Measurement of the $3s3p\ ^3P_1$ lifetime in magnesium using a magneto-optical trap

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We demonstrate an accurate method for measuring the lifetime of long-lived metastable magnetic states using a magneto-optical trap (MOT). Through optical pumping, the metastable $(3s3p)\ ^3P_1$ level is populated in a standard MOT. During the optical pumping process, a fraction of the population is captured in the magnetic quadrupole field of the MOT. When the metastable atoms decay to the $(3s^2)\ ^1S_0$ ground state they are recaptured into the MOT. In this system no alternative cascading transition is possible. The lifetime of the metastable level is measured directly as an exponential load time of the MOT. We have experimentally tested our method by measuring the lifetime of the $(3s3p)\ ^3P_1$ of ^{24}Mg. This lifetime has been measured numerous times previously, but with quite different results. Using our method we find the $(3s3p)\ ^3P_1$ lifetime to be (4.4 ± 0.2) ms. Theoretical values point toward a lower value for the lifetime.

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I. INTRODUCTION

In many spectroscopic experiments, beam line and cell measurements have played an important role, however, the workhorse of today is atom traps such as the magneto-optical trap (MOT), magnetic traps or various types of dipole traps. In these traps large numbers of atoms or molecules (10^9-10^{12}) can be stored with lifetimes exceeding 300 s at submilli-K temperatures [1–7]. Following this development a large amount of new high precision spectroscopic data on atoms and molecules has emerged. Parallel to these achievements theory has been developing new accurate models of atoms and molecules and single species and mixed species [8,9].

Compared to alkali elements collision and spectroscopic properties of the alkaline earth elements are less understood. In particular the lack of high precision radiative data prevents decisive conclusions on new accurate *ab initio* many-body calculations and models for two electron atoms [10]. Pioneering experiments on neutral Ca and Sr have provided new accurate data of the $^1P_1$, $^3P_1$, and $^3P_2$ lifetimes down to a 1% level [11–14]. These methods and the method presented in this paper are to some extent a variation of the well-known electron shelving experiments carried out with ions [15,16]. With photoassociation studies, lifetime measurements have greatly improved, yet the results are somewhat model dependent. Here we demonstrate the use a MOT for precision measurements of long-lived magnetic metastable lifetimes, by studying the metastable $(3s3p)\ ^3P_1$ state of ^{24}Mg. For this state we only consider decay to the ground state as the 0.5 mHz transition $(3s3p)\ ^3P_1 \rightarrow (3s3p)\ ^3P_0$ is strongly forbidden and suppressed. The lifetime of the $(3s3p)\ ^3P_1$ state has been measured several times, however, the various experimental results differ significantly and range from 4 ms to about 5 ms for the most recent measurement in 1992 [17–20]. Theoretical calculations support a value in the range 2.8 to 3.6 ms [10,21–23], however, not all theories were targeting high precision for this particular lifetime.

II. THEORY OF MEASUREMENT

In Fig. 1, we show the relevant energy levels and transitions for our experiment. The 285 nm transition is used for cooling the atoms, while the 457 nm transition is used to populate the metastable $(3s3p)\ ^3P_1$ state. Having cooled the atoms close to the Doppler temperature limit of approximately 2 mK, we expose the atoms to a millisecond pulse of 457 nm light. During this pulse most of the cold ensemble is transferred to the $(3s3p)\ ^3P_1$ state. Statistically about 1/3 of these can be transferred into the magnetically trapped $^3P_1$ state $|J=1, m_j=+1\rangle$ and held by the MOT quadrupole magnetic field. Experimentally, we transfer as much as 85% of the ground state by aligning the MOT to a point of nonzero magnetic field parallel to the propagation direction of the 457 nm. In this case optical pumping with circularly polarized light will only populate one of the magnetic substates $|J=1, m_j=\pm 1\rangle$ depending on the helicity of the 457 nm light and the direction of the magnetic field.

To model our system we have solved the optical Bloch equations for the three level scheme shown in Fig. 1. Special care must be taken in solving the equations numerically as the difference in the two upper state lifetimes differ by about a factor of 1000000. Figure 2 displays the calculated 285 nm fluorescence, monitored in the experiment, as a function of time. In Fig. 2(A) we have normal MOT operation, whereas in Fig. 2(B) we flash the MOT with the 457 nm pulse, and prepare a steady state population in the $(3s3p)\ ^3P_1$ state. It

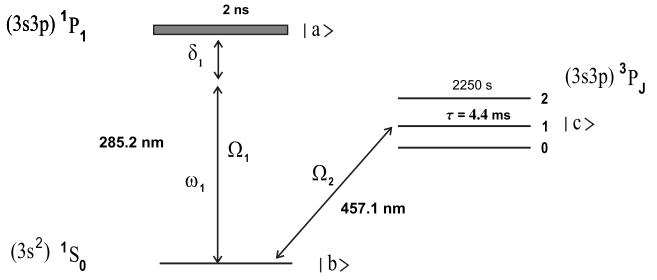

FIG. 1. Energy levels relevant for the lifetime measurement of the metastable $(3s3p)\ ^3P_1$ state of the 24 magnesium isotope. The involved levels are labeled $|a\rangle$, $|b\rangle$, and $|c\rangle$ respectively.

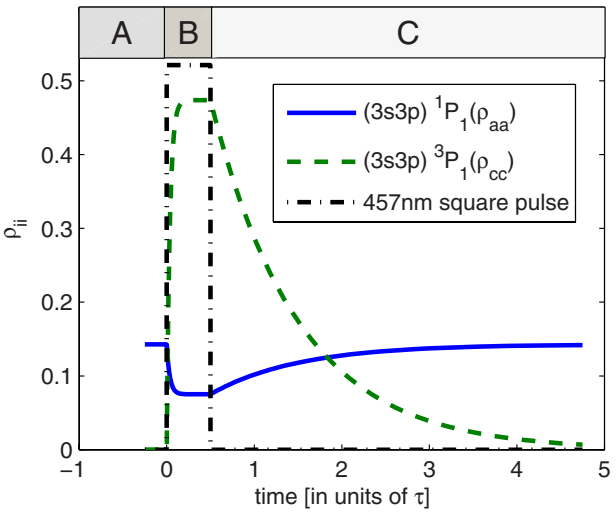

FIG. 2. (Color online) The number of atoms in the excited $(3s3p)\ ^1P_1$ (ρ_{aa}) and $(3s3p)\ ^3P_1$ (ρ_{cc}) states during the pulsed experiment. Time is in units of τ. The signal monitored in our experiment is proportional to the fluorescence determined by (ρ_{aa}).

can be shown that the 285 nm fluorescence in Fig. 2(B) decays exponentially. The time constant of this decay is controlled by the Rabi frequencies Ω_1 and Ω_2 as well as the lifetimes of the $(3s3p)\ ^1P_1$ and $(3s3p)\ ^3P_1$ states,with 2 ns and a few ms, respectively. In principle this may be used to obtain the lifetime of the $(3s3p)\ ^1P_1$ state as Fig. 2(C) yields the metastable lifetime. Practically determining the lifetime in this way is difficult as it requires precise knowledge of the involved Rabi frequencies, i.e., laser intensities at the MOT region.

In Fig. 2(C) the 285 nm fluorescence signal rises exponentially according to

$$S(t) = N_0(1 - e^{-t/\tau}), \tag{1}$$

with a time constant of the metastable lifetime τ. In the experiment $(3s3p)\ ^3P_1$ atoms decaying to the $(3s^2)\ ^1S_0$ ground state are recaptured in the MOT. Here we monitor them as they emit fluorescence at 285 nm. Three important time scales control the dynamics of the system, the metastable state lifetime of about 4 ms, the MOT lifetime 4 s and finally the magnetic trap lifetime of more than 20 s. However, the time scale of interest to this experiment is only about 40 ms $(\sim 10\tau)$, and contributions to the signal from a finite magnetic trap lifetime may safely be disregarded. By including the linear losses from the MOT due to collisions with background atoms, photoionization [24], etc., here described by α, we obtain

$$\dot{N} = \frac{N_0}{\tau}(e^{-t/\tau}) - \alpha N(t), \tag{2}$$

where N_0 is the number of metastable atoms trapped in the magnetic trap. The solution of Eq. (2) becomes

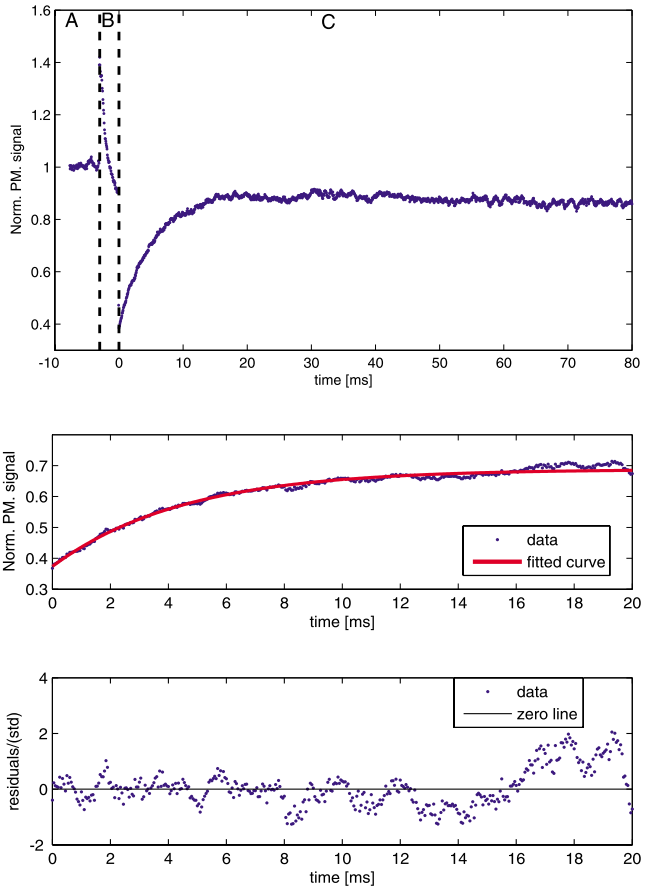

FIG. 3. (Color online) Typical MOT fluorescence measurement, here normalized to the uv power level. The interval marked with dashed lines show the 457 nm pulse. As the 457 nm light is turned on additional background photons are observed. After a load period of about 10 ms the signal is slowly decays due to the 4–5 s MOT lifetime. Lower two graphs: the red curve is a fit to Eq. (3), giving 4.4 ms. The lower part shows the residuals of the fit.

$$N(t) = \frac{\gamma_{cc}}{\gamma_{cc} - \alpha} N_0(e^{-\alpha t} - e^{-\gamma_{cc} t}), \tag{3}$$

with $\gamma_{cc} = 1/\tau$. As γ_{cc} is a factor 1000 larger than α we neglect linear losses of the magnetic trapped atoms during the time interval of the measurement $\sim 10\tau$. For time scales longer than 40 ms this is no longer true as seen in Fig. 3.

III. EXPERIMENTAL SETUP

The main part of the experimental setup has been described in Ref. [25]. A standard magneto-optical trap is intersected by a circularly polarized 457 nm light beam. The beam is overlapped with a MOT beam in the plane perpendicular to the MOT coil symmetry axis. An amplified 914 nm diode is used for generation of 457 nm light by frequency doubling in $KNbO_3$ in a standard four mirror cavity [26]. The 914 nm diode is antireflection coated with feedback from an external grating in a Littrow configuration, and it provides an output of approximately 40 mW. The amplifier consist of an AR coated broad-area laser diode (BAL). Normally we have

about 350 mW output from the 914 nm system and 125 mW of 457 nm light. The 457 nm laser is free running during the experiment (effective linewidth 1 MHz) and is tuned to resonance manually by minimizing the 285 nm fluorescence. In free running mode the 457 nm laser is stable for minutes, however, it is nevertheless tuned before each individual decay experiment. The combined power broadening of 457 and 285 nm transitions, added with the Doppler effect, makes the metastable state easily populated without the need for a narrow-linewidth 457 nm laser. In order to control the pulse of the blue light, the beam is sent through an AOM. The 457 nm beam has a diameter comparable to the UV beams (1.5 mm) used for the MOT and is sent through one of the UV mirrors overlapping the UV beam. Experiments were performed for both a single and a retro reflected beam, with no detectable difference on the outcome.

The experiment is fully controlled by computer using a sequence program in LABVIEW. Our main parameters for this experiment include: 457 nm pulse time 0.1–5 ms, coil current 100–200 A, corresponding to a coil symmetry gradient of 108–216 Gauss/cm. Just before the blue 457 nm pulse is sent we close the atomic beam shutter which remains closed during period C to prevent loading from the thermal beam source. Typically 10^7 atoms are captured in the MOT (load rate 3×10^6 s^{-1}), with a rms diameter of 1 mm and temperatures from 3–5 mK. The Doppler cooling limit for the MOT is 2 mK as sub-Doppler cooling is not supported in this system.

In our experiment, the $|J=1,m_j=+1\rangle$ atoms are captured in the MOT magnetic quadrupole field. The Zeeman energy shift yields $g_j\mu_B m_j/k_B=100.8$ μK/Gauss ($g_j=3/2$) giving 1.7 mK/mm at 172.8 Gauss/cm, our typical operating gradient on the coil symmetry axis. As the MOT temperature is 3–5 mK and the laser beam diameter a 3 mm ($1/e^2$), most $|J=1,m_j=+1\rangle$ atoms remain trapped within the MOT volume.

IV. EXPERIMENTAL RESULTS AND DISCUSSION

In Fig. 3 we present a typical experimental result with MOT operation, 457 nm flash, and reload clearly visible. Small oscillations in the fluorescence signal are visible and are due to a poor servo loop of the UV cavity. To minimize these oscillations we divide the observed fluorescence signal with a normalizing power signal taken from one of the uv beams. The dashed lines show when the 457 nm pulse is on. During this period we observe an overall increased signal due to scattered 457 nm photons on vacuum windows, etc., not being coated for this wavelength. In the reload period we recover 85% of the atoms. The last 15% may be lost due to imperfect polarization of the 457 nm light or possibly if the MOT temperature is outside our recapture range. Hot atoms trapped magnetically may decay outside the MOT capture region defined by the laser beams, here 3 mm. Our maximal range is 5.1 mK/mm (2.6 mK/mm perpendicular to the symmetry axis of the coils) and barely sufficient for a 5 mK MOT temperature. After about 10 ms we notice a weak decay of the reloaded signal. This is due to linear loss mechanisms reducing the MOT lifetime to 4–5 s, limited mainly by two photon ionization induced by the 285 nm MOT light. The lifetime of the magnetic trap is estimated to be about 20 s at our present background pressure of 10^{-9} mbar.

Figure 3, lower panel shows data from the reload period. The red curve is Eq. (3) fitted to the data giving $\tau=4.4$ ms. A total of 80 measurements were taken at different settings, exploring systematic effects (magnetic field gradient, power of the 457 nm pulse, pulse duration and polarizations) and maximum signal-to-noise ratio and recapture fraction. The measurements at maximum field gradient 200 Gauss/cm gave the best signal to noise ratio and only these 34 measurements are used in the quoted lifetime. Our measurements gave a final value of (4.4 ± 0.2) ms here quoted with the statistical uncertainty being the standard deviation divided $\sqrt{N}$, N=34. We found the standard deviation of 0.9 ms for the 34 measurements and thus an uncertainty of 0.16 ms, here rounded to 0.2 ms.

Looking at the fit in Fig. 3 middle panel, we would expect a better uncertainty than the quoted 0.2 ms. However, noise fluctuations on the same time scale as the lifetime will give shot to shot fluctuations. This could be caused by fluctuations in the laser intensity, but we rather attribute this effect to the dynamics of the atoms in the trap, however, indirectly linked to the laser intensity fluctuations. Atoms exited to $|J=1,m_j=+1\rangle$ state, experiencing an attractive potential, would perform an oscillating motion with a characteristic frequency determined by the gradient of the magnetic field and the initial temperature of the MOT. The latter is sensitive to intensity fluctuation of the laser. Simple 1D numerical simulations show, that for our setup with an estimated MOT temperature of about 3–5 mK corresponding to a mean velocity of close to 2 m/s, this would induce oscillations in the observed fluorescence peaking on time scales close to the lifetime. The amplitude of the oscillations is proportional to the number of atoms in the $|J=1,m_j=+1\rangle$, which explains why these oscillations are not observed at times much later than the lifetime. Improving the uv intensity stability by a lock, servoed using, e.g., an AOM, could to some extend lower the MOT temperature to improve the accuracy of the measurements.

In Table I we have collected experimental and theoretical investigations of the $(3s3p)\ ^3P_1$ lifetime. Experimentally, there is a good agreement between the different methods. The latest experimental value [17] seems somewhat higher compared to the present work, but still overlapping at the 1σ level. More recent theoretical calculations point toward a lower value for the lifetime. The difference seems not to be linked to a particular experimental or theoretical method, but appear as a general trend. Possible errors could be caused by accidental resonance to other excited states. For the magnesium energy levels no resonant transitions is present at half or double of the 457 nm wavelength. Closest we find $3s4p\ ^1P_1\rightarrow3s10d\ ^1D_2$ at 901.6 nm and $2p^63s^2\ ^1S_0\rightarrow3s4p\ ^1P_1$ at 202.6 nm which both can be ruled out here. Another possible mechanism could be magnetic field induced mixing to near by P states. Using first order perturbation theory we write the $^3P_1$ state in terms of $^1P_1$ and ^3P_J contributions [27]

TABLE I. Collection of most recent calculated and measured $(3s3p)\ ^3P_1$ lifetimes.

Theoretical			Experimental		
Reference	Year	τ [ms]	Reference	Year	τ [ms]
[21]	2004	2.8(1)	This work	2006	4.4(2)
[22]	2002	3.8	[17]	1992	5.3(7)
[10]	2001	3.6	[18]	1982	4.8(8)
[23]	1979	4.60(4)	[19]	1975	4.5(5)
			[20]	1975	4.0(2)

$$|^3P_1'\rangle = \alpha|^3P_0\rangle + \beta|^3P_1\rangle + \gamma|^3P_2\rangle + \delta|^1P_1\rangle. \quad (4)$$

Evaluating the involved matrix elements gives $\alpha = (3/2)^{1/2}\mu_B B/\hbar\Delta_0$, $\beta = 1$, $\gamma = (3/2)^{1/2}\mu_B B/\hbar\Delta_1$, and $\delta = (6)^{1/2}\mu_B B/\hbar\Delta_3$. Here Δ_0 is the energy difference $^3P_0$-$^3P_1$, Δ_1 is the energy difference $^3P_1$-$^3P_2$, etc, μ_B is the Bohr magneton and B the magnetic field. The Δ values are $\Delta_0 = 600$ GHz, $\Delta_1 = 1200$ GHz, and $\Delta_2 = 395.3$ THz, see Fig. 1. However, our magnetic field is below 22 Gauss within the atom cloud giving coefficients of magnitude 10^{-8} to 10^{-10} and the mixing effect can be neglected here.

V. CONCLUSION

In conclusion we have measured the metastable II4 magnesium $(3s3p)\ ^3P_1$ lifetime using a magneto-optical trap by loading the metastable atoms into a magnetic quadrupole trap. Our value (4.4 ± 0.2) ms is the most accurate to date and in good agreement with recent experimental work on an 1σ level. Theoretically, several recent calculations point toward a significantly lower value and the general discrepancy between theory and experiment is presently not understood.

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[1] R. Grimm, M. Weidemuller, and Y. B. Ovchinnikov, Adv. At., Mol., Opt. Phys. **42**, 95 (2000).
[2] J. Doyle, B. Friedrich, R. V. Krems, and F. Masnou-Seeuws, Eur. Phys. J. D **31**, 149 (2004).
[3] J. D. Weinstein, R. deCarvalho, T. Guillet, Br. Friedrich, and J. M. Doyle, Nature (London) **395**, 921 (1998).
[4] D. Egorov, W. C. Campbell, B. Friedrich, S. E. Maxwell, E. Tsikata, L. D. van Buuren, and J. M. Doyle, Eur. Phys. J. D **31**, 307 (2004).
[5] S. V. Nguyen, S. Doret, C. B. Connolly, R. A. Michniak, W. Ketterle, and J. Doyle, Phys. Rev. A **72**, 060703(R) (2005).
[6] H. L. Bethlem, G. Berden, and G. Meijer, Phys. Rev. Lett. **83**, 1558 (1999).
[7] K. M. R. van der Stam, E. D. van Ooijen, R. Meppelink, J. M. Vogels, and P. van der Straten, Rev. Sci. Instrum. **78**, 013102 (2007).
[8] J. Weiner, V. S. Bagnato, S. Zilio, and P. S. Julienne, Rev. Mod. Phys. **71**, 1 (1999).
[9] K. M. Jones, E. Tiesinga, P. D. Lett, and P. S. Julienne, Rev. Mod. Phys. **78**, 1041 (2006).
[10] S. G. Porsev, M. G. Kozlov, Yu. G. Rakhlina, and A. Derevianko, Phys. Rev. A **64**, 012508 (2001).
[11] G. Zinner, T. Binnewies, F. Riehle, and E. Tiemann, Phys. Rev. Lett. **85**, 2292 (2000).
[12] M. Yasuda and H. Katori, Phys. Rev. Lett. **92**, 153004 (2004).
[13] P. G. Mickelson, Y. N. Martinez, A. D. Saenz, S. B. Nagel, Y. C. Chen, T. C. Killian, P. Pellegrini, and R. Cote, Phys. Rev. Lett. **95**, 223002 (2005).
[14] T. Zelevinsky, M. M. Boyd, A. D. Ludlow, T. Ido, J. Ye, R. Ciurylo, P. Naidon, and P. S. Julienne, Phys. Rev. Lett. **96**, 203201 (2006).
[15] W. Nagourney, J. Sandberg, and H. Dehmelt, Phys. Rev. Lett. **56**, 2797 (1986).
[16] J. C. Bergquist, R. G. Hulet, W. M. Itano, and D. J. Wineland, Phys. Rev. Lett. **57**, 1699 (1986).
[17] A. Godone and C. Novero, Phys. Rev. A **45**, 1717 (1992).
[18] H. S. Kwong, P. L. Smith, and W. H. Parkinson, Phys. Rev. A **25**, 2629 (1982).
[19] P. S. Furcinitti, J. J. Wright, and L. C. Balling, Phys. Rev. A **12**, 1123 (1975).
[20] C. J. Mitchell, J. Phys. B **8**, 25 (1975).
[21] R. Santra, K. V. Christ, and C. H. Greene, Phys. Rev. A **69**, 042510 (2004).
[22] I. M. Savukov and W. R. Johnson, Phys. Rev. A **65**, 042503 (2002).
[23] C. Laughlin and G. A. Victor, Astrophys. J. **234**, 407 (1979).
[24] D. N. Madsen and J. W. Thomsen, J. Phys. B **33**, 4981 (2000).
[25] F. Y. Loo, A. Brusch, S. Sauge, M. Allegrini, E. Arimondo, N. Andersen, and J. W. Thomsen, J. Opt. B: Quantum Semiclassical Opt. **6**, 81 (2004).
[26] V. Ruseva and J. Hald, Appl. Opt. **42**, 5500 (2003).
[27] I. I. Sobelman, *Atomic Spectra and Radiative Transitions* (Springer-Verlag, Berlin, 1992).