Lifetime of the $1s2s\ ^3S_1$ metastable level in He-like S^{14+} measured with an electron beam ion trap

J. R. Crespo López-Urrutia,* P. Beiersdorfer, and K. Widmann
University of California Lawrence Livermore National Laboratory, Livermore, California 94550, USA
(Received 7 April 2006; published 17 July 2006)

A precision measurement of the lifetime of the lowest excited level of the He-like S^{14+} ion carried out at the Livermore EBIT-II electron beam ion trap yielded a value of (703±4) ns. Our method extends the range of lifetime measurements accessible with electron beam ion traps into the nanosecond region and improves the accuracy of currently available data for this level by an order of magnitude.

DOI: 10.1103/PhysRevA.74.012507 PACS number(s): 32.70.Cs, 32.30.Rj, 39.30.+w

I. INTRODUCTION

Accurate experimental values for the decay rates of forbidden transitions are important for the development of reliable theoretical methods for calculating atomic wave functions. Theoretical predictions of the lifetimes of metastable levels have to take into account effects that are only small perturbations for levels with electric-dipole allowed transitions, e.g., electron correlation, configuration mixing, relativistic effects, and hyperfine interactions. Even for comparatively simple systems, like two-electron ions, atomic theory is evolving with the aim to understand the role of the different contributions affecting the transition rates, such as, for example, the role of the negative energy continuum $E<-mc^2$ and the two-electron correlation [1].

Several experimental techniques allow lifetime measurements of heliumlike ions. Beam-foil spectroscopy enables studies in a lifetime range of $10^{-7}-10^{-12}$ s [2–9]. Electron cyclotron resonance ion sources and ion traps have generated data for lifetimes in the range of several milliseconds (see, for instance, Refs. [10–16]). Storage rings have been used in the range $10^{-3}-10^{0}$ s [17–19]. The TSR storage ring was used to provide measurement of the $1s2s\ ^3S_1$ metastable level of He-like C (20.589 ms±0.2%) [20], He-like N (3.91 ms±1%) [20], and He-like Be (1.80 s±3%)[21]. Electron beam ion traps can cover a lifetime range not accessible by either of these techniques, as first demonstrated by Wargelin, Beiersdorfer, and Kahn [22]; examples are the lifetimes of the $1s2s\ ^3S_1$ level in He-like Ne^{8+} (90.5 μs±1.7% [22] and 91.7 μs±0.4% [23], Mg^{10+} (13.6 μs±4%) [24], F^{7+} (276 μs±0.7%) [25], N^{5+} (3.92 ms±3% and 3.92 ms±1%) [25,26], and O^{6+} (956 μs±0.5%) [27]. The present results from the EBIT-II device in the nanosecond region overlap with data obtained by the beam-foil method. They also show the ability of the device to substantially improve the quality of the available data.

II. EXPERIMENT

The level of interest here is the lowest excited level of the heliumlike ion, the metastable $1s2s\ ^3S_1$, level. This level has the same (even) parity as the $1s^2\ ^1S_0$ ground level so that electric-dipole radiation is not possible. The level decays instead to ground by emission of $M1$ radiation.

We use the Livermore EBIT-II electron beam ion trap to generate the He-like S^{14+} ions. In this device, an electron beam of variable energy is launched axially into a magnetic field of 3 T produced by a set of Helmholtz coils and compressed by it to a diameter of approximately 50 μm. The (moving) space charge generates a radial potential and attracts positive ions. Collisions with beam electrons generate the ions and excite the metastable level. In the vertical (axial) direction, three drift tubes at different voltages create a potential well that closes the trap axially. The ions are contained in the middle drift tube in a cylindrical volume of 20 mm length and a diameter slightly larger than that of the electron beam [28].

Sulfur is injected into the trap by means of a molecular beam of SF_6. We use a continuous injection at a gas injector pressure of 10^{-7} Torr, producing a collimated beam with a neutral SF_6 density of roughly 10^8 cm^{-3}, or roughly 3×10^{-9} Torr in the trap. The electron-beam energy was varied from 3.9 to 5.1 keV. The current was around 80 mA. The SF_6 molecules are first dissociated by electron impact, and their atomic fragments are then ionized. Since the ion-ion Coulomb interaction is very strong, thermalization of the ion ensemble takes place very rapidly; thus, the lighter fluorine ions reach higher thermal velocities and escape the trapping potential more easily than the heavier sulfur ions.

Under steady-state conditions in the trap, at 3.3 keV beam energy sulfur would be mainly ionized to the hydrogenic state. However, the continuous supply of neutral SF_6 and the beam-off periods cause the time-dependent charge balance to be shifted towards the He-like charge state. We chose this beam energy to maximize the count rate under these conditions.

The lack of any other metastable states in H-like and He-like sulfur ions besides the He-like $1s2s\ ^3S_1$ level decaying through radiative transitions with an energy around 2 keV (~5 Å) allows the use of a low-resolution x-ray detector for monitoring the time-dependent x-ray signal. We register the total intensity of the $n=2\rightarrow1$ transitions with (1) a large surface area gas proportional counter [29] and (2) with a lithium-drifted silicon Si(Li) detector. The high counting efficiency of these detectors is essential for achieving good statistics. Line emission from nonmetastable levels is blended with the x-ray decay of the $1s2s\ ^3S_1$ level and is not resolved by either of the detectors. However, this emission decays much faster, and its contribution disappears after a

*Present address: Max-Planck-Institute for Nuclear Physics, D-69117 Heidelberg, Germany.

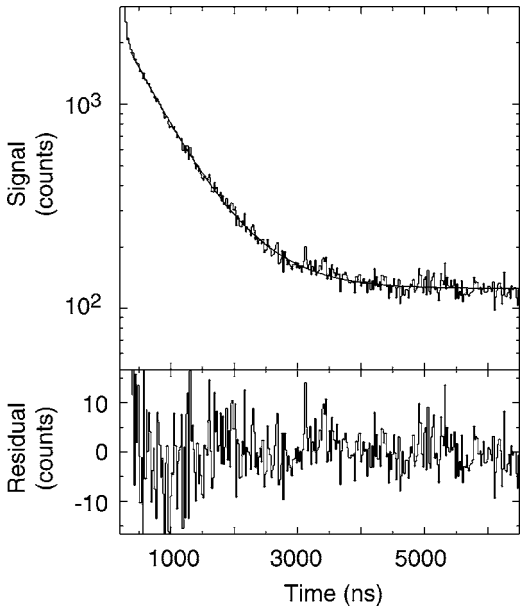

FIG. 1. Top: exponential decay fit to the He-Like S^{16+} ion data obtained with a proportional counter. Bottom: residuals of the decay fit.

time that is several orders of magnitude shorter than the lifetime of the metastable level. The energy resolution of the two detectors was still sufficient to set appropriate energy gates to discriminate against signal not produced by *K*-shell transitions in He- and H-like sulfur ions.

To observe the radiative decay of the metastable level the beam was turned off for 7 μs after staying on for 100 μs. During the beam-on period, ions are produced and excited. In the beam-off period, metastable ions decay radiatively. This cycle was repeated at 3 kHz. Time-resolved spectra are obtained by using a time-to-amplitude converter started as the beam is turned off and stopped by the arrival of the photons. The output is proportional to the delayed arrival time of the photons. Each photon arrival delay is digitized and stored in an event-mode computer. The beam current showed a measured 90%-10% decay time of less than 20 ns. When the beam is turned off, direct excitation ceases immediately, as shown by the steep shoulder appearing in the recorded x-ray signal, and the slow exponential decay of the metastables stretches clearly into the excitation-free period (cf. Fig. 1). To turn off the beam in a time much shorter than the lifetimes of interest, a negative high-voltage pulse is applied to the focus electrode of the electron gun, effectively gating the cathode emission.

Turning off the electron beam means losing the radial electrostatic potential of the electron beam space charge, which is the essential element of the electron beam ion trap method. The loss rate would have to be measured and accounted for, if ion losses occurred. Fortunately, the trap behaves without the electron beam exactly like a Penning trap—the so-called magnetic trapping mode [26]. The electrostatic potentials of the three drift tubes and the 3-T axial magnetic field confine the ions in a volume with a larger diameter than in the case when the electron beam is on. When the electron beam is turned on again, the x-ray signal does not recover to the full strength it had before the beam was turned off. Here, a reduction of around 7% of the signal was observed for 7-μs beam-off periods. In earlier measurements [27], more severe decreases were observed, as the beam-off periods were much longer (several milliseconds). This result does not mean that ions are lost from the trap. Ion cyclotron resonance measurements have shown trapping times in the magnetic mode many orders of magnitude longer than 7 μs [30–33]; also, the fact that lifetimes measured in the millisecond range agree very well with experimental results using other type of devices (e.g., Cl^{12+}, 21 ms$\pm$0.2 ms) shows that ions are not lost [34].

The reason for the reduction of the strength of the x-ray signal observed immediately after turning the electron beam back on is an expansion of the ion cloud as the electron beam is turned off due to the missing negative space charge. This expansion, caused by the ion-ion Coulomb repulsion, is governed by a slow cross-field diffusion of the ions in the presence of a strong magnetic field, which suppresses diffusion efficiently. Diffusion across magnetic field lines implies net momentum-changing collisions between the ions. Given the fact that the stored ions have similar masses and charges, the net momentum exchanged in a two-ion collision system results only in a slow drift of the center of mass of the collision system; i.e., one ion drifts out from and the other one drifts in towards the trap center. In addition, the ion density in the trap is low, and therefore the collision frequency is rather low. As a result, the ions remain within the detection volume of our detectors, even if the turn-off is as long as 1 s (orders of magnitude longer than in the present experiment), as demonstrated in [35]. Nonetheless, the slow expansion of the ion cloud results in a decrease of the overlap of the ion population with the electron beam when it is turned on again, and therefore the x-ray yield does not return to full strength instantly. The signal, however, does recover, as the cross-field diffusion is now reversed in direction by the renewed presence of the negative space charge of the electron beam.

The prompt decay of the nonmetastable levels happens simultaneously. This contribution is hidden in the prompt decay shoulder (see Fig. 1). The short time where this settling down takes place is left out of consideration in the data fits. In fact, we have studied the effect of including early times on the fit and found that only the very beginning affects the fit. The starting point did not affect the fit as long as the immediate time after the beam was turned off was excluded from the fit. The negative effect of electronic switching transients on the amplitude of the time-to-analog converter (TAC) signal is also reduced in this way.

The method of turning off the electron beam and using the magnetic trapping mode can in principle provide extremely precise lifetime data with moderate effort (use of several detectors, longer data acquisition times), as has been recently demonstrated [35]. The main contribution for a systematic error is the possible effect of competing mechanisms causing ion losses from the trap on a time scale commensurate to the metastable decay rate. We identify the most likely mechanism for losses of He-like ions in our trap as the recombination to the Li-like charge state through charge exchange with neutrals. After the electron beam is turned off, recombination of H-like and bare ions also present in the trap takes place at

TABLE I. Comparison of our measured value (703±4 ns) of the radiative lifetime of the $1s2s\ ^3S_1$ level in He-like S^{14+} with theory.

Reference	Calculated value (ns)	Difference from measured value (ns)
[1][a]	732.2	+29±4
[1][b]	721.4	+18±4
[1][c]	705.9	+3±4
[37]	698.8	−4±4
[38]	699.3	−4±4
[39]	701.3	−2±4

[a]Single-configuration Dirac-Fock approach.
[b]Multiconfiguration Dirac-Fock approach utilizing the no-pair Hamiltonian.
[c]Multiconfiguration Dirac-Fock approach including negative-energy states.

a low rate. The effect of charge exchange on lifetime measurements has been estimated in various investigations which before have concentrated on longer-lived radiative lifetimes where it matters more [26,36]. An upper bound for the rate of charge-exchange recombination at the present conditions can be estimated to be 0.5 s^{-1}, compared to a radiative decay rate of 1.4×10^6 s^{-1}. Thus recombination will not affect the lifetime measurement at the present level of precision.

By feeding series of rectangular pulses from a calibrated signal generator to the event-mode computer, the time scale was precisely calibrated to the ppm level. Since the data accumulation rate was in the order of 0.003 counts per cycle, pile-up and dead time effects are at the level of 10^{-4}.

A single exponential with constant background gave the best fit to the data obtained with the proportional counter and with the Si(Li) detector. The residuals of the fit of the proportional counter data are shown in Fig. 1. The result is τ=702 ns with a statistical uncertainty of 5 ns. Similarly, the fit of the Si(Li) detector data yields a result of τ=706 ns with a statistical uncertainty of 10 ns. The weighted average of the two independent results is τ=(703±4) ns. As discussed above, uncertainties from systematic effects, such as ion loss due to charge exchange or calibration uncertainties in reference time base, are negligibly small compared to the statistical uncertainty.

III. DISCUSSION

The present result can be compared to the earlier measurement of Bednar *et al.*, who utilized an ~50-MeV ion beam [2]. Their measured value is 706±86 ns, which is in excellent agreement with our value. The main difference is that our result is more than an order of magnitude more precise.

We can also compare our value to various theoretical calculations, as summarized in Table I. Indelicato [1] has made a systematic study of different physical processes influencing the radiative lifetime of the $1s2s\ ^3S_1$ level in heliumlike ions. Of particular concern was the effect of the negative-energy

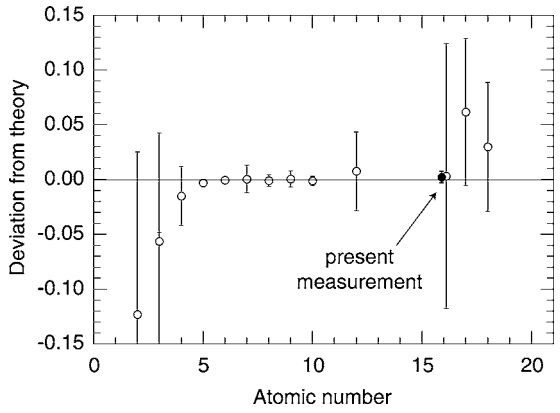

FIG. 2. Fractional difference of experimental measurements of the lifetime of the $1s2s\ ^3S_1$ level in various He-like ions and calculated values from Johnson, Plante, and Sapirstein [39], defined as $(\tau_{\text{theor}}-\tau_{\text{expt}})/\tau_{\text{theor}}$.

continuum. In the case of sulfur, he obtained lifetime values of 737.2 ns using the Dirac-Fock (DF) approach, 721.4 ns using the no-pair Hamiltonian and the multiconfiguration Dirac-Fock approach, and 705.9 ns by including the negative-energy continuum in the multiconfiguration Dirac-Fock approach [1]. Comparing with our result, it is clear that inclusion of the negative-energy continuum is indeed providing the best result, and unlike the others, this result agrees within our error bars. Agreement is also obtained with the calculation of Johnson and Lin [37] using the relativistic random phase approximation, who predicted 698.8 ns, with the calculation of Lin, Johnson, and Dalgarno [38], who also used the relativistic random phase approximation and who calculated 699.3 ns, and with the most recent relativistic calculation by Johnson, Plante, and Sapirstein [39], who obtained 701.3 ns. All three results are in excellent agreement with our measurement.

An overview of existing measurements is given in Fig. 2. Here we plot the relative difference between the theoretical and experimental values—i.e., $(\tau_{\text{theor}}-\tau_{\text{expt}})/\tau_{\text{theor}}$. The theoretical values are taken from the calculations of Johnson, Plante, and Sapirstein [39]. The plot illustrates that measurements with small uncertainty, including our measurement of heliumlike sulfur, generally are in very good agreement with the theoretical results.

IV. SUMMARY

The lifetime of the metastable $1s2s\ ^3S_1$ level in He-like S^{14+} was measured to a high precision using an electron beam ion trap in the magnetic trapping mode. The resulting lifetime value is (703±4) ns. Only a handful of other experiments have reached this level of experimental precision in the determination of lifetimes of metastable levels. The present measurement extends the range of lifetimes measurable with an electron beam ion trap, as illustrated in Fig. 2. Previously, the shortest radiative lifetime of an atomic level measured with this technique was that of heliumlike magnesium [24], which has a value near 13.6 μs. The present lifetime is in the nanosecond regime and thus lies within the

range historically only accessed by beam-foil spectroscopic methods.

ACKNOWLEDGMENTS

This work was supported in part by the NASA Astronomy and Physics Research and Analysis Program under Work Order No. S-06553-G and was performed at the University of California Lawrence Livermore National Laboratory under the auspices of the U.S. Department of Energy under Contract No. W-7405-ENG-48.

[1] P. Indelicato, Phys. Rev. Lett. **77**, 3323 (1996).
[2] J. R. Bednar, C. L. Cocke, BV. Curnutte, and R. Randall, Phys. Rev. A **11**,460 (1975).
[3] H. Gould, R. Marrus, and R. W. Schmieder, Phys. Rev. Lett. **31**, 504 (1973).
[4] C. L. Cocke, BV. Curnutte, and R. Randall, Phys. Rev. Lett. **31**, 507 (1973).
[5] R. Marrus *et al.*, Phys. Rev. A **39**, 3725 (1989).
[6] E. Träbert, Mon. Not. R. Astron. Soc. **297**, 399 (1998).
[7] R. W. Dunford *et al.*, Phys. Rev. A **41**, 4109 (1990).
[8] S. Cheng *et al.*, Phys. Rev. A **49**, 2347 (1994).
[9] A. Simionovici *et al.*, Phys. Rev. A **49**, 3553 (1994).
[10] M. H. Prior and H. A. Shugart, Phys. Rev. Lett. **27**, 902 (1971).
[11] M. H. Prior, Phys. Rev. Lett. **29**, 611 (1972).
[12] R. D. Knight and M. H. Prior, Phys. Rev. A **21**, 179 (1980).
[13] L. Yang, D. A. Church, S. Tu, and J. Jin, Phys. Rev. A **50**, 177 (1994).
[14] D. P. Moehs, D. A. Church, and R. A. Phaneuf, Rev. Sci. Instrum. **69**, 1991 (1998).
[15] D. P. Moehs, D. A. Church, M. I. Bhatti, and W. F. Berger, Phys. Rev. Lett. **85**, 38 (2001).
[16] S. J. Smith, A. Chutjian, and J. A. Lozano, Phys. Rev. A **72**, 062504 (2005).
[17] J. Doerfert, E. Träbert, A. Wolf, D. Schwalm, and O. Uwira, Phys. Rev. Lett. **78**, 4355 (1997).
[18] E. Träbert, G. Saathoff, and A. Wolf, J. Phys. B **37**, 945 (2004).
[19] E. Träbert, Phys. Scr. **T120**, 56 (2005).
[20] H. T. Schmidt *et al.*, Phys. Rev. Lett. **72**, 1616 (1994).
[21] E. Träbert, G. Gwinner, E. J. Knystautas, and A. Wolf, Can. J. Phys. **81**, 941 (2003).
[22] B. J. Wargelin, P. Beiersdorfer, and S. M. Kahn, Phys. Rev. Lett. **71**, 2196 (1993).
[23] E. Träbert *et al.*, Phys. Rev. A **60**, 2034 (1999).
[24] G. S. Stefanelli, P. Beiersdorfer, V. Decaux, and K. Widmann, Phys. Rev. A **52**, 3651 (1995).
[25] P. A. Neill, E. Träbert, P. Beiersdorfer, G. V. Brown, C. L. Harris, S. B. Utter, and K. L. Wong, Phys. Scr. **62**, 141 (2000).
[26] P. Beiersdorfer, L. Schweikhard, J. R. Crespo López-Urrutia, and K. Widmann, Rev. Sci. Instrum. **67**, 3818 (1996).
[27] J. R. Crespo López-Urrutia, P. Beiersdorfer, D. W. Savin, and K. Widmann, Phys. Rev. A **58**, 238 (1998).
[28] J. R. Crespo López-Urrutia, P. Beiersdorfer, K. Widmann, and V. Decaux, Can. J. Phys. **80**, 1687 (2002).
[29] D. Vogel, P. Beiersdorfer, V. Decaux, and K. Widmann, Rev. Sci. Instrum. **66**, 776 (1995).
[30] P. Beiersdorfer, B. Beck, S. Elliott, R. Marrs, and L. Schweikhard, Rapid Commun. Mass Spectrom. **8**, 141 (1994).
[31] P. Beiersdorfer, St. Becker, B. Beck, S. Elliott, K. Widmann, and L. Schweikhard, Nucl. Instrum. Methods Phys. Res. B **98**, 558 (1995).
[32] P. Beiersdorfer, B. Beck, St. Becker, and L. Schweikhard, Int. J. Mass Spectrom. Ion Process. **157/158**, 149 (1996).
[33] L. Schweikhard, P. Beiersdorfer, and E. Träbert, *Non-Neutral Plasmas Physics IV*, edited by F. Anderegg, L. Schweikhard, and C. F. Driscoll, AIP Conf. Proc. No. 606 (AIP, Melville, NY, 2002), pp. 164–173.
[34] E. Träbert, P. Beiersdorfer, G. Gwinner, E. H. Pinnington, and A. Wolf, Phys. Rev. A **66**, 052507 (2002).
[35] A. Lapierre *et al.*, Phys. Rev. Lett. **95**, 183001 (2005); A. Lapierre *et al.*, Phys. Rev. A **73**, 052507 (2006).
[36] P. Beiersdorfer, E. Träbert, and E. H. Pinnington, Astrophys. J. **587**, 836 (2003).
[37] W. R. Johnson and C. D. Lin, Phys. Rev. A **14**, 565 (1976).
[38] C. D. Lin, W. R. Johnson, and A. Dalgarno, Phys. Rev. A **15**, 154 (1977).
[39] W. R. Johnson, D. R. Plante, and J. Sapirstein, Adv. At., Mol., Opt. Phys. **35**, 255 (1995).