Improved electron-beam ion-trap lifetime measurement of the Ne^{8+} $1s2s\ ^3S_1$ level

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An earlier electron-beam ion-trap (EBIT) lifetime measurement of the Ne^{8+} $1s2s\,^3S_1$ level has been improved upon, reducing the uncertainties to less than the scatter in the existing theoretical calculations. The new result, 91.7 ± 0.4 μs, agrees with the previous value, but is more precise by a factor of 4. The new value distinguishes among theoretical values, as agreement is obtained only with those calculations that employ ''exact'' nonrelativistic or relativistic wave functions. Routes to measurements with even higher accuracy are discussed. [S1050-2947(99)06009-6]

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I. INTRODUCTION

Quantum-mechanical methods are applied to a wide range of atomic, many-body, and low-dimensional solid-state problems. Quantum mechanics excels in describing a plethora of phenomena, but is far from doing so in arbitrary detail. In fact, 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 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 describe many-electron systems accurately. 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 a two-electron ion. Such lifetime measurements complement measurements of atomic structure, as lifetime measurements are sensitive to details of the wave functions that are not tested by energy spectroscopy.

The $1s^2\,^1S_0$–$1s2s\,^3S_1$ transition was first observed and identified in solar spectra [1], in spite of the long-standing theoretical view [2] that this transition would proceed only as a multiphoton process. Soon afterwards, the single-photon decay mode, arising solely from relativistic effects, was shown theoretically to be the dominant decay contribution. Today this is one of the most precisely studied decays, both experimentally and theoretically.

For two-electron systems, 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 nonrelativistic wave functions and relativistic corrections to energies [3], the Dirac-Hartree-Fock (DHF) approach [4,5], the random-phase approximation (RPA) [5,6], a relativistic framework with correlation [7], relativistic wave functions in various order approximations up to the use of ''exact'' relativistic wave functions [8], and fully relativistic single-configuration or multiconfiguration Dirac-Fock (SCDF or MCDF) calculations with correlation treatment in both energies and radial wave functions [9]. The latter treatment also included effects of the negative energy continuum, which has recently become of new interest in atomic structure calculations. The various calculational results for a given ion scatter by about 1% (neglecting some results that are even farther out). Part of this scatter may be due to imperfect wave functions, and, indeed, the results from using what are probably the most perfect wave functions, whether on a nonrelativistic [3] or a relativistic basis [8], agree well with each other in the range of low nuclear charge Z.

The experimental lifetime data on this transition (from neutral He atoms to highly charged Xe^{52+} ions) span 15 orders of magnitude (see Ref. [10]), but very few of these experiments have reached a precision that is comparable with the calculational scatter. This leaves the accuracy of the various theoretical wave functions still untested. An accuracy of better than 0.5% is needed for experiments in order to test theory. This level has recently been achieved by heavy-ion storage ring work [11] and by measurements using EBIT [12]. However, the data need to be extended in the range of ions covered, because contributions such as correlation and relativistic effects scale differently with nuclear charge Z, as do the results of various calculations.

X-ray spectroscopy of He-like Ne ions in an electron-beam ion trap (EBIT) provided one of the first precise lifetime numbers (at the 1.7% level of uncertainty) on a helium-like system [13]. Here we report on new results for He-like Ne^{8+} ions. Using EBIT, but with a different technique, we have improved the level of experimental precision in order to investigate higher-order effects in atomic structure and to discriminate between current, highly developed calculations.

II. EXPERIMENT

The measurements were carried out at Lawrence Livermore National Laboratory, using the electron-beam ion trap

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EBIT II. It was operated under UHV conditions ($p < 10^{-10}$ mbar), with Ne being bled into the trap continually via a leak valve and a set of collimating apertures in a ballistic gas injection system. Every 5 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.

We performed the experiment at two different energies for the electron beam: 1.1 keV, which is below the 1196 eV ionization potential of Ne^{8+} and permitted a beam current of 25 mA, and 3.3 keV, which permitted an electron-beam current of 52 mA and thus yielded a much higher signal rate. In physics terms, this means that at the lower energy only Ne ions up to charge $q=8+$ were produced, whereas at the higher energy some $q=9+$ and bare ions ($q=10+$) were produced as well and could contribute to the observed signal after recombination.

For the lifetime measurements, EBIT was operated in a cyclic mode with a period of about 2 ms. The electron beam was switched on for about 1 ms, ionizing and exciting the ion cloud in the trap. Then the beam was switched off, and the trap was maintained as a Penning trap in the so-called magnetic trapping mode [15]. EBIT in this mode has been shown to store ions for many seconds [15,16]. The photon energy and the time of each event were recorded in ''event mode'' by the data-acquisition system [17].

The switching time of the electron beam needs to be faster than the lifetime of the level of interest. To achieve this, we combined three switching processes: The electron-beam current was reduced to zero, the focus voltage (which normally helps to concentrate the electron beam and thus to achieve a high current density) was changed to 500 V, and the beam acceleration voltage was set to zero. All have a transient of about 30 μs. Further excitation of Ne^{8+} stops when the electron energy drops below threshold. However, the x-ray signal revealed not only a prompt initial drop of the x-ray signal (from short-lived levels) on the expected time scale, but also some ringing (a wiggle or two, of steeply decreasing amplitude, with a 20 μs period, clearly seen in the higher-energy data, and almost absent in the low-energy data), which might indicate that the electron current was not yet fully stopped and the energy not yet fully reduced, possibly also reflecting floating voltages of the drift tubes that constitute the actual trap. Consequently, the data evaluation was restricted to the part of the decay curves after the first about 30 μs for all data, and even further (about 50 μs) for the data that had been recorded after using higher voltages and that were thus suffering from more prolonged transients.

In order to monitor the charge state balance in the trap, two flat-crystal spectrometers [18], with TlAP crystals, were set up to observe the 1-2 resonance and intercombination lines in the He-like ion (13.447 Å and 13.553 Å , respectively), the transition of present interest at 13.699 Å , the 1-2 Li-like DR (dielectronic recombination) satellites near 13.65 Å , and the range almost up to the 1-3 transition (11.544 Å) in the He-like ions. At the higher electron-beam energy, the 1-2 transition (Ly_α) in the H-like ion ($\lambda = 12.134$ Å) showed with an intensity of about 10% of that of the 1-2 resonance transition in Ne^{8+}. Except for the line of interest, lifetimes for the other observed lines typically are in the picosecond range.

To carry out lifetime measurements at the intended level of accuracy, the signal rate from a crystal spectrometer is too low. Instead, a thin-window, energy-dispersive, solid-state x-ray detector [Si(Li)] was employed. The Si(Li) detector combines a suitably large solid angle of observation with sufficient spectral resolution and good timing properties. The signal rate was about three orders of magnitude higher than with the crystal spectrometer. The data were stored in event mode, requiring about 2 GB of disk storage for every 100 h of data collection. At the higher electron energy, about 10^6 counts were accumulated in the decay curve part of the data. Thus a statistical reliability of 0.1% was achieved within 50 h. At the lower energy, the signal rate was lower by about a factor of 5 and a total of about 70 000 registered events corresponds to a statistical limit on the precision of the lifetime results of about 0.4%.

Another important factor for accurate lifetime measurements is the quality of the established time base. A 10 kHz function generator was triggered by the same pulses from a function generator (HP 3314A) that triggered the event mode system. Time markers were thus generated every 100 μs. About 70 000 cycles were recorded, and the centroid of each marker distribution was thus determined statistically to better than 0.4% of the 4 μs full spread of each time mark. The time markers showed an integral variation by 0.025% between runs and a matching differential nonlinearity per 100 μs interval. Because of a shortage of suitable electronics units, this measurement was done only at the end of the data runs, and we therefore double the associated probable error in our error budget.

III. DATA ANALYSIS AND RESULTS

Every three to ten data runs were summed before evaluation. Typical Si(Li) data (representative for about 25 of such sets) are shown in Fig. 1. At the lower electron beam energy, the signal rate is much lower than at the higher energy, but the signal is practically free of any background and easily represented by a single exponential of 91.69 ± 0.4 μs (1σ statistical error estimate of the mean) that reaches over more than three decades. The general storage behavior of EBIT in the magnetic trapping mode has been tested earlier [15,16]. Typical trapping times of many seconds imply corrections for the present lifetime of less than 10^{-4}, and thus are negligible here. 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 50 μs each) have a notable influence compared to the statistical uncertainty at this level. Further truncation at the beginning of the decay curve removes the statistically most meaningful data and results in larger statistical uncertainties of the rest, yielding similar lifetime results but of lower precision. No second exponential component shows in the fits. Apparently, such data, collected for a considerably longer time, would offer the prospect of even more precise lifetime data. Possible systematic errors are discussed below.

It would be advantageous if one could reach a higher statistical reliability not just by collecting for a longer time, but by doing so at a higher data rate. A higher signal rate can be achieved with a higher electron current, which is available

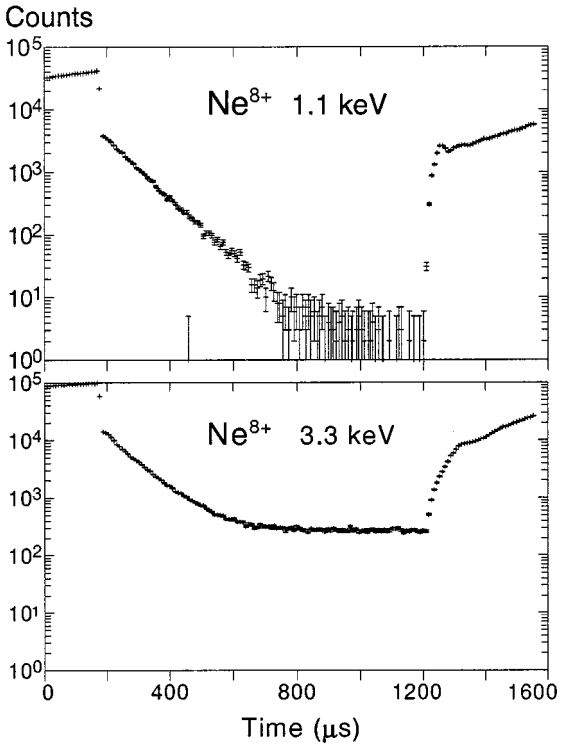

FIG. 1. Decay curve data (logarithmic scale) obtained at 1.1 keV (top) and at 3.3 keV (bottom) at the position of the $1s^2\,{}^1S_0$–$1s2s\,{}^3S_1$ transition in the Si(Li) x-ray spectrum of Ne. For the plot, the actual data have been grouped in bins of 16 channels each. The electron beam is switched off near $t=180$ μs and switched back on at 1200 μs. The initial very steep slope is due to the stopping of further excitation of levels with a short lifetime and indicates the effective switching time. The subsequent small bump in the data is evidence of some electronic ringing. Data evaluation is restricted to the later parts of the curve, between 200 and 1200 μs.

only at a higher electron beam energy. However, experiments at a higher electron energy than is necessary to produce the desired charge state (heliumlike Ne) may possibly introduce systematic errors that are not present at the lower electron beam energy. In fact, the decay curves recorded at the higher electron beam energy do have a superior statistical uncertainty (of the summed data) of almost 0.1%, but yield a lifetime (91.04 μs) that at face value is significantly different from the lower-energy data. The evident background in these data [present in both the Si(Li) (bottom of Fig. 1) and the crystal spectrometer data] limits the dynamic range of the signal to less than two decades. Such a background at the wavelength of the line of interest possibly arises from charge exchange processes (electron capture) involving Ne^{9+} or Ne^{10+} ions (neither of which are produced at the lower electron beam energy) and the rest gas in EBIT. The background has no discernible slope over the 1 ms time interval of our data and is therefore fitted by a constant function. Constrained fits with background slopes matching the multisecond storage times of ions in EBIT [15,16] did not resolve the lifetime discrepancy noted above. Again, no second exponential decay component could be retrieved from the data by the fit procedure.

The decay curves obtained with the crystal spectrometer have results with statistical uncertainties of 3% (high electron energy) and 17% (low electron energy) that are compatible with the Si(Li) data and corroborate the general appearance (with/without background), but are too weak to yield further insight.

The different lifetime fit results of the high- and low-energy runs relate to different systematic errors for the two experiments. The recognized physical and technical causes of systematic error apply mostly to the higher-energy data and need to be addressed in the course of future work. They include signal-to-background ratios, electronic switching, and unresolved satellite transitions.

The cleanliness of the practically background-free low-electron energy data with their stable fit results indicates that these data are less prone to systematic error than the higher electron beam energy data with their considerable background and the consequently much lower signal-to-background contrast. The presence of this background and the problem of establishing its slope with confidence require that one at least doubles the (statistical) uncertainty of the high-energy data.

The cleaner decay curves obtained with an electron energy not much above the excitation threshold are reminiscent of experiences with electron excitation of rare gases. There, Bennett and Kindlmann [19] showed that higher levels would be reached with higher electron-beam energies, and the subsequent cascades would easily cause a 30% error in the primary level lifetime to be extracted. However, it is well known that all cascading leads to apparent lifetimes that are longer than the true ones, whereas here we are facing a lifetime that appears shorter at high-energy excitation than just above threshold. Furthermore, the lifetime of the present level of interest is so much longer than those of reasonable cascades that such cascades would not matter much.

Spectral blends can mimic shorter or longer atomic lifetimes. However, the atomic excitation level of interest seems to be the longest-lived one in any of the systems of interest. In the He-like ion, all other low-lying levels with single-photon decay channels have lifetimes of less than a nanosecond.

At the higher electron-beam energy, the electron-beam current does not turn off completely for almost 100 μs, as was measured using an oscilloscope. Because of this and of the aforementioned electronic ringing as the beam is switched off, the excitation process does not stop as intended. Such imperfect switching may bring about a spectral contribution of prompt decays, due to the $1s2p\,{}^1P_1^o$ and $1s2p\,{}^3P_{1,2}^o$ levels in the He-like ion and the many satellite lines in the Li-like ion, to the data recorded on the decay of the long-lived $1s2s\,{}^3S_1$ level. This is because the Si(Li) detector cannot resolve these various transitions. The temporal behavior of this contribution would reflect the switching transit time for as long as the electron energy is above the excitation threshold. This in turn explains why no such contribution is seen when the original electron energy is barely above the production and excitation threshold (low-energy data), but also how such a spectral admixture of a more prompt decay is possible in the high-energy case. Though this contribution ought to die out with the truncation of early data channels, the present data do not show any such trend of

TABLE I. Results of calculations and measurements of the lifetime of the $1s2s\,^3S_1$ level in Ne^{8+}.

Reference	Comment	Lifetime (μs)
Theory		
Drake [3]	nonrel. wave functions	92.00
Johnson and Lin [4]	DHF	91.07
Johnson and Lin [5]	DHF	104.07
	RPA	90.91
Lin, Johnson and Dalgarno [6]	RRPA	90.91
Johnson, Plante and Sapirstein [8]	MCDF ''exact'' wave functions	91.58
Indelicato [9]	SCDF	99.35
	MCDF (no pair)	95.84
	MCDF (e^+e^-)	92.70
Experiment		
Wargelin *et al.* [13]	Electronic trapping mode	90.5 ± 1.5
This work	Magnetic trapping mode	91.7 ± 0.4

the average lifetime with truncation. Of course, the statistical significance of the truncated data is lower than that of the full data sample, and with progressing truncation it rapidly approaches that of the lower-statistics low-energy data. The additional constant decay contribution (present even in the spectrally well-resolved data from the crystal spectrometer) also hinders an evaluation with the full precision desired.

The shorter apparent atomic lifetime found at higher electron energies might also relate to an indirect effect of the beam energy, which is a higher electron-beam current, and the consequently higher density of electrons and ions (and thus higher collision frequency and possibly higher temperature) in the trap. Collisional quenching may affect long-level lifetimes in two ways, for the individual atom [a perturbed (damped) oscillator usually is reduced in lifetime] or for the number of atoms in the observed sample. Collisions that result in ion loss will reduce the number of ions in the trap. As discussed above, the typical ion storage time constants in EBIT [15,16] are so much higher than the wanted atomic lifetime that any correction would be insignificant. In the present experiment, varying pressure conditions (injecting Ne into EBIT at higher pressures, or opening a valve to a leaking spectrometer) affected the ratio of wanted events to those of the spectrally resolved satellite lines rather adversely. The Si(Li) spectra are not resolved enough to distinguish all of such satellites spectroscopically. However, the only recognized correlation of the extracted atomic lifetime is not with pressure (in this operating range), but that of high-energy versus low-energy data runs. This hypothesis of a density effect could in future experiments be tested by a variation of the electron-beam current at a given electron energy.

Taking systematic errors into account, we find a lifetime from the low-energy data of $[91.69\pm0.4$ (statistical) ±0.05 (systematic)] μs and from the high-energy data of $[91.04\pm0.2$ (statistical) ±0.5 (systematic)] μs. We adopt the result of the low-energy Si(Li) measurement as our final result, because here the systematic errors are under best control. For future precision measurements, however, it would be desirable to work at higher data rates and better statistical accuracy as offered by the high-energy measurements, once the systematics are better understood.

IV. DISCUSSION

The previous EBIT lifetime measurement on Ne^{8+} [13] (and Mg^{10+} [14]) had been obtained by different techniques: The level of interest is isolated enough so that the electron

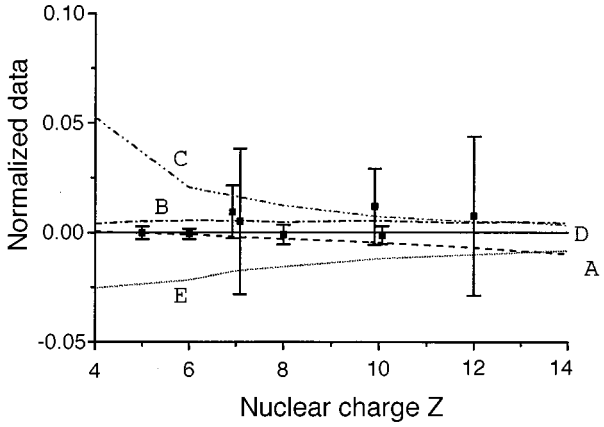

FIG. 2. Scaled experimental and selected theoretical transition rate data on the $1s^2\,^1S_0$–$s2s^3S_1$ transition in light He-like ions. The data are presented by the difference from the theory data by Johnson, Plante, and Sapirstein [8] and normalized to the latter. Experimental data for B^{3+} are from Schmidt [20], for C^{4+} from Schmidt *et al.* [11], for N^{5+} from Schmidt *et al.* [11] and Beiersdorfer *et al.* [15], for O^{6+} from Crespo López-Urrutia *et al.* [12], for Ne^{8+} from Wargelin *et al.* [13] and this work, and for Mg^{10+} from Stefanelli *et al.* [14]. Theory data are *A* (- -) [3], *B* (- · -) [4], *C* (- · · -) [6], *D* (—) [8], and *E* (. . .) [9]. Of the theory data shown, those from Drake [3] are based on nonrelativistic wave functions and are therefore not expected to work well for higher nuclear charge. For Ne they are still inside the experimental error bar, at the low side. The data given by Lin *et al.* [6] run on the high side while those by Indelicato [9] are rather low. 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 [8] is excellent, not only for Ne, but for all experimental data displayed.

beam energy could be set alternatingly just above and below the excitation threshold, while always actively producing a population of heliumlike ions in the trap. For Mg, the different Li-like satellite contributions were explicitly taken into account, while for Ne the energy modulation was sufficient to avoid the production of satellite lines in the measurement time interval. The present technique is much simpler, in that collisional excitation stops when the electron beam is switched off completely.

The new lifetime result, 91.7 ± 0.4 μs, is within the 1σ error bar of the previous data ($90.5 \pm 1.5 \mu$s). Obviously the present technique with its cleaner data (and supposedly fewer systematic corrections) is the better suited one for future precision measurements. At the present level of technology and EBIT performance, measurements of about 250 h of continuous running will be needed to reach 0.1% statistical uncertainty.

The new lifetime result is in the same range as the theoretically predicted data (Table I, Fig. 2). The new level of uncertainty 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, by now exclude the earlier calculations by Johnson and collaborators [4–6], as well as the calculations by Indelicato [9]. This, however, does not invalidate the negative energy state contribution included in the latter calculation. This contribution is much smaller than the discrepancy between the overall result of Indelicato's and the other calculations, so that different shortcomings may be suspected. Within this range of elements, the oldest calculation by Drake [3] (using very precise nonrelativistic wave functions) and the more recent fully relativistic calculation by Johnson *et al.* [8] (using ''exact'' wave functions) come closest to the experimental results. The latter is in excellent agreement with our result (and with the earlier data for lower nuclear charges). Following the isoelectronic trend, the nonrelativistic calculation by Drake is expected to fail at higher nuclear charges. The difference between these two calculations is larger than our present error bars for all elements heavier than Ne, so that decisive measurements seem in reach. A pathway to such better data has now been demonstrated.

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