Two-photon Doppler cooling of alkaline-earth-metal and ytterbium atoms

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The possibility of laser cooling of alkaline-earth-metal atoms and ytterbium atoms using a two-photon transition is analyzed. We consider a $^1S_0$-$^1S_0$ transition with excitation in near resonance with the $^1P_1$ level. This greatly increases the two-photon transition rate, allowing an effective transfer of momentum. The experimental implementation of this technique is discussed and we show that for calcium, for example, two-photon cooling can be used to achieve a Doppler limit of 123 μK. The efficiency of this cooling scheme and the main loss mechanisms are analyzed.

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I. INTRODUCTION

Laser cooling of neutral atoms and ions has played a fundamental role in many fields such as metrology, atom and quantum optics, and Bose-Einstein condensation. For a strong and closed single-photon atomic transition (linewidth $\Delta\nu$ of a few megahertz), a high scattering rate can quickly reduce the atomic temperature (or velocity) down to a ''Doppler'' limit given by the spontaneous emission rate, typically in the millikelvin range. The multilevel atomic structure is essential in other cooling techniques, such as polarization gradient [1,2], which allow the achievement of microkelvin temperatures. Although these techniques have been widely applied in metal-alkaline elements, they are not applicable to the alkaline-earth-metal and ytterbium atoms because of their simpler level structure with nondegenerate ground states and no hyperfine structure. Thus, until recently, the smaller temperatures achieved with these elements were in the millikelvin range, given by the Doppler limit of the resonant $^1S_0$-$^1P_1$ cooling transition. Microkelvin temperatures, close to the recoil limit, have been achieved for strontium by Doppler cooling using the narrower $^1S_0$-$^3P_1$ intercombination transition ($\gamma/2\pi=7.6$ kHz) [3] and also for ytterbium ($\gamma/2\pi=182$ kHz) [4], an alkaline-earth-metal-like atom. Cooling using the intercombination transition has also been demonstrated for calcium, where the relatively long lifetime of the $^3P_1$ level ($\tau=0.38$ ms, $\gamma/2\pi=408$ Hz) had to be reduced by coupling it to another level [5,6] in order to make the cooling process effective. This ''quenching'' cooling scheme has also been used several years ago to cool mercury ions to the zero-point energy of a trap [7]. With the exception of ytterbium, cooling on the intercombination transition was used as a second stage after initial precooling to millikelvin temperatures with the strong $^1S_0$-$^1P_1$ dipole transition. Although this narrow line cooling can reach temperatures near the recoil limit, only a small fraction of the atoms cooled by the strong $^1S_0$-$^1P_1$ line can be transferred to the microkelvin regime. For calcium, for example, only about 15% of the atoms captured in a MOT were transferred to lower temperatures by quenching cooling [5,6]. Another possibility for obtaining cold calcium atoms was recently demonstrated by optically pumping a fraction of atoms captured in a MOT into the metastable $^3P_2$ state, and application of a second stage of laser cooling on the $^3P_2$-$^3D_3$ transition to produce metastable calcium at microkelvin temperatures [8].

In this paper, we present a scheme for laser cooling of alkaline-earth-metal and ytterbium atoms using a two-photon $^1S_0$-$^1S_0$ transition. Although laser cooling involving two photons takes place, for example, in Raman cooling [9] and quenching cooling, so far no two-photon Doppler cooling has been demonstrated. We show that two-photon cooling can be quite efficient and might be used as a second cooling stage after precooling with the conventional method. The experimental setup should be much simpler than in quenching cooling, requiring only one additional laser with linewidth of the order of a megahertz. In addition, a significant temperature reduction, with respect to the one-photon Doppler limit, can be achieved. In Sec. II, we describe the feasibility of two-photon cooling and present the calculations of the transition rates for the one-photon $^1S_0$-$^1P_1$ and two-photon $^1S_0$-$^1S_0$ cooling transitions. In Sec. III, we present an analysis of the temperature limit that can be achieved by this cooling scheme. Sec. IV discuss the main loss mechanisms, considering ^{40}Ca as an example. Finally, the conclusions are summarized in Sec. V.

II. TWO-PHOTON TRANSITION RATES

Let us consider the interaction of an atom with two copropagating laser beams, with wave numbers $k_1=2\pi/\lambda_1$ and $k_2=2\pi/\lambda_2$ and frequencies ω_1 and ω_2. Figure 1(a) shows a diagram of the relevant level structure of alkaline-earth-metal and ytterbium atoms. The ground state is represented by $|g\rangle=(ns^2)^1S_0$ and the excited states are $|r\rangle=(nsnp)^1P_1$ and $|e\rangle=[ns(n+1)s]^1S_0$. From level $|e\rangle$, the atoms quickly decay to the ground state by spontaneous emission via the intermediate state $|r\rangle$ with relaxation rates γ_2 and γ_1. Table I presents some relevant parameters for the most abundant isotopes of Mg, Ca, Sr, and Yb.

We see that for all these atoms, the first laser is in the blue or ultraviolet region, while the other laser is at the near infrared region. The resonant saturation intensity for the blue

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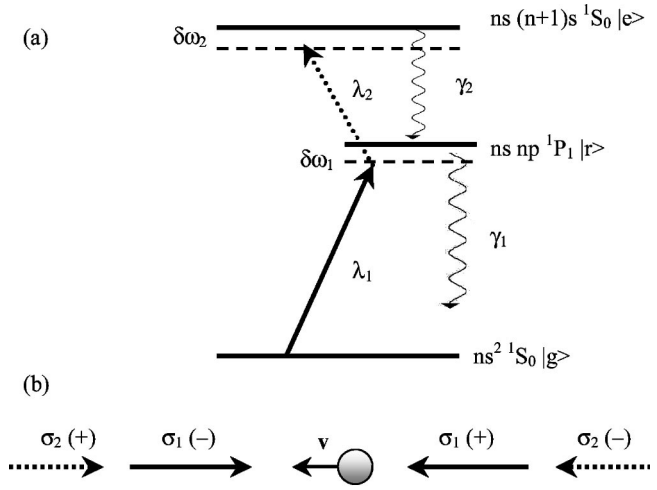

FIG. 1. (a) Energy-level diagram for two-photon cooling in alkaline-earth-metal and Yb atoms. (b) Configuration for a one-dimensional two-photon optical molasses.

transition $^1S_0$-$^1P_1$ is I_{1s}, while I_{2s} is the one for the infrared transition $^1P_1$-$^1S_0$. The recoil velocity, after two-photon absorption, is given by $v_r=\hbar(k_1+k_2)/M$, where M is the atomic mass. The recoil temperature is then $T_r=Mv_r^2/k_B$, where k_B is the Boltzmann constant. Both are obviously higher than in the case of the single-photon $^1S_0$-$^1P_1$ cooling transition.

Two-photon transition rates are dependent on the light beam intensities and detunings from real levels and usually are much smaller than single-photon dipole allowed transition ones. Nevertheless, they can be quite strong for spectroscopic purposes, allowing the implementation of powerful Doppler-free techniques [10], widely used over the past 30 yrs. However, 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. The general expression for the two-photon transition rate between levels $|g\rangle$ and $|e\rangle$ is given by Refs. [11,12]:

$$\Gamma_{ge}=\left|\sum_r \frac{\langle e|H_2|r\rangle\langle r|H_1|g\rangle}{\hbar\Delta_r-i(\hbar\gamma_1/2)}\right|^2 \frac{\gamma_2}{[\Delta_2^2+(\gamma_2/2)^2]}, \quad (1)$$

where the matrix elements of the interaction Hamiltonian between the atomic levels are $\langle e|H_2|r\rangle$ and $\langle r|H_1|g\rangle$. The detunings, including the Doppler shifts, are $\Delta_r=\omega_{gr}-\omega_1+k_1v$ and $\Delta_2=\omega_{eg}-\omega_1-\omega_2+(k_1+k_2)v$, with the Bohr frequencies between the atomic levels $|l\rangle$ and $|m\rangle$ represented by ω_{lm}. The laser detuning relative to level $|r\rangle$ is defined as $\delta\omega_1=\omega_1-\omega_{gr}$. The first term of the expression involves a sum on the intermediary levels $|r\rangle$, which are coupled to $|g\rangle$ by allowed single-photon transitions. The second factor gives the spectral line profile of the two-photon transition of a single atom, which has a two-photon detuning $\delta\omega_2=\omega_1+\omega_2-\omega_{eg}$. In order to preserve angular momentum in a $\Delta J=0$ transition $^1S_0$-$^1S_0$, we are also assuming $\sigma_1(-)$ and $\sigma_2(+)$ polarizations for the two laser beams at λ_1 and λ_2, respectively [see Fig. 1(b)]. Although $\sigma_1(+)$ and $\sigma_2(-)$ photons can be absorbed from opposite directions, this possibility will not be considered here. Polarization selection rules for two-photon transitions are discussed in Refs. [10,13]. For alkaline-earth metals and ytterbium, Eq. (1) can be written as

TABLE I. Parameters of interest for one- and two-photon cooling in some alkaline-earth-metal and Yb atoms.

Parameter	^{24}Mg	^{40}Ca	^{88}Sr	^{174}Yb
λ_1 (nm)	285.2	422.8	460.7	398.8
$\gamma_1/2\pi$ (MHz)	80.95	34.63	4.48	28.01
I_{1s} (mW/cm^2)	456.0	59.9	6.0	57.7
λ_2 (nm)	1182.8	1034.4	1130	1077.3
$\gamma_2/2\pi$ (MHz)	4.14	4.77	2.96	4.81
I_{2s} (mW/cm^2)	0.3	0.6	0.3	0.5
v_r (cm/s)	7.14	3.31	1.39	0.79
T_r (μK)	14.92	5.30	2.04	1.31

$$\Gamma_{ge}=\frac{4S_1S_2\gamma_2}{[1+S_1+(2\Delta_r/\gamma_1)^2][1+S_1S_2+(2\Delta_2/\gamma_2)^2]}, \quad (2)$$

where the sum over intermediary levels $|r\rangle$ is reduced to only one term, involving the real level $^1P_1$. We used in this equation the saturation parameters $S_1=2|\Omega_1|^2/\gamma_1^2=I_1(\omega_1)/I_{1s}$ and $S_2=2|\Omega_2|^2/\gamma_2^2=I_2(\omega_2)/I_{2s}$, where $\Omega_j=-\mu_jE_j(\omega_j)/\hbar$ $(j=1,2)$ are the Rabi frequencies associated with the applied fields $E_j(\omega_j)$ and the dipole operators μ_j. We have introduced in this last equation the possibility of saturation of the one- and two-photon transitions.

The transition rate Γ_{gr} for the strong one-photon $^1S_0$-$^1P_1$ dipole transition is given by [14]

$$\Gamma_{gr}=\frac{S_1(\gamma_1/2)}{[1+S_1+(2\Delta_r/\gamma_1)^2]}, \quad (3)$$

Figures 2(a) and 2(b) show the transition rates $\Gamma_{ge}/2\pi$ and $\Gamma_{gr}/2\pi$, as a function of the one-photon detuning $|\Delta_r|$, normalized by γ_1, for a two-photon detuning $|\Delta_2|=\gamma_2/2$, and saturation parameters $S_1=0.1$ and $S_2=3$. The calculations were made for magnesium (dashed), calcium (solid), strontium (dash-dot), and ytterbium (doted curve). The chosen saturation parameters made the one- and two-photon transition rates of the same magnitude. For ^{40}Ca, for example, they correspond to (see Table I) $I_1(\omega_1)\approx 6$ mW/cm^2 and $I_2(\omega_2)\approx 2$ mW/cm^2. In fact, over 100 mW of laser radiation at 422.8 nm can be generated by frequency doubling near infrared diode lasers [15] or a Ti:sapphire laser [16]. Radia-

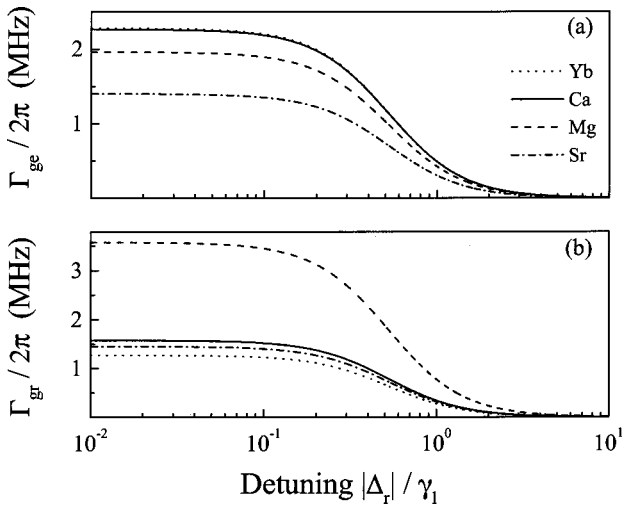

FIG. 2. Transition rates for two-photon (a) and one-photon (b) cooling processes in some alkaline-earth-metal and ytterbium atoms, as a function of the normalized detuning $|\Delta_r|/\gamma_1$. The two-photon detuning is $\Delta_2=-\gamma_2/2$, and the saturation parameters are $S_1=0.1$ and $S_2=3$.

tion at 1034 nm can be generated with a Ti:sapphire laser or with a Yb:YAG (yttrium aluminum garnet) laser. With this last option, powers over 500 mW are readily achieved. Even at low power levels, the two-photon transition rate can still be significant. We can see in Fig. 2(a) that for $|\Delta_r|<\gamma_1$, the two-photon transition rate is about 1 MHz, therefore a little smaller than the rates of the conventional cooling transitions of metal-alkaline atoms. If we assume, for example, red detuning of the incident lasers, $\Delta_r=-\gamma_1/2$ and $\Delta_2=-\gamma_2/2$, the two-photon transition rate will be $\Gamma_{ge}/2\pi=1.2$ MHz for calcium. For these parameters, the single-photon transition rate is $\Gamma_{gr}/2\pi=826$ kHz.

The two-photon transition rate can be much higher than the one-photon rate if the saturation parameter S_2 is much higher than S_1. However, as we will see in the following section, increasing the second laser intensity results in an increase of the equilibrium temperature due to the heating caused by spontaneous emission.

III. DOPPLER-COOLING LIMIT

The temperature limit in laser cooling is determined by a balance between damping forces and heating due to spontaneous emission. For a two-level system, considering a one-photon process this Doppler limit is $k_BT_D=\hbar\gamma_1/2$, while the recoil limit is given by $k_BT_r=\hbar^2k_1^2/M$. In general, $T_D\gg T_r$, for one-photon cooling transitions of alkali metals.

A. Two-photon process

The Doppler temperature limit can be estimated by the balance between heating and cooling, giving $k_BT_D=D/\alpha$ [14], where D is the diffusion constant and α is the damping coefficient. Let us consider one atom interacting with two copropagating $\sigma_2(+)$ and $\sigma_1(-)$ laser beams [see Fig. 1(b)]. The direct excitation from $|g\rangle$ to $|e\rangle$ by the simultaneous absorption of two photons, $\omega_1+\omega_2$, occurs with a rate Γ_{ge}, given by Eq. (2). From the upper level $|e\rangle$, the atom spontaneously decays to the intermediate $|r\rangle$ level with a rate γ_2 and, from this level, with a rate γ_1 to the ground state. On an average, the time taken by one atom to absorb simultaneously two photons and go back to the ground state by spontaneous cascade decay is given by

$$\Delta t=\Gamma_{ge}^{-1}+\gamma_2^{-1}+\gamma_1^{-1}. \tag{4}$$

If the atom is moving in the opposite direction of the laser beams [left side of Fig. 1(b)], then a mean radiation force F_+ can be written as the ratio between the momentum change $\Delta p=\hbar(k_1+k_2)$ and the time interval Δt

$$F_+=\frac{4\hbar(k_1+k_2)S_1S_2\gamma_2}{(1+S_1+4x_1^2)(1+S_1S_2+4x_2^2)+4S_1S_2(1+\gamma_2/\gamma_1)}$$

$$\times\left\{1-\frac{8(1+S_1S_2+4x_2^2)(x_1k_1v/\gamma_1)+8(1+S_1+4x_1^2)[x_2(k_1+k_2)v/\gamma_2]}{(1+S_1+4x_1^2)(1+S_1S_2+4x_2^2)+4S_1S_2(1+\gamma_2/\gamma_1)}\right\}, \tag{5}$$

where $x_i=\delta\omega_i/\gamma_i$ $(i=1,2)$ are the detunings normalized by the natural linewidths of the transitions, and we have disregarded terms of the order of $(kv)^2/\gamma^2$. Adding one pair of $\sigma_1(+)$ and $\sigma_2(-)$ laser beams counterpropagating to the first ones [right side of Fig. 1(b)], an additional force F_- will act on the atom, and we have a configuration of a one-dimensional (1D) two-photon optical molasses. In this configuration, the radiation pressure reduces to

$$F=F_++F_-=-\alpha_2v, \tag{6}$$

where the damping coefficient α_2 is given by

$$\alpha_2 = \frac{-64\hbar(k_1+k_2)S_1S_2\gamma_2}{\{(1+2S_1+4x_1^2)(1+4S_1S_2+4x_2^2)+4S_1S_2(1+\gamma_2/\gamma_1)\}^2} \times\{(x_1k_1/\gamma_1)[1+4S_1S_2+4x_2^2]+[x_2(k_1+k_2)/\gamma_2][1+2S_1+4x_1^2]\}, \quad (7)$$

which leads to a kinetic-energy losing rate $(dE/dt)_{cool} = Fv = -\alpha_2 v^2$. The damping coefficient is positive for red detunings and implies in a damping force for all velocities, if $\delta\omega_1, \delta\omega_2 < 0$, similar to the case of "one-photon optical molasses." For low intensities and far from saturation of the transitions ($S_1 \ll 1$, $S_1S_2 \ll 1$), we have

$$\alpha_2 = \frac{-64\hbar(k_1+k_2)S_1S_2\gamma_2}{(1+4x_1^2)(1+4x_2^2)}\left\{\frac{x_1k_1/\gamma_1}{1+4x_1^2}+\frac{x_2(k_1+k_2)/\gamma_2}{1+4x_2^2}\right\}. \quad (8)$$

The momentum diffusion constant in a two-photon molasses, D_2, can be estimated through a straightforward generalization of the analysis of the random-walk process in one-photon molasses made by Lett *et al.* [17]. Although the damping force reduces the average velocity of the atoms to zero, the fluctuations of this force produce heating due to the spontaneous emission, which leads to a spread in the mean-square momentum,

$$d(p^2)/dt = 2\hbar^2(k_1^2+k_2^2)\Gamma_{ge} = 2D_2. \quad (9)$$

In analogy to Ref. [17], the resultant momentum diffusion constant in a two-photon molasses is

$$D_2 = \frac{4\hbar^2(k_1^2+k_2^2)(2S_1)(2S_2)\gamma_2}{(1+2S_1+4x_1^2)(1+4S_1S_2+4x_2^2)}, \quad (10)$$

where the saturation of the one- and two-photon transitions by the two pairs of counterpropagating beams was taken into account, and we have disregarded the Doppler shifts $|v| \ll \gamma_1/k_1,\ \gamma_2/(k_1+k_2)$.

The rate of increase of kinetic energy in this two-photon molasses is given by $(dE/dt)_{heat} = d(p^2/2M)/dt = D_2/M$. When the equilibrium is reached, the heating and cooling rates are equal, that is, $(dE/dt)_{cool} + (dE/dt)_{heat} = 0$. Therefore, using Eqs. (8) and (10), the Doppler temperature is estimated as

$$k_BT_D = \frac{\hbar\gamma_1}{2}\frac{(k_1^2+k_2^2)}{2(k_1+k_2)}\left\{\frac{k_1|x_1|}{1+4x_1^2}+\frac{(k_1+k_2)|x_2|\gamma_1/\gamma_2}{1+4x_2^2}\right\}^{-1}. \quad (11)$$

We recognize the first term on Eq. (11) as the one-photon Doppler limit $T_1 = \hbar\gamma_1/2k_B$, which is 1.94 mK, 831 μK, 108 μK, and 672 μK, respectively, for magnesium, calcium, strontium, and ytterbium atoms. The detunings that minimize the two-photon Doppler temperature are $x_i = -1/2$ ($\delta\omega_1 = -\gamma_1/2$, $\delta\omega_2 = -\gamma_2/2$), giving the minimum value

$$k_BT_{min} = \hbar\gamma_1\frac{(k_1^2+k_2^2)}{(k_1+k_2)[k_1+(k_1+k_2)(\gamma_1/\gamma_2)]}. \quad (12)$$

Using the parameters of Table I, we obtain the following temperatures: $T_{min} = 131$ μK, 123 μK, 57 μK, and 124 μK, respectively, for magnesium, calcium, strontium, and ytterbium. These minimum values of temperature are very close for these elements because the linewidths of the second transition, γ_2, are very similar (see Table I). This does not happen for the one-photon Doppler limit. The predicted ratio between the one- and two-photon Doppler temperatures will be then $\hbar\gamma_1/(2k_BT_{min}) = 14.8$ (Mg), 6.8 (Ca), 1.9 (Sr), and 5.4 (Yb) (Table II). Equations (11) and (12) are valid only for low laser intensities ($S_1 \ll 1$, $S_1S_2 \ll 1$). If we take into account higher intensities, as a result there is an increase in temperatures, as in the case of one-photon optical molasses [17].

B. Combined one- and two-photon cooling processes

Since the one-photon $^1S_0$-$^1P_1$ and two-photon $^1S_0$-$^1S_0$ transitions occur simultaneously, we should consider both cooling processes jointly. Depending on the detunings and intensities of the incident laser beams, one process can dominate the other. The case of one-photon optical molasses was discussed in several references, and results in the following coefficients [14,17]:

$$\alpha_1 = \frac{-8\hbar k_1^2S_1x_1}{(1+2S_1+4x_1^2)^2}, \quad (13)$$

$$D_1 = \frac{\hbar^2k_1^2S_1\gamma_1}{(1+2S_1+4x_1^2)}, \quad (14)$$

taking into account the saturation of the first transition involving the blue laser at frequency ω_1. The effective damping and diffusion coefficients that jointly take into account the one- and two-photon cooling processes are

$$\alpha_{eff} = \alpha_1 + \alpha_2, \quad (15)$$

TABLE II. Predicted parameters for the two-photon Doppler cooling.

Element	T_{min} (μK)	T_1/T_{min}
^{24}Mg	131	14.8
^{40}Ca	123	6.8
^{88}Sr	57	1.9
^{174}Yb	124	5.4

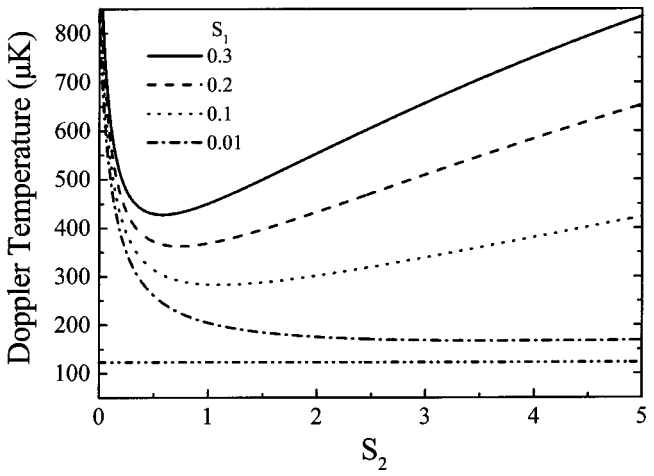

FIG. 3. Doppler temperatures for calcium atoms, $T_D(\mu K)$, as a function of saturation parameter S_2 for some fixed values of S_1 (0.3, 0.2, 0.1, and 0.01) and detunings $\delta\omega_2 = -\gamma_2/2$, $\delta\omega_1 = -\gamma_1/2$. The minimum value of Doppler temperature achieved is 123 μK (dash dot-dotted curve).

$$D_{eff} = D_1 + D_2, \tag{16}$$

and the Doppler equilibrium temperature is obtained by $k_B T_D = D_{eff}/\alpha_{eff}$. Figure 3 presents this temperature for calcium atoms, as a function of the infrared saturation parameter S_2, for several values of the first laser saturation parameter ($S_1 = 0.3$, 0.2, 0.1, and 0.01). The laser detunings were assumed to be at the optimum values $\delta\omega_2 = -\gamma_2/2$ and $\delta\omega_1 = -\gamma_1/2$. Some features in Fig. 3 call our attention. 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: $\hbar\gamma_1/2k_B = 831$ μK for calcium. However, for low intensities of the blue laser ($S_1 \ll 1$) and with the increase in the S_2 parameter, the Doppler temperature is quite reduced, tending to the minimum value of 123 μK (dash dot-dotted curve; see also Table II). This minimum value of temperature is close to the Doppler limit that would be associated only with the second transition, that is, $\hbar\gamma_2/2k_B = 115$ μK for calcium atoms. As already mentioned, this new Doppler limit is 6.8 times smaller than the one-photon Doppler limit of the blue transition. This happens due to the increase in the damping coefficient for the combined cooling processes. For the other elements of Table II, this reduction factor varies considerably, depending on the values of the linewidths γ_1 and γ_2.

IV. EFFICIENCY OF THE COOLING PROCESS AND MAIN LOSS MECHANISMS

In this section, we discuss the main loss mechanisms that can limit the efficiency of the Doppler cooling scheme discussed here. It is important to know the population fraction in the excited states $|r\rangle$ and $|e\rangle$, which can be lost by spontaneous emission into metastable levels that do not participate in the cooling process, or through photoionization induced by the incident laser beams.

Figure 4 shows some possible loss channels that can limit

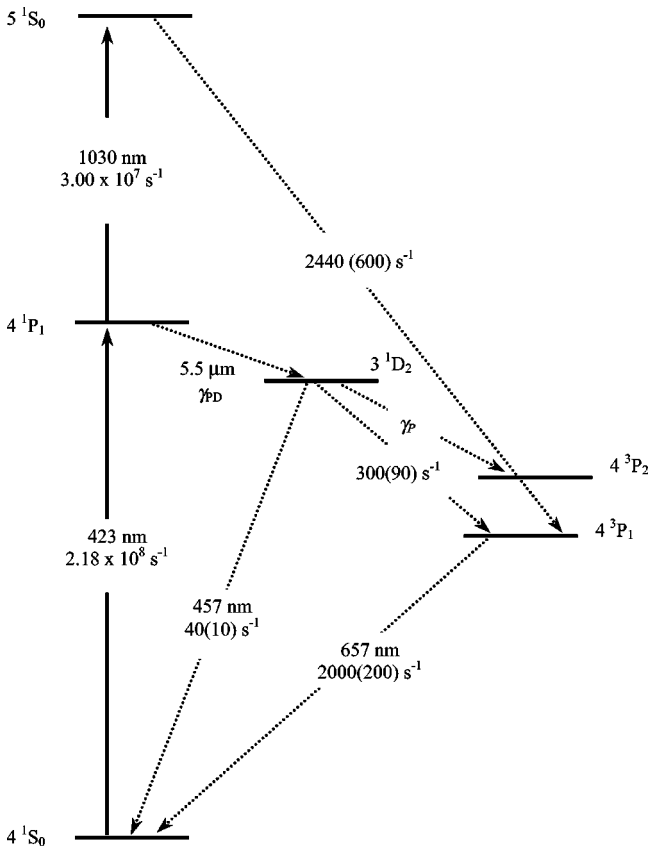

FIG. 4. Energy levels of the calcium atom, showing the main loss channels that limit the efficiency of the one- and two-photon cooling processes. The decay rate from 3^1D_2 to 4^3P_2 is $\gamma_P = 89 \times 10^3$ s^{-1} [23].

the cooling process, again using calcium as an example. One of these channels is the spontaneous decay from the $(4s4p)^1P_1$ to the $(3d4p)^1D_2$ level, at a rate $\gamma_{PD} = 2180$ s^{-1} [18]. For magnesium, the $^1D_2$ state is above the $^1P_1$ level, so this loss channel is not present. Another is the direct spontaneous decay from the $(4s5s)^1S_0$ level to the $^3P_1$ level, at 553 nm, which occurs at a rate of 2440(600) s^{-1} [5]. A third loss channel is due to photoionization of the excited state $(4s5s)^1S_0$, which can be connected to the $(3d6p)^1P_1$ level above the ionization limit by a photon at 423 nm. The photoionization rate for this process is given by $\Gamma_{ion} = \sigma_{ion}(\omega_1) \times I(\omega_1)/(\hbar\omega_1)$ [19], where $\sigma_{ion}(\omega_1)$ is the photoionization cross section at the blue laser frequency. Although this cross section has not been reported in the literature, we do not expect an expressive photoionization rate for calcium because this process can be considered far off resonance. The branching ratio of the ionization process, with respect to the spontaneous decay, to the intermediate level $|r\rangle$, and from this to the ground state should be very low because $\Gamma_{ion} \ll \gamma_1\ \gamma_2$ for small values of intensity of the blue laser. This is not the case of magnesium, where photoionization is an important loss channel.

The atomic populations of the excited states [Fig. 1(a)] can be calculated using the density-matrix formalism applied to a three-level atom interacting with two laser beams [20]. The optical Bloch equations (OBE's) for the density-matrix

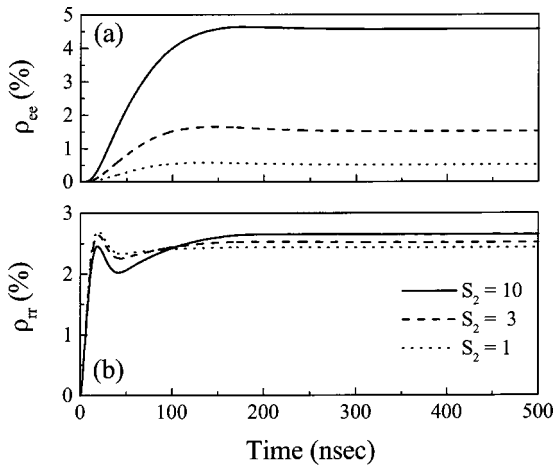

FIG. 5. Population distributions of the excited states (a) $-\rho_{ee}$, (b) $-\rho_{rr}$, for calcium atoms in the two-photon cooling process for $S_1=0.1$ and $S_2=1$, 3, and 10.

elements with the terms ρ_{ij} $(i,j=g,r,e)$ varying slowly in the rotating-wave approximation are given by

$$\frac{\partial\rho_{ee}}{\partial t}=-\gamma_2\rho_{ee}-i\frac{\Omega_2}{2}(\rho_{er}-\rho_{re}),$$

$$\frac{\partial\rho_{rr}}{\partial t}=-\gamma_1\rho_{rr}+\gamma_2\rho_{ee}-i\frac{\Omega_1}{2}(\rho_{rg}-\rho_{gr})+i\frac{\Omega_2}{2}(\rho_{er}-\rho_{re}),$$

$$\frac{\partial\rho_{gg}}{\partial t}=\gamma_1\rho_{rr}+i\frac{\Omega_1}{2}(\rho_{rg}-\rho_{gr}),$$

$$\frac{\partial\rho_{er}}{\partial t}=-[(\gamma_1+\gamma_2)/2-i(\delta\omega_2-\delta\omega_1)]\rho_{er}-i\frac{\Omega_1}{2}\rho_{eg}-i\frac{\Omega_2}{2}(\rho_{ee}-\rho_{rr}),$$

$$\frac{\partial\rho_{eg}}{\partial t}=-[\gamma_2/2-i\delta\omega_2]\rho_{eg}-i\frac{\Omega_1}{2}\rho_{er}+i\frac{\Omega_2}{2}\rho_{rg},$$

$$\frac{\partial\rho_{rg}}{\partial t}=-[\gamma_1/2-i\delta\omega_1]\rho_{rg}-i\frac{\Omega_1}{2}(\rho_{rr}-\rho_{gg})+i\frac{\Omega_2}{2}\rho_{eg}, \tag{17}$$

where atomic coherences obey the relation $\rho_{ij}^*=\rho_{ji}$. We have assumed in these equations that the linewidths of the incident laser beams are smaller than the half natural widths of the atomic transitions, that is, $\Delta\nu_1<\gamma_1/2$, $\Delta\nu_2<\gamma_2/2$. A linewidth of about a megahertz would satisfy this condition, and is typical of, for example, a commercially available Ti:sapphire laser.

The coupled OBE's in Eqs. (17) are numerically solved by integrating over time t via a Runge-Kutta fourth-order method with the initial conditions: $\rho_{ij}(t=0)=\delta_{ig}$. The populations ρ_{ee} and ρ_{rr} for calcium atoms can be seen in Figs. 5(a) and 5(b), respectively, where the blue laser saturation parameter is $S_1=0.1$, while the infrared saturation parameters are $S_2=1$ (doted), 3 (dashed), and 10 (solid line). We can observe in Fig. 5 that the increase in the infrared laser intensity results in an increase in the excited-state populations due to optical pumping. However, in the steady-state regime $(t\to\infty)$, the percentage of excited atoms is low ($<10\%$), corresponding to a high fraction of atoms in the ground state.

An important parameter that characterizes the dynamics of the cooling process is the cooling time τ_{cool}. This time for the two-photon cooling can be calculated by the relationship between the kinetic energy and its loss rate:

$$\tau_{cool}=-\frac{E}{(dE/dt)_{cool}}=\frac{M}{2\alpha_{eff}}. \tag{18}$$

From Eq. (15), the last expression implies a decrease in the cooling time, in comparison to one-photon cooling: $\tau_{cool}=(M/2\alpha_1)1/(1+\alpha_2/\alpha_1)<M/2\alpha_1$. For calcium, we have $\tau_{cool}\approx 2$ μs, for $\delta\omega_i=-\gamma_i/2$, $S_1=0.1$, and $S_2=3$. Since this time is much shorter than the storage time, dictated by optical pumping into the metastable $^1D_2$ state [8], two-photon cooling should be a fast and efficient process.

Another important parameter is the capture velocity, which is of the order of $\gamma_1/k_1=14.7$ m/s for the one-photon cooling process in calcium atoms [14]. In general, the average force given in Eq. (6) reduces to $F=-\alpha_2 v/[1+(v/v_c)^2]$, and the capture velocity for the two-photon cooling is a little smaller, of the order of $v_c=(\gamma_1+\gamma_2)/(2k_1+2k_2)$ for detunings $|\delta\omega_i|=\gamma_i/2$. For calcium, $v_c=6$ m/s is much larger than the mean velocity of the atoms, already cooled in the first cooling process: $v_{rms}=\sqrt{\hbar\gamma_1/(2M)}=41.6$ cm/s. This fact $(v_{rms}\ll v_c)$ implies that the initial number of atoms that were captured in the first cooling stage should be transferred completely to the second one, resulting in a significant number of colder atoms. In other words, the transfer efficiency from the first (one-photon) to the second (two-photon) stage should be basically 100%.

V. CONCLUSIONS AND PROSPECTS

We proposed a possibility to reach microkelvin temperatures in laser cooling of alkaline-earth-metal and ytterbium atoms, using a two-photon $^1S_0$-$^1S_0$ transition. After discussing the implementation of this technique and the calculation of the transition rates, we analyzed the experimental cases of atomic interaction with a pair of copropagating laser beams and two-photon optical molasses. We considered excitation with laser beams at frequencies ω_1 and ω_2, in near resonance with the $^1P_1$ state, to enhance the two-photon scattering rate. Doppler limits of 131 μK (Mg), 123 μK (Ca), 57 μK (Sr), and 124 μK (Yb) have been found at optimum detunings and laser powers. Since the optimum detuning for the blue $^1S_0$-$^1P_1$ transition is $\gamma_1/2$, two-photon cooling can be simply applied as a second stage, just by adding the second near-infrared laser. This will bring all atoms to substantially lower temperatures (for example, from 831 to 123 μK for calcium). With such 100% efficiency, any loss of atoms in this second cooling stage should not be expected. It is

important to point out that two-photon cooling is considerably simpler than ''quenching'' cooling, requiring only one extra laser, with a linewidth of about a megahertz. The two-photon cooling scheme should be applicable to a 3D optical molasses or a magneto-optical trap, in this case taking advantage of the magnetic intermediate state $^1P_1$. This will be investigated in a future work. We also expect to make the experimental implementation of this technique with the addition of the 1034-nm laser in our 3D calcium optical molasses and MOT [21]. Finally, the lower temperatures achieved by two-photon cooling can be also very useful for efficient loading of an optical dipole trap, which might be an important step towards the achievement of all-optical Bose-Einstein condensates [22] for alkaline-earth-metal and ytterbium atoms.

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