PHYSICAL REVIEW A **72**, 051403(R) (2005)

Two-photon cooling of magnesium atoms

N. Malossi, S. Damkjær, P. L. Hansen, L. B. Jacobsen, L. Kindt, S. Sauge, and J. W. Thomsen*
The Niels Bohr Institute, Universitetsparken 5, 2100 Copenhagen, Denmark

F. C. Cruz
Instituto de Fisica Gleb Wataghin, UNICAMP, Campinas, 13083-970, Brazil and JILA, NIST and University of Colorado, Boulder, Colorado, 80309-0440, USA

M. Allegrini and E. Arimondo
Dipartimento di Fisica Enrico Fermi, Università di Pisa, L. Pontecorvo 3, I-56127 Pisa, Italy
(Received 8 May 2005; published 16 November 2005)

A two-photon mechanism for cooling atoms below the Doppler temperature is analyzed. We consider the magnesium ladder system $(3s^2)^1S_0 \rightarrow (3s3p)^1P_1$ at 285.2 nm followed by the $(3s3p)^1P_1 \rightarrow (3s3d)^1D_2$ transition at 880.7 nm. For the ladder system quantum coherence effects may become important. Combined with the basic two-level Doppler cooling process this allows for reduction of the atomic sample temperature by more than a factor of 10 over a broad frequency range. First experimental evidence for the two-photon cooling process is presented and compared to model calculations. Agreement between theory and experiment is excellent. In addition, by properly choosing the Rabi frequencies of the two optical transitions a velocity independent atomic dark state is observed.

DOI: 10.1103/PhysRevA.72.051403 PACS number(s): 32.80.Pj, 42.50.Vk, 32.80.Wr

The interest in laser cooling of alkaline-earth-metal atoms has increased significantly over the past years. One of the main reasons is the use of those elements for improved frequency standards and optical clocks [1–4]. For this purpose the ultranarrow intercombination $^1S_0 \rightarrow {}^3P_{1,0}$ lines are promising candidates. Their linewidths range from a few kHz to mHz depending on the specific element and isotope. For example, the $^1S_0 \rightarrow {}^3P_1$ linewidth increases from 31 Hz in Mg to 7.6 kHz in Sr due to a larger spin-orbit coupling with an increasing nuclear charge. The $^1S_0 \rightarrow {}^3P_0$ is strongly forbidden in even isotopes [5] but weakly allowed in odd isotopes [6], leading to mHz linewidths. Another very attractive feature of group two elements is the simpler internal structure of the most abundant (bosonic) isotopes, with no hyperfine levels. Efforts have concentrated on magnesium, calcium, strontium, and ytterbium [1–22], while, to our knowledge, beryllium and barium have not yet been cooled or trapped. Ytterbium is an alkaline-earth-metal–like atom that has been Bose condensed [21] using all-optical methods. Because the $^1S_0$ ground state of alkaline-earth-metal atoms is nondegenerate, sub-Doppler cooling is not possible on the resonance transition and the temperature is often limited to a few millidegrees Kelvin. Other cooling strategies have been suggested, and for some elements, successfully employed. The most direct method was demonstrated for strontium atoms where the narrow $^1S_0 \rightarrow {}^3P_1$ intercombination line was used to cool the atomic ensemble to the nanokelvin regime [19]. It has also been exploited for cooling ytterbium [20]. For magnesium and calcium, this method is not straightforward since the intercombination line cooling force is too weak to counteract gravity. One alternative is the so-called quench cooling [13], which consists in reducing the apparent lifetime of the triplet state by coupling it to a fast decaying singlet state using an extra laser. Quench cooling has been demonstrated for calcium [14,15] with an atom transfer efficiency of 20% and temperatures of about 7 μK. For magnesium, quench cooling seems difficult to implement.

In this paper we demonstrate an alternative cooling strategy using a two-color laser excitation on the ladder system: a resonant excitation $(3s^2)^1S_0 \rightarrow (3s3p)^1P_1$ transition at 285.2 nm followed by the $(3s3p)^1P_1 \rightarrow (3s3d)^1D_2$ transition at 880.7 nm (see Fig. 1). The two-photon cooling process involves the atomic coherence between the terminal states of the ladder scheme. As a key requirement, the laser frequency must be close to the two-photon resonance $\delta_1 + \delta_2 = 0$. If both lasers are detuned from the single-photon resonances, the two-photon resonance determines an effective two-level system with an associated Doppler limit set by the lifetime of the uppermost ladder state. In another complex regime both

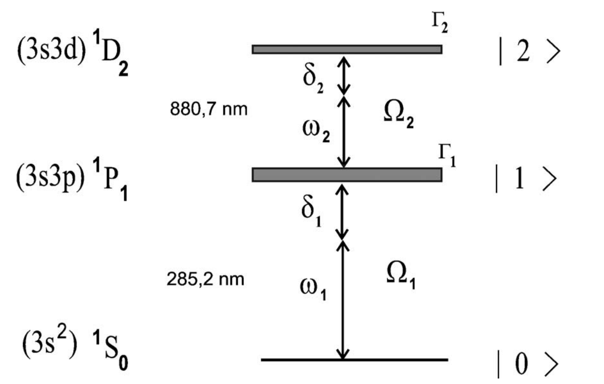

FIG. 1. Energy levels relevant for the two-photon transition $(3s^2)^1S_0 \rightarrow (3s3p)^1P_1 \rightarrow (3s3d)^1D_2$ in ^{24}Mg.

*Electronic address: jwt@fys.ku.dk

TABLE I. Characteristic numbers for the transitions of Fig. 1.

Parameter	${}^1S_0 \rightarrow {}^1P_1$	${}^1P_1 \rightarrow {}^1D_2$
Wavelength	285.21 nm	880.68 nm
Linewidth $\Gamma_i/2\pi$	78.8 MHz	2.2 MHz
Saturation intensity $I_{s,i}$	446 mW/cm^2	426 μW/cm^2
Doppler Temperature $\Gamma_i/2k_B$	1.9 mK	53.5 μK

one- and two-photon resonances are important. We have explored experimentally a coherent cooling regime where the uv laser is not far detuned from the single-photon resonance, but the ir laser is far detuned yet the frequency sum is nearly zero. Two-photon excitation allows for production of atomic coherences that lead to a broad frequency region over which cooling takes place. However, the additional laser can also produce heating through an increased momentum diffusion. Therefore the final temperature achieved by the presence of the additional laser excitation requires a careful analysis.

The two-photon excitation of ^{24}Mg is illustrated in Fig. 1. The levels are $|0\rangle = (3s^2)\,{}^1S_0$, $|1\rangle = (3s3p)\,{}^1P_1$, and $|2\rangle = (3s3d)\,{}^1D_2$. Relevant numbers for the two transitions are listed in Table I. From the $(3s3d)\,{}^1D_2$ state the atom can spontaneously decay to the $(3s3p)\,{}^1P_1$ state at a rate of $\Gamma_2 = 1.4 \times 10^7$ s^{-1} and further to the ground state at a rate $\Gamma_1 = 4.95 \times 10^8$ s^{-1}. Unlike other alkaline-earth-metal systems, the magnesium ladder system is an ideal closed three-level system. Firstly, no lower-lying D states contribute to the single-photon excitation as for Ca, Sr, and Ba. Secondly, all other optical decay channels from the $(3s3d)\,{}^1D_2$ state have vanishing transition rates [22]. The laser-atom interaction is described by the Rabi frequency Ω_i and the saturation parameter $s_i = \Omega_i^2/(\delta_i^2 + \Omega_i^2)$, with $i = (1,2)$ for the two transitions. For laser cooling, the first (uv) and second (ir) transitions are usually excited by counterpropagating beams. However, emphasizing the effect played by induced coherences of the upper transition, we consider only a single-beam, running-wave configuration for the infrared transition. Conventional Doppler cooling in a three-level ladder configuration has been investigated in [23]; however, the coherences between the two extreme states $|0\rangle$ and $|2\rangle$ were not considered. Three-level systems are characterized by the presence of additional coherence between the two extreme levels, $|0\rangle$ and $|2\rangle$ in Fig. 1. The role of this additional coherence on the laser cooling process has been the subject of several investigations. Two-color excitation in a three-level Λ system, with a large role played by the low-frequency coherence, has been examined in detail for the laser cooling of ions [24,25]. More recently ion cooling based on the atomic preparation into a dark state was investigated [26]. For neutral atoms the attention was concentrated on the ladder configuration. A detailed investigation was applied to metastable helium [27]. The helium two-color excitation produced a strong force, damping the atomic motion, but the final atomic temperature was larger than that reached in the Doppler cooling for the lower two-level system. This higher temperature is simply due to the shorter lifetime of the upper state compared to the intermediate level. Another important application of the two-color ladder excitation is quench cooling using the narrow intercombination line (Mg, Ca). In this case the ladder system is similar, but the excited state $|1\rangle$ has a decay rate Γ_1 significantly smaller than the upper state decay rate Γ_2. Accordingly, the population and optical coherences related to state $|2\rangle$ may be eliminated adiabatically and the laser cooling may be described through an effective two-level atom with an effective damping rate $\Gamma_1^{\mathrm{eff}} = \Gamma_1 + (\Omega_2^2/\Gamma_2)[\Gamma_2^2/(4\delta_2^2 + \Gamma_2^2)]$ [13]. This effective rate leads to a stronger cooling force acting on the lower levels, a result that also applies to the helium two-color case.

For the magnesium case with $\Gamma_1 \gg \Gamma_2$, it is the population and coherences related to the intermediate $|1\rangle$ state that may be eliminated adiabatically, leading to an effective two-level system for the $|0\rangle$ and $|2\rangle$ states, coupled by the two-photon Rabi frequency $\Omega_{\mathrm{tp}} = \Omega_1\Omega_2/|\delta_1|$. Provided that the one- and two-photon frequencies are well separated, we obtain the following Doppler limited temperature:

$$k_B T_{\mathrm{tp}} = \frac{\hbar}{2}\left(\Gamma_2 + (\Gamma_1 + \Gamma_2)\frac{\frac{\Omega_1^2}{2}}{\frac{(\Gamma_1 + \Gamma_2)^2}{2} + \delta_1^2}\right). \tag{1}$$

This temperature can be significantly lower compared to the $|0\rangle$–$|1\rangle$ Doppler temperature. The effective two-level system approach is not valid when both the one-photon and two-photon contributions are present. Investigation in this regime was based on the exact solution of the three-level density matrix equations. The force, damping, and diffusion acting on the atoms may be derived from (here for the running wave configuration)

$$F = \frac{\hbar k_1 \Gamma_1}{2}[\rho_{11}(\delta_1 - k_1 v, \delta_2 - k_2 v) - \rho_{11}(\delta_1 + k_1 v, \delta_2 + k_2 v)] \tag{2}$$

and the diffusion in the case of low saturation of both levels

$$D = D_1 + D_2 = 2\hbar^2 k_1^2 \Gamma_1 \rho_{11} + 2\hbar^2 k_2^2 \Gamma_2 \rho_{22}, \tag{3}$$

where ρ_{11} and ρ_{22} are the population of the intermediate and upper level, respectively. The temperature is calculated from $T = D/\alpha$, where $\alpha = (dF/dv)|_{v=0}$ is the damping coefficient [28]. In Fig. 2 we display the temperature T relative to the single photon $|0\rangle$–$|1\rangle$ Doppler temperature T_D, as a function of ir detuning. We observe a clear cooling effect over a broad frequency range compared to the natural linewidth $\Gamma_2 = 2.2$ MHz. The minimum temperature corresponds to 1/5 of the Doppler temperature associated with the uv transition. Near the two-photon resonance the cooling force is affected as a dark state is formed. Within a three-level ladder system the presence of a dark state coupled to the ir+uv laser radiation plays a key role in the excitation process. The dark state wave function $|\psi_{\mathrm{NC}}\rangle$ [29] is given by

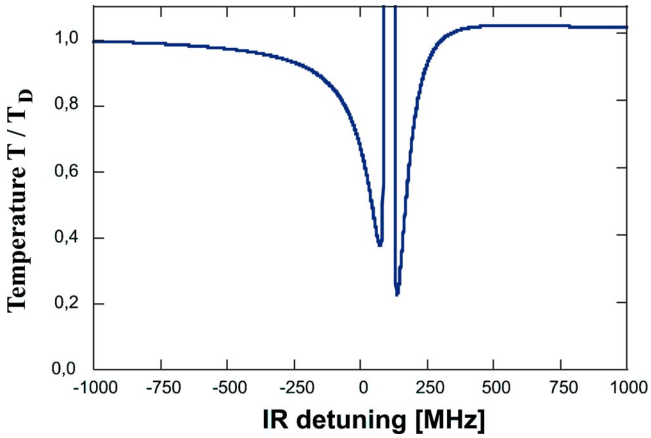

FIG. 2. (Color online) Calculated temperature T relative to the $|0\rangle$–$|1\rangle$ Doppler temperature T_D as a function of ir detuning. The uv detuning is $\delta_1 = -100$ MHz and the Rabi frequencies $\Omega_1 = 40$ MHz and $\Omega_2 = 120$ MHz, respectively.

$$|\psi_{\mathrm{NC}}\rangle = \frac{1}{\sqrt{\Omega_1^2 + \Omega_2^2}}[\Omega_2|0\rangle - \Omega_1|2\rangle]. \tag{4}$$

For this wave function the coupling to the intermediate $|1\rangle$ state vanishes and uv fluorescence is suppressed. In addition, for the dark state laser cooling on the uv transition is no more active. The time evolution of the atomic population prepared into the dark state is determined by the imaginary part of the smallest eigenvalue obtained by diagonalizing the Hamiltonian

$$H = \hbar \begin{pmatrix} \delta_1 + \delta_2 - i\frac{\Gamma_2}{2} & -\frac{\Omega_2}{2} & 0 \\ -\frac{\Omega_2}{2} & \delta_1 - i\frac{\Gamma_1}{2} & -\frac{\Omega_1}{2} \\ 0 & -\frac{\Omega_1}{2} & 0 \end{pmatrix} \tag{5}$$

representing the one- and two-photon excitation in the dressed state picture. In Fig. 3 we show the lifetimes of the three eigenstates along with the ac stark energy spectrum as a function of the ir laser detuning. Generally, the dark state is formed at $\delta_1 + \delta_2 = 0$ with a frequency width determined by Ω_1, Ω_2. The eigenvalues have a negligible dependence on the atomic velocity, excluding the presence of laser cooling based on velocity-dependent coherent population trapping.

The experimental setup has been described previously [9]. A series of experiments were performed to investigated the MOT fluorescence and shape as a function of the ir laser detuning and power. Atom fluorescence from 10^8 atoms is recorded by a photomultiplier only sensitive to uv. For spatial monitoring of the MOT we use a standard charge-coupled device (CCD) camera. Initially we performed spectroscopy measurements using a focused ir laser beam on the MOT. The ir beam was copropagating with one of the uv MOT laser beams and having the same polarization, but not retroreflected. The waist size was 200 μm and the power 80 mW, giving an ir Rabi frequency of $\Omega_2 = 600$ MHz. The

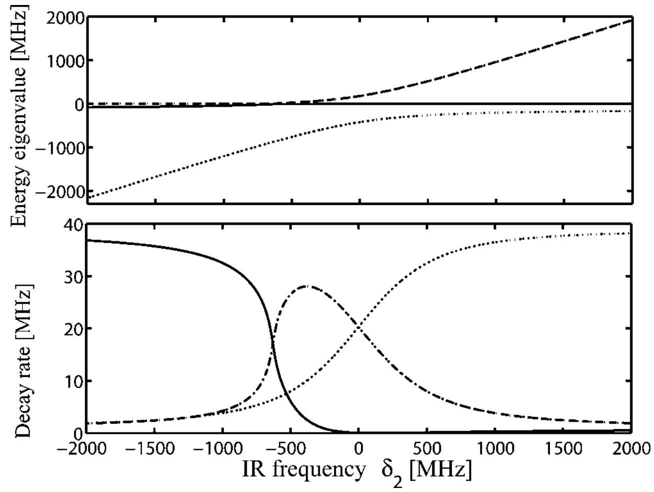

FIG. 3. Energy eigenvalues and decay rates obtained from diagonalizing Eq. (5) for $\delta_1 = -100$ MHz, $\Omega_1 = 40$ MHz, and $\Omega_2 = 600$ MHz. These values correspond to the experiment decribed below. The dark state (solid line) is present for a ir frequency detuning of about 0–600 MHz.

uv Rabi frequency was $\Omega_1 = 40$ MHz if all six uv beams are taken into account. According to Fig. 3 we expect a strong suppression of the uv fluorescence in this regime as a dark state is formed. From Eq. (4) the system is mainly trapped in the ground state $|\psi_{\mathrm{NC}}\rangle \sim |0\rangle$. Figure 4 shows the MOT with and without the ir laser beam. We observe a 200-μm tunnel in the MOT image, corresponding to suppression of fluorescence, when illuminated with the ir beam.

Using the single ir beam geometry we investigated cooling effects. For these experiments the ir laser was expanded to about 2 mm and $\Omega_2 = 120$ MHz. The uv Rabi frequency was set to $\Omega_1 = 40$ MHz. By monitoring the uv fluorescence and MOT z, x radii (z is the symmetry axis of the MOT coils) as a function of ir laser detuning we were able to identify a significant reduction in the MOT size. The reduction in both x and z radii was identical. Progressing in a similar way as for Sr, Ca [11,18], by using the equipartition theorem $\frac{1}{2}\kappa\langle r^2\rangle = \frac{1}{2}k_B T$, we estimated a reduction in temperature by

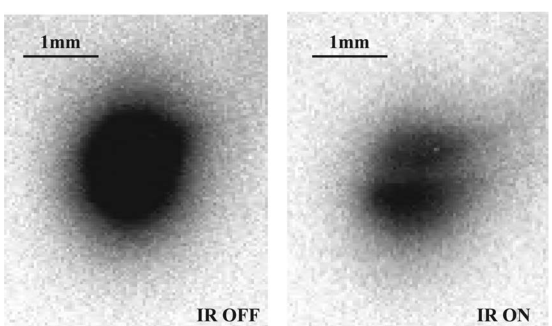

FIG. 4. Images of the MOT without the horizontal ir beam and with the horizontal ir beam, at $\Omega_1 = 40$ MHz and $\Omega_2 = 600$ MHz. The ir beam size of 200 μm is reproduced in the right image, the bright tunnel through the image (bright color means less fluorescence). The laser detunings were $\delta_1 = -100$ MHz and $\delta_2 = 400$ MHz.

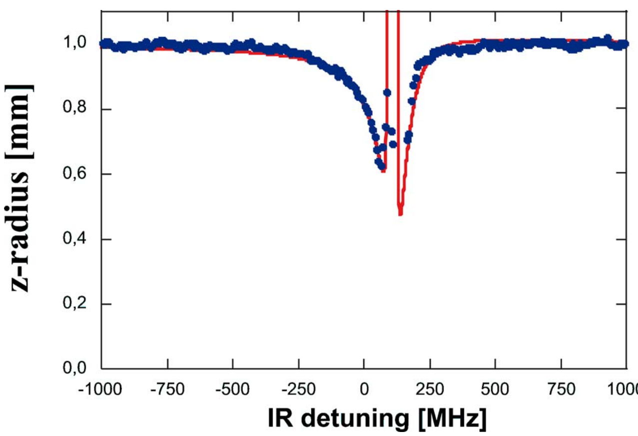

FIG. 5. (Color online) Experimental data for the MOT radius compared with the theoretical prediction presented in Fig. 2. The radius is related to the temperature by using the equipartition theorem. No fitting parameters are introduced, merely experimental determined values of $\delta_1 = -100$ MHz and Rabi frequencies of $\Omega_1 = 40$ MHz, $\Omega_2 = 120$ MHz, respectively.

more than a factor of 2.5 relative to the uv Doppler value. Mechanical effects of the ir beam were excluded by the constant level of uv fluorescence and by monitoring the position of the MOT during the experiment. Figure 5 shows a comparison between the theoretically predicted radius and the measured radius. No fitting parameters are introduced in the model, only experimentally determined values for laser detunings and Rabi frequencies. Additional heating is observed around the two-photon resonance where as dark state is formed. For very low uv and ir intensities calculations predict a temperature reduction of a factor of 10.

In conclusion, we have identified a cooling process working in a detuning range far from the usual two-photon Doppler regime. The basic principle, easy to implement, is to modify the quantum coherence of the initial two level system through an additional transition, in this case by using a ladder scheme. The dependence on the atomic and experimental parameters is an important issue. However, the coherence cooling regime was interpreted through numerical simulations only, and a better understanding of the cooling and heating processes requires further investigation. Temperatures relative to the Doppler temperature are reduced by more than a factor of 2.5 in the present setup, and theoretical predictions show that a factor 10 is possible. The present technique will also apply to other alkaline-earth-metal systems, such as calcium. Finally, we predict that the cooling mechanism described should also be effective in a V- and lambda-type of system using the intercombination line.

J.W.T. acknowledges financial support from the Danish Natural Research Council and the Carlsberg Foundation. We acknowledge support from the INTAS-01-0155 program. F.C.C. acknowledges the support of the Brazilian agencies FAPESP, CNPq, and CAPES, the contribution of Wictor C. Magno in the initial stage of this work, and discussions with Marcos C. Oliveira. We thank Nils Andersen and Giovanna Morigi for a careful reading of the manuscript.

[1] F. Ruschewitz *et al.*, Phys. Rev. Lett. **80**, 3173 (1998).
[2] T. Kisters *et al.*, Appl. Phys. B: Lasers Opt. **59**, 89 (1994).
[3] T. Udem *et al.*, Phys. Rev. Lett. **86**, 4996 (2001).
[4] C. W. Oates *et al.*, Opt. Lett. **25**, 1603 (2000).
[5] R. Santra, K. V. Christ, and C. H. Greene, Phys. Rev. A **69**, 042510 (2004).
[6] M. Takamoto and H. Katori, Phys. Rev. Lett. **91**, 223001 (2003).
[7] K. Sengstock *et al.*, Appl. Phys. B: Lasers Opt. **59**, 99 (1994).
[8] M. Machholm, P. S. Julienne, and K.-A. Suominen, Phys. Rev. A **59**, R4113 (1999).
[9] F. Y. Loo *et al.*, J. Opt. B: Quantum Semiclassical Opt. **6**, 81 (2004).
[10] T. P. Dinneen *et al.*, Phys. Rev. A **59**, 1216 (1999).
[11] X. Xu *et al.*, Phys. Rev. A **66**, 011401(R) (2002).
[12] X. Xu *et al.*, Phys. Rev. Lett. **90**, 193002 (2003).
[13] T. E. Mehlstäubler *et al.*, J. Opt. B: Quantum Semiclassical Opt. **5**, S183 (2003).
[14] T. Binnewies *et al.*, Phys. Rev. Lett. **87**, 123002 (2001).
[15] E. A. Curtis, C. W. Oates, and L. Hollberg, Phys. Rev. A **64**, 031403(R) (2001).
[16] J. Grünert and A. Hemmerich, Phys. Rev. A **65**, 041401(R) (2002).
[17] R. L. Cavasso-Filho *et al.*, J. Opt. Soc. Am. B **20**, 994 (2003).
[18] R. L. Cavasso Filho *et al.*, Appl. Phys. B: Lasers Opt. **78**, 49 (2004).
[19] H. Katori *et al.*, Phys. Rev. Lett. **82**, 1116 (1999).
[20] T. Kuwamoto *et al.*, Phys. Rev. A **60**, R745 (1999).
[21] Y. Takasu *et al.*, Phys. Rev. Lett. **91**, 040404 (2003).
[22] S. G. Porsev *et al.*, Phys. Rev. A **64**, 012508 (2001).
[23] W. C. Magno *et al.*, Phys. Rev. A **67**, 043407 (2003).
[24] M. Lindberg and J. Javanainen J. Opt. Soc. Am. B **3**, 1008 (1986).
[25] I. Marzoli *et al.*, Phys. Rev. A **49**, 2771 (1994).
[26] G. Morigi, J. Eschner, and C. H. Keitel, Phys. Rev. Lett. **85**, 4458 (2000).
[27] W. Rooijakkers *et al.*, Phys. Rev. Lett. **74**, 3348 (1995); Phys. Rev. A **56**, 3083 (1997).
[28] P. D. Lett *et al.*, J. Opt. Soc. Am. B **6**, 2084 (1989).
[29] E. Arimondo, in *Progress in Optics*, edited by E. Wolf (Elsevier, Amsterdam, 1996), Vol. 35, p. 25.