Measurement of the N^+ $2s2p^3$ $^5S_2^o$ level lifetime using a heavy-ion storage ring

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The slow electric-dipole intercombination decay of the $2s2p^3\,{}^5S_2^o$ level in the C-like ion N^+, which appears in the auroral airglow spectrum, has been optically observed from ions circulating in a storage ring. A result of 5.88 ± 0.03 ms was obtained for the natural lifetime of this level. This represents an improvement in precision by a factor of 10 compared to previous radio-frequency and electrostatic ion trap work and is in slight disagreement with the latest theoretical values.
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I. INTRODUCTION

Auroral spectra, excited by particle bombardment of the upper atmosphere, show an emission feature near 214 nm. This emission was first identified as belonging to the radical NO, but was later found to be dominated by the $2s^2 2p^2\,{}^3P_{1,2} - 2s2p^3\,{}^5S_2^o$ intercombination (spin-changing electric dipole) transition of N II (the spectrum of the atomic ion N^+) [1–5]. Theoretical calculations of the transition probability [6–8] subsequently corroborated this interpretation qualitatively.

The level of interest, $2s2p^3\,{}^5S_2^o$, is the lowest excited level above the $2s^2 2p^2$ ground configuration in six-electron ions of the carbon isoelectronic sequence (see Fig. 1). The $^5S_2^o$ level mixes slightly with the $^3P_2^o$ and $^1D_2^o$ levels of the $2s2p^3$ configuration, which opens up the electric-dipole ($E1$) intercombination decay channels to the $2s^2 2p^2\,{}^3P_{1,2}$ levels (air wavelengths 213.90 and 214.28 nm [9]) and also (weakly, near 317 nm) to the $2s^2 2p^2\,{}^1D_2$ level [6].

Early experiments using radio-frequency ion traps [10–12] have measured lifetimes in the range suggested by various calculations. Working with an electrostatic ion trap, Calamai and Johnson [13] obtained the most precise lifetime result so far, $\tau = 5.4 \pm 0.3$ ms.

The $2s2p^3\ {}^5S_2^o$ level in C-like atomic systems and its decay branches have puzzled theorists and experimenters for quite a while. All the earlier calculations required adjustment of the results through calibrating with experimental atomic structure data. Only very recently have such calculations been developed far enough so that the required adjustments became small. Concerning the branching ratio in neutral carbon, finally experiment [14] and latest calculations converged.

For N^+ a number of calculations have been done (for references and detailed discussions of the theory problems see [15–17]). We note that extensive calculations achieved accurate results on the singlet and triplet levels and their decay probabilities, but yielded a lifetime prediction (6.45 ms) for the quintet level that almost coincides with that from early, simpler calculations and bears the authors' cautionary statement of being inaccurate [15]. This level then was reexamined systematically by Hibbert and co-workers [16,17]. Their computations increased in complexity by a progressively finer tuning of *ab initio* level energies to gross structure and to fine structure effects; also, an increase in the number of orbitals from just the $n=2$ states up to and including the $n=6$ states. In this process, the calculated lifetime changed by more than a factor of 2. The result from this set of calculations was a lifetime prediction of 5.43 ms, which matches the latest electrostatic ion trap result [13]. While error estimates for the theory indicated a 1% uncertainty fron the convergence behavior, doubts about the validity limits of the fine-tuning procedure led to the assumption of a larger error estimate, of about 5%.

In the same isoelectronic sequence, there are ion trap lifetime data on the next heavier ion, O^{2+}, by Johnson *et al.* [18], as well as recent theory results [19,20], which agree

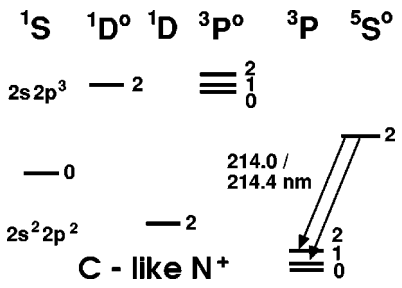

FIG. 1. Selection of the lowest levels of a C-like ion, indicating the $2s^2 2p^2\,{}^3P_{1,2} - 2s2p^3\,{}^5S_2^o$ $E1$ intercombination transitions of present interest.

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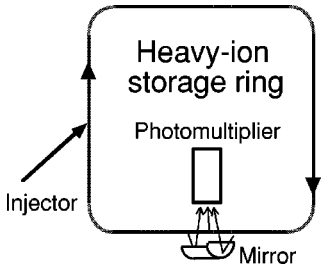

FIG. 2. Schematics of ion storage ring and optical detection aided by a light collecting mirror.

within the 7% (2σ) experimental uncertainty. This apparent agreement of experiment and theory, however, might be deceptive. For a related case, the $3s3p^3\,{}^5S_2^o$ level lifetime in the Si isoelectronic sequence, the theoretical problems were recognized and solved in a largely satisfying way only after an experiment [21] provided a benchmark at a low ionization stage system (see discussions in [22,23]). Therefore, an experimental check on the data for N^+, with an improved technique and the prospect of notably smaller uncertainties, seemed worthwhile. The 6-ms lifetime in N^+ lies in an excellent range for such measurement at existing heavy-ion storage rings.

II. EXPERIMENT

Our experiment employed a heavy-ion storage ring (TSR at Heidelberg) using the procedures described previously [24,25]. The basic layout is sketched in Fig. 2. Nitrogen ions were produced in the gas stripper of the tandem injector from CN^- ions extracted from the ion source. At an injector terminal voltage of 2.04 MeV, the energy of the N^+ ion fragments from the breakup of the CN molecule (by collisions with the stripper gas) is close to 1.1 MeV. At this energy, about 20% of the nitrogen ions are expected to leave the stripper as singly charged ions [26,27] and their total ion energy after the injector is 3.14 MeV. Experiments were done at this ion energy and also at 6.28 MeV. The lower of the two ion beam energies was chosen to maximize the yield of the desired charge state (and, it was hoped, also that of the excited level of interest) in the gas stripper of the tandem accelerator while keeping in mind the ion beam transmission to the storage ring. At the higher energy, the expected stripper yield of the desired charge state ions is substantially lower, but the ion beam transmission is higher so that higher ion currents can be stored. Furthermore, we were thus able to study possible systematic effects by significantly changing the ion beam and storage ring parameters.

The ion source was operated in a pulsed mode, so that for each operating cycle (every 200 ms) an ion beam pulse of a few milliseconds in length was extracted. This pulse length is much longer than is required to inject and store the ions and thus effectively a dc (continuous) beam is injected into the ring. In the storage ring the circulating ion beam was stacked, so that injection could proceed over about 30 turns. At 3.14 MeV, a current of 12 μA was stored in the ring, and at 6.28 MeV, the stored current reached 25 μA.

The storage ring was used basically as a large ion trap. The wide bundle of circulating ions was not cooled because expected cooling times for such low-charge state ions would be prohibitively long compared to the lifetime of the level of interest. The coasting ion beam was stored for 200 ms while optical observation took place. After this time interval the stored ions were dumped and the storage ring was refilled with fresh ions. This storage cycle matched about 30 atomic lifetimes of the level of interest, so that the optical decay curve could be observed well into the background.

Ions that experience charge-changing collisions leave the observation volume and this loss is monitored via the total beam current. The cross sections are small at MeV energies, in contrast to the much larger cross sections at the eV energies typical of more conventional ion traps. The ion beam lifetime depends on the background gas pressure of a few times 10^{-11} mbar in the storage ring; this pressure is lower than that typical for traditional radio-frequency or electrostatic ion traps by several orders of magnitude. The beam lifetime was found to be in the range of 7–9 s at both the lower and the higher ion energy. A small correction due to the beam loss rate has to be applied to the observed photon signal decay rate in order to obtain the natural lifetime. Under the present conditions, however, with a (roughly) 6-ms atomic lifetime, this is a minor (0.1%) effect. The collisional ionization cross sections for metastable ions are usually larger than those for ground state ions, by the ratio of the different binding energies. We neglect this difference, the beam loss rate representing already a small correction only. Fluorescence decay curves were also measured after deliberately worsening the local pressure by up to one order of magnitude. This was accomplished by switching off vacuum pumps in the storage ring and a reduction of the ion beam storage lifetime by about a factor of 3 was observed.

A few-percent fraction of the ion beam was expected to be in excited levels; the fraction originates from the stripping and excitation processes taking place in the gas stripper of the injector. The ion beam travels about 100 m from the injector to the ion storage ring, which at these ion energies takes 10 to 15 μs, comparable to the 8-μs revolution time of ions in the ring (at the lower energy). The full injection and storage process takes about 0.3 ms, a time interval short compared to the expected radiative lifetime of interest of about 6 ms, but long compared to most allowed cascade transitions. Higher-lying levels to which ions can be excited in the gas stripper mostly have lifetimes much shorter than the travel time to the storage ring; hence their population will have decayed to the ground state or to low-lying metastable and long-lived levels before observation begins and only ground configuration ions and those excited to the $2s2p^3\,{}^5S_2^o$ level will be left. Very high-lying hydrogenic levels with their very long natural lifetimes are expected to be quenched via Stark mixing in the motional electric field experienced by the ions in the magnetic fields of the beam transport system. Exceptional cascades might be those from other, rather low-lying levels of particular symmetries that have lifetimes as long as or even longer than the ones of interest. Candidates for such relatively low-lying, extremely long-lived levels would be the highest total angular momentum J levels of the $2s2p^2 3d$ configuration. Some of these can only decay by one or more $M1$ (forbidden) transitions within the shell before they can cascade feed the present level of interest. However, we do not find reproducible signs of such a contribution in our data. In any case, the primary lifetime is not affected

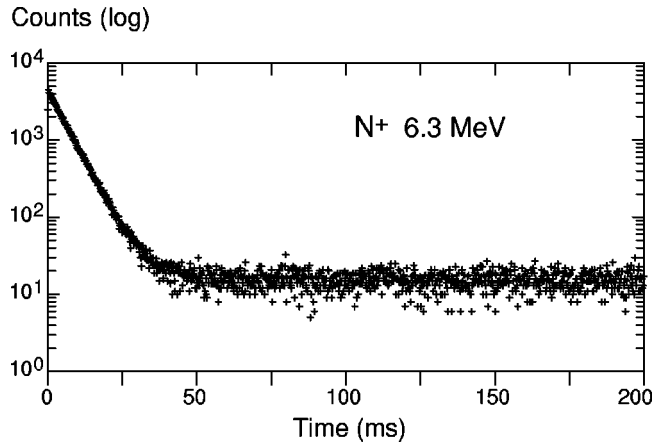

FIG. 3. Photon signal obtained with N^+ ions. The displayed data represent a typical 1.5-h run that collected data for 5.7 s per channel.

by the possible presence of very weak cascade components with time constants in the second range.

We used side-on observation (Fig. 2), through a sapphire window positioned at a distance of 5 cm from the average ion trajectory. In order to boost the signal rate, a light collection system was employed [24]. This simple trough-shaped reflector (oriented along the beam trajectory) enhances light collection mostly across the beam.

The light was detected by a 25-mm window diameter, EMR 541 *Q* photomultiplier tube with a dark rate of 1 count per second. This detector was used in combination with an interference filter with center wavelength 215 nm, transmission 40.6%, and bandwidth 42 nm. The two intercombination lines of interest in N^+ are at 214 nm, only 0.4 nm apart, and were not resolved from each other. Each detection cycle began within about 1 ms after injection and events were sorted into 1000 bins of 0.2 ms width each, fully covering each storage cycle.

III. DATA EVALUATION AND RESULTS

The high beam current values reached for N^+ ions yielded data sets with more than 2000 counts in the peak channel within an hour or two. The total signal reached up to order 10^5 counts per data set. A sample data set is shown in Fig. 3. No ion beam related background was found and the background in the signal curves was entirely due to the detector dark rate and electronics noise. Furthermore, the data show no discernible and reproducible cascade contamination. A number of such data sets were collected and evaluated individually. Most of these decay curves (8 at the lower, 12 at the higher energy) are sufficient to determine a lifetime value to better than 1%. The data were analyzed in raw form, subtracting a small correction for the ion beam decay rate from the obtained fluorescence decay rates; the correction for relativistic time dilation ($\gamma = 1.000\,24$ and $1.000\,48$, respectively) is negligible. Nonlinear least-squares fits of one and two exponentials were made on the full data sets and also on various subsets: These included truncation of the background tail, yielding no observable differences, and sequential truncation of up to 100 early data channels. This last option resulted in the largest variation of the lifetime results. Truncating the first few channels removes data channels that might be affected by the injection process and the subsequent stabilization of the coasting ion beam in the storage ring. With starting channels after the first 4 ms, the lifetime results nevertheless varied only within the statistical uncertainty of the individual data set. For a starting channel 4 ms after the injection, weighted averages of the corrected fluorescence decay rate obtained in fits of single exponentials to the individual data sets are $170.5 \pm 0.4\ s^{-1}$ at the low ion energy at the base vacuum pressure, $169.7 \pm 0.9\ s^{-1}$ at the same energy and the increased vacuum pressure (see above), and $169.8 \pm 0.3\ s^{-1}$ at the high ion energy and base vacuum pressure. The subtracted beam decay rate amounts to $0.13\ s^{-1}$ (or 0.08%, which we take as the maximum error due to pressure effects) at low pressure (both ion energies) and $0.4\ s^{-1}$ at high pressure. The results for individual data sets within each class agree within the statistical errors. Fits with two exponentials yielded spurious indications of a second exponential component in some of the data sets. The variation of the fit results indicates an uncertainty of 0.5%, which may be due to this or to cascade tail remnants.

TABLE I. Lifetime value τ for the $2s2p^3\,^5S_2^o$ level in the ion N^+.

τ (ms)	Ref.
Theory	
6.4	[6]
5.8	[7]
3.2 ± 1.6	[8]
6.45	[15]
5.43 ± 0.3	[16]
5.43 ± 0.3	[17]
Experiment using a radio-frequency ion trap	
4.2 ± 0.6	[10]
5.7 ± 0.6	[11]
Experiment using an electrostatic ion trap	
5.4 ± 0.3	[13]
Experiment using a heavy-ion storage ring	
5.88 ± 0.03	this work

The comparison of the data at two ion energies that are different by a factor of 2 (velocities and magnetic fields differing by a factor of $\sqrt{2}$, motional electric fields by a factor of 2) shows time constants that agree well within the error bars. Because the level of interest is fairly isolated and without fine-structure splitting, the expected influence of fine-structure mixing collisions, which might be mediated by the magnetic fields of the ion beam transport and storage system [24,25], is less than 10^{-7}.

From the results at the two ion energies we derive the final result for the radiative decay rate as their weighted mean. The above uncertainties combine to our final error of $\pm 0.5\%$ (1σ), while the purely statistical error amounts to only $\pm 0.2\%$. This yields for the natural lifetime of the $2s2p^3\,^5S_2^o$ level the value of 5.88 ± 0.03 ms.

IV. DISCUSSION

As listed in Table I, our lifetime result is slightly outside of the most recent experimental and theoretical error ranges, but in agreement with the earlier experimental finding of Johnson *et al.* [11]. We consider the improvement in precision and accuracy achieved by our experiment being due to a combination of a number of factors such as the spatial separation and independent optimization of ion production, selection, storage, and observation, as well as the use of better vacuum conditions and a less problematic regime of ion energies, causing lower perturbation by collisions.

In conclusion, our experiment demonstrates that optical detection at a heavy ion storage ring provides a precise way of measuring millisecond atomic lifetimes. The result obtained on the $2s2p^3\,{}^5S_2^o$ level lifetime in the C-like N^+ ion improves by one order of magnitude on the available experimental data from conventional ion traps and challenges the most recent theoretical data in terms of both precision and accuracy.

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