Metastable Ion Lifetime Studies Utilizing a Heavy-Ion Storage Ring: Measurements on Be^-

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The measurement of long (~10 μs–100 ms) ionic autodetachment lifetimes utilizing a heavy-ion storage ring is demonstrated with 10–125 keV beams of Be^-. The lifetime of the metastable $Be^-(2s2p^2\,^4P_{3/2})$ state was found to be 45 ± 5 μs, which represents an order-of-magnitude improvement in experimental accuracy over an earlier direct-beam-line experiment. This new result is a factor of 3 shorter than the recent theoretical value of 129 μs. New perspectives for storage-ring-based measurements of a variety of metastable lifetimes, both autoionizing and radiative, are briefly discussed.

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Storage rings have long been a tool of nuclear and particle physics. Recently storage rings have also been constructed for atomic and laser physics experiments [1]. Heavy ions can be stored for several seconds to facilitate the long-time observation of particles. A direct application of the long storage times is the measurement of ~10 μs–100 ms autodetachment (in general, autoionization) lifetimes of ions which are almost inaccessible via traditional beam techniques. The measurements of structural or dynamical properties of weakly bound negative-ion systems provide an excellent test of calculations on many-body systems in atomic physics and in particular the prominent role of electron correlation effects (for recent reviews, see Refs. [2,3]). In the present Letter we report on the possibilities of studying the lifetimes of long-lived metastable ions by utilizing a heavy-ion storage ring. The $Be^-(2s2p^2\,^4P)$ ion, with only five electrons, has been chosen as a model case, whereas detailed results on $He^-(1s2s2p\,^4P_J)$ and $Ca^-(4s4p^2\,^4P_J)$ will be reported elsewhere.

The Be^- ion was already known to exist more than 25 years ago [4]. The structural identification of the metastable $Be^-(2s2p^2\,^4P)$ ion (see Fig. 1) was the result of a series of experimental [5,6] and theoretical studies [7–10]. The absolute term value of Be^-, $2s2p^2\,^4P$, is known from theoretical calculations [11]. Properties such as the fine structure [10] and the lifetimes [12–14] of the three ^4P_J levels have also been considered theoretically. The ^4P_J levels decay via the spin-orbit-induced Coulomb autodetachment to the neutral $Be(^1S)$ ground state, with lifetimes predicted to be in the 0.1 μs to 2 ms region [12–14]. Bae and Peterson [5] in a beam-line experiment reported that the decay of metastable $Be^-(2s2p^2\,^4P_J)$ exhibits at least two components, one with a lifetime ~100 μs, which now can be attributed to the $^4P_{3/2}$ level, with the second decay being ~10 μs in duration.

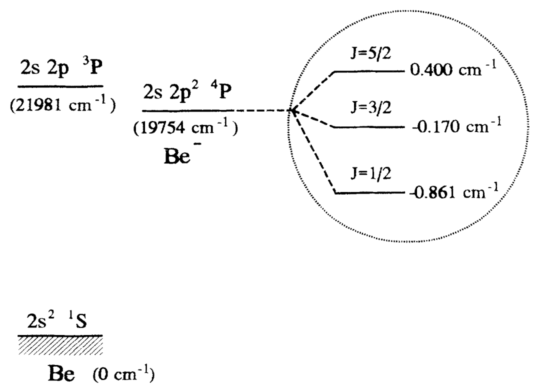

FIG. 1. Simplified energy-level diagram of Be and Be^-. The $Be^-(^4P)$ state is bound with respect to $Be(^3P)$ by 276 meV [11]. The theoretically predicted fine structure (J) splitting of the $Be^-(^4P)$ state [10] is illustrated in the inset.

Our experiments were conducted on the Aarhus Storage Ring in Denmark (ASTRID). The ring, which is kept at a vacuum of $\sim 3\times10^{-11}$ torr, has four straight sections of 10 m length, four bending magnets, and four groups of four correcting quadrupole magnets. Earlier experiments have characterized the storage times of a variety of stable negative ions with different electron affinities. All storage times were found to be in the range 1–5 s [15], indicating that lifetimes of metastable ions in the range 10 μs to 100 ms can be measured. The short-time limit is set by the round-trip time of the ions in the ring; the long-time limit, by the vacuum. A sketch of the storage ring setup for lifetime measurements is shown in Fig. 2. The positive ions are extracted from an ion source, accelerated to 10–125 keV energies, mass and charge-state analyzed, and passed through a 4-cm-long Na-vapor cell. After charge exchange, a negative-ion beam of ~20 nA is injected into the storage ring. The decay is measured with a 25-mm-diam tandem-channel-plate detector which monitors fast neutral atoms produced along one of the straight sections of the ring. This approach allowed monitoring the decay for several lifetimes of the metastable beam with a significant signal-to-noise ratio.

A typical plot of the neutral-atom signal versus time is shown in Fig. 3. The time interval between the data points depends on the operation mode of the storage ring [15]. In several of our runs, the detector was saturated initially due to a high count rate, but after several revolu-

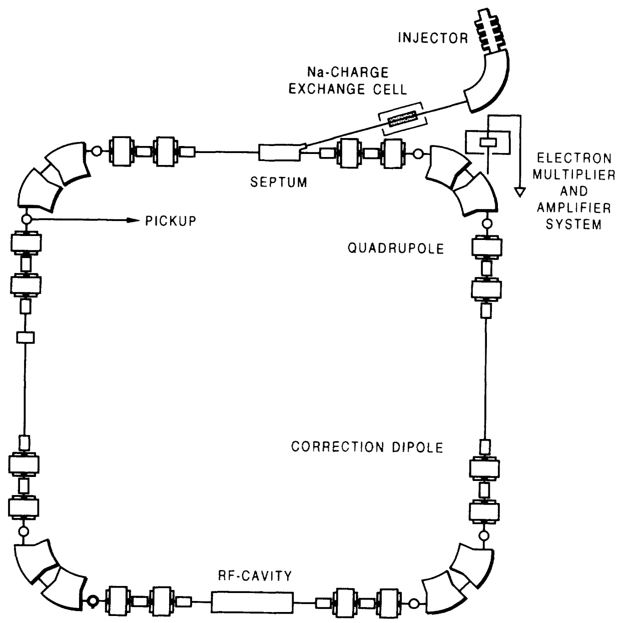

FIG. 2. Schematic of the Aarhus heavy-ion storage ring, showing the ion injection system which is equipped with a metal-vapor charge-exchange cell, and the single-particle detector setup at one corner of the ring.

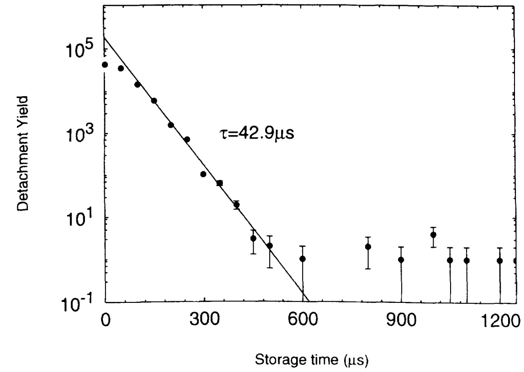

FIG. 3. An example of a plot of the autodetachment yield vs time on a semilogarithmic scale for a stored Be^- beam at 50 keV. The straight line represents the exponential fit. Statistical error bars are shown for experimental points for which the error bars are larger than the size of the dots. As discussed in the text, data from the first couple of revolutions are not utilized. For times greater than 600 μs, only electronic noise from the tandem-channel-plate detector is seen.

tions of the ring single-particle measurements without pileup could be performed. The oscillatory behavior overlaying the decreasing exponential is due to betatron oscillations with an amplitude $\sim$10–15 mm in the ring, which result in a varying detection efficiency of neutral atoms because of the finite size of our detector. The data could be accurately fitted by a single exponential, representing primarily the decay of the $^9P_{3/2}$ level, whereas the $J=\frac{5}{2}$ and $J=\frac{1}{2}$ levels have much shorter lifetimes [12–14]. The ability to obtain a good signal-to-noise ratio out to several lifetimes allows a determination with low statistical uncertainty, and effectively averages out the betatron oscillations in the ring. Slit scattering is virtually nonexistent after a couple of revolutions in the ring, as only particles which are well confined to the near-harmonic-oscillator potential remain circulating. Several checks were performed on the integrity of the data. The vacuum was degraded by a factor of more than 10 without any significant change in the measured lifetime. Tests for potential systematic effects comprise storage at several energies, wide variation in the beam current, and the demonstration of reproducibility of measured lifetimes from run to run.

The effect of magnetic fields in the ring cannot in general be neglected. The field in the dipole magnets is strong enough to mix magnetic substrates from different fine-structure components with the same M_J quantum number. In first-order perturbation theory, the mixing amplitudes will be proportional to the magnetic field strength (Zeeman coupling); thus the decay rate is expected to depend quadratically on the field strength [16], i.e., linearly on the beam energy at sufficiently low energies. Figure 4 shows the experimental results. It is apparent that the decay rate is well represented by a linear function of beam energy. The linearity allows a simple extrapolation to determine the zero-field limit. A least-squares fit to the data points with the form $\Gamma(E)=\Gamma_0(1+\beta E)$ gives $\beta=(1.5\pm0.7)\times10^{-3}$ keV^{-1} and $\Gamma_0=22.2\pm1.5$ ms^{-1} corresponding to a lifetime of $\tau_0(J=\frac{3}{2})=45\pm3$ μs. Here we assume that the long-lived state is, in accordance with theory [14], due to $J=\frac{3}{2}$. By in addition incorporating an estimate of potential systematic uncertainties, our final result for the $J=\frac{3}{2}$ lifetime is 45 ± 5 μs.

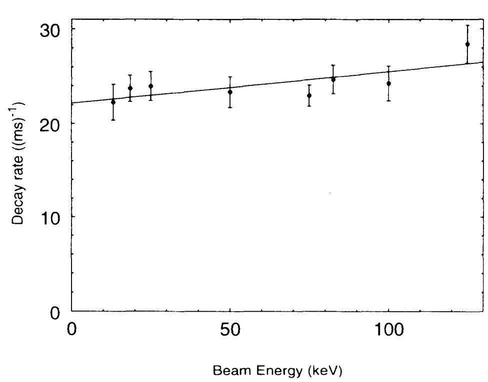

FIG. 4. Measured Be^- decay rate vs ion beam energy. Each point represents the average of several independent measurements, and the error bars indicate the statistical uncertainty of the mean. The finite slope is due to magnetic-field-induced fine-structure (J) mixing effects. The zero-field $J=\frac{3}{2}$ decay rate is 22.2 ms^{-1}, corresponding to a lifetime of 45 μs.

A simple analysis shows that the weak-field approximation assumed above is valid for magnetic fields B much smaller than a characteristic Zeeman parameter B_0,

$$B \ll B_0 = (1/a)(\Delta E/\mu_B),$$

where ΔE is a characteristic fine-structure splitting, a is a corresponding coupling constant (of the order of unity), and μ_B is the Bohr magneton. In $\mathrm{Be}^-(2s2p^{2\,4}P)$, where the fine-structure splittings have been predicted theoretically [10] to be 0.57 cm^{-1} ($\frac{5}{2}$-$\frac{3}{2}$) and 0.69 cm^{-1} ($\frac{3}{2}$-$\frac{1}{2}$), B_0 is of the order of 10 kG, which is much larger than the field applied (~300–1200 G) in the storage ring.

The magnetic-field-induced mixing depends in general on the M_J value of the level considered. This implies that magnetic sublevels are depleted at different rates and in turn that corresponding components in principle could be observed in the decay curve. This effect is, however, eliminated by the action of small inhomogeneities in the applied magnetic fields, and in particular by the in-plane component of the field in the quadrupole magnets on the straight sections of ASTRID. The quadrupoles are 30 cm long, and a field ~1 G is typically utilized to keep the beam in focus. A simple estimate shows that a transverse field component of that order is sufficient to effectively randomize the population over magnetic sublevels with respect to the direction of the strong fields of the bending magnets.

A general account of the field mixing effect requires a diagonalization of the Zeeman Hamiltonian and a dynamic treatment of nonadiabatic effects. Assuming that the magnetic fields in the ring can be considered to be switched on and off adiabatically along a beam trajectory (which should be a good assumption at present kinetic energies), we find in the weak-field limit that the effective decay rate of the $\mathrm{Be}^-(^4P_{3/2})$ state as a function of beam energy in keV is given by

$$\Gamma_{\mathrm{eff}}(E) = \Gamma_{3/2} + [(\Gamma_{5/2} - \Gamma_{3/2})\alpha_{5/2} + (\Gamma_{1/2} - \Gamma_{3/2})\alpha_{1/2}]E\,.$$

The constants α depend on the ring configuration and for ASTRID are given by $\alpha_{5/2} = 4.99 \times 10^{-6}$ keV^{-1} and $\alpha_{1/2} = 3.06 \times 10^{-6}$ keV^{-1}. The experimental slope parameter may accordingly be used to interrelate the widths of the two shorter-lived states. We find

$$\Gamma_{5/2} + 0.63\Gamma_{1/2} = 6.7 \pm 3.5\ \mu\mathrm{s}^{-1}\,.$$

The rather large uncertainty (1 standard deviation) related to the slope parameter, however, limits the applicability of this relationship between $\Gamma_{5/2}$ and $\Gamma_{1/2}$ as a critical test of theoretical calculations for $\mathrm{Be}^-(2s2p^{2\,4}P)$. An additional assumption about the relative widths is needed to extract lifetimes for $J=\frac{1}{2}$ and $\frac{5}{2}$ separately. *If*, for example, $\Gamma_{1/2} \sim \Gamma_{5/2}$, as predicted theoretically [14], then $\tau(J=\frac{1}{2},\frac{5}{2}) \sim 0.25 \pm 0.15$ μs. An overview of the various lifetimes, both experimental and theoretical, is given in Table I. The theoretical lifetimes of Brage and Froese Fischer [14] are about a factor of 3 larger than our experimental results. This is acceptable considering that the main purpose of the theoretical study [14] was to identify the different decay modes for metastable negative ions rather than optimizing the calculation of the specific lifetimes. Note in particular that the ratio between long and short lifetimes is correctly reproduced.

Since Be^- is bound by 276 meV with respect to the $\mathrm{Be}(2s2p\,^3P)$ threshold [11], blackbody radiation is not expected to have any significant influence on the decay rate even of the long-lived $J=\frac{3}{2}$ component. This was confirmed in separate measurements where the temperature of one-half of the storage ring was raised to 383 K, without observing any change in the lifetime. However, for longer intrinsic lifetimes, and particularly for electron affinities less than 100 meV, blackbody-radiation-induced photodetachment cannot be neglected. In fact, in our experiments on storage of Ca^- beams, blackbody-radiation effects played a dominating role by inducing photodetachment of *ground-state* $\mathrm{Ca}^-(4s^24p\,^2P)$ ions which are bound by only ~18 meV [17]. This effect complicated experiments aimed at measurements of the lifetime of the long-lived *metastable* $\mathrm{Ca}^-(4s4p^{2\,4}P_J)$ state [18]. Moreover, such blackbody-radiation effects should be taken into account in the experimental determination of the lifetime of $\mathrm{He}^-(1s2s2p\,^4P_J)$. As will be discussed thoroughly in a separate article, we find that this effect, which was neglected in earlier experiments [16], is ~20% for $\mathrm{He}^-(^4P_{5/2})$ at room temperature.

Our experimental approach to the determination of lifetimes should be applicable to a variety of positive and negative ions, including molecular and cluster species. The storage ring can also be a very sensitive tool in the search for a small fractional component of an autodetaching species in a beam. Consider an ion beam containing primarily a ground-state component, and say with an ~10^{-4} metastable fraction with a lifetime of ~100 μs. If the stable component storage time is ~10 s, the initial neutral-atom signal from the metastable decay would exceed that from the collisionally detached stable com-

TABLE I. Lifetime values in μs for $\mathrm{Be}^-(2s2p^{2\,4}P_J)$.

Reference	$J=\frac{1}{2}$	$J=\frac{3}{2}$	$J=\frac{5}{2}$
Theory			
Aspromallis and co-workers [12,13]	0.08	2000	1.0
Brage and Froese Fischer [14]	0.86	129	0.94
Experiment			
Bae and Peterson [5]	~10	~100	~10
Present work	0.25 ± 0.15 [a]	45 ± 5	0.25 ± 0.15 [a]

[a]Based on the assumption that the lifetimes of the $J=\frac{1}{2}$ and $\frac{5}{2}$ levels are essentially identical. See discussion in text.

ponent by an order of magnitude. In addition, states which are metastable with respect to radiative decay can be studied via neutral-particle detection by performing *time-delayed saturated photodetachment*. Here the time between ion injection and the triggering of a Q-switched laser is varied, and the laser wavelength is chosen so that the excited state is detached by absorption of a single photon, while detachment of the ground state represents a multiphoton process.

In conclusion, storage ring technology is shown to offer possibilities for detailed atomic structure studies in previously inaccessible regimes. In the present work, this capability was demonstrated with an accurate lifetime determination for the long-lived $Be^-(^4P_{3/2})$ state. Moreover, studies of magnetic-field-induced quenching effects were shown to provide additional information on the decay rates of the shorter-lived $J=\frac{1}{2}$ and $\frac{5}{2}$ levels of the Be^- ion.

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