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Magneto-optical trapping of barium

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Laser cooling and trapping of the heavy alkaline-earth-metal element barium has been achieved based on the strong $6s^2\ {}^1S_0$-$6s6p\ {}^1P_1$ transition. The excited state decays to a large fraction into metastable D states. Two schemes were implemented where three additional laser-driven transitions provide closed cooling cycles. This results in a fraction of 50(20)% of the trapped atoms in one of the metastable states. A total efficiency of $0.4(1)\times10^{-2}$ for slowing a thermal atomic beam and capturing atoms into a magneto-optical trap was obtained. Trap lifetimes of more than 1.5 s were observed. They are shortened at high laser intensities by three photon photoionization losses. The developed scheme can be transferred to other systems with large leakage from the cooling transition, such as Ra which is interesting for sensitive fundamental symmetry research.

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Laser cooled atoms have become a vital tool for a variety of fundamental and applied experiments, e.g., Bose-Einstein condensation, high-precision measurements, optical frequency standards, and studies on fundamental symmetries. To date, the list of optically trapped elements includes all alkaline metals [1], noble gases in metastable states [2] except Rn, alkaline-earth-metal elements (Mg [3], Ca [4], Sr [4], and Ra [5]) and several other elements, e.g., Cr [6], Er [7], Ag [8] Yb [9], Hg [10], and Cd [11]. Laser cooling and trapping relies on narrow-band optical excitation in an almost closed subset of atomic states. In general, this requires driving of more than one optical transition. Extending laser cooling to other elements requires the selection of an efficient laser cooling scheme which accounts for peculiarities of their atomic level structure. The main motivation for trapping Ba is that its chemical analog Ra is uniquely sensitive to fundamental symmetry violation such as permanent electric dipole moments (EDMs) [12] or measurements of atomic parity violation (APV) [13].

In Ba or Ra the strong $ns^2\ {}^1S_0$-$nsnp\ {}^1P_1$ transitions, $n=6$ and 7, offer large optical forces. However, in both cases the substantial branching of 0.3% of the $nsnp\ {}^1P_1$ state into metastable D states requires quantitative repumping via several transitions [14] (Fig. 1). Only in Ra, the $7s^2\ {}^1S_0$-$7s7p\ {}^3P_1$ intercombination line is an alternative transition for laser cooling. The advantages of using this transition are the lower Doppler cooling limit and the simplicity of repumping. Although only a fraction of less than 4×10^{-5} is lost from this cooling cycle, the rather long lifetime of 422(20) ns of the $7s7p\ {}^3P_1$ state [15] results in a cooling force which is smaller by 2 orders of magnitude than the strong $7s^2\ {}^1S_0$-$7s7p\ {}^1P_1$ transition. Capturing of Ra in a magneto-optical trap (MOT) was reported from a Zeeman slowed atomic beam with 7×10^{-7} efficiency [5].

The $ns^2\ {}^1S_0$-$nsnp\ {}^1P_1$ transitions, $n=3,\ldots,5$, are used for laser cooling and trapping of lighter alkaline-earth-metal elements, where the branching to metastable D states is much smaller than for Ra and Ba. On average, an atom is transferred to one of the metastable D states after scattering of only $A_{\text{leak}}^{\text{Ba}}=330(30)$ photons at wavelength λ_1 for Ba (Fig. 1 and Table I). This corresponds to a velocity change of 1.8(2) m/s only. For Ra the leak rate is $A_{\text{leak}}^{\text{Ra}}=350(50)$ and the velocity change is 0.9 m/s. The largest leak from a cooling cycle of previously trapped elements of $A_{\text{leak}}^{\text{Cr}}=2500$ was given for Cr [6]. Because of its lighter mass Cr could be loaded into a MOT without repumping; however, the efficiency could be increased by 2 orders of magnitude with repumping from the D states. In contrast, for Ba no trapping can be expected without effective repumping from all three low-lying D states, i.e., $6s5d\ {}^1D_2$, ${}^3D_1$, and ${}^3D_2$ with lifetimes of 0.25, 60 s and even longer, respectively [18]. In this work two different repumping schemes were implemented for Ba, where repumping via low-lying states minimized optical pumping into additional states. The first scheme uses only the $6s6p\ {}^1P_1$ level as the intermediate state. This constitutes a closed five-level manifold ($6s^2\ {}^1S_0$, $6s6p\ {}^1P_1$, $6s5d\ {}^1D_2$, $6s5d\ {}^3D_2$, and $6s5d\ {}^3D_1$) involving transitions at the infrared wavelengths λ_{ir1}, λ_{ir2}, and λ_{ir3} (Table I). The common excited state for the cooling and the repumping transitions leads to multiple coherent Raman resonances [14]. In the limit of high intensities the populations in all five states become equal. This reduces the maximum optical force by a factor of 2/5 compared to an ideal closed two-level system. In atoms with nuclear spin different hyperfine states can be employed for cooling and repumping. The second scheme repumps the $6s5d\ {}^3D_1$ state via the $5d6p\ {}^3D_1^\circ$

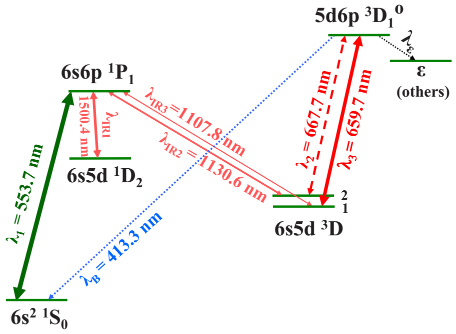

FIG. 1. (Color online) Low-lying energy levels of atomic Ba relevant for laser cooling. Full lines indicate laser-driven transitions and dashed lines show spontaneous decay channels.

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TABLE I. Vacuum wavelengths λ and experimental transition rates A_{ik} for barium.

Upper level	Lower level	Label	λ (nm)	A_{ik} ([10^8] s^{-1})
$6s6p\ ^1P_1$	$6s^2\ ^1S_0$	λ_1	553.7	1.19(1)[a]
	$6s5d\ ^1D_2$	λ_{ir1}	1500.4	0.0025(2)[a]
	$6s5d\ ^3D_2$	λ_{ir2}	1130.6	0.0011(2)[a]
	$6s5d\ ^3D_1$	λ_{ir3}	1107.8	0.000031(5)[a]
$5d6p\ ^3D_1^\circ$	$6s^2\ ^1S_0$	λ_B	413.3	0.013(1)[b]
	$6s5d\ ^3D_2$	λ_2	667.7	0.17(2)[a]
	$6s5d\ ^3D_1$	λ_3	659.7	0.38(2)[a]
	others		≥3000	0.011(2)[b]

[a] From Ref. [16].
[b] This work [17].

state using light at wavelength λ_3. This strong transition exhibits only a 2.0(4)% leak to further states. The main contribution arises from the $5d^2\ ^3F_2$ state, which according to a recent calculation [19] cascades to 94(3)% into one of the states of the cooling manifold. The observed number of trapped atoms was the same for both cooling schemes.

In the experiment, a Ba atomic beam was produced from an isotopically enriched sample of $^{138}BaCO_3$ which was mixed with Zr powder as a reducing agent in a resistively heated oven at 780(40) K temperature (Fig. 2). The beam entered a straight section, where it was overlapped with counterpropagating deceleration laser beams at wavelengths λ_1, λ_{ir1}, and λ_{ir2}. The laser beams were focused into the 1-mm-diameter oven orifice. The divergence of the laser beams was typically 5 mrad, their diameter 600 mm downstream of the oven is 3 mm and the typical laser powers were 12 mW at wavelength λ_1, 5 mW at λ_{ir1}, and 35 mW at λ_{ir2}. The detunings from the resonances were −290(2), −60(10), and −90(10) MHz, respectively. These parameters permitted slowing of atoms with velocities up to 150 m/s. Additional repumping from the $6s5d\ ^3D_1$ state increased the flux of slow atoms by 60%.

At the end of the slowing region three mutually orthogonal beams of 12 mm diameter of up to 15 mW power at wavelength λ_1 were retroreflected into themselves. Circular polarization for the MOT beams was produced by a set of $\lambda/4$ plates. The six beams crossed in the minimum of a magnetic field produced by a pair of coils in anti-Helmholtz configuration. The field gradient along the axis of the two coils reached up to 36 G/cm. The frequency detuning from resonance δ_{trap} of the trapping light was tunable between −200 and 50 MHz by acousto-optical modulators (AOMs). Three custom-made fiber lasers (Koheras) at the wavelengths λ_{ir1}(5 mW), λ_{ir2}(25 mW), and λ_{ir3}(60 mW) of typically 5 mm diameter are overlapped with the central region of the trap. Their detunings from resonance were kept well below the natural linewidth of 18 MHz of these transitions to achieve optimal repumping for the trapped atoms. For the second repumping scheme a laser beam at wavelength λ_3 of 10 mm diameter and 5 mW light power is copropagating with the MOT beams. The fluorescence from the central region of the trap was collected by a 60 mm focal length and focused on an aperture of 2.0(5) mm diameter to select the field of view. Fluorescence at the wavelengths λ_1 and λ_B were detected simultaneously by two photomultiplier tubes equipped with interference filters of 10 nm spectral bandwidth.

The fluorescence rate from trapped atoms R_1 increased for small negative frequency detunings δ_{trap} of the trapping laser beams at wavelength λ_1 [Fig. 3(a)]. Typical trap populations N_{MOT} up to 10^6 atoms were achieved. A lowest temperature for the trapped cloud of 5.4(7) mK was observed at a detuning of −15 MHz and an intensity of $0.2I_s$, where $I_s = 14.1\ mW/cm^2$ is the saturation intensity. The vertical MOT beam, which was orthogonal to the atomic beam, produced a Zeeman broadened fluorescence signal R_{beam} which is used to estimate the flux of atoms in the atomic beam. A comparison of the signal rates for these two conditions yields the fraction of the atomic beam which was captured in the MOT. The scattering rates γ_1 from the MOT and γ_{beam} from

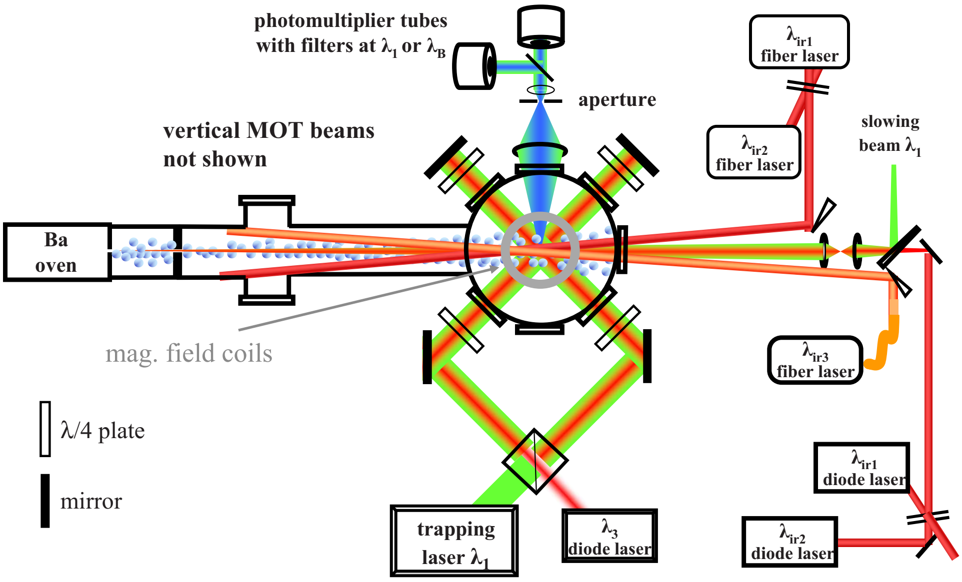

FIG. 2. (Color online) Setup for laser cooling and trapping of barium.

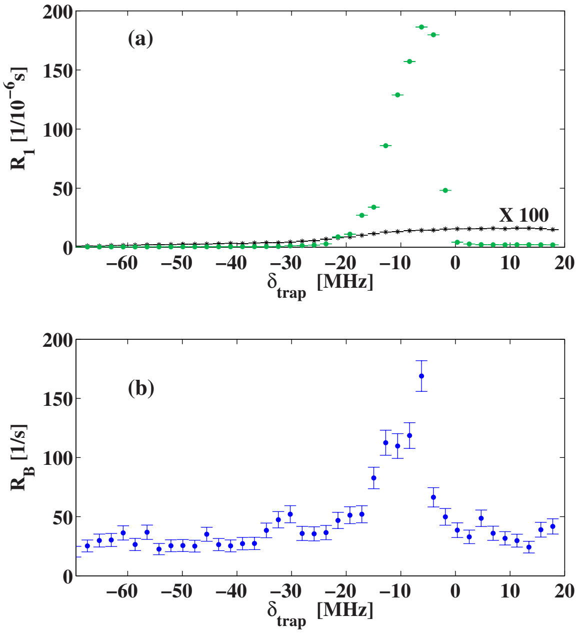

FIG. 3. (Color online) Signals from trapped atoms as a function of detuning of the trapping laser light at wavelength λ_1. (a) Fluorescence at wavelength λ_1. The black points is the Doppler-free fluorescence signal arising from the MOT laser beam which is orthogonal to the atomic beam. (b) Fluorescence at wavelength λ_B detected simultaneously. The trap lifetime was $\tau_{\mathrm{MOT}}=0.15(4)$ s.

the atomic beam are estimated from the known laser intensities and detunings. The collection efficiency is

$$\epsilon = \frac{R_1}{R_{\mathrm{beam}}} \frac{\Delta t}{\tau_{\mathrm{MOT}}} \frac{\gamma_{\mathrm{beam}}}{\gamma_1}, \tag{1}$$

where Δt is the average time of flight of thermal atoms through the light collection region. An efficiency of $\epsilon = 0.4(1) \times 10^{-2}$ was determined.

When the $6s5d\ ^3D_1$ state is repumped via the $5d6p\ ^3D_1^{\circ}$ state, fluorescence rate at wavelength λ_B was observed [Fig. 3(b)]. The rate was used to determine the fraction of atoms ρ_D in the metastable D states. The rate R_B is

$$R_B = \epsilon_B B_B B_{\mathrm{ir3}} \gamma_1 N_{\mathrm{MOT}}, \tag{2}$$

where ϵ_B is the detection efficiency for a photon at wavelength λ_B, $B_B=2.2(2)\%$ is the decay fraction of the $5d6p\ ^3D_1^{\circ}$ state to the ground state, and $B_{\mathrm{ir3}}=4.2(4)\times 10^{-5}$ is the decay fraction of the $6s6p\ ^1P_1$ state to the $6s5d\ ^3D_1$ state. Similarly the rate R_1 at wavelength λ_1 is

$$R_1 = \epsilon_1 \gamma_1 N_{\mathrm{MOT}}(1-\rho_D). \tag{3}$$

The ratio of the detection efficiencies was experimentally determined to be $\epsilon_B:\epsilon_1 = 1:0.8(1)$. A large fraction in the metastable states of $\rho_D=0.5(2)$ was found as it is expected for due to the coherent Raman transitions.

The trap population N_{MOT} depends on the intensity of the slowing beam at wavelength λ_1 (Fig. 4). The detuning of the slowing laser beam was $\delta_s=-260$ MHz, corresponding to a velocity class of 145 m/s. The loading rate increased up to a cooling beam power of 11 mW which corresponds to $2.7 I_{\mathrm{rms}}$. A further increase in the cooling beam power yielded a smaller flux into the MOT. This is caused by stopping atoms in the beam before they reach the trapping region. Thus, the velocity change in the slowing section was larger than 150 m/s. The average deceleration exceeded 1.7×10^4 m/s^2. Stopping can be avoided by a weak laser beam at wavelength λ_1 copropagating with the atomic beam, which would define a finite-end velocity [20].

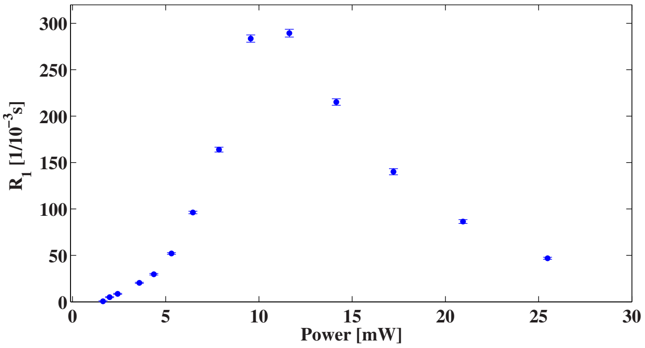

FIG. 4. (Color online) Dependence of the trap population N_{MOT} on the laser power of the deceleration beam at wavelength λ_1. The decrease at higher intensities was due to stopping of atoms before they reached the trapping region.

The lifetime τ_{MOT} of the trapped sample depends strongly on the intensity of the trapping laser beams at wavelength λ_1 (Fig. 5). A third-order process is observed for the losses as a function of laser intensity I with a rate constant of $\beta = 0.20(3)\ \mathrm{s}^{-1} I_s^{-3}$. This could be explained with the assumption of three-photon ionization as the main loss mechanism. The overlap of all laser beams in the trap region was crucial. The MOT lifetime τ_{MOT}^0 of up to 1.5 s was limited in the experiments by the intensity in the repumping laser beams. Atoms in the metastable states escape from the trap since they do not experience any trapping force. Similar effects have been observed in Ca [21].

We have realized laser cooling of Ba both in a closed five-level subsystem and in a six-level system with a small leak. These are the minimal subsets of levels for laser cooling of Ba in the ground state. Efficient deceleration of the atomic beam is achieved with counterpropagating lasers at high intensities. Further improvements can be expected from

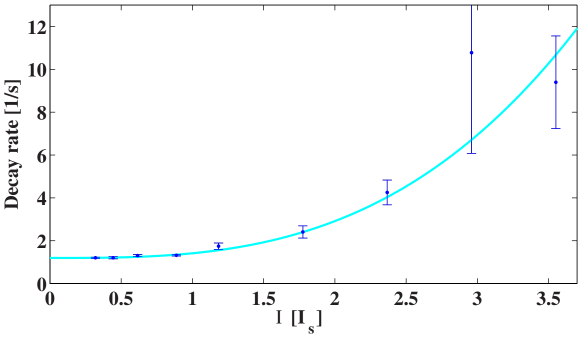

FIG. 5. (Color online) Decay rate from trapped sample as a function of MOT laser intensity. The solid line is $1/\tau_{\mathrm{MOT}} = 1/\tau_{\mathrm{MOT}}^0 + \beta I^3$, where $1/\tau_{\mathrm{MOT}}^0$ is the decay rate at intensity $I=0$ and β is the rate constant.

frequency broadening of the deceleration lasers, e.g., with electro-optical modulation [22]. This would enlarge the velocity acceptance of the deceleration and a larger fraction of the atomic beam velocity distribution can be stopped. As a note, a Zeeman slower is not applicable in such multilevel atomic laser cooling systems, for which repumping during slowing is required, because the changing Doppler shifts cannot be compensated by single magnetic field for all transitions at the same time.

The described laser cooling with a complex cooling cycle appears particularly well suited for efficient collection of rare isotopes in a MOT with similar level schemes, i.e., Ra isotopes. For Ra the strong optical transition $7s^2\ {}^1S_0$-$7s7p\ {}^1P_1$ yields a large optical force which permits a short slowing section for an atomic beam, while the intercombination line $7s^2\ {}^1S_0$-$7s7p\ {}^3P_1$ can be used to confine atoms in a MOT [5] which promises also lower temperature. The strong motivation for trapping Ra arises because such samples allow for novel precision measurements within the standard model in particle physics and searches for physics beyond it [5,23]. Atomic structure calculations for heavy alkaline-earth-metal atoms to evaluate their sensitivity to new physics are performed by several groups [24]. Recent computations show that Ra offers an enhancement of about 500 times due to nuclear effects [25] and 40 000 times due to the unique atomic level structure for nucleon or electron EDMs [12]. In addition, APV-induced effects are 100 times larger for the weak charge and 10^3 times larger for the nuclear anapole moment than in other systems [13]. All Ra isotopes with nuclear spin $I \neq 0$ have short lifetimes (e.g., ^{225}Ra, $\tau = 14.8$ d) and are only available in small quantities and require experiments in the proximity of an isotope production facility. Sensitive experimental searches for EDMs, which would establish simultaneous breaking for the symmetries charge conjugation (C) and parity (P) without strangeness, and measurements of APV in Ra require the developed trapping techniques.

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