

A linear Paul trap for radiative lifetime measurements on ions

M. Schultz-Johanning, R. Schnabel, and M. Kock^a

Institut für Atom- und Molekülphysik, Abt. Plasmaphysik, Universität Hannover, Callinstr. 38, 30167 Hannover, Germany

Received: 28 May 1998 / Revised: 10 September 1998 / Accepted: 26 October 1998

Abstract. A linear ion trap has been designed for collimation and storage of ions from an effusive particle beam generated in a hollow cathode discharge. The motivation is to measure radiative lifetimes on multiply ionized atoms by using the time-resolved laser-induced fluorescence technique. The trap has been tested on iron and tungsten ions both as an ion guide and also as a storing device. Radiative lifetimes have been measured in the trap with a remarkably higher signal-to-noise ratio than in the original effusive atom beam.

PACS. 32.50.+d Fluorescence; phosphorescence (including quenching) – 32.80.Pj Optical cooling of atoms; trapping

1 Introduction

There is still a growing need for transition probabilities of lines of ionized atoms in astrophysics because of the interpretation of satellite-borne UV and vacuum-UV spectra, and in fusion research because of the investigation and handling of plasma impurities like tungsten.

The standard technique for obtaining reliable transition probabilities is the combination of lifetime measurements with branching ratios from the corresponding level [1,2]. The lifetimes of excited atomic or ionic states are preferably measured with the time-resolved laser-induced fluorescence (TRLIF) technique provided the number of atoms or ions in the interaction volume is sufficiently high in order to obtain decay curves with a good signal-to-noise ratio.

We use a hollow cathode discharge for the sputtering of the metal component into the plasma and extracting an effusive atomic beam into the low-pressure measuring chamber. In this procedure the number of ions in the interaction volume is small per se and is further reduced by the divergence of the beam. Our attempt to measure lifetimes especially on W II levels therefore failed.

In the present paper we show how to overcome this problem. The small number of ions present in the beam are confined in an ion guide or accumulated in an ion trap. The stored ions themselves can be further ionized by laser radiation or by an electron beam for lifetime measurements on multiply ionized atoms.

We developed a linear Paul trap which in conjunction with collisional cooling performs as an ion guide in its simplest mode of operation. The quadrupole has also been used as a real trap by limiting the axial movement of the

ions. With such a trap we have already measured successfully lifetimes on W II and Fe II.

2 A linear Paul trap: Model and design

In the fifties Paul [3] developed an ion trap in which the charged particles could be stabilized by a high-frequency electric field [4]. The stabilization of the charged particles in an rf field oscillating with angular frequency ω can be understood with the principle of an effective potential. In this adiabatic approximation the motion of an ion can be described as split into a drift or secular part and an oscillatory part. Since the oscillatory part is solely given by the rf field, the drift is composed of the static electric field and a local variation in the field strength of the rf field which is also called field-gradient force. The latter part propels the ions into the region of lower ac field strength and is responsible for the stabilizing property of an ion trap [5–7].

In linear quadrupoles the ion stability can best be understood by analytically solving the equations of motion (Mathieus equation). However, if we replace the hyperbolic electrodes by cylindrical rods as shown in Figure 1 there are no longer analytical solutions. For a general form of the electrodes the potential and the trajectories have to be calculated numerically. For the potential the procedure of successive over-relaxation [8] or an approximation by line charges [6] can be applied. The trajectories can be calculated with the well-known Runge-Kutta method (see Ref. [8], p. 710).

The stability characteristics are no longer independent of the initial values, they have to be calculated separately for each trajectory. The quadratic approximation of the potential around the origin, however, allows furthermore

^a e-mail: kock@pmp.uni-hannover.de

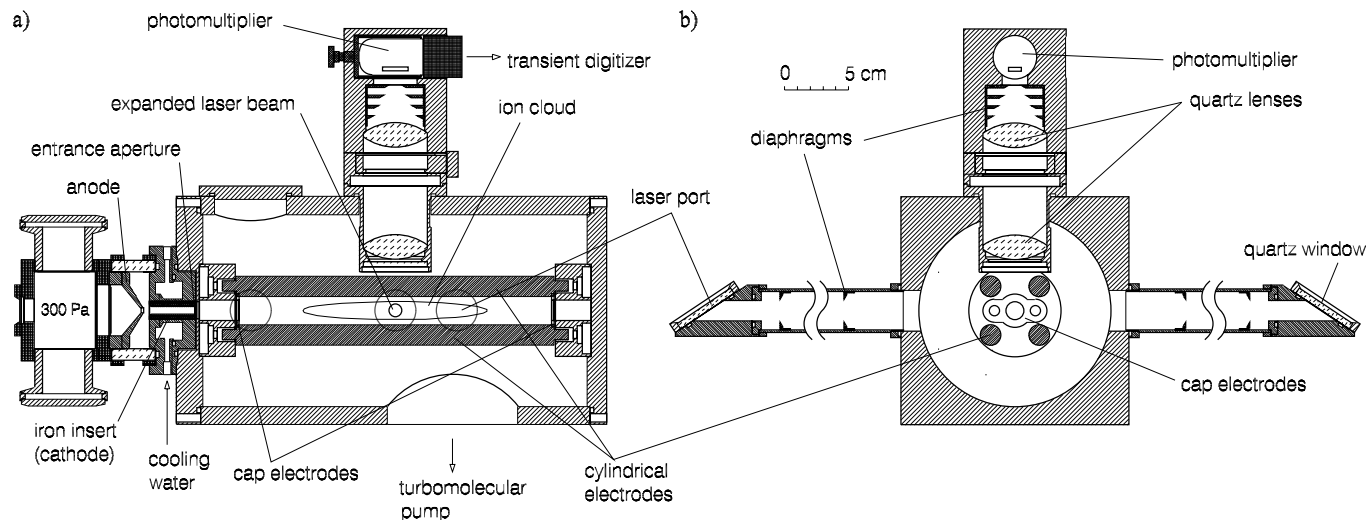


Fig. 1. LIF Apparatus. (a) Cross-sectional view of the linear Paul trap flanged to the hollow cathode. The fluorescence photons are imaged perpendicular to the laser beam onto a photomultiplier tube. Three laser ports can be seen. (b) View from the front of the apparatus.

a calculation of the stability in a way analogous to the ideal quadrupole. That has been done for tungsten and iron as the elements to be stored.

Linear Paul traps have been described elsewhere [9–11]. Our ion trap consists of four copper-made cylindrical rods as guiding electrodes and two cap electrodes at the ends of the trap [12]. The actual electrode configuration is given in Figure 1. The rods have a length of 300 mm, a diameter of 16 mm and a spacing between the surfaces of 38 mm diagonally. We applied a large rod spacing in contrast to the ideal case ([5], p. 117), in order to optimize our light gathering optics. The housing is made of aluminium. Through ports laser radiation can be directed into the ion cloud. The LIF photons are imaged onto a fast photomultiplier. Scattered light has been carefully excluded by means of a stack of diaphragms and by graphiting all aluminium parts. All insulators are concealed so that no ions from the discharge or from the ion cloud can hit them and hence falsify the trap potential.

The two cap electrodes limit the axial extent of the ion trajectories whereby the ions reach the trap through an orifice in one of the electrodes. The discharge potentials can be screened by a fine wire mesh set into the openings.

The trap electronics mainly consists of the power supply for the ac voltage of the quadrupole electrodes and dc sources for the end caps. A sinus generator supplies ac voltages of less than 0.5 V at frequencies up to 800 kHz. This signal is amplified to $48 V_{eff}$ at a 120 W output power. A transformer supplies the ac signals of up to 500 V to the rod electrodes. If only a specific ion species is wanted a dc voltage has to be superimposed [6].

When applying an rf voltage to the rod electrodes the trap operates as an ion guide. A real trapping of ions from the discharge can only be achieved with additional dc voltages applied to the cap electrodes. If the ions are not generated in the trap itself cooling is a prerequisite.

3 Experimental arrangement

The linear Paul trap is one part of our time-resolved laser-induced fluorescence (TRLIF) experiment as described elsewhere [13,19]. The tungsten atoms and ions are generated in a modified version of our high-current hollow cathode [15] shown in Figure 1. The cylindrical cathode insert of 35 mm length and a central bore of 6 mm is sealed off to the low-pressure side by a 0.3 mm aperture. Within the discharge the buffer gas pressure is typically 300 Pa so that the trap is filled continuously through the aperture by a pressure gradient. The discharge is operated in neon with currents between 0.3 A and 1 A. Neon is also used as a cooling gas fed in through a separate gas inlet.

A Quanta Ray DCR 11–3 Nd:YAG laser-pumped dye laser (Lambda Physik: LPD 3002) produces laser pulses of 4–5 ns duration (FWHM) with a spectral bandwidth of 0.2 cm^{-1} and a repetition rate of 10 Hz. The range 220–280 nm has been obtained by frequency doubling with BBO and KDP crystals leading to a pulse duration of 3–4 ns (see Fig. 3). The laser beam is crossed with the ion cloud/beam in the Paul trap (see Fig. 1). There are three laser ports at our disposal. The first port is 4 cm behind the entrance aperture, the second is in the middle of the trap 15 cm behind and the last one is 19 cm behind the aperture.

Perpendicular to both beams the fluorescence photons are imaged by a lens system onto the photocathode of a multiplier. The angle between the direction of polarization of the laser pulses and the direction of the detected fluorescence is set to the so-called magic angle of 54.7° to exclude the influence of collisional disalignment or hyperfine structure quantum beats [17] which can falsify the lifetime measurements. We no longer need optical filters for excluding unwanted light because scattered laser light is sufficiently suppressed by a stack of diaphragms, and light from the discharge itself is excluded by the 0.3 mm aperture. Our multiplier (Hamamatsu R2496) has a finite

Table 1. W II radiative lifetimes.

Level	Energy (cm^{-1})	Transition $\lambda_{exc.}$ (nm)	Lifetime (ns)	
			Ref. [18]	this work
${}^6F_{1/2}^0$	36165.35	276.4261	14.0(7)	14.42(14)
${}^2S_{1/2}^0$	38576.32	269.7706	11.3(6)	11.88(7)

rise-time of 700 ps and a non-ideal response function with ringing due to the electrical circuitry. The ringing cannot be smoothed without a loss in time-resolution. We therefore measure the response function separately and include it in our evaluation procedure (see also Refs. [13,14]).

In addition, we measure the temporal shape of our laser pulse by means of a fast photodiode (Hamamatsu R1328U-52) having a rise-time of 60 ps. This arrangement is also used to trigger the fluorescence signal. Moreover, the shape of the laser pulse enters our evaluation procedure. The fluorescence curves are recorded time-resolved by means of a fast digitizing oscilloscope (Tektronix TDS 680 B) with an analog bandwidth of 1 GHz and a real-time scanning rate of 2×5 Gigasamples. This instrument allows the recording of a full decay curve, and simultaneously the recording of the temporal laser pulse with a 200 ps time increment which is comparable with the rise-time of the oscilloscope. In the present experiment we typically added 500 single shots for obtaining a flawless signal-to-noise ratio. It is worth mentioning that the measuring time is less than one minute. On each level we made at least 5 independent measurements on up to three transitions using differing buffer gas conditions and laser pulse energies.

4 Results and discussion

We tested our linear Paul trap as an ion guide, but also as a trap for singly ionized iron and tungsten ions. The figure of merit is demonstrated in Figure 2 where we compare a TRLIF signal obtained on guided/trapped ions with a signal from a switched-off trap, however, still with the discharge operating. For both signals 500 single shots at 5 Gigasamples have been summed. Although the latter signal is enlarged by a factor of 20 in the figure the huge improvement in the LIF signals is evident.

When applying dc potentials to the cap electrodes the LIF signals are further increased by up to 50%. However, at present no serious attempt has been made for a long-time trapping. Probably, this will be important when multiply ionized species come to be investigated.

Evaluation of our LIF signals is done by fitting an exponential decay curve which is convoluted with both the laser pulse shape and the response function of our photomultiplier. This procedure is described in reference [13], additionally we have extended it to handle saturation in the fluorescence signals by solving an appropriate rate equation system. Figure 3 demonstrates the excellent matching between measuring points and fitted curve even if

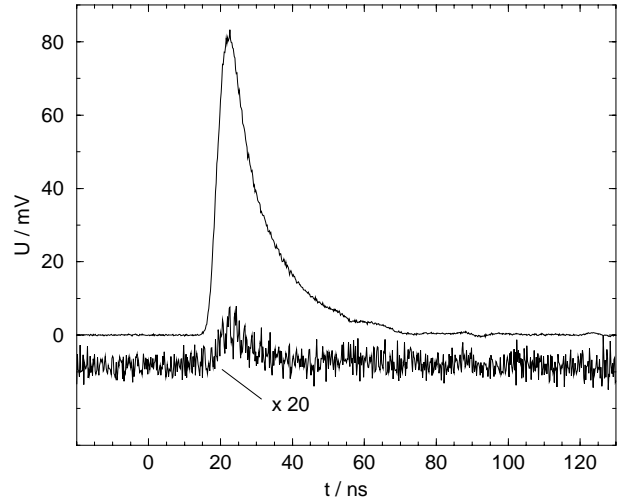


Fig. 2. LIF signals obtained at the first laser port close to the entrance aperture with and without guiding of the ions. The lower curve has been 20 times enlarged and slightly shifted.

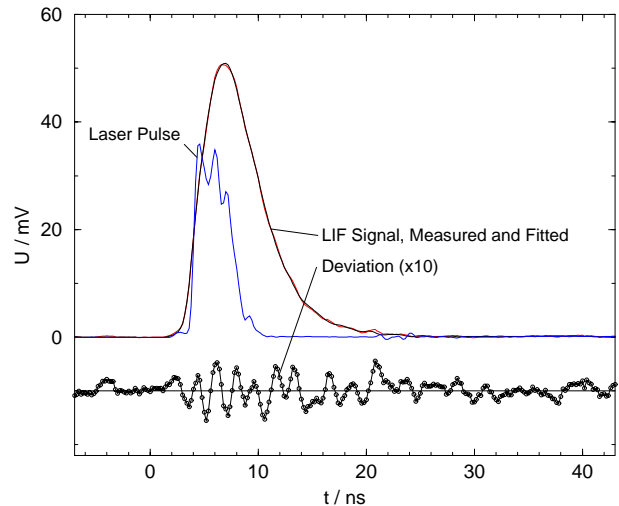


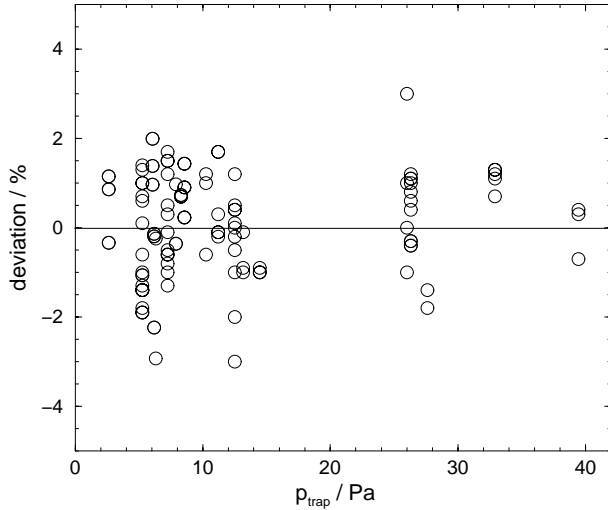
Fig. 3. Time-resolved LIF signal on the Fe II ${}^4D_{7/2}^0$ level, measured and fitted, together with the exciting laser pulse. The fitted curve matches the measured points in an excellent way, giving a lifetime of $\tau = 2.97 \pm 0.02$ ns.

the lifetime is slightly shorter than the FWHM of the laser pulse. We tested the quality of our lifetime measurements on the linear trap by means of two W II and seven Fe II level lifetimes which are listed in Tables 1 and 2, respectively, together with literature data. The consistency is excellent for both elements.

It is common practice to perform lifetime measurements at gas pressures as low as possible to exclude quenching collisions. We found, however, that pressures of the cooling gas between 3 Pa and 40 Pa are necessary for an efficient trapping of the ions. Below 3 Pa the cooling effect is too small, above 40 Pa the collisions begin to disturb trapping. On the other hand an influence on the measured lifetime from quenching by neon atoms should only occur in a pressure regime above several 1000 Pa [17]. Figure 4 shows the deviation of measured lifetimes from

Table 2. Fe II radiative lifetimes.

Level	Energy (cm^{-1})	Transitions			Lifetimes(ns)			
		$\lambda_{exc.}$ (nm)			Ref. [20]	Ref. [21]	Ref. [22]	this work
${}^6D_{7/2}^0$	38 660.04	258.5876	261.1874	263.1324	3.68 ± 0.07		3.75 ± 0.2	3.64 ± 0.09
${}^6D_{5/2}^0$	38 858.96	259.8370	261.7618	263.1048	3.63 ± 0.08		3.7 ± 0.2	3.70 ± 0.05
${}^6D_{3/2}^0$	39 013.21	260.7088	262.0409	262.8294	3.83 ± 0.10		3.7 ± 0.2	3.73 ± 0.07
${}^4D_{7/2}^0$	44 446.88	224.9180				3.02 ± 0.07	3.1 ± 0.2	2.97 ± 0.02
${}^4D_{5/2}^0$	44 784.76	225.1556	226.5995			3.10 ± 0.08	3.1 ± 0.2	2.90 ± 0.06
${}^4D_{3/2}^0$	45 044.17	226.2688					3.0 ± 0.2	2.91 ± 0.09
${}^4F_{5/2}^0$	45 079.88	225.0936	226.0860			3.75 ± 0.14	3.7 ± 0.2	3.55 ± 0.08

**Fig. 4.** Deviation of measured lifetimes plotted *versus* cooling gas pressure. No pressure dependence has been found in the pressure range used in the experiments.

our averaged lifetime values over the pressure range used in our experiments. Obviously, no pressure-dependent effects are present in our data.

In conclusion we can state that our linear Paul trap is simple in operation and a good ion collector increasing the number of ions in the interaction volume appreciably. Moreover, the large scale ion guide allows a remarkable reduction of scattered light from the discharge by placing the interaction volume 15 cm away from the entrance aperture. At this distance we do not see any fluorescence from neutrals, so that wrong identifications, blends on the transitions and also quenching by the neutrals can be excluded. Because of the reduced scattered light, LIF signals of a higher quality can be obtained from smaller ion densities. With our ion trap we have no problems in measuring level lifetimes of the second spectra. A larger set of W II lifetimes has already been finished. In a next step we intend to produce W III ions by radiative ionization in the trap itself.

The combination of a hollow cathode with a linear ion trap makes available lifetime measurements on nearly all elements in different stages of ionization.

This project is financially supported by Deutsche Forschungsgemeinschaft DFG. We thank E. Schulz-Gulde for a critical reading of the manuscript.

References

1. M.C.E. Huber, R.J. Sandeman, Rep. Prog. Phys. **49**, 397 (1986).
2. A. Bard, M. Kock, Astron. Astrophys. **282**, 1014 (1994).
3. W. Paul, O. Osberghaus, E. Fischer, *Ein Ionenkäfig* (Forschungsbericht des Wirtschafts- und Verkehrsministeriums Nordrhein-Westfalen, 1998).
4. E. Fischer, Z. Phys. **156**, 1 (1959).
5. P.H. Dawson, *Quadrupole Mass Spectrometry and its Applications* (Elsevier Scient. Publ. Company, Amsterdam, 1976).
6. D. Gerlich, *Advances in Chemical Physics*, Vol. LXXXII (John Wiley & Sons, New York, 1992), p. 27, p. 66.
7. R.E. March, R.J. Hughes, *Quadrupole Storage Mass Spectroscopy* (J. Wiley & Sons, New York, 1989).
8. W.H. Press, S.A. Teukolsky, W.T. Vetterling, B.P. Flannery, *Numerical Recipes in C*, 2nd edn. (Cambridge University Press, 1992), p. 863.
9. J.D. Prestage, G.J. Dick, L. Maleki, J. Appl. Phys. **66**, 1013 (1989).
10. M.G. Raizen, J.M. Gilligan, J.C. Bergquist, W.M. Itano, D.J. Wineland, J. Mod. Opt. **39**, 233 (1992).
11. P.T.H. Fisk, M.A. Lawn, C. Collins, Appl. Phys. B **57**, 287 (1993).
12. M. Schultz, Diploma thesis, Universität Hannover (1998).
13. D. Engelke, A. Bard, M. Kock, Z. Phys. D **27**, 325 (1993).
14. R. Schnabel, A. Bard, M. Kock, Z. Phys. D **34**, 223 (1995).
15. K. Danzmann, M. Guenther, J. Fischer, M. Kock, M. Kühne, Appl. Opt. **27**, 4947 (1988).
16. M. van Lessen, R. Schnabel, M. Kock, J. Phys. B: At. Mol. Opt. Phys. **31**, 1931 (1998).
17. P. Hannaford, R.M. Lowe, Opt. Eng. **22**, 532 (1983).
18. M. Kwiatkowski, F. Neumann, K. Werner, P. Zimmermann, Phys. Lett. A **108**, 49 (1984).
19. R. Schnabel, M. Kock, Z. Phys. D **41**, 31 (1997).
20. E. Biemont, M. Baudoux, R.L. Kurucz, W. Ansbacher, E.H. Pinnington, Astron. Astrophys. **249**, 539 (1991).
21. B. Guo, W. Ansbacher, E.H. Pinnington, Q. Ji, R.W. Berends, Phys. Rev. A **46**, 641 (1992).
22. P. Hannaford, R.M. Lowe, N. Grevesse, A. Noels, Astron. Astrophys. **259**, 301 (1992).