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Axialisation, cooling and quenching of Ba^+ ions in a Penning trap

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Abstract. Collisions of ions, stored in a Penning trap, with neutral background molecules usually lead to rapid ion loss from the trap unless the ions are excited by the sum of the frequencies of the reduced cyclotron and magnetron motion. Then the ions are cooled by collisions and are driven to the trap centre leading to substantial increase of the storage time. Furthermore in a three level system including a long living metastable state collisions deexcite this metastable state and increase the population density in the ionic ground state. In a laser spectroscopic experiment we demonstrate the advantages of collisions on Ba^+ ions stored in a Penning trap. The combined action of metastable state quenching, axialisation and cooling leads to a significantly enhanced laser induced fluorescence signal. The cross section for collisional relaxation of the ground state Zeeman levels has been determined and we find that it is of the same order of magnitude as quenching cross section for a metastable state. Cooling and increased signal strength allows us to observe extremely narrow resonances and motional sidebands in microwave induced Zeeman transitions in the Ba^+ electronic ground level.

PACS. 32.80.Pj Optical cooling of atoms; trapping – 32.60.+i Zeeman and Stark effects – 34.50.-s Scattering of atoms, molecules, and ions

1 Introduction

g_J-factors of atoms and ions are an important source of information about the wave functions of those systems since they include relativistic and radiative contributions. A comparison of experimental values to those obtained by theoretical calculations can act as a good test of the different theoretical approaches. Of particular interest in this respect are the atoms or ions of alkali - like structures since wave functions for these cases can be obtained with great precision.

Recently we performed g_J-factor measurements in the $6S_{1/2}$-ground state [1] and the long living $5D_{3/2}$-metastable state [2] of Ba^+. The laser - microwave double resonance technique was used to observe induced Zeeman - transitions on $^{138}Ba^+$ ions stored in a Penning trap. These experiments resulted in values for the g_J-factors which were accurate to 3×10^{-7} and 5×10^{-7} in the $6S_{1/2}$- and $5D_{3/2}$-states, respectively. Further improvement in accuracy was not possible because after laser excitation from the ground state the ions are quickly pumped into Zeeman components of the $5D_{3/2}$-state, which were not subject to our investigations. The radiative lifetime of this state has recently been determined to 79.8 s [3]. Although in our case at a residual pressure around 10^{-9} mbar the effective lifetime was reduced to a few seconds by collisional quenching, repopulation of the ground state by decay from the metastable state was very slow. Consequently the signal-to-noise ratio in our previous experiments was poor and averaging times of about 20 min were required to obtain sharp microwave induced Zeeman resonances. This time competes with the ion's storage time of the same order of magnitude and ion loss during the measuring time affected the line shape.

In principle the ion population in the ground state could be substantially enhanced by repumping the ions from the metastable states by lasers resonantly tuned to the $5D_{3/2}-6P_{1/2}$ transition. While this has been successfully performed on ions confined in a Paul trap, it would require 4 additional lasers in the case of a Penning trap, since at our magnetic field of 3 Tesla the $5D_{3/2}-6P_{1/2}$ transition is split into several Zeeman components. The separation between these components ranges up to 40 GHz which makes repumping by a single laser and its sidebands generated with the presently available techniques very difficult.

The easiest way to relax the ion population from the metastable states would be if they collide with the buffer gas molecules. This has been performed successfully in Paul traps [4]. In those cases collisions of heavy ions with light buffer gas molecules dampen the ions

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oscillation amplitude and lead to reduced ion temperatures and increased storage times. In a Penning trap, however, collisions with neutral atoms increase the radius of the so-called magnetron orbit, which describes the motion of the centre of the ions cyclotron orbit around the traps centre, and the ions get lost rapidly.

2 Collisional cooling in a Penning trap

Ion loss from a Penning trap by collisions with neutral background atoms can be avoided if the magnetron motion is coupled to the cyclotron motion by an additional radio frequency field at the sum frequency of the two eigenfrequencies $\omega_c^{'}$ and ω_m:

$$\omega_c' = \omega_c/2 + [\omega_c^2/4 - \omega_z^2/2]^{1/2} \quad (1)$$

$$\omega_m = \omega_c/2 - [\omega_c^2/4 - \omega_z^2/2]^{1/2}. \quad (2)$$

Here $\omega_c = (e/m)B$ is the free ions cyclotron frequency and $\omega_z = (eV/mr_0^2)^{1/2}$ the axial oscillation frequency in a trap of radius r_0 at an applied potential V. This method has been developed and described in detail by G. Bollen and coworkers [5] and applied successfully to high precision mass measurements in a Penning trap [6]. It also has been used for axialization of ions in Fourier transform ion cyclotron resonance mass spectrometry [7]. It relies on the fact that energy is continuously dissipated by collisions from the cyclotron motion of the ions. The radiofrequency field at $\omega = \omega_c' + \omega_m$, applied in a quadrupolar geometry in the radial plane of the trap, couples the magnetron motion to the cyclotron motion. Detailed studies of the ion trajectories [6] show that the magnetron radius decreases with time and the ions concentrate near the centre of the trap. Finally the ions assume the temperature of the environment. This technique is closely related to the motional sideband cooling introduced by Dehmelt for trapped electrons [8], where the cyclotron energy is dissipated by synchrotron radiation in a strong magnetic field.

We have observed the advantageous effect of collisions on the laser induced fluorescence of a stored cloud of Ba^+ ions. We used the same apparatus as described in references [1,2]. The ions were confined in a trap of 1.3 cm radius placed at the centre of a superconducting solenoid of 3 Tesla field strength. They were excited by a Nitrogen pumped pulsed dye laser of 30 Hz repetition rate. The azimuthal quadrupolar field at $\omega_c = \omega_c^{'} + \omega_m$ (322.8 kHz) was applied between two adjacent segments of the ring electrode, which was divided into 4 quadrants, of which the opposite ones were electrically connected. The laser induced fluorescence after excitation of the $6S_{1/2}$–$6P_{1/2}$ resonance transition (493.4 nm) was observed perpendicular to the exciting laser beam through a mesh endcap electrode at the $6P_{1/2}$–$5D_{3/2}$ transition (649.6 nm). Figure 1 shows the observed spectrum (a) under UHV conditions ($p = 10^{-9}$ mbar) and (b) at 8.3×10^{-7} mbar N_2 background pressure. Both spectra were taken with 1 s dwell time per channel. Because of the laser polarisation only 2 of the possible 4 Zeeman components are visible. A similar result was obtained when we use He as buffer gas, however at a one order of magnitude higher pressure. As the combined result of axialization, which increases the spatial overlap between the ion cloud and the laser beam, and the collisional deexcitation of the metastable D-state we observe an increase of the fluorescence intensity by about one order of magnitude. In the higher pressure range between 10^{-6} and 3×10^{-5} mbar N_2 (10^{-6}–5×10^{-4} mbar for He) we did not observe a significant change of the signal amplitude.

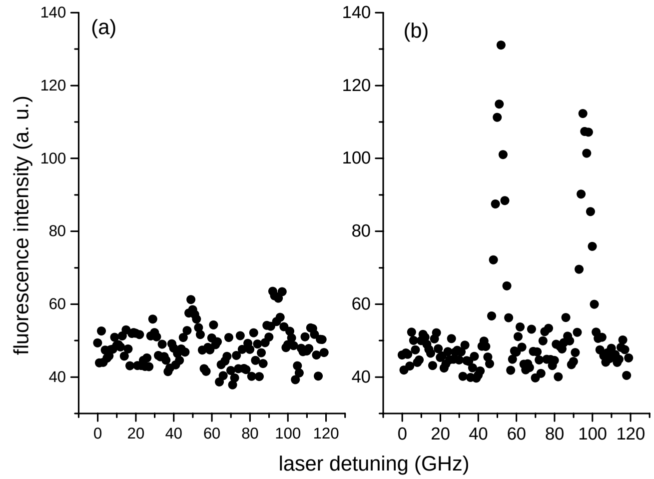

Fig. 1. Laser induced fluorescence from a stored cloud of $^{138}Ba^+$ ions in a Penning trap at 3 Tesla magnetic field strength. Excitation wavelength: 493.4 nm, detection wavelength: 649.6 nm. (a) Spectrum under UHV conditions ($p = 10^{-9}$ mbar); (b) 8.3×10^{-7} mbar N_2 as buffer gas. Because of the laser polarisation only 2 of 4 possible Zeeman components are visible.

As a side effect we found that the storage time of the Ba^+ ion cloud was extended from previously typical 20 min under UHV conditions to more than a few hours at the conditions of axialization. This is an indication of the reduced ion temperature.

3 Relaxation measurements

A possible drawback of collisions for Zeeman spectroscopy could be relaxation processes between the ground state Zeeman levels. In Laser-microwave double resonance experiments for g-factor determination one of the ground state Zeeman levels is depopulated by selective laser excitation. Induced Zeeman transitions are observed by an increase of the fluorescence intensity at resonance. Collision induced relaxation between the Zeeman levels would destroy the optical pumping effect. We have measured the influence of relaxing collisions on the signal strength of an induced Zeeman transition. The laser was tuned to depopulate one of the two magnetic sublevels in the ground state of $^{138}Ba^+$ and microwaves were blown into the trap to induce a $\Delta m_J = 1$ transition. To make sure that we obtain the maximum microwave induced signal strength we

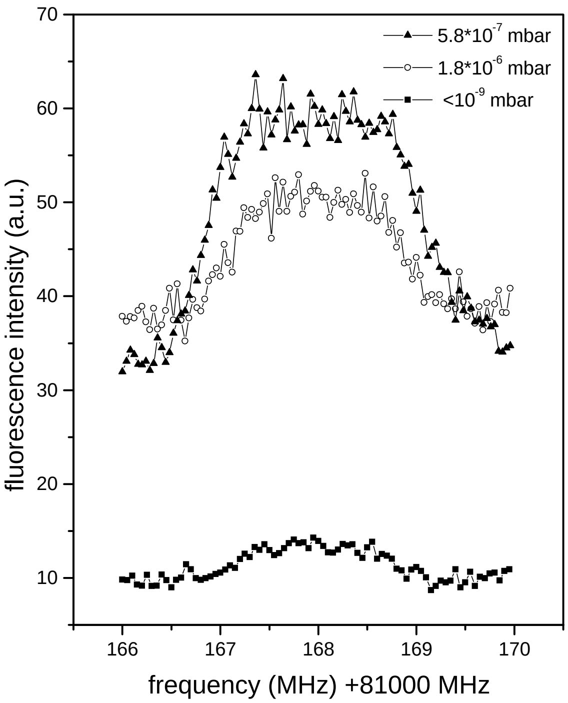

Fig. 2. Microwave induced Zeeman transition in the $6S_{1/2}$ ground state of $^{138}Ba^+$ at different N_2 background pressures. The microwave power was set above saturation level.

set the microwave power well above the saturation level and consequently we obtain a broad spectral line. Figure 2 shows the result of such a measurement. Under UHV conditions the signal is very small because most of the ions are trapped in the metastable 3D state. At 5×10^{-7} mbar of N_2 we observe a strong microwave induced signal at a somewhat increased background. At 1.8×10^{-6} mbar the signal strength does no longer increase while the collisional induced background becomes very significant. We define a signal strength P as the ratio of the microwave induced count rate to the total count rate. Figure 3 shows the measured values of P at different N_2 pressures.

The value of P at different background pressures can be calculated by the solution of the rate equation which describes the population of the different state populations under the influence of laser excitation, microwave radiation and collisions. If we assume microwave saturation between the two ground state Zeeman levels and laser saturation for the 6S−6P excitation at our pulsed laser power of more than 1 kW and include the known branching ratio B for the decay of the excited 6P state into the 6S and 5D states ($B = 2.77$, [9]) we obtain

$$P - 1 = \frac{1 - e^{\gamma_R \tau}}{e^{\gamma_R \tau} - 1 + A + C + C e^{\gamma_Q \tau}(1 - 2e^{\gamma_R \tau})} \cdot \qquad (3)$$

Here γ_Q is the quenching rate of the metastable 5D-level by collisions, $A = (1/3)(B/B+1)$, $C = 1/(B+1)$. τ is the time between two consecutive laser pulses. γ_R is the collisional relaxation rate between the Zeeman levels, which is proportional to the pressure. We take values of γ_Q from reference [4] and make a least squares fit to our data point leaving γ_R as free parameter. Setting

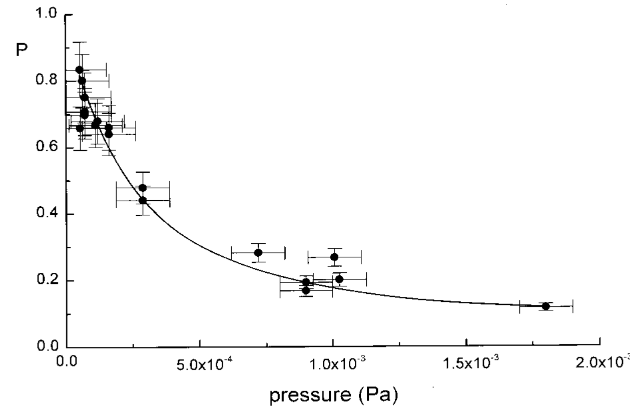

Fig. 3. Signal strength P, defined as ratio of microwave induced count rate to total count rate, at different N_2 Background pressures. The solid line is a least squares fit according to equation (3) with the collisional induced relaxatin rate γ_R as free parameter

$$\gamma_R = \frac{pv\sigma_R}{kT} \qquad (4)$$

where p is the buffer gas pressure, v the relative velocity between ions and neutral atoms at room temperature and σ_R the relaxation cross section, we obtain from the fit

$$\sigma_R(\mathrm{He}) = 5.9(1.5) \times 10^{-18}\ \mathrm{cm}^2$$
$$\sigma_R(\mathrm{N_2}) = 1.3(0.3) \times 10^{-16}\ \mathrm{cm}^2.$$

This is of the same order as the quenching cross sections for the metastable 5D state, which we have determined earlier [4] to

$$\sigma_Q(\mathrm{He}) = 1.4(0.5) \times 10^{-18}\ \mathrm{cm}^2$$
$$\sigma_Q(\mathrm{N_2}) = 2.5(0.6) \times 10^{-16}\ \mathrm{cm}^2.$$

This indicates that quenching and relaxation are competing processes and the pressure range for double resonance experiments has to be chosen carefully to exploit the advantages of increased population density by metastable state quenching without loss of microwave induced signal strength by ground state relaxation. Our results, shown in Figure 2, demonstrates that it is possible to operate the trap under favourable conditions.

4 Zeeman transitions

We have used the increased signal strength under buffer gas collisions to induce ground state Zeeman transitions at low microwave power. Figure 4 shows an example of our measurements. By the combined action of buffer gas cooling and axialization the ions fullfill the Dicke criterion that their oscillation amplitude is smaller than

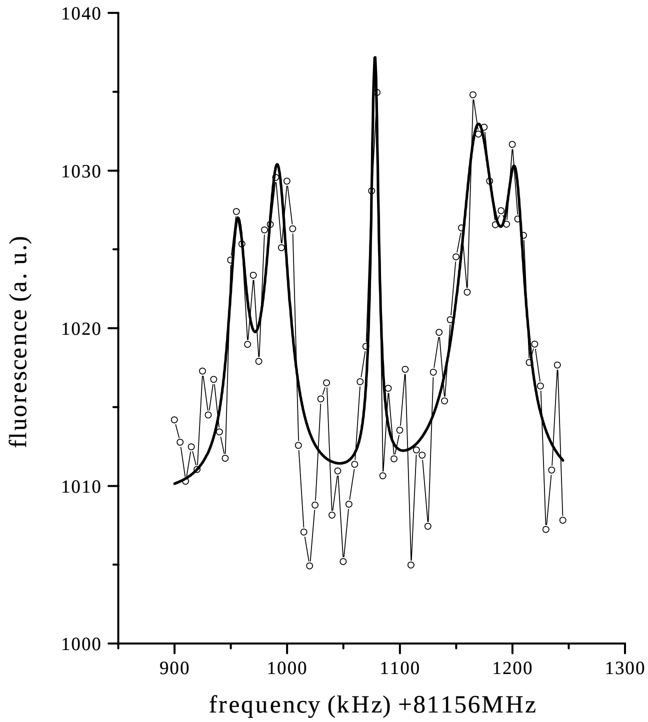

Fig. 4. Microwave induced Zeeman transition in the $6S_{1/2}$ ground state of $^{138}Ba^+$ at low microwave power. The spectrum shows an unshifted carrier and Doppler sidebands at a combination of ion oscillation frequencies. A Lorentzian fit to the central line gave a narrowest observed line width of 3.2 kHz at a total frequency of 81 GHz and a fractional uncertainty of the line centre of 1.2×10^{-9}.

the wavelength of the radiation (3.7 mm at 81 GHz). An unshifted carrier appears at the Zeeman transition frequency and symmetric sidebands at the ions oscillation frequencies. The sidebands which are visible in our spectrum appear at the frequency $\omega_c{}' - \omega_z$, ω_z being the axial oscillation frequency. This sideband has the maximum amplitude if we assume that our trap acts as microwave cavity and the dominant mode is TE_{013} [10,11]. The fluorescence minimum at the centre of the sidebands is due to the high microwave amplitudes [12]. The narrowest line of the central carrier which we have obtained so far has a full width of 3.2 kHz at 81 Ghz and a Lorentzian fit to the data points gives a statistical uncertainty of the line centre to 100 Hz. This corresponds to a fractional uncertainty of only 1.2×10^{-9}. It represents an improvement of more than 2 orders of magnitude compared to our previous measurements [1,2]. If the magnetic field strength at the ions position can be measured with similar precision by a determination of the cyclotron resonance frequency of electrons stored in the same trap we would obtain a very accurate g_J-factor for the ground state of $^{138}Ba^+$. This is not of great interest at present since calculation of the g-factor by including relativistic and radiative corrections is limited to a precision of only 10^{-6} [13]. More interesting may be measurements on odd isotopes of Ba^+. Here the Zeeman transition frequency contains the contribution of the nuclear g-factor, which could be determine with some 10^{-4} relative uncertainty. The extension to unstable isotopes, as successfully demonstrated in Paul traps [14], could give for the first time accurate directly determined values of nuclear magnetic moments of radioactive isotopes.

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