# Radiative Lifetime of the Long-Lived $1s2s\ ^3S_1$ State in Heliumlike Neon by Electron-Beam Excitation of Trapped Ions

B. J. Wargelin,[1,*] P. Beiersdorfer,[2] and S. M. Kahn[1]
[1]Department of Physics and Space Sciences Laboratory, University of California, Berkeley, California 94720
[2]Lawrence Livermore National Laboratory, University of California, P.O. Box 808, Livermore, California 94550
(Received 19 October 1992)

We report a technique for measuring the lifetimes of long-lived excited levels in highly charged ions that fall into the range from $10^{-2}$ to $10^{-7}$ sec, inaccessible to existing methods. Employing a fast-switching electron beam to produce and excite electrostatically trapped ions, lifetimes are determined by observing the fluorescent decay of metastable levels. A value of $90.5 \pm 1.5$ $\mu$sec is obtained for the $1s2s\ ^3S_1 \rightarrow 1s^2\ ^1S_0$ transition in heliumlike neon, in good agreement with theoretical predictions of 91.1 and 92.0 $\mu$sec.

PACS numbers: 32.70.Fw, 35.80.+s, 34.50.Fa, 34.80.Kw

Lifetime measurements provide important tests of atomic theory because radiative rates, unlike energy levels, are sensitive to the long-range behavior of atomic wave functions. These include tests of relativistic effects [1], two-photon decay [2], and the hyperfine interaction [3,4]. Moreover, precise knowledge of the lifetimes of long-lived metastable levels is crucial for proper application of density diagnostics used in astrophysics and plasma sciences. For neutral or singly charged ions, photon-fluorescence techniques may be used [5], while the accelerator beam-foil method has been very successful in measuring radiative decays shorter than a few hundred nsec in highly charged ions [6]. Additionally, retrapping of ions from an ion source was recently used for measuring metastable levels in highly charged ions whose lifetime exceeds several milliseconds [7]. For the specific case of the $1s2s\ ^3S_1$ metastable level in He-like ions, these methods have been employed to measure lifetimes in ions with $Z \leq 3$ [8,9] (for which $\tau \geq 58.6$ sec) and $Z \geq 16$ [1,3,10–14] ($\tau \leq 706$ nsec). Thus, a gap exists for heliumlike ions with $4 \leq Z \leq 15$ and metastable lifetimes between $10^{+1}$ and $10^{-6}$ sec, leaving He-like density diagnostics [15] between $10^6$ and $10^{14}$ cm$^{-3}$ untested.

Here we describe the first technique to measure excited-state lifetimes in multiply charged ions in the 5-order-of-magnitude range that cannot be measured with existing techniques, because the levels decay too slowly to be measured with beam-foil techniques, too fast to be studied by retrapping of ions from an ion source, and are too energetic to be excited by photopumping. It relies on high-resolution spectroscopy of stationary, electrostatically trapped ions excited by a monoenergetic electron beam. Our method of excitation is thus the inverse of the beam-foil method, and the radiative lifetime is determined from the temporal decay of the photon signal instead of from its spatial variation along a beam line. We illustrate this new technique for the case of $Ne^{8+}$ and determine the radiative lifetime of the $^3S_1$ level to be $90.5 \pm 1.5$ $\mu$sec. The precision of our measurement surpasses that of any other published $^3S_1$ lifetime, thus demonstrating the accuracy of the method.

This experiment consisted of three separate measurements performed on the Livermore electron-beam ion trap (EBIT) [16]. The beam energy was switched (1–2 kHz) between 960 and 750 eV, above and below the 905 eV excitation threshold of the $^3S_1$ level. The higher beam energy excites only the $n=2$ level, so corrections for radiative cascades from higher levels are unnecessary. No lines are excited at the lower energy.

A vacuum flat crystal spectrometer (FCS) employing a thallium-hydrogen-phthalate crystal ($2d=25.76$ Å) recorded the resulting $2 \rightarrow 1$ spectrum, shown in Fig. 1, which includes the resonance line $w$ ($1s2p\ ^1P_1 \rightarrow 1s^2\ ^1S_0$), the intercombination blend of $x$ and $y$ ($1s2p\ ^3P_{2,1} \rightarrow 1s^2\ ^1S_0$), the forbidden line $z$ ($1s2s\ ^3S_1 \rightarrow 1s^2\ ^1S_0$), and the lithiumlike resonance line $q$ ($1s2s2p\ ^2P_{3/2} \rightarrow 1s^2 2s\ ^2S_{1/2}$). The $xy$ blend is dominated by $y$, since the $^3P_2$ level has only a 2% branching ratio for decay via $x$. For two of the three measurements, we also used a Si(Li) detector which provided a much higher count rate, although the much lower spectral resolution

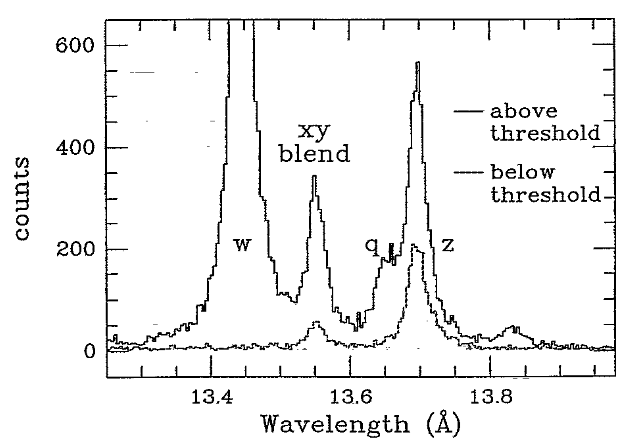


FIG. 1. Spectra of He-like Ne as seen by the FCS. Note how only $xy$ and $z$ remain when the beam energy is lowered below $z$'s excitation threshold. Both spectra are integrations of the FCS data shown in Fig. 2: "above threshold" from time $-150$ to 0 $\mu$sec and "below threshold" from 20 to 420 $\mu$sec.


0031-9007/93/71(14)/2196(4)$06.00

meant that $w$, $x$, $y$, $q$, and $z$ could not be individually resolved. The Si(Li) data also had a much higher background.

A data acquisition system recorded the spectrum as a function of time with a resolution of 1.6 $\mu$sec. The temporal behavior of the individual lines is shown in Fig. 2. The origin of the time axis is the point at which the electron beam energy is switched from 960 to 750 eV. The slew rate is 10 eV/$\mu$sec, and it thus takes about 20 $\mu$sec for excitation of $w$ and $q$ to cease. Line $z$ and the Si(Li) $2\rightarrow 1$ peak both show prominent exponential tails. A weaker tail also appears on the $xy$ blend. Other time-resolved spectra taken when no Ne was present showed that there were no other long-lived transitions and that the background was constant in time.

The $xy$ tail exists because of collisional quenching of the ${}^3S_1$ lifetime at the effective electron density of the measurement, i.e, because of transfer of an electron from the $1s2s\,{}^3S_1$ to $1s2p\,{}^3P_{2,1,0}$ levels. The ${}^3P_2$ and ${}^3P_0$ levels immediately decay back to ${}^3S_1$ (with branching ratios of 97.9% and 100%, respectively), but the ${}^3P_1$ level decays to ground via $y$ photon emission (branching ratio 96.5%).

One can eliminate collisional quenching by sufficiently reducing the beam current and thus the effective electron density. Alternatively, the effects of this collisional transfer may be accounted for by solving

$$dn_{\rm meta}/dt = -n_{\rm meta}(1/\tau_z + 1/\tau_{\text{coll-}xy} + 1/\tau_{\text{coll-}g}) , \quad (1)$$

and

$$dn_{xy}/dt = -n_{xy}/\tau_{xy} + n_{\rm meta}/\tau_{\text{coll-}xy} , \quad (2)$$

where $n_{\rm meta}$ is the number of metastable ${}^3S_1$ ions, $n_{xy}$ is the number of ${}^3P$ ions that decay to ground via $x$ and $y$,

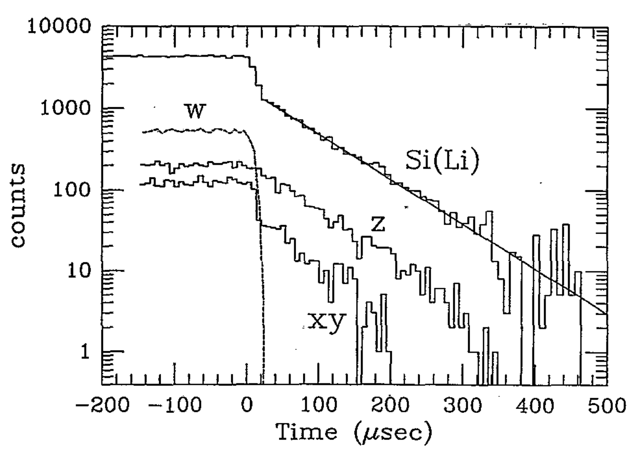


FIG. 2. Line intensities as a function of time (background subtracted). The FCS data ($w$, $xy$, and $z$) are from measurement 1, while the Si(Li) data is from 2. Time 0 marks where the electron-beam energy starts to drop from 960 to 750 eV. Excitation of the resonance line $w$ ceases after about 20 $\mu$sec. The exponential decay of the other lines beyond this point is apparent.

$\tau_z$ is the radiative lifetime of $z$, $\tau_{xy}$ is an average of the radiative lifetimes of $x$ and $y$, $\tau_{\text{coll-}xy}$ is the time scale for collisional redistribution from ${}^3S_1$ to ${}^3P$ that results in the production of an $x$ or $y$ photon, and $\tau_{\text{coll-}g}$ is the time scale for collisions from ${}^3S_1$ to any other levels that then decay to ground. Solving these equations and using the fact that the radiative lifetimes for $x$ (0.44 $\mu$sec) and $y$ (0.19 nsec) [17] are negligible compared to that for $z$ (the very small intensity of $x$ makes its lifetime effectively negligible), we obtain

$$\tau_z/\tau_{\text{coll-}xy} = I_{xy}/I_z , \quad (3)$$

and

$$1/\tau_{\rm decay} = 1/\tau_z + 1/\tau_{\text{coll-}xy} + 1/\tau_{\text{coll-}g} , \quad (4)$$

where $I_z$ and $I_{xy}$ are line intensities during decay and $\tau_{\rm decay}$ is the decay constant of the tails on both $xy$ and $z$. Combining Eqs. (3) and (4) we get

$$\tau_z = \tau_{\rm decay}[1 + (1+k) I_{xy}/I_z] , \quad (5)$$

where the constant $k = \tau_{\text{coll-}xy}/\tau_{\text{coll-}g} \ll 1$ is independent of density. Thus, the radiative lifetime of $z$ is equal to the decay constant of the exponential tails on $xy$ and $z$ multiplied by a correction factor arising from the collisional redistribution of ${}^3S_1$ to other levels (mostly ${}^3P$) that then decay to ground via photon emission.

We conducted measurements at the three effective electron densities listed in Table I. Each density was determined from the ratio $I_{xy}/I_z$ using a detailed model of the level populations in heliumlike neon constructed with the Hebrew University–Lawrence Livermore Atomic Code package (HULLAC) [18]. The populations were calculated from a balance of all collisional and radiative processes connecting the $n=2,3$ excited levels as well as the ground state.

Starting at the point where excitation of $w$ had ceased, the decay curves were fitted with an exponential term $Ae^{-t/\tau_{\rm decay}}$ plus a constant background. Standard $\chi^2$ methods were used to obtain the fits and their uncertainties, and double checked using the C statistic discussed by Cash [19]. A typical fit is shown in Fig. 2. The intensity ratios $I_{xy}/I_z$ for each measurement were obtained using the number of $xy$ and $z$ counts in the FCS exponential tails (with background subtracted), and then correcting for small differences in spectrometer efficiency between $xy$ and $z$. The results are listed in Table I.

Looking at Eq. (5), the uncertainty in our determination of $\tau_z$ includes contributions from statistical errors in $\tau_{\rm decay}$ and $I_{xy}/I_z$, systematic uncertainties in the measurement of these quantities, and uncertainties in the theoretical calculation of $k$. As we show in the following, all these possible errors are small, the largest arising from statistical uncertainties.

Because the electron-beam density varies with radius and because a fraction of each ion's orbit may lie outside the beam [20], we actually measure an average of $\tau_{\rm decay}$

TABLE I. Results of lifetime measurements and associated uncertainties.

| Measurement | 1 | 2 | 3 |
|---|---|---|---|
| Spectrometer | FCS | Si(Li) | Si(Li) |
| Electron density ($cm^{-3}$) | $8.7\times10^{11}$ | $5.7\times10^{11}$ | $2.9\times10^{11}$ |
| Total counts: background | 2352:86 | 21 324:8424 | 19 093:8240 |
| $I_{xy}/I_z$ (from FCS) | | | |
| Fit results | $0.218\pm0.010$ | $0.142\pm0.014$ | $0.073\pm0.011$ |
| Correction for spectrometer efficiency | $+0.006\pm0.004$ | $+0.004\pm0.003$ | $+0.002\pm0.001$ |
| Correction for polarization | $-0.005\pm0.002$ | $-0.004\pm0.001$ | $-0.003\pm0.001$ |
| Final value | $0.218\pm0.012$ | $0.141\pm0.014$ | $0.072\pm0.011$ |
| $\tau_{decay}$ ($\mu$sec) | | | |
| Fit results | $74.33\pm2.28$ | $78.56\pm1.68$ | $83.94\pm2.14$ |
| Timing calibration | $+0.00\pm0.38$ | $+0.00\pm0.50$ | $+0.00\pm0.53$ |
| Correction for $n_e$ profile | $+0.16\pm0.16$ | $+0.09\pm0.09$ | $+0.03\pm0.03$ |
| Correction for polarization | $-0.56\pm0.17$ | $+0.55\pm0.16$ | $+0.41\pm0.12$ |
| Correction for ion loss | $+0.05\pm0.05$ | $+0.05\pm0.05$ | $+0.05\pm0.05$ |
| Final value | $73.98\pm2.32$ | $79.25\pm1.76$ | $84.43\pm2.20$ |
| $\tau_z$ ($\mu$sec) | | | |
| Using Eq. (5) | $90.27\pm2.96$ | $90.54\pm2.32$ | $90.59\pm2.54$ |
| Uncertainty in $k$ | $+0.00\pm0.03$ | $+0.00\pm0.02$ | $+0.00\pm0.01$ |
| Final value | $90.27\pm2.96$ | $90.54\pm2.32$ | $90.59\pm2.54$ |

over different densities. This means that the decay curve we fit is a superposition of exponentials with different decay constants. Likewise, the intensity ratio $I_{xy}/I_z$ we measure is an average of many slightly different intensity ratios. Fitting multiexponentials to the data cannot resolve this effect, however, because of its small size. Modeling the superposition assuming a Gaussian beam profile, we find that fitting only one $\tau_{decay}$ to the decay curve and using the *effective* $I_{xy}/I_z$ will lead to an underestimate of $\tau_z$. We correct for this by increasing $\tau_{decay}$ by 0.16 (0.09, 0.03) $\mu$sec for measurement 1 (2, 3) and including an uncertainty of equal magnitude, as shown in Table I.

The interpretation of $I_{xy}/I_z$ and $\tau_{decay}$ is also affected by the polarization and anisotropy of the emitted line radiation since electron collisions in EBIT are nearly unidirectional [21]. Using the formalism of Alder and Steffen [22] and magnetic sublevel calculations from the code of Zhang, Sampson, and Clark [23], we compute the polarization of $z$ at $t=0$ to be $-10.5\%$ ($-11.7\%$, $-13.1\%$) in measurement 1 (2, 3) and conservatively assume a 30% relative uncertainty. Polarization of $z$ has opposite effects on the FCS and Si(Li) data. It decreases $I_z$ in the FCS spectra because the reflectivity of the TAP crystal for the perpendicular polarization component is less than that for the parallel component by a factor of 4.5 [24]; it increases $I_z$ in the Si(Li) data because the emission of negatively polarized magnetic dipole lines is enhanced perpendicular to the beam. The degree of polarization and anisotropy decreases slowly in time (fastest for the highest density measurement) because of collisional redistribution. $x$ and $y$ are assumed unpolarized. Modeling the time dependence of the polarization and using the formalism described in [25] to correct for polarization effects, we determine that the apparent values of $I_{xy}/I_z$ are too large by 2.5%, 3.1%, and 3.8%, respectively, and find that the apparent values of $\tau_{decay}$ are too high by $0.56\pm0.17$ $\mu$sec for measurement 1 and too low by $0.55\pm0.16$ ($0.41\pm0.12$) $\mu$sec for measurement 2 (3).

The apparent $\tau_{decay}$ may also be affected by the loss of $^3S_1$ states due to recombination and ion escape from the trap region. We estimated the time scale for these losses by observing Lyman-$\alpha$ emission from hydrogenlike neon ions. A population of H-like ions was first created by raising the beam energy above the ionization potential of He-like ions (1196 eV) but below that for ionization to bare nuclei (1362 eV). The beam energy was then dropped to just above the excitation energy of Ly-$\alpha$ (1022 eV). Under these conditions, the Ly-$\alpha$ intensity is proportional to the number of H-like ions in the trap, and can be used to measure the loss rate of H-like ions. We conclude that the time scale for loss of excited He-like ions is at least 60 msec, and that the required correction to $\tau_{decay}$ is no more than 0.10 $\mu$sec; we use $0.05\pm0.05$ $\mu$sec.

The final values for $\tau_{decay}$ and $I_{xy}/I_z$ are listed in Table I. Their uncertainties are calculated from the quadrature sum of statistical errors and the correction term uncertainties discussed above, including a 0.51% (0.63%) uncertainty in the timing calibration for measurement 1 (2 and 3).

The constant $k$, which appears in Eq. (5), depends on the time scale for collisions from $^3S_1$ to levels that then decay to ground other than through $x$ or $y$. We calculat-

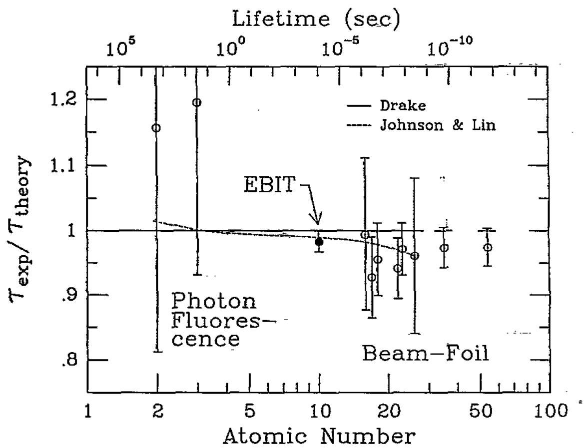


FIG. 3. Comparison of theoretical and experimental lifetimes for the $^3S_1$ level. All data are normalized to values calculated by Drake [26] except Br and Xe which are from Drake [26] with relativistic corrections (cf. Refs. [1,14]). Values calculated by Johnson and Lin [27] are indicated by a dashed line. Experimental values and error limits are from Refs. [8] (He), [9] (Li), [11] (S, Cl), [13] (Ar), [12] (Ti), [3] (V, Fe), [14] (Br), and [1] (Xe).

ed this with the HULLAC modeling package which yields a value of 0.0082, with an uncertainty of 20%. Using this value, we obtained the $\tau_z$'s shown in Table I.

Taking a weighted average we obtain a final result of $\tau_z = 90.49 \pm 1.48$ $\mu$sec, which has the smallest percentage uncertainty of any published $^3S_1$ lifetime measurement (see Fig. 3). This is in reasonable agreement with the value of 92.0 $\mu$sec calculated by Drake [26], and even better agreement with the more recent result of 91.1 $\mu$sec by Johnson and Lin [27] who included higher order relativistic terms in their calculation. This measurement represents the first test of a lifetime used in heliumlike density diagnostics applicable to stellar atmospheres, in this case, densities of $10^{11}$–$10^{13}$ cm$^{-3}$.

The discussion of errors shows that statistical uncertainties were dominant in this $Ne^{8+}$ measurement, but systematic errors may be much more important in other experiments. In particular, ion loss from the trap on time scales of roughly 100 msec (varying with the ionic species) places an upper limit on measurable decay rates. A lower limit of a few nsec is set by the slew rate of the electron-beam power supply. This represents a temporal range of some 8 orders of magnitude that is now available to measure lifetimes in highly charged ions. Five of those decades cannot be explored with any other existing method.

We gratefully acknowledge A. Osterheld for providing HULLAC atomic modeling results and K. Reed for calculating magnetic sublevel excitation cross sections. This work is supported by the California Space Institute and by NASA's X-Ray Astronomy Research and Analysis Program under Grant No. NAGW-2688 and was performed at LLNL under the auspices of the U.S. Department of Energy under Contract No. W-7405-ENG-48.

*Present address: Harvard-Smithsonian Center for Astrophysics, 60 Garden St., Cambridge, Massachusetts 02138.

[1] R. Marrus *et al.*, Phys. Rev. A **39**, 3725 (1989).
[2] R. Marrus *et al.*, Phys. Rev. Lett. **56**, 1683 (1986); R. W. Dunford *et al.*, Phys. Rev. Lett. **62**, 2809 (1989); R. W. Dunford *et al.*, Phys. Rev. A **38**, 5423 (1988).
[3] H. Gould, R. Marrus, and P. J. Mohr, Phys. Rev. Lett. **33**, 676 (1974).
[4] R. W. Dunford *et al.*, Phys. Rev. A **44**, 764 (1991).
[5] J. C. Bergquist *et al.*, Phys. Rev. Lett. **55**, 1567 (1985); W. M. Itano, J. C. Bergquist, R. G. Hulet, and D. J. Wineland, Phys. Rev. Lett. **59**, 2732 (1987).
[6] I. A. Sellin and D. J. Pegg, *Beam-Foil Spectroscopy* (Plenum, New York, 1972), Vol. 1.
[7] L. Yang and D. A. Church, in *Proceedings of the VIth International Conference on the Physics of Highly Charged Ions*, edited by P. Richard *et al.*, AIP Conf. Proc. No. 274 (AIP, New York, 1993), p. 549.
[8] H. W. Moos and J. R. Woodworth, Phys. Rev. Lett. **30**, 775 (1973); J. R. Woodworth and H. W. Moos, Phys. Rev. A **12**, 2455 (1975).
[9] R. D. Knight and M. H. Prior, Phys. Rev. A **21**, 179 (1980).
[10] C. L. Cocke, B. Curnutte, and R. Randall, Phys. Rev. Lett. **31**, 507 (1973).
[11] J. R. Bednar *et al.*, Phys. Rev. A **11**, 460 (1975).
[12] H. Gould, R. Marrus, and R. W. Schmieder, Phys. Rev. Lett. **31**, 504 (1973).
[13] G. Hurbricht and E. Träbert, Z. Phys. D **7**, 243 (1987).
[14] R. W. Dunford *et al.*, Phys. Rev. A **41**, 4109 (1990).
[15] A. K. Pradhan and J. M. Shull, Astrophys. J. **249**, 821 (1981).
[16] M. A. Levine *et al.*, Nucl. Instrum. Methods Phys. Res., Sect. B **43**, 431 (1989).
[17] G. W. F. Drake, Astrophys. J. **158**, 1199 (1969); **163**, 439(E) (1971); G. W. F. Drake and A. Dalgarno, Astrophys. J. **157**, 459 (1969).
[18] A. Bar-Shalom, M. Klapisch, and J. Oreg, Phys. Rev. A **38**, 1773 (1988).
[19] W. Cash, Astrophys. J. **228**, 939 (1979).
[20] M. B. Schneider *et al.*, in *Proceedings of the International Symposium on Electron Beam Ion Sources and their Applications*, edited by A. Hershcovitch, AIP Conf. Proc. No. 188 (AIP, New York, 1989), p. 145.
[21] J. R. Henderson *et al.*, Phys. Rev. Lett. **65**, 705 (1990).
[22] K. Alder and R. M. Steffen, in *The Electromagnetic Interaction in Nuclear Spectroscopy*, edited by W. D. Hamilton (American Elsevier, New York, 1975), pp. 1–38 and pp. 505–582.
[23] H. L. Zhang, D. H. Sampson, and R. E. H. Clark, Phys. Rev. A **41**, 198 (1990).
[24] E. Gullikson (private communication).
[25] P. Beiersdorfer *et al.*, Phys. Rev. A **46**, 3812 (1992).
[26] G. W. F. Drake, Phys. Rev. A **3**, 908 (1971).
[27] W. R. Johnson and C. Lin, Phys. Rev. A **9**, 1486 (1974).