Efficient photoionization for barium ion trapping using a dipole-allowed resonant two-photon transition

G. Leschhorn · T. Hasegawa · T. Schaetz

Abstract Two efficient and isotope-selective resonant two-photon ionization techniques for loading barium ions into radio-frequency (RF)-traps are demonstrated. The scheme of using the strong dipole-allowed transition \(6s^2 \, ^1S_0 \rightarrow 6s6p \, ^1P_1\) at \(\lambda = 553\) nm as a first step towards ionization is compared to the established technique of using a weak intercombination line \(6s^2 \, ^1S_0 \rightarrow 5d6p \, ^3D_1\), \(\lambda = 413\) nm). An increase of two orders of magnitude in the ionization efficiency is found favoring the transition at 553 nm. This technique can be implemented using commercial all-solid-state laser systems and is expected to be advantageous compared to other narrowband photoionization schemes of barium in cases where highest efficiency and isotope-selectivity are required.

1 Introduction

Ion traps have become an important tool in a growing number of fields, like precision measurements of fundamental constants\[1\], testing general relativity\[2\], frequency standards\[3\], quantum information processing\[4\] [5], quantum simulation\[6\] [7] or mass spectrometry\[8\]. During the last decade, the state-of-the-art loading of ion traps has changed from electron beam bombardment to photoionization of neutral atoms inside the trapping region. The main reasons are a higher efficiency and the isotope selectivity of most of the photoionization schemes presented so far. Additional beneficial effects, like strongly reduced static charging of dielectrics\[9\], lower motional heating rates of the ions after loading due to the suppression of patch potentials\[10\] and easier loading of surface-electrode traps with a substantially reduced trap depth\[11\] is of great advantage in the laboratories. Loading of Paul traps by narrowband cw photoionization\[12\] has been achieved by several groups on a growing number of atomic species (Ba\[13\] [14], Mg\[15\], Ca\[9\] [16] [17], Yb\[18\], Sr\[19\] [20], In\[21\] etc.). Several of these schemes have been applied to photoionization loading techniques that substitute the resistively heated atomic oven as source of neutral atoms. Instead, non-evaporating sources, for instance laser ablation targets\[22\] or magneto-optical traps\[23\] are used. These techniques additionally increase the efficiency and the advantages of photoionization loading and were mainly developed to load micro-structured surface traps. Furthermore, non-isotope selective loading schemes, e.g. using ultra-short pulses\[24\] or laser ablation\[25\] have been realized which share the advantage of being applicable to most laser-cooled ion species with a further increased efficiency.

In this paper, a novel loading technique for barium by isotope-selective photoionization using a dipole-allowed two-photon transition is demonstrated. A substantial increase in ion loading efficiency compared to established schemes\[26\] is found. The presented technique promises to provide substantial advantages in experiments that depend on maximizing the efficiency or...
on a fast, controlled and minimal invasive reloading of ions.

2 Experiment

The ion trap used for the experiments presented in this paper consists of four cylindrical, gold plated copper rods of 2 mm diameter for radial confinement and two additional ring shaped electrodes around the rods providing axial confinement. The rods are arranged in a quadrupole configuration with a minimal distance of 1.12 mm between the trap center and the rod surface. An RF voltage at $f_{RF} = 2\pi \times 6.8$ MHz is applied on the rod-electrodes. For $^{138}$Ba$^+$ the typically used radial secular frequency amounts to $2\pi \times 470$ kHz. The 5 mm-thick ring electrodes that are centered around the trap axis are spaced by 15 mm and have an outer diameter of 24 mm and an inner diameter of 8 mm. A DC voltage applied to these ring electrodes leads to a static, nearly harmonic confinement potential along the axis of the linear Paul trap. The trap setup is housed in an ultra-high vacuum chamber at a pressure of 2 Pa. 

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In scheme B, a 553.7 nm photon excites the $^1P_1$ state of $2\pi \times 18.9$ MHz natural linewidth, corresponding to 8.4 ns lifetime. Another photon at 413 nm excites the atom above the ionization threshold at 5.21 eV. The other ionization scheme (scheme A) works with two photons at 413 nm. The first one couples the ground state with the 5d6p $^3D_1$ level. The second photon excites the electron deeply into the continuum. Note that in scheme A, the atoms are optically pumped (wavy line) to metastable states because the spontaneous emission rate to the states 6s5d $^2D_1$ (3.8 $\times$ 10$^5$ s$^{-1}$ to $^2D_1$, 1.9 $\times$ 10$^7$ s$^{-1}$ to $^2D_2$) is larger than that to the ground state (1.5 $\times$ 10$^6$ s$^{-1}$). Laser cooling on the singly charged ion is done on the $A$ system spanned by the $^2S_{1/2}$, $^2P_{1/2}$ and $^2D_{5/2}$ levels, using photons near 493 nm and near 560 nm.

Neutral barium atoms are provided by a thermal beam evaporated into the trapping region by resistively heating a tantalum tube of 1 mm inner diameter filled with barium. A heating current of 3.15 A results in an atomic barium beam encloses an angle of approximately 100 $\pm$ 5° with the direction of laser propagation and
the transition frequency is thus red-shifted. The atomic beam is collimated by a 200 μm wide slit aperture at a distance of 4 cm from the trap axis in the direction perpendicular to the laser beam to ensure a minimal contamination of the trap electrodes with barium.

After ionization, the $^{138}$Ba$^+$ ions are laser-cooled on the $^2S_{1/2} \rightarrow ^2P_{1/2}$ transition at 493 nm. Figure 3(a) shows the relevant electronic levels and transitions of singly ionized barium. The upper state decays with a branching ratio of approximately 3:1 into the ground and a metastable $^2D_{3/2}$ state[30]. The population in the D state is re-pumped to the P state by an additional laser at 650 nm to approach a closed cycling transition for laser cooling. In addition, a weak magnetic field (0.5 mT) perpendicular to the linear polarization of the laser beam is used to prevent dark states in the Zeeman manifold of the metastable state. The 493 nm light is produced by frequency-doubling a commercial narrow bandwidth (different to the first step in scheme A) operating in the broadening of the spectral line can be found in section 2. In the present setup, the atoms have a velocity component along the propagation direction of the laser beams, and hence the transition is Doppler-broadened and the maximum loading rate is Doppler-shifted towards lower frequencies. The center of the unshifted atomic resonance is indicated by an arrow in the figure.

The experimentally derived loading rates of scheme A (B) are shown in dependence of the laser power in Fig. 4 (A: blue circles and B: red squares). The data of scheme A (scheme B) show in principle a dependency on the square of the laser power at 413 nm and 553 nm), as to be expected in a process requiring two photons. In scheme B, the laser power of the second laser (413 nm) is kept equal to that of the first laser (553 nm) and therefore, the two schemes are comparable in terms of laser power. This is justified, because scheme B does in principle not require a separate laser system at 413 nm as chosen in the current experiment for simplicity, having the laser source of scheme A available. As shown in [13,14], the transition into the continuum does not have to be of narrow bandwidth (different to the first step in scheme A and B) and can be replaced by an incoherent light source. The solid lines present the results of the calculations based on the model. For the given parameters, the loading rate of scheme B is already about 40 times larger than that of scheme A. As already mentioned in section 2, the beam waists of the two lasers used in scheme B differ considerably at the trap center. Atoms can only be ionized when both required beams overlap, so the lifetime of the excited state is much shorter than the typical transit time of the atom through the beam (see also section 4 and the caption of Fig. 4).
Taking the currently not contributing amount of laser power within the 413 nm beam for scheme B into account, a lower bound for the additionally increase in ionization efficiency can be derived. For the comparison of the two schemes, a homogeneous atom density in the trap volume and an equal probability to trap an ionized atom is assumed. Imposing a flat top profile for both beams of different waists would lead to an underestimation of scheme B in the comparison of efficiencies by a factor of \((w_{553}/w_{413})^2 \approx 4\). Considering, more realistically, gaussian beams and assuming a concentric overlap, the measured ionization rate achieved via scheme B has to be multiplied at least with a factor of 2.5 for comparison with scheme A. This allows to derive an increase of the total efficiency via scheme B by two orders of magnitude.

In Fig. 5, the dependence of the loading rate on the power of the first laser (553 nm) is shown when the power of the second laser is kept again constant at 1 mW. Taking saturation effects into account allows to explain the deviation from a linear dependency of the laser power at 553 nm. The power dependence is similar to that described in reference [13] (Fig.6) and follows in principle a dependency on the square root of the power,
as to be expected for an increased saturation of the transition. The solid lines represent the results of the calculations based on the model and will be explained in detail in the next section.

4 Model

In the present setup, the Doppler broadening of the spectral lines is caused by the temperature $T$ of the atomic ensemble in combination with the angle $\theta$ between the atomic and the laser beam. The derived spectral line of scheme A (Fig. 3(a)) has a full-width-at-half-maximum (FWHM) of 110 MHz. If this effect was caused by Doppler broadening at the given angle $\theta$, the FWHM achieved via scheme B should amount to 82 MHz. The experimental result, however, shows an even larger width (Fig. 3(b)). This effect is caused by power broadening, because the transition dipole moment $\mu$ exploited in scheme B is much larger than in scheme A. This is in agreement with the non-linear dependency of the loading rate depicted as a function of laser power in Fig. 5. Here a model is introduced in order to describe and explain the broadening of the spectral lines and the power dependence of the ion loading rate for scheme B with a set of three parameters ($\theta$, $T$, $\tau$) to obtain the results presented in Figs. 4 and 5. The red dashed curve is the result of fitting the model with parameter $\alpha$ and the frequency set at the maximum loading rate. The red solid line is the result of the fit when a detuning of -50 MHz from this frequency is assumed (see main text).

$$\Delta = -\frac{2 \left( (v_x t - X_0)^2 + (v_y t)^2 \right)}{w_2^2},$$

where $\alpha$ is a constant of proportionality (see also caption of Fig. 4). The peak intensity is related to the laser power $P_1$ by $I_1 = 2P_1/(\pi w_2^2)$.

The population $\rho$ of an atom in the intermediate state is assumed to be stationary. The lifetime in the intermediate state (inverse of the natural linewidth $\Gamma$) is much shorter than the typical transit time of the atom across the laser beam. For example, assuming a Maxwell-Boltzmann distribution for $T = 600$ K, the most probable speed of Ba atoms is about 200 m/s, and the transit time through the laser beam ($2w_1 = 460\mu m$) amounts to 2.3 $\mu$s, which is much longer than the lifetime of the $6s6p^1P_1$ state (8 ns). Therefore, $\rho$ can be expressed as

$$\rho = \frac{1}{4} \frac{\Omega_0 \left( v_x t, v_y t \right)^2}{(\delta - kv_z)^2 + k^2 + \Omega_0(v_x, v_y)^2},$$

where $\delta$ is the detuning of the laser from the atomic resonance frequency at rest, $k$ is the wavenumber of the laser beam driving the transition at the Rabi frequency $\Omega_0$ which is a function of the spatial coordinates:

$$\Omega_0 (x, y)^2 = \frac{2\mu^2}{\hbar^2\varepsilon_0 c} I_1 \exp \left\{ -\frac{2}{w_1^2} (x - X_0)^2 + y^2 \right\}. \quad (3)$$
In Eq. (3), $\hbar$ is the reduced Planck constant, $\varepsilon_0$ is the electric constant and $c$ is the speed of light in vacuum.

The total ion loading rate is obtained by integrating over the atomic velocity for an appropriate velocity distribution function $f(v_r, v_y)$:

$$\eta_0 = \int_0^\infty v_r dv_r \int_{-\infty}^\infty dv_y \int_0^{r+\delta} d\theta f(v_r, v_y),$$

$$\eta(v_r, v_y, v_r \cos \theta).$$

Here the coordinates $(r$ and $\theta)$, defined as $x = r \sin \theta$ and $z = r \cos \theta$, are introduced and $v_x$ and $v_z$ are replaced by $v_r \sin \theta$ and $v_r \cos \theta$, respectively. The azimuthal angle $\theta$ is the angle included by the direction of the atomic and the laser beam. For a thermal beam of atoms with mass $M$, a Maxwell-Boltzmann distribution can be assumed:

$$f(v_r, v_y) = \frac{1}{N} \exp \left( -M \frac{v_r^2 + v_y^2}{2k_B T} \right),$$

where $k_B$ is the Boltzmann constant and $N$ is a normalization factor. In the present experimental setup, the slit aperture collimates the beam in the $y$-direction. However, the diverging angle of the atomic beam in the $x$-$z$-plane is negligible compared to the velocity distribution of atoms in a thermal beam. Therefore, the divergence $\delta \theta$ in Eq. (4) approaches zero and $\theta$ is assumed to be at one value in the following.

In the case of scheme A the spontaneous emission from the intermediate state $(5d6p^4D_1)$ mainly leads to metastable states such as $6s5d^3D_{1,2}$. The maximal spontaneous emission rate from $5d6p^4D_1$ to the ground state $(6s^2 1S_0)$ amounts to $1.5 \times 10^6$ s$^{-1}$, whereas that to $6s5d^3D_1$ is $3.8 \times 10^7$ s$^{-1}$ and that to $6s5d^3D_2$ is $1.9 \times 10^6$ s$^{-1}$. Therefore, the atoms are optically pumped to the metastable states, and the stationary value of $\rho$ approaches zero. By changing the definition of $\rho$ to be the transition probability from the ground state to the intermediate state, it is possible to use the same formulae as in Eqs. (13), except for the value of the constant of proportionality $\alpha$. Thus, applying the present model does not allow for comparing the absolute value of the loading rates achieved via the two schemes.

By substituting Eqs. (13) and (5) into Eq. (4), the theoretical curves in Fig. 3 are obtained. In the case of scheme A, the temperature of the atomic beam is assumed to be $T=600$ K, the spontaneous emission rate is $5.85 \times 10^6$ s$^{-1}$, and the parameters of the laser providing the first and the second step of ionization are identical. For scheme B, the temperature is also set to $T=600$ K, but the spontaneous emission rate is $1.2 \times 10^6$ s$^{-1}$, and the parameters of the first and second step lasers are set independently. In the calculation, the parameters $\theta$, $\alpha$, and the center frequency of the resonance are left as fitting parameters for the data presented in Fig. 3(a) and $\theta = 98.3$ degrees is deduced. The center of the resonance is pointed at by an arrow in the figure. In Fig. 3(b) $\theta$ is fixed to the obtained value above, and only the center frequency of the resonance and $\alpha$ remain as fitting parameters.

Using the same model, the results of the derivation for the dependency of the ion loading rate on the power is also depicted in Figs. 4 and 5. Here the only fitting parameter is $\alpha$, and the detuning is set to the frequency at which the loading rate is at its maximum. In Fig. 5 however, the red dashed curve derived from the model does not fit the experimental result at small laser power very well. Scrutinizing the experimental data, it turned out that the laser frequency has drifted a few tens of megahertz to the red of the frequency for the peak loading rate. Assuming a detuning of $-50$ MHz from the frequency of maximal loading rate, the result of the model (red solid line) is in better agreement with the experimental findings.

5 Conclusions and outlook

In conclusion, this work demonstrated a novel, isotope-selective resonant two-photon photoionization scheme for barium using a dipole-allowed transition. The loading rates into a linear Paul trap was compared to an established scheme using a weak intercombination line. An increase in efficiency by two orders of magnitude was found. This allows to enhance the advantages of photoionization for barium like minimal charge build-up on insulators compared to electron bombardment ionization or patch potentials caused by a contamination of the trap electrodes. Reducing these effects might have a great impact in experiments that rely on high efficiency such as in cavity quantum-electrodynamics with ion coulomb crystals[33], one-dimensional surface traps for quantum information processing[11], two-dimensional trapping arrays for quantum simulations[35,36] or the recently realized optical trapping of ions[37]. The direct, efficient photoionization of an optically trapped barium atom reduces detrimental charging effects, followed by a loss of the ion out of the shallow, compared to RF-potentials, optical potential. This might allow, for example, to study cold-chemistry processes[38] and to simultaneously avoid micro-motion that normally occurs in conventional ion traps[30]. In addition, providing loading rates of $10^4$ ions/s are required to allow for experiments at repetition rates of the order of 0.1-1 kHz, for example, exploring controlled molecular ions that are sympathetically cooled via Ba ions. The devices combining the species, a storage ring[40,41,42] or
a molecular conveyor belt \[43\], will substantially depend on the sufficient efficiency of the Ba-ion source.

The presented technique can be further advanced by exploiting the tuneability of the 413 nm laser and its proximity to the ionization threshold. An increase in efficiency is expected by tuning the laser on resonance with a transition to either a field ionized Rydberg state \[14\] or with an autoionization resonance \[44,45\].

The presented technique can be simplified because the dye laser used to generate the 553 nm light can be substituted by a low maintenance, all-solid-state laser system.

Very recently, a commercial frequency-doubled diode laser used to generate the 553 nm light can be substituted by a low maintenance, all-solid-state laser system.

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