Two-photon and electromagnetically-induced-transparency-assisted Doppler cooling in a three-level cascade system

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Laser cooling is theoretically investigated in a cascade three-level scheme, where the excited state of a laser-driven transition is coupled by a second laser to a top, more stable level, as for alkaline-earth-metal atoms. The second laser action modifies the atomic scattering cross section and produces temperatures lower than those reached by Doppler cooling on the lower transition. When multiphoton processes due to the second laser are relevant, an electromagnetic-induced transparency modifies the absorption of the first laser, and the final temperature is controlled by the second laser parameters. When the intermediate state is only virtually excited, the dynamics is dominated by the two-photon process and the final temperature is determined by the spontaneous decay rate of the top state.

Ultracold temperatures in atomic gases are reached by means of laser cooling [1], evaporative cooling [2], sympathetic cooling [3], or stochastic cooling [4]. In order to decrease the kinetic energy associated with the atomic center-of-mass motion, laser cooling is based on the exchange of momentum between laser light and a closed atomic (or molecular) system consisting of a few active levels. In Doppler cooling, the action of light scattering on the atomic motion can be described by means of a force [5]. The basic ingredients are (i) an atomic cross section with a resonance enhancing the photon absorption and the force acting on the atoms; (ii) a dependence of the absorption process on the atomic momentum leading to a dependence of the force on the momentum. If the resulting force damps the atomic momentum, the cooling process compresses the atomic momenta into a narrow distribution, from which one extracts the laser cooling temperature.

Laser cooling techniques have been demonstrated to be very efficient for alkali-metal atoms, where temperatures down to several hundreds of nanoKelvins have been reached [1]. Much lower efficiencies have been achieved for other atomic systems, and in particular for alkaline-earth-metal atoms. One attractive feature of group-II elements is the simple internal structure with no hyperfine levels for the most abundant bosonic isotopes, which make them particularly attractive for improved frequency standards and optical clocks. However, because the ground state is nondegenerate, sub-Doppler cooling is not possible and the temperature for Doppler cooling on the resonance line is typically limited to a few milliKelvins. For these elements other cooling strategies have been employed, such as cooling on the intercombination singlet-triplet line, pioneered in [6], or quench cooling, modifying the lifetime of a metastable state by coupling it to a fast-decaying transition, implemented for neutral calcium atoms in [7,8], and first applied to enhance sideband cooling of ions [9].

An alternative cooling strategy employs a two-color laser excitation of a three-level cascade configuration and is based on mixing by laser radiation the fast decaying singlet first excited state with another state having a longer lifetime. This mixing cooling uses the two-photon atomic coherence created between the ground and top states of the cascade. Because the Doppler-cooling temperature depends inversely on the excited state lifetime, a lower temperature may be produced by the excited state mixing. The scheme has flexible handles in the frequency and intensity of the mixing laser. This scheme was initially explored on metastable helium with limited efficiency [10,11]. Theoretical analyses on alkaline-earth-metal and ytterbium atoms [12] were followed by experimental evidence of improved laser cooling for magnesium atoms [13,14]. These experiments showed that the temperatures reached by the excited state mixing are lower than the Doppler temperature determined by the lifetime of the singlet first excited state, and identified signatures hinting to dynamics significantly determined by the two-photon atomic coherence.

In this Rapid Communication we present a theoretical analysis of laser cooling on a cascade-level scheme with two-color laser excitation. We determine the laser requirements for reaching low temperatures, and thereby single out the role of atomic coherence on the cooling dynamics, identifying the regimes when it is a key ingredient for reaching efficient cooling. The atomic level configuration we consider is found in group-II elements (see the first and second columns in Table I), and it is composed by a stable state (ground state), coupled by a laser to an excited level (intermediate state), which itself is coupled by a second laser to a higher energy state (top state). The intermediate state decays more rapidly than the top state, and the second laser modifies the absorption on the lower transition by the coupling to the upper transition. This configuration supports the creation of a stationary atomic coherence between the ground and top state when the two-photon transition is driven close to resonance [15].

Depending on the laser detunings we identify two different regimes, which characterize the cooling dynamics. In the first one the formation of atomic coherence between the ground and top states critically affects the properties of the

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force by modifying the lower transition absorption, and temperatures lower than the Doppler limit on the lower transition are achieved. We denote this regime as cooling assisted by electromagnetic-induced transparency (EIT), similar to that realized in a Λ configuration [17]. The second regime is characterized by two-photon processes coupling the ground and top states, while the intermediate state is only virtually involved. The dynamics corresponds thus to that of an effective two-level system with the linewidth of the upper state. This process produces a temperature lower than that of the EIT-assisted cooling and essentially limited by the lifetime of the upper state.

The theoretical analysis is based on the time evolution of the atomic momentum and kinetic energy following the model discussed in Refs. [18–20]. We restrict our study to one dimension, and consider an atom of mass M and momentum p (along the x axis), with internal levels $|0\rangle, |1\rangle, |2\rangle$ with increasing energies $0, \hbar\omega_{01}, \hbar\omega_{02}$, where $|1\rangle$ ($|2\rangle$) decays radiatively into $|0\rangle$ ($|1\rangle$) at rate Γ_1 (Γ_2). The Hamiltonian for the atom interacting with two laser fields at frequencies ω_1, ω_2 and wave vectors k_1, k_2, respectively, is $\mathcal{H} = \mathcal{H}_{at} + V_L + W$, where

$$\mathcal{H}_{at} = \frac{p^2}{2M} - \hbar\delta_1|1\rangle\langle1| - \hbar(\delta_1+\delta_2)|2\rangle\langle2|,$$

$$V_L = \hbar\Omega_1|1\rangle\langle0|\cos(k_1 x) + \hbar\Omega_2|2\rangle\langle1|\cos(k_2 x) + \text{H.c.},$$

with detunings $\delta_j = \omega_j - \omega_{j0}$ ($j = 1, 2$), Rabi frequencies Ω_1 and Ω_2. W describes the coupling to the modes of the electromagnetic field in the vacuum. We follow the time evolution of an atom, which at $t=0$ is in the state $|0, p\rangle$ with energy $E_0(p) = p^2/(2M)$, under the assumption of weak Rabi frequency Ω_1, so that we can treat the coupling of state $|0\rangle$ to state $|1\rangle$ in perturbation theory [21]. The scattering processes causing a change of the atomic momentum are the following:

(i) Absorption of one photon with momentum $\hbar k_1$ along the x axis on the transition $|0\rangle \rightarrow |1\rangle$ followed by emission of a photon with momentum $\hbar k_{1s}$. The final atomic state is $|0, p'\rangle = |0, p+\Delta p_1\rangle$ with $\Delta p_1 = \hbar k_1(1 - \hat{e}_1 \cdot \hat{x})$, where $\hat{e}_1$ denotes the direction of photon emission. The corresponding scattering rate is $\mathcal{W}_1(p, \hat{e}_1) = \mathcal{P}_1(\hat{e}_1)\mathcal{R}_1(p)$, where $\mathcal{P}_1(\hat{e}_1)$ is the spatial pattern of spontaneous emission for transition $|1\rangle \rightarrow |0\rangle$, normalized to unity. The dependence $\mathcal{R}_1(p)$ on atomic momentum is evaluated using the approach in [21], and takes the value

$$\mathcal{R}_1(p) = \frac{\Gamma_1\Omega_1^2}{8}\left|\frac{\delta_1' + \delta_2' + i\Gamma_2/2}{(\delta_1'+\delta_2'+i\Gamma_2/2)(\delta_1'+i\Gamma_1/2) - \Omega_2^2/4}\right|^2 \tag{1}$$

with $\delta_1' = -k_1 p/M + \delta_1$ and $\delta_2' = -k_2 p/M + \delta_2$. The scattering rate for absorption of one photon with momentum $-\hbar k_1$ is analogously evaluated. Around the two-photon resonance, $\delta_1 + \delta_2 = 0$, the scattering cross section exhibits an asymmetric Fano-like structure as a function of p, typical of interference processes [21]. This asymmetry is controlled by δ_1 and Ω_2, and affects critically the gradient of the cross section at $p = 0$, and thus the force exerted on the atom at slow velocities, as found in [17].

TABLE I. Parameters for cascade atomic transitions.

Parameter	Mg	Ca	Cs
1st Trans.	$3^1S_0 \rightarrow 3^1P_1$	$4^1S_0 \rightarrow 4^1P_1$	$6^2S_{1/2} \rightarrow 6^2P_{3/2}$
λ_1 (nm)	285.29	422.79	852.12
$\Gamma_1/2\pi$ (MHz)	78.8	34.7	5.2
$T_{D,1}$ (mK)	1.9	0.833	0.125
2nd Trans.	$3^1P_1 \rightarrow 3^1D_2$	$4^1P_1 \rightarrow 5^1S_0$	$6^2P_{3/2} \rightarrow 10^2D_{5/2}$
λ_2 (nm)	880.92	1034.66	563.68
$\Gamma_2/2\pi$ (MHz)	2.0	5.3 [8]	0.49 [16]
$T_{D,2}$ (μK)	48	127	12

(ii) The atom undergoes two absorption processes at frequencies ω_1 and ω_2 followed by emission of a photon of wave vector k_{2s} and a second of wave vector k_{1s} such that the atom is pumped back to state $|0\rangle$. After this process, the atom is found in the state $|0, p'\rangle = |0, p+\Delta p_2\rangle$ with $\Delta p_2 = \hbar k_1(\pm1 - \hat{e}_1\cdot\hat{x}) + \hbar k_2(\pm1 - \hat{e}_2\cdot\hat{x})$ where $\hat{e}_2$ denotes the direction of emission of the k_{2s} photon. The scattering rate for each process is $\mathcal{W}_2(p, \hat{e}_1, \hat{e}_2) = \mathcal{P}_1(\hat{e}_1)\mathcal{P}_2(\hat{e}_2)\mathcal{R}_2(p)$. Here $\mathcal{P}_2(\hat{e}_2)$ is the spatial pattern for spontaneous emission on $|2\rangle \rightarrow |1\rangle$ normalized to unity and $\mathcal{R}_2(p)$ is the convolution of transition amplitudes, where energy conservation is imposed on the scattering process, and it vanishes when Γ_2 or Ω_2 are set to zero.

A differential change of the mean kinetic energy $\langle E \rangle$ is given by the sum of the energy changes due to each scattering process weighted by their corresponding rates [18]. The equation for the evolution of $\langle E \rangle$ is found taking into account all absorption-emission paths of copropagating and counter-propagating photons, and has the form [22]

$$\frac{d}{dt}\langle E \rangle = \int dp f(p)\frac{\hbar p}{M}\{k_1[\mathcal{R}_1(p) - \mathcal{R}_1(-p)]$$
$$+ (k_1+k_2)[\mathcal{R}_2(p) - \mathcal{R}_2(-p)]\} + H(\sigma_1, \sigma_2). \tag{2}$$

Here $f(p)$ is the momentum distribution and term

$$H(\sigma_1, \sigma_2) = \sigma_1(1+\chi_1)\frac{\hbar^2 k_1^2}{M}$$
$$+ 2\sigma_2\left((1+\chi_1)\frac{\hbar^2 k_1^2}{M} + (1+\chi_2)\frac{\hbar^2 k_2^2}{M}\right) \tag{3}$$

describes the heating process, with $\chi_i = \int d\hat{e}_i \mathcal{P}_i(\hat{e}_i)(\hat{e}_i\cdot\hat{x})^2$ and $\sigma_i = \int dp f(p)\mathcal{R}_{i,}(p)$ ($i = 1, 2$). The final temperature is found by considering the latest stages of cooling, when the Doppler shift is assumed to be smaller than the natural width of the optical transitions [18,19]. By expanding the rates $\mathcal{R}_i(\pm p) = \mathcal{R}_i(0) \pm \mathcal{R}_i'(0)p$, $\mathcal{R}_i'(0) = d\mathcal{R}_i(p)/dp|_{p=0}$, and $\sigma_i \sim \mathcal{R}_i(0)$, we obtain

$$\frac{d}{dt}\langle E \rangle = -2\alpha\langle E \rangle + H^o, \tag{4}$$

where α is the cooling rate

$$\alpha = -2\hbar k_1 \mathcal{R}_1'(0) - 2\hbar(k_1+k_2)\mathcal{R}_2'(0) \tag{5}$$

and $H^o = H(\mathcal{R}_1(0), \mathcal{R}_2(0))$ is the heating rate. The temperature T is operationally defined by the relation $k_B T/2 = \langle E \rangle_{st}$,

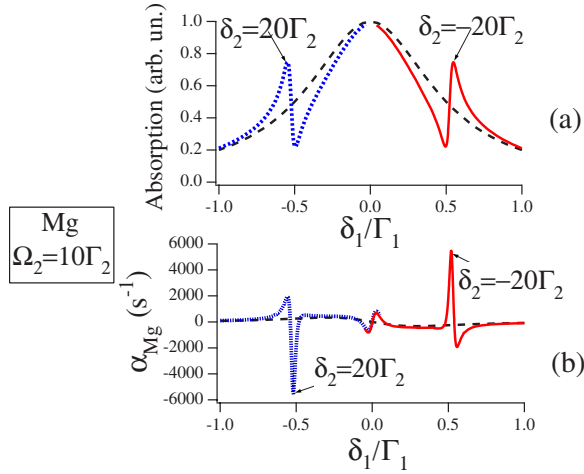FIG. 1. (Color online) (a) Absorption coefficient (arbitrary units) and (b) cooling rate α_{Mg} versus detuning δ_1 for the regime of EIT-assisted cooling on the Mg transition in Table I. The parameters are $\Omega_1 = 0.01\Gamma_1$ and $\Omega_2 = 0$ (dashed line), $\Omega_2 = 10\Gamma_2$ and $\delta_2 = -20\Gamma_2$ (solid red line), $\Omega_2 = 10\Gamma_2$ and $\delta_2 = 20\Gamma_2$ (dotted blue line). The parameters Ω_2 and δ_2 correspond to the experimental conditions of [13]. The minima of the absorption coefficient at $\delta_1 \approx \pm 0.5\Gamma_1$ are at the two-photon resonance. They are due to atomic coherence between the ground and top states of the cascade and give rise to a substantial modification of the cooling rate.

where k_B the Boltzmann constant and $\langle E \rangle_{st}$ is the mean energy, steady-state solution of Eq. (4). One finds

$$k_B T = H^o / \alpha, \tag{6}$$

which depends through H^o on the parameters χ_i determined by the spatial pattern of the spontaneous emission and equal to 2/5 for a dipole emission. For Doppler cooling on the lower transition the value $\chi_1 = 1$ reproduces the temperature reached in three-dimensional cooling [20].

The evaluation of the final temperature T requires the knowledge of term $\mathcal{R}_2(p)$, which allows for an explicit analytical form only in certain limiting cases. Therefore, in our numerical analysis we have linked $\mathcal{R}_1, \mathcal{R}_2$ to the steady-state solution of the optical Bloch equations for the atomic density matrix ρ [11],

$$\mathcal{R}_1(p) = \Gamma_1 \rho_{11}^{st}(p) - \Gamma_2 \rho_{22}^{st}(p), \quad \mathcal{R}_2(p) = \Gamma_2 \rho_{22}^{st}(p), \tag{7}$$

where the steady state populations ρ_{ii}^{st} of the three atomic levels depend on the atomic momentum p due to the Doppler effect. The results obtained with this method agree with the prediction obtained using Eq. (1) when Γ_2 (and thus $\mathcal{R}_2$) is zero [23].

We have evaluated the cooling temperatures as a function of laser and atomic parameters for the different species listed in Table I. $T_{D1,2}$ denotes the Doppler cooling limit temperature $\hbar\Gamma_{1,2}/(2k_B)$ for a two-level system with the upper lifetime equal to that of the $|1\rangle$ or $|2\rangle$ state, respectively. In order to simplify the theoretical treatment we treat all optical transitions as closed.

Figure 1(a) displays the absorption coefficient of Mg atoms (where $\Gamma_1/\Gamma_2 \approx 39$) for Doppler cooling on the lower transition (dashed line) and for EIT-assisted cooling with a
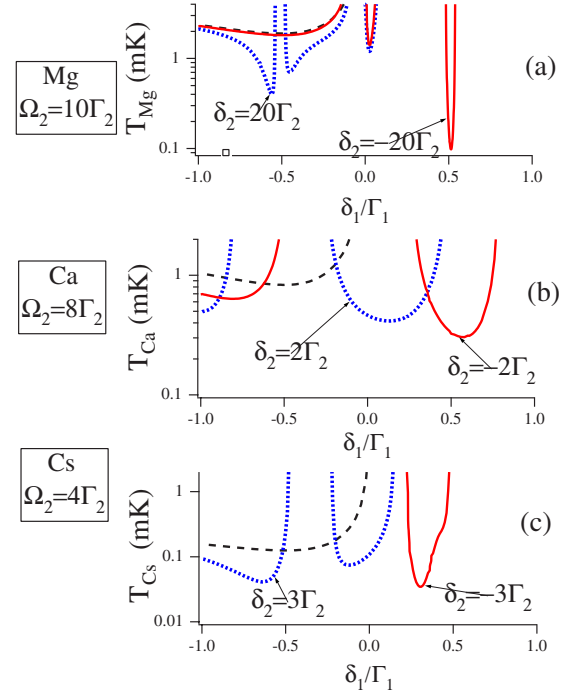

FIG. 2. (Color online) Temperatures T in mK versus detuning δ_1 for EIT-assisted cooling of (a) Mg, (b) Ca, and (c) Cs, using the parameters in Table I, for $\chi_1 = \chi_2 = 1$ as for a three-dimensional cooling geometry. The hyperfine structure of Cs is not considered. The other parameters are $\Omega_1 = 0.01\Gamma_1$, where Γ_1 depends on the atomic species, Ω_2 and δ_2 are specified in the figures. The continuous red (dotted blue) lines indicate the temperatures obtained for negative (positive) values of δ_2, the dashed line shows the Doppler cooling temperature obtained when $\Omega_2 = 0$.

strong near resonant coupling laser on the upper transition (continuous and dotted lines). The coupling laser modifies strongly the lower transition absorption profile around $\delta_1 + \delta_2 \approx 0$. For the chosen parameters the absorption profile contains a narrow and deep structure at positive or negative detuning δ_1, depending on the sign of δ_2. In presence of the coupling laser, the cooling rate α_{Mg}, proportional to the derivative of the absorption coefficient with respect to the frequency, presents deep and narrow structures, as in Fig. 1(b). The larger values of α in Fig. 1(b) correspond to lower cooling temperatures, as shown in Fig. 2(a), see also Eq. (6). For $\delta_2 > 0$ the EIT process increases the cooling rate by twenty times, and reduces the Doppler limit temperature by a factor of 5. Figures 2(b) and 2(c) display similar behaviors for the Ca and Cs transitions. The Ca case confirms a peculiar feature of the Mg results: the lowest temperatures are reached about the two-photon resonance for positive detuning δ_1, corresponding to the largest increase in the damping rate as shown in Fig. 1(b). A similar dependence was already observed in the EIT cooling of [17]. EIT-assisted cooling at positive δ_1 is difficult to realize because its efficiency is sensitive to changes of Ω_2, which may occur along the atomic sample: in regions where $\Omega_2 \sim 0$ the atoms are heated by the laser coupling to the lower transition. For the explored parameters of Mg and Ca, the temperatures reached in this EIT-assisted cooling are not very close to the Doppler cooling limit on the upper transition. Interestingly, in Cs low
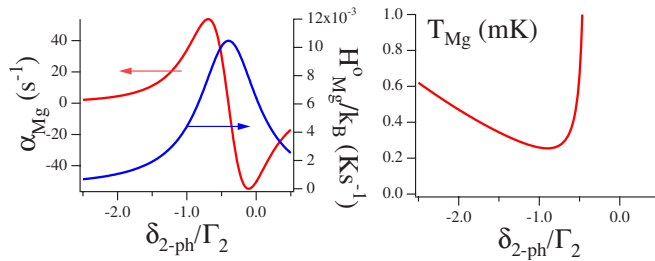
FIG. 3. (Color online) Two-photon cooling of Mg. (Left) Cooling rate α_{Mg} and heating rate H^o_{Mg}, and (right) temperature T_{Mg} (right) versus the two-photon detuning $\delta_{2\text{-ph}} = \delta_1 + \delta_2$. The parameters are $\Omega_1 = 0.01\Gamma_1$, $\delta_1 = -40\Gamma_1$, and $\Omega_2 = 50\Gamma_2$.

temperatures also are achieved using a laser at short wavelength on the upper transition, corresponding to a large value of k_2 and contributing to an increase in the last term in the heating rate of Eq. (3).

Tuning the intermediate state $|1\rangle$ far-off resonance, while keeping the two lasers close to two-photon resonance, one switches from the EIT-assisted cooling regime, where atomic coherence plays an important role on the cooling dynamics, to the two-photon cooling regime, where the dynamics is essentially described by an effective two-level system composed by the states $|0\rangle$ and $|2\rangle$. Here, cooling is expected to produce a temperature close to the Doppler limit T_{D2} on the upper level. This is confirmed by the numerical analysis displayed in Fig. 3 for Mg, where the ω_1 laser is far detuned by 40 linewidths from the $|0\rangle \rightarrow |1\rangle$ transition and the frequency sum $\delta_{2\text{-ph}} = \delta_1 + \delta_2$ is scanned over a small frequency interval determined by the Γ_2 decay rate. Temperatures $T \sim T_{D2}$, and much smaller than T_{D1}, are obtained for different values of

δ_1 and Ω_2. The application of this scheme requires a control of the laser sum frequency $\delta_1 + \delta_2$ with a precision determined by Γ_2, which is challenging in the case of an ultraviolet laser as required for Mg. An analysis of the velocity capture range Δv_c shows that a lower temperature is reached at the expense of the lower efficiency in the collection of the cooled atoms. This can be seen by comparing the capture range of the EIT process $\Delta v_c \sim 0.2\Gamma_1/k_1$, as derived from the frequency range of Fig. 1 where the damping rate is significantly different from that of the Doppler cooling process, with the velocity capture range of two-photon cooling $\Delta v_c \sim \Gamma_2/k_2$, as derived from Fig. 3. Moreover, the numerical results also point out that in EIT-assisted cooling the cooling rate α is much larger than in the two-photon cooling.

In conclusion, we have analyzed laser cooling based on two-color excitation of an atomic cascade, when the upper state has a longer lifetime. This scheme is complementary to that of quenching cooling where the stability of top and intermediate states is exchanged. We have discussed two regimes, EIT-assisted and two-photon cooling. Lower temperatures may be reached using two-photon excitations, with the intermediate state excited only virtually, while in the EIT-assisted scheme a larger capture range and a more flexible combination with the single photon Doppler cooling enhance the cooling performance.

Note added in proof: Recently, we learned of a related work by Dunn *et al.* [24].

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