, we should require $N_c p \leq 1$. For the experimentally relevant parameters of Fig. 4 we find, in the limit $\Gamma \rightarrow 0$, $N_c p \simeq$ 0.5 and $N_c p \approx 0.04$ for the cases of "commensurable" and "incommensurable" frequencies, respectively.

In summary, we have derived a formula describing the energy reabsorbed and the reabsorption probability for trapped atoms at sufficiently high temperatures, where quantum statistical effects are negligible, and in the optically dilute regime. The formula reflects the effects of the trap geometry as well as of the atom motion. In the free space limit ($\Gamma \gg \omega$) the reabsorption probability scales as $1/(k_0\Delta r)^2$. It can be strongly reduced by decreasing Γ/ω ,

and deforming the trap. In the *festina lente* regime the reabsorption probability scales as $1/(k_0\Delta r)^3$ for traps with commensurable frequencies. For incommensurable trap frequencies one can reduce this probability even further.

We acknowledge discussions with I. Bouchoule, J. Dalibard, C. Salomon, and P. Zoller, and we thank C. Herzog and A. Sinatra for a careful reading of the manuscript. M. L. and Y. C. thank the University of Innsbruck for hospitality.

- [1] D. W. Sesko, T. G. Walker, and C. E. Wieman, J. Opt. Soc. Am. B **8**, 946 (1991).
- [2] M. Olshan'ii, Y. Castin, and J. Dalibard, in *Proceedings of the 12th International Conference on Laser Spectroscopy,* edited by M. Inguscio, M. Allegrini, and A. Lasso (World Scientific, Singapore, 1996).
- [3] A. M. Smith and K. Burnett, J. Opt. Soc. Am. B **9**, 1256 (1992).
- [4] K. Ellinger, J. Cooper, and P. Zoller, Phys. Rev. A **49**, 3909 (1994).
- [5] M. H. Anderson *et al.,* Science **269**, 189 (1995); K. B. Davis *et al.,* Phys. Rev. Lett. **75**, 3969 (1995); C. C. Bradley, C. A. Sackett, and R. G. Hulet, *ibid.* **78**, 985 (1997).
- [6] C. J. Myatt, E. A. Burt, R. W. Ghrist, E. A. Cornell, and C. E. Wieman, Phys. Rev. Lett. **78**, 586 (1997).
- [7] D. Boiron, A. Michaud, P. Lemonde, Y. Castin, C. Salomon, S. Weyers, K. Szymaniec, L. Cognet, and A. Clairon, Phys. Rev. A **53**, R3734 (1996).
- [8] M. Rauner, M. Schiffer, S. Kuppens, G. Wokurka, G. Birkl, K. Sengstock, and W. Ertmer, *Laser Spectroscopy XIII* (Springer-Verlag, Heidelberg, 1997).
- [9] T. Müller-Seydlitz, M. Hartl, B. Breyger, H. Hansel, C. Keller, A. Schnetz, R.J.C. Spreeuw, T. Pfau, and J. Mlynek, Phys. Rev. Lett. **78**, 1038 (1997).
- [10] J. Lawall, S. Kulin, B. Saubamea, N. Bigelow, M. Leduc, and C. Cohen-Tannoudji, Phys. Rev. Lett. **75**, 4194 (1995).
- [11] H.J. Lee, C.S. Adams, M. Kasevich, and S. Chu, Phys. Rev. Lett. **76**, 2658 (1996), and references therein.
- [12] U. Janicke and M. Wilkens, Europhys. Lett. **35**, 561 (1996).
- [13] J. I. Cirac, M. Lewenstein, and P. Zoller, Europhys. Lett. **35**, 647 (1996).
- [14] See, for example, L. Allen and J. H. Eberly, *Optical Resonance and Two-Level Atoms* (Dover, New York, 1975).
- [15] R. H. Lehmberg, Phys. Rev. A **2**, 883 (1970).
- [16] T. W. Hijmans, G. V. Shlyapnikov, and A. L. Burin [Phys. Rev. A **54**, 4332 (1996)] have shown that for a farblue detuned pumping laser, the short range dipoledipole interaction gives a contribution to the reabsorption probability $\approx 1/(k_0\Delta r)^3$, where Δr is the typical distance between the two atoms. These authors have suggested that this effect can be suppressed by using a far red detuned laser tuned between two molecular levels. [See also K. Burnett, P. S. Julienne, and K.-A. Suominen, Phys. Rev. Lett. **77**, 1416 (1996).]