

# Two-Photon Doppler Cooling of Calcium Atoms

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## Abstract

*A new possibility of laser cooling of Calcium atoms using a two-photon transition is analyzed. We investigate the possibility of Doppler cooling of Calcium using its two-photon  $(4s^2) ^1S_0 - (4s5s) ^1S_0$  transition, with excitation in near resonance with the  $(4s4p) ^1P_1$  level. The two-photon absorption of laser beams at 423 and 1034 nm greatly increases the two-photon rate, allowing an effective transfer of momentum. The experimental implementation of this technique is discussed and we show that two-photon cooling can be used to achieve a temperature limit of about 1/7 of the one-photon Doppler limit reached in a conventional MOT.*

## Introduction

In metal-alkali elements the hyperfine structure is the basis of sub-Doppler techniques, as polarization gradient cooling, which allows the achievement of microKelvin and sub-microKelvin temperatures. These techniques are not applicable to alkaline-earths and, therefore, until a few years ago the smaller temperatures achieved with these elements were in the miliKelvin range. Recently, microKelvin temperatures, close to the recoil limit, have been achieved for Strontium by Doppler cooling using the narrow  $^1S_0 - ^3P_1$  intercombination transition ( $\gamma/2\pi = 7.6$  kHz) [1], and also for Ytterbium ( $\gamma/2\pi = 182$  kHz) [2], which has a level structure similar to Calcium. Cooling using the intercombination transition has also been demonstrated for Calcium, where the relatively long lifetime of the  $^3P_1$  level (0.38ms) had to be reduced by coupling it to another level, in order to make the cooling process effective [3,4]. With the exception of Ytterbium, cooling on the intercombination transition was used as a second stage, after initial pre-cooling to milliKelvin temperatures with the strong  $^1S_0 - ^1P_1$  dipole transition. We will consider the possibility of performing two-photon Doppler cooling of atomic neutral Calcium, using its  $(4s^2) ^1S_0 - (4s5s) ^1S_0$  transition. Although two photon transitions rates can be high enough for spectroscopic purposes, allowing the implementation of powerful spectroscopic Doppler-free techniques [5], in order to reduce the atomic velocity they must be high enough to allow an effective and fast transfer of momentum from the light fields to the atom. We show that for Calcium, two-photon cooling is possible and can be used to achieve a Doppler limit near 123  $\mu$ K, about seven times smaller than the Doppler limit achieved with the  $^1S_0 - ^1P_1$  resonant transition.

## Results and Discussions

We will consider the two-photon transition, at frequency  $\omega_{eg}$ , between the ground state  $|g\rangle = (4s^2) ^1S_0$  and the excited state  $|e\rangle = (4s5s) ^1S_0$  (Fig.1). From the  $(4s5s) ^1S_0$  level the atoms quickly decay to the ground state by spontaneous emission via the  $|r\rangle = (4s4p) ^1P_1$  state, with rates of  $\gamma_e = 3.00 \times 10^7 \text{ s}^{-1}$  and  $\gamma = 2.18 \times 10^8 \text{ s}^{-1}$ . Let us consider the experimental case of one atom interacting with two copropagating laser beams, with wavevectors  $k_1 = 2\pi/\lambda_1$  and  $k_2 = 2\pi/\lambda_2$ , and frequencies  $\omega_1$  and  $\omega_2$ . We will consider excitation with photons at  $\lambda_1 = 423$  nm in near resonance with the strong dipole single-photon  $^1S_0 - ^1P_1$  transition, and a second infrared laser at  $\lambda_2 = 1034$  nm (Fig. 1).

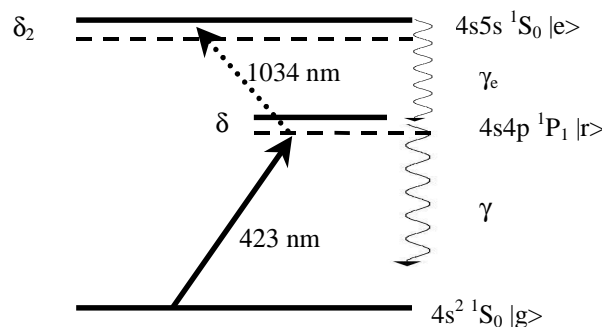


Figure 1. Energy level diagram for two-photon cooling in Ca atoms.

In order to preserve angular momentum in a  $\Delta J = 0$  transition ( $^1S_0 - ^1S_0$ ), we are also assuming  $\sigma_+$  and  $\sigma_-$  polarizations for the two beams at 423 and 1034 nm respectively. The expression for the two-photon transition rate between levels  $|g\rangle$  and  $|e\rangle$  is [6]:

$$\Gamma_{ge} = \frac{|\Omega_1|^2 \cdot |\Omega_2|^2}{\delta^2 + (\gamma/2)^2} \cdot \frac{\gamma_e}{\delta_2^2 + (\gamma_e/2)^2}, \quad (1)$$

where  $\Omega_j$  ( $j = 1, 2$ ) are the Rabi frequencies of the transitions, which can be written in terms of the saturation parameter  $S_j$  and the relaxation rates:  $S_1 = 2 \cdot |\Omega_1|^2 / \gamma = I_1(\omega_1) / I_{1s}$ ,  $S_2 = 2 \cdot |\Omega_2|^2 / \gamma_e = I_2(\omega_2) / I_{2s}$ . For Calcium we have  $I_{1s} \approx 60 \text{ mW/cm}^2$  ( $4s^2 \ ^1S_0 - 4s4p \ ^1P_1$  transition) and  $I_{2s} = 0.6 \text{ mW/cm}^2$  ( $4s4p \ ^1P_1 - 4s5s \ ^1S_0$  transition). Equation (1) can be rewritten with the above definitions as:

$$\Gamma_{ge} = \frac{4 \cdot S_1 \cdot S_2}{1 + S_1 + (2 \cdot \delta / \gamma)^2} \cdot \frac{\gamma_e}{1 + S_1 \cdot S_2 + (2 \cdot \delta_2 / \gamma_e)^2}, \quad (2)$$

where we have included in the last equation the possibility of saturation of the one- and two-photon transitions. Even at low power levels the two-photon transition rate can still be significant. For example, if we suppose  $S_1 = 0.1$  and  $S_2 = 3$ , and assume red detuning of the incident lasers,  $\delta = -\gamma/2$  and  $\delta_2 = -\gamma_e/2$ , the two-photon transition rate will be  $\Gamma_{ge}/2\pi = 1.2 \text{ MHz}$ . The single-photon ( $4s^2 \ ^1S_0 - 4s4p \ ^1P_1$ ) transition rate is:

$$\Gamma_{gr} = \frac{S_1}{1 + S_1 + (2 \cdot \delta / \gamma)^2} \cdot \frac{\gamma}{2}, \quad (3)$$

and for the above parameters  $\Gamma_{gr}/2\pi = 826 \text{ kHz}$ , and the ratio between two-photon and single-photon rates is  $\Gamma_{ge}/\Gamma_{gr} = 1.5$ .

Lets us now consider one stationary atom interacting with the copropagating  $\sigma_+$  and  $\sigma_-$  laser beams. The direct excitation from  $|g\rangle$  to  $|e\rangle$  by the simultaneous absorption of two photons occurs with a rate  $\Gamma_{ge}$ , given by Eq.(2). From the upper level  $|e\rangle$ , the atom spontaneously decays to the intermediate  $|r\rangle$  level with a rate  $\gamma_e$  and, from this level, with a rate  $\gamma$  to the ground state. On average, the time it takes one atom to absorb simultaneously two photons and go back to the ground state by this spontaneous cascade decay is given by:

$$t = \Gamma_{eg}^{-1} + \gamma_e^{-1} + \gamma^{-1}. \quad (4)$$

As the direction of the spontaneous emitted photons is random, the mean spontaneous radiation force in this process is given simply by the ratio of the momentum change  $\hbar(k_1 + k_2)$  and the time  $t$ . If the atom is moving with a velocity  $-v$  in the direction of the laser beams, this mean radiation force can be written, disregarding terms on the order of  $(kv)^2/\gamma^2$ , as:

$$\frac{dp}{dt} = \frac{4 \cdot \hbar \cdot (k_1 + k_2) \cdot S_1 S_2 \cdot \gamma_e}{[1 + S_1 + (2\delta/\gamma)^2] \cdot [1 + S_1 S_2 + (2\delta_2/\gamma_e)^2] + 4S_1 S_2 \cdot (1 + \gamma_e/\gamma)} \times \left\{ 1 - 8 \cdot \frac{[1 + S_1 S_2 + (2\delta_2/\gamma_e)^2] \cdot (\delta \cdot k_1 \cdot v/\gamma^2) + [1 + S_1 + (2\delta/\gamma)^2] \cdot [\delta_2 \cdot (k_1 + k_2) \cdot v/\gamma_e^2]}{[1 + S_1 + (2\delta/\gamma)^2] \cdot [1 + S_1 S_2 + (2\delta_2/\gamma_e)^2] + 4S_1 S_2 \cdot (1 + \gamma_e/\gamma)} \right\}. \quad (5)$$

Adding one pair of  $k_1$  and  $k_2$  laser beams counterpropagating to the first ones, we have a configuration of a two-photon one-dimension optical molasses. In this configuration, the radiation pressure reduces to  $dp/dt = -\alpha_2 v$ , where the damping coefficient  $\alpha_2$  is given by:

$$\alpha_2 = \frac{-64 \cdot \hbar \cdot (k_1 + k_2) \cdot S_1 S_2 \cdot \gamma_e}{\left\{ [1 + 2S_1 + (2\delta/\gamma)^2] \cdot [1 + 4S_1 S_2 + (2\delta_2/\gamma_e)^2] + 4S_1 S_2 \cdot (1 + \gamma_e/\gamma) \right\}^2} \times$$

$$\times \left\{ \frac{\delta}{\gamma} \cdot \frac{k_1}{\gamma} \cdot [1 + 4S_1 S_2 + (2\delta_2/\gamma_e)^2] + \frac{\delta_2}{\gamma_e} \cdot \frac{k_1 + k_2}{\gamma_e} \cdot [1 + 2S_1 + (2\delta/\gamma)^2] \right\} \quad (6)$$

Lets us consider the heating due to the spontaneous emission processes. The damping force reduces the average velocity of the atoms to zero, but the random nature of the spontaneous emission leads to a non-zero mean square velocity. Through a straightforward generalization of the analysis of the random walk process in “one-photon molasses” [7], we see that the momentum diffusion constant in a “two-photon molasses” is:

$$D_2 = \frac{4 \cdot \hbar^2 \cdot (k_1^2 + k_2^2) \cdot (2S_1) \cdot (2S_2) \cdot \gamma_e}{[1 + 2S_1 + (2\delta/\gamma)^2][1 + 4S_1 S_2 + (2\delta_2/\gamma_e)^2]} \quad (7)$$

The Doppler equilibrium temperature is obtained by  $k_B T = D_2 / \alpha_2$  [8]:

$$k_B T = \frac{\hbar \gamma}{2} \cdot \frac{(k_1^2 + k_2^2)}{2 \cdot (k_1 + k_2)} \cdot \left\{ \frac{|\delta|}{\gamma} \cdot \frac{k_1}{[1 + (2\delta/\gamma)^2]} + \frac{|\delta_2|}{\gamma_e} \cdot \frac{\gamma}{\gamma_e} \cdot \frac{(k_1 + k_2)}{[1 + (2\delta_2/\gamma_e)^2]} \right\}^{-1} \quad (8)$$

which is valid for low intensities ( $S_1 \ll 1$ ,  $S_1 S_2 \ll 1$ ). When we take into account higher intensities of the laser beams, the damping coefficient and the momentum diffusion constant become more complicated and the result is an increase of the temperature with increasing intensity, as in the case of “one-photon optical molasses” [7]. We recognize the first term on Eq.(8),  $\hbar \gamma / 2$ , as the one-photon Doppler limit, which for Calcium is 831  $\mu$ K. The minimum temperature in the “two-photon” molasses occurs for the laser detunings  $\delta_2 = -\gamma_e/2$  and  $\delta = -\gamma/2$ , and is 123  $\mu$ K for Calcium.

We should consider both one- and two-photon cooling processes jointly, since the  $^1S_0 - ^1P_1$  and  $^1S_0 - ^1S_0$  transitions occurs simultaneously. Depending on the detunings and intensities of the incident laser beams, one process can dominate the other. The effective damping and diffusion coefficients that jointly take into account the one- and two-photon cooling processes can be written as:

$$\alpha_{eff} = \alpha_1 + \alpha_2 \quad , \quad D_{eff} = D_1 + D_2 \quad , \quad (9)$$

where the coefficients  $\alpha_1$  and  $D_1$  for the case of “one-photon optical molasses” were discussed in several references [7,8]. The Doppler equilibrium temperature is obtained by  $k_B T_D = D_{eff} / \alpha_{eff}$ .

Figure 2 presents the equilibrium temperature as a function of the saturation parameter  $S_2$ , for several values of the first laser saturation parameter:  $S_1 = 0.3$  (solid), 0.2 (dash), 0.1 (dot) and 0.01 (dash-dotted curve), and for optimum laser detunings of  $\delta = -\gamma/2$  and  $\delta_2 = -\gamma_e/2$ . The dash dot-dotted curve is the minimum value achieved for the two-photon cooling process, 123  $\mu$ K for Calcium atoms. The limit of low intensities of the infrared laser ( $S_2 \ll 1$ ) results in a temperature expected on the basis of the two-level atom theory:  $\hbar \gamma / 2 k_B = 831 \mu$ K for Calcium. However, for low intensities of the blue laser ( $S_1 \ll 1$ ) and with the increase of the  $S_2$  parameter, the Doppler temperature is quite reduced.

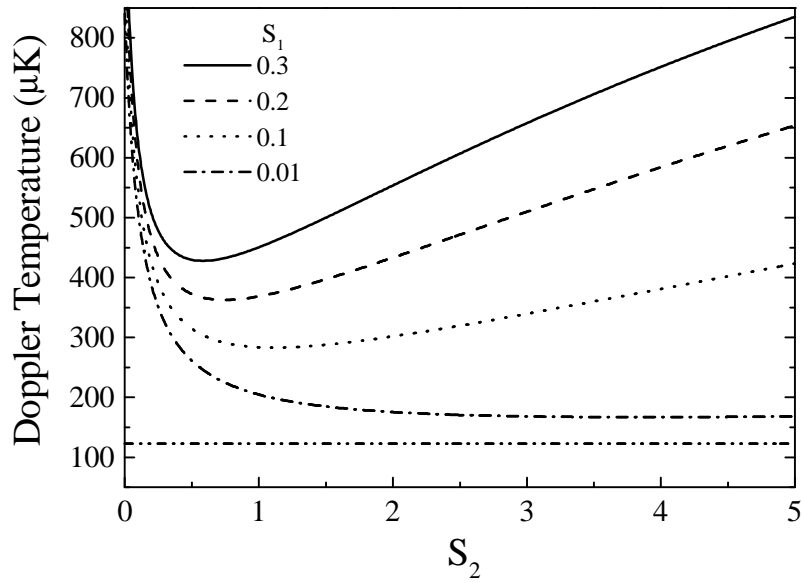


Figure 2. Doppler temperatures for Calcium atoms,  $T_D$  ( $\mu\text{K}$ ), as a function of saturation parameter  $S_2$ , for some fixed values of  $S_1$  (0.3, 0.2, 0.1 and 0.01) and detunings  $\delta\omega_2 = -\gamma_2/2$ ,  $\delta\omega_1 = -\gamma_1/2$ . The minimum value of Doppler temperature achieved is 123  $\mu\text{K}$  (dash dot-dotted curve).

## Conclusions

We have discussed the possibility of two-photon laser cooling of Calcium, using a  $^1S_0 - ^1S_0$  transition. We considered excitation with laser beams at 423 and 1034 nm, to take advantage of the  $^1P_1$  state as a virtual level to enhance the two-photon scattering rate. A Doppler cooling limit near 123  $\mu\text{K}$  has been found at optimum conditions. This technique can be efficiently used as a second stage of cooling, allowing 100 % transfer efficiency from the first stage. It is also simpler than “quench” cooling and can be useful for subsequent loading of an optical dipole trap. This might be an important step towards a possible all-optical achievement of a Bose-Einstein condensate for Calcium.

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