Precise Atomic Lifetimes Measured at a Heavy-Ion Storage Ring

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1 Technique

A new experimental technique, derived from beam-foil spectroscopy, has been developed for the measurement of long atomic lifetimes in ions. The ions are produced in a standard ion source and then, after a first acceleration step, collisionally excited in the gas or foil stripper of an accelerator. A beam of charge-state selected MeV-ions is transported to and then stored in the clean ultra-high vacuum conditions of a heavy-ion storage ring. This storage has been combined with straightforward optical detection (Fig. 1) to make precise and accurate atomic lifetime measurements in the millisecond range. As any element can be collisionally excited, the method is very versatile and may be used to study a number of cases which are of fundamental atomic structure or astrophysical and terrestrial plasma diagnostic interest.

A major advantage of the new technique is that the ion excitation on one hand and the trapping and detection on the other are spatially separated and can be individually optimized. The experiment works in fixed geometry. Thus there are no mechanically moving parts and no explicit dependence of the decay curves on the ion beam velocity. All that is needed to reach high precision are a reliable clock frequency, a measurement of the ion beam storage time (to correct the photon signal for ion beam losses during the measurement cycle), and a photon data rate somewhat higher than the detector dark rate. We employed a solar-blind photomultiplier and an interference filter to suppress possible stray light from ion getter pumps in the vacuum vessel. There is no need for higher spectral selectivity, as the transition of interest is often the only one of extreme longevity of the upper level in a given ionization stage of the element under study. Thanks to this longevity, measurements may begin after higher-lying levels have decayed to the ground state or to the metastable level of interest, and consequently there is practically no cascade problem. No ion-beam related background was detected. Further improvements would be possible by increasing the detection efficiency, for example by using several photomultipliers in parallel.

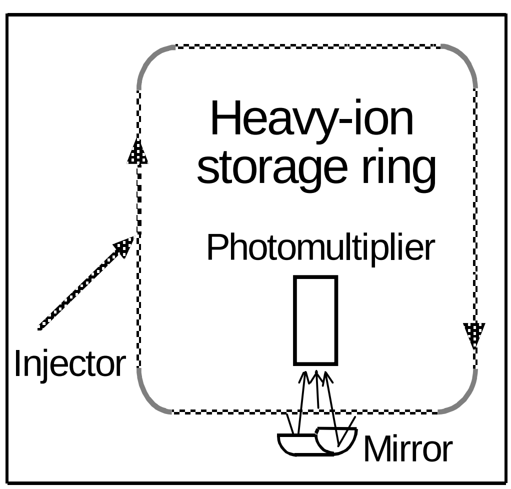

Figure 1: Schematics of the experimental arrangement

The new technique has been applied to several intercombination (E1, $\Delta S=1$) transitions and to a few forbidden (M1) transitions. The selection of test cases matches the wavelength range (λ 190 to 270 nm) of our low-noise photomultiplier (with a dark rate of about 1 count/s) and the vacuum vessel window material and wavelength filter ranges.

2 Cases Studied

2.1 Intercombination Transitions

Among the systems studied are the $2s^2\ {}^1S_0 - 2s2p\ {}^3P^0_1$ intercombination transition in the Be-like ions B^+ and C^{2+}, the $3s^2\ {}^1S_0 - 2s2p\ {}^3P^0_1$ transition in the Mg-like ion Al^+, and the $2s^2 2p^2\ {}^3P_{1,2} - 2s2p^3\ {}^5S^0_2$ transitions in C-like N^+.

The $2s^2\ {}^1S_0 - 2s2p\ {}^3P^0_1$ intercombination transition in the doubly charged carbon ion (Be-like) is of particular interest as a test case for fundamental atomic structure calculations and for some astrophysical applications (see Fig. 2). After the notable scatter of the early theoretical data, we note that some later calculational results seemingly followed experiment, even as early data from radiofrequency ion trap experiments varied considerably and appear mutually contradictive. These earlier lifetime results are now being surpassed in accuracy and precision because of the much cleaner experimental trapping conditions available at the heavy-ion storage ring, by the new experimental result of ($\tau = 9.714 \pm 0.013$ ms) [1].

In the most recent calculations by the groups at Belfast and Nashville, however, very different theoretical approaches yielded rather similar results and were quoted with intrinsic error estimates in the few-percent range. These similarities, however, were furthered by semiempirical adjustments to experimental energy level and fine structure data. No fully *ab initio* calculation of this system so far comes close to the experimental precision and accuracy. An example is set by the case of B^+: Two approaches (configuration interaction [2] and MCHF/MCDF [3] calculations) led to lifetime results for the $2s2p\ {}^3P^0_1$ level which were claimed to have 2% uncertainties but differed from each other by about 6%. This discrepancy boils down to the use of different experimental values for the $2s2p\ {}^3P^0$ fine structure intervals. With the same (right) choice of atomic structure reference data, the lifetime calculations perfectly agree with each other and with experiment ($\tau = 97.7 \pm 1$ ms). Assuming the validity of the theoretical treatment, our lifetime measurement with its poor spectral resolution can thus be seen as an indirect, but sensitive test on difficult to obtain experimental fine structure data.

The intercombination decay of the $3s3p\ {}^3P^0_1$ level in Mg-like Al^+ may be seen as an analog to the above two ions in the Be sequence. In this case, our experiment ($\tau = 305 \pm 10$ μs) [4] confirms the lifetime result of an earlier radiofrequency ion trap study [5] (which used laser ablation of Al and thus operated at a lower ambient pressure than is possible with gases). However, the latest calculations do not match the experimental data, and the results of earlier calculations have been stated with considerable uncertainties.

For the $2s^2 2p^2\ {}^3P_{1,2} - 2s2p^3\ {}^5S^0_2$ transition in C-like N^+, which appears in auroral spectra of the upper atmosphere, a series of calculations and ion trap measurements finally led to lifetime data which agreed with each other and were quoted with about 6% uncertainty. Our storage ring data ($\tau = 5.87 \pm 0.03$ ms) [6] reach 0.5% uncertainty and are just outside the previously given 5% (1σ) error ranges of both the latest ion trap data and the most advanced theoretical treatment [7].

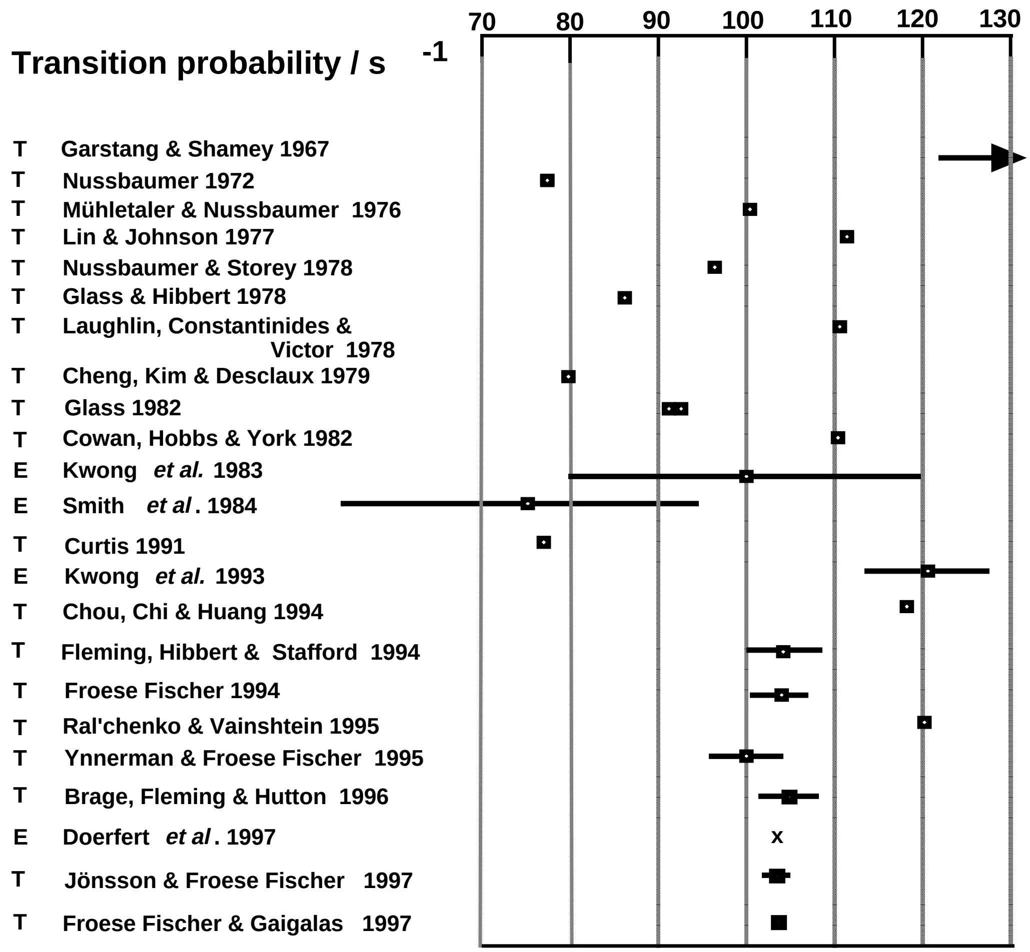

Figure 2: Intercombination transition probability in the C^{2+} ion. T Theory, E Experimental data. Our experiment is denoted "Doerfert *et al.* 1997" [1].

2.2 Forbidden Transitions

Of forbidden (M1) transitions we have studied test cases of interconnects within the ground state complex of several ions. In F-like Sc^{12+} our experiment aimed at the forbidden transition which connects the $2s^2\,2p^5\ {}^2P^0_{3/2,1/2}$ fine structure levels. The signal statistics was limited by the achievable ion beam current and thus the result limited to 3% precision. The available calculation [8] matches the experimental transition rate result of (1000 ± 30) s^{-1} [4] only after a 2% correction for experimental fine structure data. For the much longer lived $2s^2 2p^2$ and $2s^2 2p^4\ {}^1D_2$ levels in C-like Si^{8+} and O-like Si^{6+} our storage ring data corroborate theory on the forbidden transition rates [9,10] at the 3% level, yielding lifetimes of 38.8 ± 0.5 ms and 65 ± 3 ms, respectively [6]. The experimental precision on these long atomic lifetimes is limited by the ion beam dynamics after injection into the storage ring: Although the long-term beam storage time constant may be as high as 40 s (which would cause little trouble), the initial transitory behavior of the ion beam necessitates larger corrections with notable uncertainties.

3 Conclusion

The lifetimes already measured with the new technique range from 0.3 ms to 100 ms. Depending on the experimental conditions (it is mostly the available ion beam current which determines the signal rate), the results have errors in the range 3% to 0.13%. At such a level of precision, several of the new benchmark data test theory. Concerning intercombination transition rates, our data are found to corroborate only the latest, most extensive calculations. Even these, however, have not been obtained fully *ab initio*, but use various adjustments to experimental atomic structure reference data. Concerning forbidden transition probabilities, theory seems much better off. Here, however, the predicted transition rates strongly depend on the assumed level splittings, and theories falling short by a few percent on this account easily yield 10% errors of the lifetime results. Such errors are clearly resolvable by the new experimental lifetime measurement technique.

Judging from our test cases, the forbidden transition rates apparently are under better theoretical control than the intercombination transition rates (after managing the term differences), probably because rather similar wavefunctions of initial and final states are involved and not the more dissimilar ones of the intercombination transition problem. However, we note that earlier on there were order-of-magnitude differences between differently calculated forbidden transition rates.

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