

Improved Electron-Beam Ion-Trap Lifetime Measurement of the $1s2s\ ^3S_1$ Level in N^{5+} and F^{7+}

P. A. Neill*, E. Träbert†, P. Beiersdorfer, G. V. Brown, C. L. Harris*, S. B. Utter and K. L. Wong

Department of Physics and Space Technology, Lawrence Livermore National Laboratory, Livermore, CA 94551, U.S.A.

Received January 10, 2000; accepted in revised form January 31, 2000

PACS Ref: 32.70.Cs; 32.30.Rj; 39.90.+d

Abstract

Earlier measurements using a heavy-ion storage ring and an electron beam ion trap (EBIT) for the determination of the $N^{5+}\ 1s2s\ ^3S_1$ level lifetime are improved upon by new EBIT work. The new result, 3.94 ± 0.05 ms, agrees with the previous values, but is more precise. A corresponding measurement on F^{7+} yields a lifetime of 276 ± 2 μ s. The new values corroborate the isoelectronic trend of the most precise data for this isoelectronic sequence and thus help distinguish among theoretical predictions.

1. Introduction

Quantum mechanical methods are applied to a wide range of atomic, many-body, and low-dimensional solid state problems. While many phenomena can be described and understood qualitatively this way, quantum mechanical methods often fail to yield precise numbers. Concerning atoms, the level of precision reached is excellent only for one-electron systems. There are partial successes for systems with two or three electrons, but notable limitations appear beyond. Experiments testing the accuracy of predictions for two-electron systems, the simplest many-electron systems, are of special importance for the development of approaches to accurately describe any many-electron system. The present experiment aims at a precise lifetime measurement of the purely relativistic, magnetic dipole (M1) transition $1s^2\ ^1S_0 - 1s2s\ ^3S_1$ in two-electron ions of N ($Z=7$) and F ($Z=9$). Such lifetime measurements complement measurements of atomic structure, because lifetime measurements are sensitive to details of the wavefunctions that are not tested by energy spectroscopy.

The $1s^2\ ^1S_0 - 1s2s\ ^3S_1$ transition was first observed and identified in solar spectra. Soon after, it was shown that the single-photon decay mode, arising solely from relativistic effects, is the dominant decay contribution. Meanwhile this is one of the most precisely studied decays, both experimentally and theoretically. Precise predictions for the $1s^2\ ^1S_0 - 1s2s\ ^3S_1$ transition rate involve large-scale calculations that aim at an extensive treatment of electron correlation. The various techniques used include non-relativistic wave functions and energies with relativistic effects included in the transition operator [1], the Dirac-Hartree-Fock (DHF) approach [2,3], the Random-Phase Approximation (RPA) [3,4], a relativistic framework with correlation [5],

relativistic wavefunctions in various order approximations up to the use of “exact” relativistic wavefunctions [6], and fully relativistic single- or multi-configuration Dirac-Fock (SCDF/MCDF) calculations with correlation treatment in both energies and radial wave functions [7]. The latter treatment also included effects of the negative energy continuum, which has recently become of new interest in atomic structure calculations [8,9]. The various calculational results for a given low- Z (nuclear charge Z) ion scatter by several percent. Part of this scatter may be due to imperfect wavefunctions, and, indeed, the results from using what are probably the most perfect wavefunctions, whether on a non-relativistic [1] or a relativistic basis [6], agree well with each other in the low- Z range. However, their results diverge with increasing values of Z , as relativity plays a growing role. The leading relativistic correction term, a factor $(1+1.07(\alpha Z)^2)$, has been calculated by Lin [10]. After this correction, the old calculational results by Drake come very close to the latest by the Johnson group, at least for the range of nuclear charges of present interest.

The experimental lifetime data on this transition (from neutral He atoms to highly charged Xe^{52+} ions) span 15 orders of magnitude (see [11]), but very few of these experiments have reached a precision that is comparable with the calculational scatter. At high Z , relativistic effects are so large that they are tested to first order even by experiments with a typical uncertainty of 5%. In the middle of the range, hardly any experiment reaches this precision, and new experimental schemes are needed to obtain data with meaningful precision. In the low- Z range, say from $Z=5$ to $Z=12$, some recent experiments at a heavy-ion storage ring [12,13] and by using EBIT [14,15] have reached a precision of better than 0.5% and thus excluded all but those two calculations that use practically exact wavefunctions. The storage ring experiment has almost reached its short-lifetime limit at $Z=7$, while for EBIT the 4 ms lifetime in N^{5+} is fairly long. Consequently, the previous data for heliumlike ions of N from the storage ring and from EBIT [12,16] were less precise than those for other ions. Even in the absence of improved precision, new data for other elements can support the statistical reliability of the presently available data sample. Atomic structure contributions like correlation and relativistic effects scale differently with nuclear charge Z , and so do the results of various calculations. A dense set of reference data ties down such variations, and we set out to obtain such data for N and F.

* Permanent address: Department of Physics, University of Nevada Reno, Reno NV 89557, USA

† Present address: I.P.N.E., Université de Liège, Belgium; E-mail: elmar@mbox.pne.ulg.ac.be; Permanent address: Fakultät für Physik und Astronomie, Ruhr-Universität Bochum, D-44780 Bochum, Germany; E-mail: Traebert@ep3.ruhr-uni-bochum.de

2. Experiment

The measurements were carried out at Lawrence Livermore National Laboratory, using the electron beam ion trap EBIT-II. It was operated under UHV conditions ($p < 10^{-10}$ mbar). For the measurements on nitrogen, N_2 was being bled into the trap continually, via a leak valve and a set of collimating apertures in a ballistic gas injection system. We performed the experiment at various energies of the electron beam, from the observed production threshold of the excited level of interest in the N^{5+} ion at 410 eV to 600 eV, which is beyond the ionization limit (552 eV) of N^{5+} and thus an energy at which the next higher charge state ions, N^{6+} , are also produced. The injector pressure was varied by a factor of two. As the electron beam current under such low-energy conditions (for EBIT) was only of the order of a few mA, no current density variation (as discussed earlier [15]) was possible.

For the lifetime measurements, EBIT was operated in a cyclic mode. Every 0.1 s the accumulated ion cloud was purged from the trap (by lowering a drift tube potential) in order to avoid the build-up of high- Z contaminants. The electron beam was switched on for about 0.03 ms, ionizing and exciting the ion cloud in the trap. Then the electron beam was switched off, and the trap was maintained as a Penning trap in the so-called magnetic trapping mode [16]. EBIT in this mode has been shown to store ions for many seconds [16,19]. The switching time of the electron beam needs to be faster than the lifetime of the level of interest. To achieve this, we combined two switching processes: The electron beam current was reduced to zero, and the beam acceleration voltage was set to zero. All have a transient of about 30 μ s. Further excitation of N^{5+} stops when the electron energy drops below threshold. The data evaluation was restricted to the part of the decay curves after the first about 5 ms for N in order to avoid possible stray influences of the switching processes on the decay curves.

In order to monitor the charge state balance in the trap, a flat-field grating spectrograph, equipped with a charge-coupled device (CCD) camera, was set up to observe the wavelength range from 2 to 8 nm, in which the 1-2 resonance and intercombination lines in the He-like and H-like ions of N are to be found. The spectra confirmed the dominance of nitrogen (in contrast to C and O ions under other conditions) in the trap, as well as the presence of ions either only up to charge state N^{5+} or to N^{6+} , depending on the electron beam energy. At the electron beam energies necessary for the production, but not high enough to effect further ionization of He-like N^{5+} , only the ground state transitions in this ion appeared in the soft-X-ray spectra.

For the N^{5+} lifetime measurement proper, a thin-window, energy-dispersive, solid-state X-ray detector [intrinsic germanium (IGLET)] was employed. The detector combines a low energy threshold with sufficient spectral resolution and good timing properties. The true signal rate for N^{5+} was about 15000 counts per hour. The photon energy and the time of each event were recorded in "event mode" by the data acquisition system. In total, about 10^5 counts were accumulated in the decay curve part of the data, corresponding to an overall statistical reliability of 0.3%. A decisive factor for the lifetime measurement is the reference time base. For this purpose, a 100 kHz frequency generator

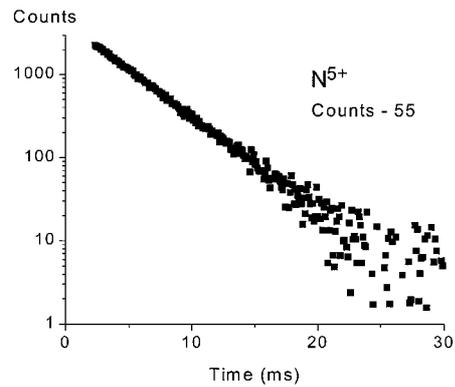


Fig. 1. Decay curve data (logarithmic scale) obtained with 450 eV-electron beam energy at the position of the $1s^2\ ^1S_0-1s2s\ ^3S_1$ transition in the IGLET X-ray spectrum of N. The data were accumulated over 7 hours.

was used to provide clock pulses. The number of clock pulses in the time interval between receiving a reference signal (starting the trap cycle) and the arrival of an X-ray signal pulse was stored as the time information in the event mode system. As the generator was not synchronized with the trap, this introduces a time jitter of 10 μ s that is negligible for lifetime measurements in the millisecond range.

The lifetime value extracted from the decay curve data (Fig. 1) turned out to vary as a function of the start channel of the evaluation. Consequently, an increasing number of initial channels were left out until the fit solution stabilized. The foreign contribution must be somewhat shorter lived, because the apparent lifetime extracted from the full curve was shorter than the eventually reached one by several percent. Such effect could be expected from unresolved spectral blends, for example with the $1s2p\ ^3P^o$ levels (which are repopulated by cascades), that cannot be resolved by a solid state X-ray detector. While the flat-field grazing incidence spectrometer available for spectroscopic checks could resolve those lines, it is neither equipped with a time-resolving detector nor would the spectrometer efficiency be sufficient to obtain statistically meaningful data if it was. The truncation of the decay curve implies a loss of statistical data quality, so that the final result could not be determined to much better than 1%.

The fluorine experiment was run at an electron energy of about 2 keV, that is more than 1 keV above the ionization limit of F^{7+} (954 eV). A sample decay curve is shown in Fig. 2. As observed with Ne^{8+} [15] since, under such conditions the signal-to-background ratio suffers from a notable tail, which we ascribe to recombination events. The signal statistics was good enough for a statistical precision of the lifetime result of 0.7%. In this measurement, the time stamping of each event was achieved by measuring the height reached by a linearly increasing ramp voltage when the photon signal arrived. Test pulses from a frequency generator were afterwards injected into the same measurement system at two different frequencies and reproduced the time base to 10^{-4} .

3. Data analysis and results

Typical X-ray data of N (representative for six of such sets) are shown in Fig. 1. The signal contains some background

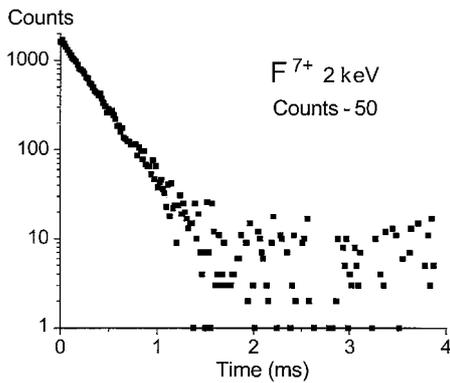


Fig. 2. Decay curve data (logarithmic scale) obtained at 2 keV electron beam energy at the position of the $1s^2\ ^1S_0-1s2s\ ^3S_1$ transition in the Si(Li) X-ray spectrum of F.

because of the low photon energy that causes spill-over of electronic noise into the signal pulse height range. Also, there is the possibility of recombination events from collisions with other atoms. Both contributions are almost time-independent in the range of our measurements, so that in the analysis the background was treated as flat. Typical trapping times of many seconds as observed under clean EBIT conditions [16,19] would imply corrections for the present lifetime of the order of 10^{-3} , and thus would be negligible here. However, in the course of another experiment a measurement of charge exchange decay times was done close to the calendar date of the nitrogen measurement and thus close to the actual operating conditions of the present experiment. In that other experiment, bare and H-like Fe ions experienced a charge exchange decay time of (0.38 ± 0.04) s while similar ions of Ar showed a charge exchange decay time of (0.52 ± 0.03) s. The trend of these charge-exchange limited ion storage times (for H-like ions) corresponds to the roughly linear dependence of the charge-exchange cross section on charge state q [20]. Extrapolating these data to N^{5+} ions, a storage time of 1.38 s is to be expected which implies the presence of a higher residual gas pressure than found under optimum conditions, and which translates to a systematic shift of the atomic lifetime by 0.3%. The effect on F^{7+} with its much shorter atomic lifetime is presently negligible.

The counting statistical uncertainty dominates the error budget. Neither the time base (0.05%) nor the variation of the fit results when truncating early or late parts of the decay curve data (by up to 5 ms for N, 200 μ s for F) have a notable influence compared to the statistical uncertainty at this level. Taking quantifiable and tested systematic errors into account, we find a $1s2s\ ^3S_1$ level lifetime of (3.94 ± 0.03) ms for N^{5+} and of (274 ± 2) μ s for F^{7+} .

However, in the course of our studies we found reason to suspect the presence of additional systematic errors that we have not been able to quantify. These observations are as follows. For both ions, truncation of early and late data channels up to more than one lifetime was tried in order to check for hidden systematic errors. In F this procedure yielded a stable lifetime result, but in N we encountered a systematic lengthening of the lifetime result by about 2% before it reached the aforementioned value. By this time, however, the counting statistics worsened so that further

truncation would be progressively losing significance. Some of the variability is connected to the signal-to-noise ratio, that is measurements at lower gas injection pressure suffered from a lower signal rate without a corresponding reduction of the noise rate. While by visual judgement of the two-dimensional event mode map (photon energy versus time) the data used for constructing the decay curve were sufficiently well separated from the noise range, there might have been some spillover. Also, within the energy resolution of the X-ray detector we could not distinguish the photon energies of He-like N^{5+} from $n \rightarrow 1$ X-rays of C ions generated in charge-transfer recombination of bare or hydrogenlike C ions, which furthermore may have a functional time dependence. K-shell X-rays from high n, l levels represent an appreciable fraction of the photons generated in charge transfer reactions at the low-collision energy conditions found in EBIT [17]. In contrast, $n = 2 \rightarrow 1$ K-shell X-rays in C ions did not exceed the noise spectrum energies (which is why this experiment did not cover C^{4+} as well). The slight contamination of EBIT with C is unavoidable, but it is not known how little of this is actually present. After trying a number of model assumptions, from a second exponential decay component to a background that increases with time, we make a conservative estimate and enlarge our total uncertainty for N from ± 0.03 ms to ± 0.05 ms.

For F the signal was spectrally cleaner. However, the decay curves featured a high and practically constant tail that limited the dynamic range of the decay curve data. Unfortunately, fitting this tail by a second (slow) decay component or by a constant does not prevent systematic error. In the measurements on Ne^{8+} [15] it was found that the primary lifetime result under comparable conditions was shorter by 0.8% than in case of a measurement at an electron beam energy not much above the production threshold of the He-like ion, not yet producing the H-like ion. Then the tail was absent, the dynamic range was much larger, and the decay curves appeared cleaner. Lacking a measurement under similar conditions for F, we introduce an estimate of a systematic shift of 0.6%, to (276 ± 2) μ s.

4. Discussion

The new lifetime result for N lies within the 1σ error bars of the previous data [12,16], but is more precise. The result for F^{7+} agrees very well with the isoelectronic trend, but is not quite as significant as the measurements for O^{6+} [14] and Ne^{8+} [15] (Table 1, Fig. 3). The new level of uncertainty for F is better than the 1% scatter of the various predictions. The available precision data on elements B to Ne, from heavy-ion storage ring or EBIT, exclude the earlier calculations by Johnson and collaborators [2–4], as well as the calculations by Indelicato [7]. Within this range of elements, the oldest calculation, by Drake [1] (using very precise nonrelativistic wavefunctions) and the more recent fully relativistic calculation by Johnson *et al.* [6] (using “exact” wavefunctions) are fully compatible with the experimental results. Drake’s original results are still within our present range of uncertainty, but they become virtually indistinguishable from Johnson’s latest results (in this range) by inclusion of the leading term of the relativistic correction

Table I. Results of calculations and measurements of the lifetime of the $1s2s\ ^3S_1$ level in N^{5+} and F^{7+} .

Reference	Comment	Lifetime (ms)	
		Ion	Ion
Theory		N^{5+}	F^{7+}
Drake [1]	non-rel. wave functions	3.949	0.2772
Drake [1]	with relativistic correction [10]	3.938	0.2759
Johnson and Lin [2]	DHF	3.920	0.2747
Johnson and Lin [3]	DHF	4.750	0.3138
	RPA	3.885	0.2737
Johnson, Plante, Sapirstein [6]	MCDF “exact” wave functions	3.942	0.2762
Indelicato [7]	MCDF (no pair)	4.205	
	MCDF (e^+e^-)	4.012	
Experiment			
Schmidt <i>et al.</i> [12]	Heavy-ion storage ring	3.905 ± 0.05	
Beiersdorfer <i>et al.</i> [16]	EBIT	3.92 ± 0.13	
This work	EBIT	3.94 ± 0.05	0.276 ± 0.002

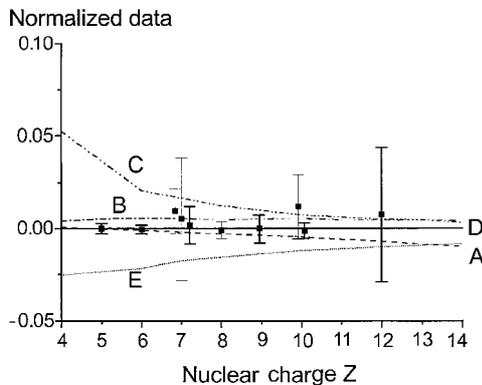


Fig. 3. Scaled experimental and selected theoretical transition rate data on the $1s^2\ ^1S_0 - 1s2s\ ^3S_1$ transition in light He-like ions. The data are presented by the difference to the theory data by Johnson, Plante and Sapirstein [6] and normalized to the latter. Experimental data for B^{3+} are from Schmidt [13], for C^{4+} from Schmidt *et al.* [12], for N^{5+} from Schmidt *et al.* [12], Beiersdorfer *et al.* [16] and this work, for O^{6+} from Crespo López-Urrutia *et al.* [14], for F^{7+} from this work, for Ne^{8+} from Träbert *et al.* [15], and for Mg^{10+} from Stefanelli *et al.* [18]. Theory data are A (---) [1], B (- · -) [2], C (- · · -) [4], D (—) [6] and E (···) [7]. Of the theory data shown, those from Drake [1] (curve A) are based on non-relativistic wavefunctions and are therefore not expected to work as well for higher nuclear charge. For Ne they are still inside the experimental error bar, at the low side. To within the stated error bars, agreement of the experimental results with the plot reference data (horizontal line at zero) by Johnson, Plante and Sapirstein [6] (curve D) is excellent, for all experimental data displayed. A leading-term relativistic correction, however, brings curve A into practical agreement with curve D. All other theoretical results shown are now of mere historical interest.

as prescribed by Lin [10]. Decisive tests distinguishing between these two calculations (after the aforementioned relativistic correction) in the present range of nuclear charge will need experiments that are more precise than 0.1%, which is not presently in reach.

5. Acknowledgments

We are happy to acknowledge the dedicated technical support by Dan Nelson, Phil D'Antonio, and Ed Magee. We are also grateful to G. W. F. Drake (Windsor) for insightful advice and pointing out ref. [10]. The work at Lawrence Livermore National Laboratory was performed under the auspices of the Department of Energy under Contract No. W-7405-Eng-48. E.T. acknowledges travel support from the German Research Association (DFG).

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