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3d $^2D_{5/2}$ lifetime in laser cooled Ca^+: Influence of cooling laser power

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Abstract. We have measured the lifetime of the metastable $3D_{5/2}$ level in Ca^+ using the "quantum jump" technique on a single stored and laser cooled ion in a linear Paul trap. We found a linear dependence of the measured decay rate on the power of the laser which repumps the ions from the long lived $3D_{3/2}$ level. This can be explained by off-resonant depletion of the $3D_{5/2}$ level. The proper lifetime of this level is obtained by a linear extrapolation of the measured lifetime to zero laser power. We obtain 1100(18) ms in agreement with theoretical calculations. The observed systematic change of the decay rate resolves discrepancies between earlier experiments in which this effect had not been considered. Measurements on a linear chain of 10 laser cooled ions showed unexpected frequent coincidences of quantum jumps within our observation time of 20 ms. This indicates a so far unexplained correlation between the ions in the chain at large distances.

PACS. 32.70.Fw Absolute and relative intensities – 42.50.Vk Mechanical effects of light on atoms, molecules, electrons, and ions – 42.50.Fx Cooperative phenomena; superradiance and superfluorescence

1 Introduction

Metastable atomic states have lifetimes in the range of a few ms up to years [1] because they cannot decay *via* electric dipole transitions to the ground state. Such states are attractive to realize more accurate frequency standards since they provide transitions with narrow linewidth. Experimental problems in lifetime measurements are the sensitivity of these levels to perturbations. Collisional quenching and finestructure mixing can lead to changes in the observed lifetime. The ion trap is an ideal instrument to investigate lifetimes because one can prepare a single particle in a well controlled environment *i.e.* low background gas pressure of typically 10^{-10} mbar, the absence of electrical fields and no collisions with the walls. The storage time of a single particle under these conditions can be as long as month and does not impose any limits on the measurement even for very long lifetimes. It is, however, often difficult to detect weak electric quadrupole or magnetic dipole radiation from the decay of these states. Therefore an indirect detection scheme is applied known as the shelving technique introduced by Dehmelt [2]. For a single ion one can determine the lifetime from the observation of quantum jumps. Yu *et al.* [3] have demonstrated this for the $3D_{5/2}$ state in the Ba^+ ion with a rather long radiative lifetime of 79.8(4.6) s. Similar experiments have been carried out earlier by Bergquist *et al.* [4] on Hg^+, Urabe *et al.* [5] and Ritter *et al.* [6] for a single trapped Ca^+ ion.

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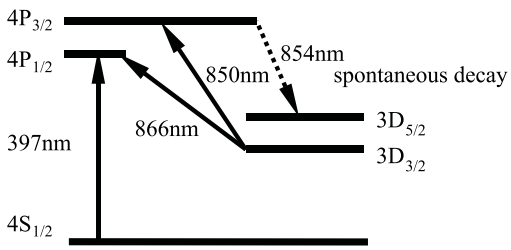

Fig. 1. Partial level scheme of Ca^+.

The typical experimental accuracy is on the percent level for ion trap based lifetime measurements.

In addition to spectroscopic applications the lifetimes of metastable states are of theoretical interest because they provide a good test for calculated wavefunctions and the influence of the interaction between the electrons in the shell can be investigated. The use of advanced approximation models has also lead to very precise theoretical values with an accuracy comparable to experiments.

2 Calcium

The Ca^+ ion has been addressed by numerous groups of theoreticians and experimentalists. It has a lambda shaped level scheme with two low lying metastable 3D states (Fig. 1). From the theoretical point of view Ca^+ is interesting because of the closed shell plus a single

Table 1. Theoretical and experimental values for the lifetime of the $3D_{5/2}$ level.

Experimental lifetimes [s]:	
Urabe [5]	1.08(22)
Ritter *et al.* [6]	0.969(21)
Arbes *et al.* [12]	1.24(39)
Knoop *et al.* [13]	0.994(38)
Gudjons *et al.* [14]	1.064(17)
Lidberg *et al.* [15]	1.09(5)
Arbes *et al.* [17]	1.054(61)
this work	1.100(18)

Theoretical lifetimes [s]:	
Liaw [7]	1.045
Guet *et al.* [8]	1.236
Vaeck *et al.* [9]	1.14
Biemont *et al.* [10]	1.07
Ali *et al.* [11]	0.95
Brage *et al.* [20]	1.16

valence electron. Hence there is no outer correlation which makes calculations easier but core effects already play an important role. The electric field of the valence electron causes a core polarization leading to a modified nuclear electric field seen by the outermost electron. This effect has been treated in different ways using well-known approaches such as the many body perturbation theory (MBPT), multiconfiguration Hartree-Fock (MCHF) or the Brueckner approximation. All the above mentioned methods lead to results differing by about 10% from each other. Regarding the $3D_{5/2}$ level, which is of interest for our experiment, the Brueckner approximation used by Liaw [7] leads to a lifetime of 1.045 s, Guet and Johnson [8] obtain 1.236 s using MBPT, the MCHF based calculation by Vaeck *et al.* [9] leads to a value of 1.14 s. The most recent calculation by Biemont and Zeippen [10] using the Superstructure code program gives 1.07 s. Ali *et al.* [11] report a value of 0.95 s using the multiconfiguration Dirac-Fock method. The comparison between theory and experiment provides a test for the quality of each method.

In recent years several groups have experimentally investigated the lifetime of the $3D_{5/2}$ level in Ca^+ using Paul traps. Arbes *et al.* [12] used a hot stored ion cloud. A similar method was employed by Knoop *et al.* [13]. A laser cooled ion cloud, which reduces systematic effects from interaction with the background gas, was used by Gudjons *et al.* [14]. Quantum jumps on a single laser cooled Ca^+ ion were first employed by Urabe *et al.* [5] and later substantially improved by Ritter *et al.* [6]. Table 1 lists the most accurate experimental values together with the theoretical predictions mentioned above. As can be seen from Table 1, the experimental values differ by several standard deviations of the quoted uncertainties, which makes the comparison of different theoretical models difficult. Although a recent experiment performed on a storage ring [15] supports the values reported in [14] the accuracy is not sufficient to decide between the different trap experiments. In order to resolve the discrepancy and to check for possible systematic shifts of the decay rate we have set up a new experiment, using the shelving technique on a single laser cooled ion.

3 Experiment

We trap a single Ca^+ ion in a linear Paul trap consisting of four cylindrical rods. Each of them is divided into three sections of 15 mm length. The rod diameter is 6 mm and the closest distance between opposing electrodes r_0 is 2.66 mm. A radiofrequency field between diagonal opposing rods provides the radial trapping force while the axial trapping is achieved by an additional DC field applied to the outer segments. A single ion is supposed to be on the field free trap axis. However it can be displaced from this position due to contact potentials. They can arise from the deposition of calcium on the rods when we create the ions by ionizing an atomic beam. We compensate them with additional DC fields that can be applied to each of the rod segments individually. Typically our trapping field has an amplitude of 100 V and the driving field frequency is 2 MHz. The trap is enclosed in a vacuum chamber under UHV conditions (typically 10^{-10} mbar). The pressure is determined using a quadrupole mass spectrometer. The laser radiation for the UV transition is generated by an intracavity frequency doubled Ti:Sa laser. For the IR transitions we used extended cavity diode lasers. The lasers are parallel overlapping and focused into the trap with a slight angle with respect to the trap axis. Brewster windows made of uncoated quartz are used to reduce scattered light. The fluorescence is observed perpendicular to the trap axis. The light is collimated by a lens system inside the vacuum chamber and focused to an intensified CCD camera by a second lens outside. Single ions as well as ion crystals can be observed spatially resolved which has been demonstrated earlier [16]. A sketch of the setup is given in Figure 2.

For the lifetime measurement we prepare a single ion and compensate the residual micromotion. The storage time exceeds one day. Cooling is provided by a red detuned laser at 397 nm. A second laser at 866 nm is needed for repumping from the long lived $3D_{3/2}$ state into which the excited ion may decay. Both lasers run continuously and we observe fluorescence photons at a rate of a few kHz. For a time in the order of 1 second we apply a third laser at the $4P_{3/2}$–$3D_{3/2}$ transition. By decay of the $4P_{3/2}$ level the ion may fall into the metastable $3D_{5/2}$ state and the fluorescence vanishes. After the decay back to the ground state we observe fluorescence again (Fig. 3). For observation we have used a CCD camera instead of a photomultiplier in order to be able to observe heating effects or collisions. These would result in an increase of the micromotion amplitude which can be easily observed. Our camera has

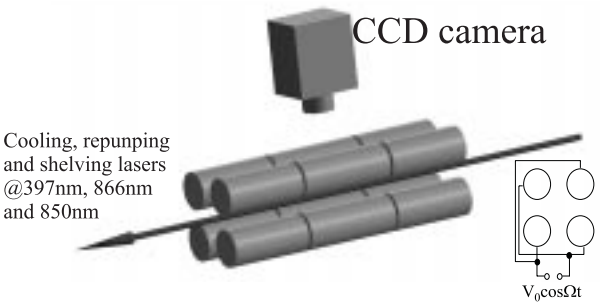

Fig. 2. Experimental setup showing the segmented trap electrodes, the cooling and the repumping lasers parallel to the trap axis and the shelving laser which excites ions to the $4P_{3/2}$ level from where they decay into the metastable $3D_{5/2}$ state.

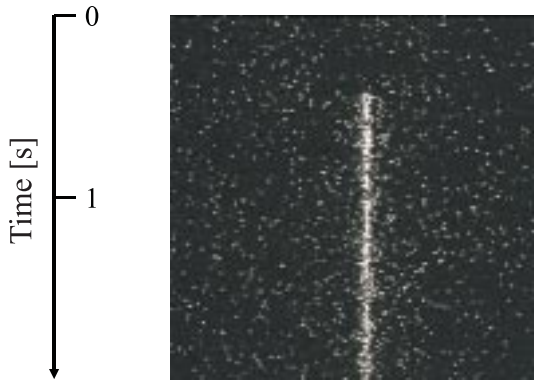

Fig. 3. Picture of a single laser cooled ion taken by a CCD camera. A shelving laser at the $3D_{3/2}$–$4P_{3/2}$ transition pumps the ion into the metastable $3D_{5/2}$ level. At $t = 0$ the shelving laser is turned off and the ion becomes visible when it returns to the cooling cycle by spontaneous decay. Dwell time of the camera is 20 ms.

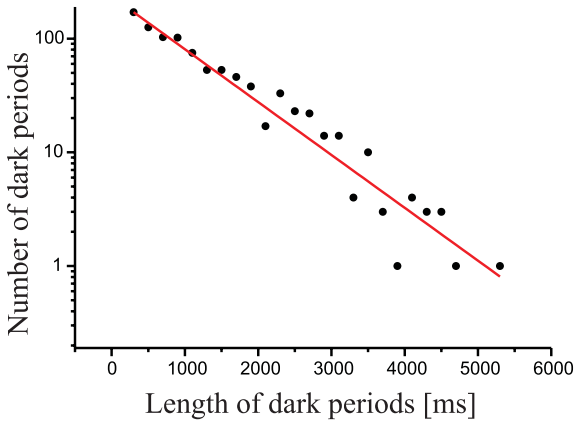

Fig. 4. Histogram of the dark periods for a single ion. The experimental data are fitted by an exponential.

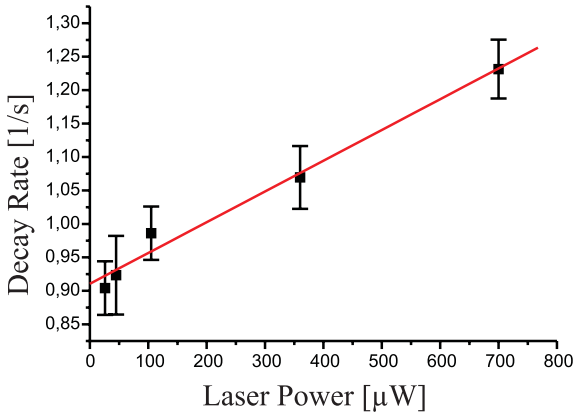

Fig. 5. $3D_{5/2}$ lifetime for a single trapped ion at different powers of the $3D_{3/2}$–$4P_{1/2}$ repumping laser. The radiative lifetime of the $3D_{5/2}$ level (1100(18) ms) is obtained by a linear extrapolation to zero laser power.

a time resolution of 20 ms. One measurement cycle consists of the pumping time into the D level by the shelving laser of 1 second and a readout time of the camera of 5 seconds after blocking the shelving laser. We repeat this sequence many times and we determine the time intervals where no fluorescence is observed after blocking the shelving laser. Several thousand of such measurements have been performed for different laser powers. From the distribution of the dark times we get the lifetime by fitting an exponential decay to the data (Fig. 4).

4 Results and discussion

When we varied the power of the cooling lasers we found a linear dependence of the decay rate on the repumping laser power at 866 nm (Fig. 5). This observation can be explained when we assume that the $3D_{5/2}$ level is depleted to a certain extend by the repumping laser. This may happen off-resonant through the the $4P_{3/2}$–$3D_{5/2}$ transition at 854 nm. The wavelength difference of 12 nm corresponds to 5×10^{-6} of the natural width of this transition, arising from the 7 ns radiative lifetime of the $4P_{3/2}$ level. We focus our repumping laser of nominal 5 mW total output power to a spot size of approximately 100 μm at the position of the ion. This corresponds to about 100 times the saturation intensity of the $4P_{3/2}$–$3D_{3/2}$ line. To estimate the influence of the off-resonant excitation of the decay rate Γ on the $3D_{5/2}$ level we assume that the transition matrix element for the $3D_{5/2}$–$4P_{3/2}$ line is of the same order of magnitude as for the $3D_{3/2}$–$4P_{1/2}$ line. Since the excitation strength reduces with the square of the detuning from resonance, the probability of excitation of the $3D_{5/2}$–$4P_{3/2}$ line by the laser $3D_{3/2}$–$4P_{1/2}$ radiation is approximately by $a = 2 \times 10^9$ times reduced compared to the repumping line. The decay rate Γ of the $3D_{5/2}$ level then is modified by the corresponding admixture of the $4P_{3/2}$ level

$$\begin{aligned}\Gamma &= \Gamma(D_{5/2}) + a\Gamma(P_{3/2}) \\ &= 0.91\ s^{-1} + (2 \times 10^{-9})(1.4 \times 10^8\ s^{-1}) \\ &= 1.19\ s^{-1}. \end{aligned} \quad (1)$$

This gives the order of magnitude for the observed reduction of the lifetime. A more careful analysis would require

a better knowledge of the laser spot size at the ions position as well as a consideration of the spatial laser power distribution over the residual ion's oscillation amplitude. Since the admixture amplitude a is proportional to the laser power, we expect a linear dependence on the laser intensity as it is in fact observed. To check the above given assumption we varied the cooling laser power at 397 nm by more than one order of magnitude and found no variation of the measured decay rate outside the statistical variation. The radiative lifetime of the unperturbed $3D_{5/2}$ level is obtained by a linear extrapolation of the measured decay rates to zero laser power. We obtain a lifetime of

$$\tau = \frac{1}{\Gamma} = 1100(18) \text{ ms}. \quad (2)$$

The quoted error is one standard deviation. The slight discrepancy to our previous result [14] obtained on a laser cooled ion cloud may be easily explained by the fact that the spatial ion cloud distribution was of the same order or even larger than the laser spot size, and the average laser power, experienced by the ions, is smaller than in the case of a single ion. This results in an only small reduction of the lifetime. The result reported in [6] on a single ion could be reproduced when we apply the same laser power as in that experiment. A systematic uncertainty of the measured decay rate by collision induced deexcitation of the $3D_{5/2}$ level into the ground state or the $3D_{3/2}$ finestructure level can be excluded at our level of precision. Calculations of the decay rate using the measured cross sections [12] under our background gas pressure of 10^{-10} mbar and the assumption that the residual gas is H_2, which has the largest cross section of all likely molecules in the background gas, leads to a reduction of the decay rate in the order of 1×10^{-3}. This is one order of magnitude below our statistical uncertainty.

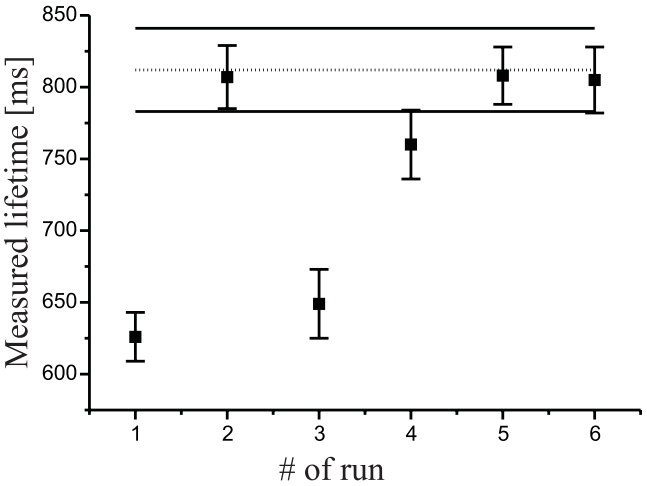

Fig. 6. Lifetime obtained with 10 laser cooled and crystallized ions at a repumping laser power of 700 μW. The picture shows results of different runs under the same experimental conditions. For comparison the lifetime measured for a single ion (horizontal dotted line) and the 1σ standard deviation, obtained at the same repumping laser power, is given.

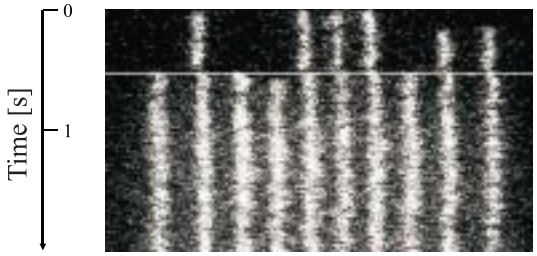

Fig. 7. Picture of 10 ions. Three ions (#1, 3, 8 from left) perform a quantum jump within the same opening time of the camera (20 ms). The horizontal line is given to guide the eye.

5 Measurements in ion chains

The simultaneous observation of several ions by a CCD camera allows to reduce the total data acquisition time and thus reduces the statistical uncertainty. We have attempted to observe quantum jumps in a chain of 10 ions, crystallized and aligned along the trap axis. As expected the statistical error of the $3D_{5/2}$ decay time was smaller as in the case of single ion observation for comparable measuring times. The experiments showed, however, unexpected scatter of the results for different runs under seemingly identical conditions. Trapping parameters, background pressure, laser intensity and laser focal spot size were held constant to the best of our ability. In contrast the results of the lifetime measurements differed for different runs in some cases by as much as 30%, while the 1σ standard deviation for a run was about 5%. Figure 6 shows a typical example. The results are compared with the single ion value for the same power of the repumping laser. While in some cases the decay constants agree within the statistical errors, sometimes substantially shorter lifetimes have been obtained. In the latter cases we found that quantum jumps for different ions in a chain appeared simultaneously within the dwell time of 20 ms of our CCD camera. Two- or three-fold coincidences appeared much more frequently than expected statistically. Figure 7 shows a typical example were three ions performed a quantum jump in the same 20 ms time interval. As evident from this example the coincidences were not restricted to adjacent ions but often appeared at large distances within the chain. A similar observation of frequently coinciding quantum jumps has been reported previously by Toschek and coworkers [18]. This observation indicates a long range interaction between the ions in the linear crystal. This is somewhat surprising considering the large interionic distance of about 10 μm in the chain. Superfluorescence as observed in a two ion crystal [19] can be excluded because it plays a significant role only at an ion distance of the order of $\lambda/2$ of the decay photons (729 nm). Additional measurements using different ion numbers in the chain are required to allow better insight into a possible interaction mechanism. Since we cannot explain our observation at this moment we do not include the measurements on the linear ion chain in our result for the $3D_{5/2}$ lifetime.

6 Conclusion

The lifetime of the $3d\ ^2D_{5/2}$ state of Ca^+ has been measured with an uncertainty of 1.5%. We believe that we have reduced discrepancies between previous measurements by proper consideration of off-resonant depletion of the $D_{5/2}$ state by the repumping laser, which is needed for the "electron shelving" method on a single stored and cooled ion. Further improvement would be possible when the data acquisition time is increased. Then all experimental conditions have to be kept constant over a time of a few hours. This may be necessary when further progress in calculation methods leads to improved theoretical values for the lifetime.

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References

1. M. Roberts, P. Taylor, G.P. Barwood, P. Gill, H. Klein, W.R.C. Rowley, Phys. Rev. Lett. **78**, 1876 (1997).
2. H.G. Dehmelt, Bull. Am. Phys. Soc. **20**, 60 (1975).
3. N. Yu, W. Nagourney, H.G. Dehmelt, Phys. Rev. Lett. **78**, 4898 (1997).
4. J.C. Bergquist, R. Hulet, W.M. Itano, D.J. Wineland, Phys. Rev. Lett. **57**, 1699 (1986).
5. S. Urabe, K. Hayasaka, M. Watanabe, H. Imajo, R. Ohmukai, R. Hayashi, Appl. Phys. B **57**, 367 (1993).
6. G. Ritter, U. Eichmann, J. Phys. B **30**, L141 (1997).
7. S. Liaw, Phys. Rev. A **51**, R1723 (1995).
8. C. Guet, W.R. Johnson, Phys. Rev. A **37**, 307 (1988).
9. N. Vaeck, M. Godefroid, C. Froese Fischer, Phys. Rev. A **46**, 3704 (1992).
10. E. Biemont, C.J. Zeippen, Comm. At. Mol. Phys. **33**, 29 (1996).
11. M.A. Ali, Y. Kim, Phys. Rev. A **38**, 3992 (1988).
12. F. Arbes, T. Gudjons, F. Kurth, G. Werth, F. Marin, M. Inguscio, Z. Phys. D **25**, 295 (1993).
13. M. Knoop, M. Vedel, F. Vedel, Phys. Rev. A **52**, 3763 (1995).
14. T. Gudjons, B. Hilbert, P. Seibert, G.Werth, Europhys. Lett. **33**, 595 (1996).
15. J. Lidberg, A. Al-Khalili, L.-O. Norlin, P. Royen, X. Tordoir, S. Mannervik, J. Phys. B **32**, 757 (1999).
16. W. Alt, M. Block, P. Seibert, G. Werth, Phys. Rev. A **58**, R23 (1998).
17. F. Arbes, M. Benzing, T. Gudjons, F. Kurth, G. Werth, Z. Phys. D **29**, 159 (1994).
18. T. Sauter, R. Blatt, W. Neuhauser, P.E. Toschek, Opt. Commun. **60**, 287 (1986).
19. R.G. DeVoe, R.G. Brewer, Phys. Rev. Lett. **76**, 2049 (1996).
20. T. Brage, C. Froese Fischer, N. Vaeck, M. Godefroid, A. Hibbert, Phys. Scripta T **48**, 533 (1993).