Lifetime measurement of He⁻ utilizing an electrostatic ion storage ring

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The electrostatic heavy-ion storage ring, ELISA is utilized to measure the lifetime of the metastable $1s2s2p$ ⁴ $P_{5/2}$ level of the He⁻ ion together with the average lifetime of the ⁴ $P_{1/2}$ and ⁴ $P_{3/2}$ levels. This storage ring allows lifetime measurements to be performed without the influence of magnetic fields, and at temperatures below -50 °C, which reduces the photodetachment of He⁻ due to blackbody radiation. The lifetime of the ⁴ $P_{5/2}$ level is determined to be 365 \pm 3 μ s, in agreement with, but more accurate than, a previous magnetic storage ring measurement, and 6% longer than the lifetime recently reported utilizing an electrostatic ion trap [Wolf *et al.*, Phys. Rev. A 59, 267 (1999)]. The average lifetime of the ${}^4P_{1/2}$ and ${}^4P_{3/2}$ levels is determined to be 11.1 \pm 0.3 μ s, in agreement with theoretical predictions, but about 25% longer than the reported value from the electrostatic ion trap experiment.

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I. INTRODUCTION

The introduction of heavy-ion storage rings as a tool to study structural and dynamical properties of positive and negative ions has proven to be very beneficial (for recent reviews, see Refs. $[1-4]$. Conventional heavy-ion storage rings use magnetic bending and focusing elements to store the ions. One of the consequences of this is that the magnetic field can mix magnetic substates from different, but closelying, fine-structure components with the same M_I quantum number. This effect complicates both the measurements and the interpretation of the data in lifetime studies $[5]$. On the technical side, magnetic rings are usually large-scale devices with limited access to control, for instance, the temperature in connection with the study of the influence of blackbody radiation-induced photodetachment of weakly bound negative ions $[6]$. These difficulties have, among others, been avoided in the newly developed electrostatic storage ring ELISA (Electrostatic Ion Storage Ring, Aarhus) [7], in which heavy ions are stored using purely electrostatic deflection and focusing elements. An important fundamental difference between an electrostatic ring and its magnetic counterpart is that the strength of the bending forces ''felt'' by the ions depends on their kinetic energy and not on their momentum. This means that high-intensity beams can be used to set up the storage ring before turning to the desired lowintensity beam of a different mass as long as the charge to kinetic energy ratio is conserved, a feature which was very important in the first ELISA experiments $[8,9]$. Being a small ''table-top'' ring, ELISA is to some extent complementary to an ion trap in which ions are stored at very low energy and with no preferred direction of motion. The advantage of a storage ring is that the ion beam circulates at a well-defined kinetic energy and direction in space, which gives easy access to both the primary beam and possible decay products. A ''hybrid'' storage device, also based on purely electrostatic fields, has recently been developed by Zajfman *et al.* $[10]$, in which ions with an energy of a few keV are stored between two electrostatic mirrors.

In the class of metastable negative ions which do not form

a stable ground state, He^- represents the simplest system. It is formed in the $1s2s2p$ ⁴P state, which is bound by only 77 meV [11] relative to the $1s2s³S$ excited state of neutral helium. Due to the different strengths of the couplings to the continuum, there exist a differential metastability among the three fine-structure levels of the $1s2s2p^{4}P$ state. The *J* $=$ 5/2 component has a lifetime more than one order of magnitude longer than those of the two other components (*J* $=3/2$ and $1/2$). According to theoretical calculations by Brage and Froese Fischer [12], all the ${}^{4}P_J$ levels decay primarily via two-body relativistic interactions, i.e., autodetachment by spin-spin and spin-other-orbit interactions. The *J* $=1/2$ and $3/2$ levels can decay to the neutral He ground state either by direct relativistic or induced Coulomb autodetachment by emitting a p -wave electron (the latter by mixing with the fast decaying ${}^{2}P_{1/2,3/2}$ levels of the $1s2s2p$ configuration), whereas the $J = 5/2$ level only can decay by direct relativistic autodetachment by emitting an *f*-wave electron, resulting in a much longer lifetime.

Being a special prototype system, the dynamics of He have attracted a large amount of experimental $[5,13-18]$ and theoretical $[12,19-23]$ attention, and the reported lifetimes are summarized in Table I. Theoretical lifetimes of the ${}^{4}P_{5/2}$ level have been predicted in the range of $266-550$ μ s using various models, whereas only one calculation exists for the $^{4}P_{3/2}$ and $^{4}P_{1/2}$ levels, yielding 11.8 and 10.7 μ s, respectively [12]. The theoretical results depend critically on the precise form of the wave functions in both the initial and final states, and for the short-lived components $(J=3/2$ and $1/2$) the predictions are further complicated due to the coupling between the ${}^{4}P$ and ${}^{2}P$ levels of the same configuration. However, for the $J=5/2$ level an interaction Hamiltonian is explicitly known which simplifies the task. The most accurate calculation is considered to be that of Miecznik, Brage, and Froese Fischer [23], yielding $\tau_{5/2}$ =345 ± 10 μ s. This is in good agreement with the two most recent measurements of $\tau_{5/2}$ of 350 ± 15 and 343 ± 10 μ s conducted by Andersen *et al.* [5] using the heavy-ion storage ring Aarhus Storage Ring, Demmark (ASTRID), and by

Lifetime (μs)					
Determination	$J = 1/2$	Average	$J = 3/2$	$J = 5/2$	References
Experimental		18.2			$[13]$
		11.5 ± 5		345 ± 90	$[14]$
	16±4		10 ± 2	500 ± 200	$[15]$
		$9 \pm \frac{5}{3}$			$[16]$
		16.7 ± 2.5			$[17]$
			12 ± 2	350 ± 15	$[5]$
		8.9 ± 0.2		343 ± 10	$\lceil 18 \rceil$
		11.1 ± 0.3		365 ± 3	This work
Theoretical				266	$[19]$
				303/550	$[20]$
				455	$[21]$
				497	$[22]$
	10.7		11.8	405	$[12]$
				345 ± 10	$[23]$

TABLE I. Experimental and theoretical lifetimes of the three different fine-structure levels of He⁻. The column labeled "Average" represents the average values of $J=1/2$ and 3/2.

Wolf *et al.* [18] utilizing the new heavy-ion electrostatic trap device mentioned above, respectively.

In the ASTRID experiment $[5]$, the measured lifetime had to be corrected for the magnetic-field effect from the dipole and quadrupole magnets. This was done by measuring the lifetime of the ${}^{4}P_{5/2}$ level as a function of the beam energy, i.e., at different values of the confining magnetic fields in the ring. The negative-ion beam was produced from $He⁺$ ions at injection energy by two-electron capture in a Na vapor cell. After each injection, the neutral-atom signal stemming from autodetachment of the circulating ions was recorded as a function of time in one corner of the ring. A two-parameter fit based on a theoretical expression incorporating the Zeeman mixing in the dipole magnets was finally applied to extrapolate the data to zero magnetic field. This fitting procedure provided additional information on the lifetime of the ${}^{4}P_{3/2}$ level (12±2 μ s; see Table I), and thus the apparent disadvantage of the magnetically induced mixing effects in view of obtaining the lifetime of the ${}^{4}P_{5/2}$ level turned out to be an advantageous tool to explore the lifetime of another fine-structure component. The influence of blackbody radiation from the surrounding vacuum chamber on the measured lifetime of the weakly bound He^- ion was another important correction. Generally, for negative ions with electron affinities below \sim 250 meV, detachment due to blackbody radiation limits the storage time, depending on the actual binding energy, the temperature, and the photodetachment cross section [6]. In order to investigate this effect, the temperature dependence of the apparent ${}^{4}P_{5/2}$ lifetime was tested by heating one-half of the ASTRID storage ring to \sim 390 K, and the observed lifetimes were consistently reduced at all beam energies. At room temperature, it was found that blackbody radiation reduces the lifetime by \sim 20%.

In the most recent experiment on the He^- lifetimes performed by Wolf *et al.* [18], the application of purely electrostatic fields made magnetic-field-mixing effects negligible. A 4.2 -keV He $^-$ beam was injected into a new type of electrostatic ion trap. In short, the trap consists of two sets of electrodes working as two electrostatic mirrors between which the ions bounce back and forth. Neutral He atoms produced either in collisions with residual-gas particles or in the autodetachment process leave the trap through one of the electrodes, leading to a 50% detection in one end. The recorded signal showed two exponential decays, of which the fastdecaying component was assigned to the lifetime of the weighted average of the ${}^{4}P_{3/2}$ and ${}^{4}P_{1/2}$ levels of He⁻, and the slow-decaying component to the ${}^{4}P_{5/2}$ level. In the latter case, a value of 290 ± 2 μ s was found for the measured lifetime at room temperature. To correct for photodetachment due to blackbody radiation, the decay rate of 0.534 ms^{-1} from Ref. [5] was subtracted, yielding the final lifetime of 343 ± 10 μ s. The short-lived decay component, which is almost independent of the blackbody radiation, was found to have a lifetime of $\tau_{\langle 3/2,1/2\rangle}=8.9\pm0.2$ μ s. This value is shorter than in most previous experiments (see Table I), and more than 25% below the weighted average of the theoretical values reported by Brage and Fischer $[12]$, determined to be 11.4 μ s.

The electrostatic storage ring ELISA seems well suited for an investigation of the lifetimes of the $He^ (1s2s2p⁴P_J)$ levels, since it is possible to avoid the magnetic-field effects affecting the original $J = 5/2$ data from the magnetic storage ring $[5]$. Furthermore, operating the ring from room temperature to below -50 °C will make it possible to study the influence of blackbody radiationinduced photodetachment over a wide temperature range, leading to a more accurate $J = 5/2$ lifetime. The much smaller circumference of ELISA as compared to the magnetic storage ring ASTRID will also allow a measurement of the average lifetime for the $J=3/2$ and $1/2$ components, and thereby provide a test of the recent electrostatic ion trap value of 8.9 ± 0.2 μs [18].

FIG. 1. Schematic diagram of ELISA with injector. The He⁻ beam was injected from the lower left side, and neutral particles were detected by a MCP detector located at the upper left side.

II. LIFETIME MEASUREMENTS

A. Experimental procedure

The present experiment dealing with the He^- lifetime has been conducted with the purely electrostatic storage ring ELISA [7] shown in Fig. 1. The race-track-shaped ring has a circumference of 7.62 m and an average working pressure of 2×10^{-11} mbar. The electrostatic lattice confining the ions is defined by two 160° cylindrical deflectors, each having a 10° parallel-plate deflector and a quadrupole doublet for horizontal and vertical focusing on each side. In the two field-free straight sections of the ring, eight positionsensitive pickups are placed to monitor the circulating ions. Positive helium ions were created and extracted from a plasma ion source, and subsequently accelerated to the injection energy of 22 keV. After mass- and charge-state analyses in a separator magnet, the beam was passed through a 4-cmlong K vapor cell, where \sim 1% of the ions underwent twoelectron capture, producing a He^- beam of a few nA. The beam was chopped to match the circumference of ELISA, which in the present study corresponded to a pulse length of around 8 μ s. At injection, the first 10° deflector in ELISA was kept at zero potential while the ions passed through. The voltage needed for storage of the ions was then rapidly switched on at the moment when the front of the injected pulse had made one turn. Injection was performed with a repetition rate of 25 Hz.

Neutral He atoms created in the straight sections, either by rest-gas collisions or by autodetachment, will pass undeflected through the 10° deflectors and thus leave the ring. A microchannel plate (MCP) detector positioned opposite the injection side (see Fig. 1) counts the number of neutrals leaving the ring as a function of time after injection. The experimental conditions with respect to the He^- beam can be varied as far as, e.g., the beam energy, beam intensity, rest-gas pressure, and temperature of the ring are concerned. The temperature variation is the key point in order to explore the influence of blackbody radiation on the measured He^- lifetimes. As opposed to the ASTRID experiment $[5]$, the small size of ELISA makes it possible to cool the entire vacuum system down below -50 °C simply by building an isolating box around the ring and cooling the inside by inlet of liquid nitrogen. The temperature is measured at ten different positions around the ring using the average value as a representative value.

B. Experimental data

Figure 2 shows a typical neutral-atom signal versus time at room temperature. Two distinct exponential decays are

FIG. 2. Neutral He signal at room temperature from the MCP as a function of time after injection, here accumulated over 3528 fillings of the ring. The separation between two consecutive points is 8 μ s, corresponding to the revolution time in ELISA at 22 keV. The solid line is a fit to the data (see the text).

clearly observed, superimposed on a constant background consistent with the noise level of the detector. The experimental data, which cover an elapsed time of almost 3 ms after injection, are fitted with the sum of two exponentials and a constant, the result of which is indicated by the solid line in Fig. 2. A fitting value of 10.9 ± 0.1 μ s for the fast decay is assigned to the weighted average lifetime of the $^{4}P_{1/2}$ and $^{4}P_{3/2}$ levels of He⁻, whereas the slow decay stems from the ⁴ $P_{5/2}$ level, yielding 306.1 \pm 0.6 μ s. The uncertainties are purely statistical from the applied least-squares fit. However, for the $J=5/2$ component, the uncertainty has a larger contribution from small oscillations in the data points from revolution to revolution which is due to a nonuniform gain in the MCP detector. In a storage ring, a nonideally injected beam has an inherent small-amplitude oscillation, the so-called betatron oscillation, which results in a movement of the neutral beam spot on the detector. In the fitting procedure, these oscillations imply that $\tau_{5/2}$ changes a few thousandths depending on the range of the fit. For the data in Fig. 2, this results in a final value of $\tau_{5/2}$ =306.1±2.0 μ s.

Several tests were performed to check the robustness of the experimental data. The vacuum was degraded by an order of magnitude by switching off the four ion pumps in ELISA which, as expected from the earlier ASTRID experiment $[5]$, did not influence the measured lifetimes since the collisionally induced lifetime is several seconds. Furthermore, any dependence on the beam intensity was found to be negligible by changing the K-vapor density. The experimental results were reproduced at beam energies from 22 down to 4 keV, excluding any influence from for instance stray magnetic fields, or a possible stripping of the He^- ions by the electric fields of the lattice in ELISA. The lower limit in electric-field strength which could give rise to a reduction in the He^- lifetime due to static field detachment, can be estimated using the theory of electric-field dissociation applied by Nadeau and Litherland in studies of binding energies of

FIG. 3. He^{-4} $P_{5/2}$ lifetime data at eight different temperatures of ELISA. \circ , measured data. \bullet , data corrected for blackbodyradiation detachment. —, mean weighted value (MWV) of the corrected data. \cdots , uncertainty bands on the MWV. - - -, estimated data from the MWV with blackbody-radiation detachment included.

negative ions $\lceil 24 \rceil$ (and references therein). The survival fraction will be 100% as long as the He^- ions are exposed to field strengths below 5 MV/m during their entire storage time of several milliseconds. In ELISA, the maximum field strength amounts to less than 0.2 MV/m in the 160° cylindrical deflectors, which furthermore covers only \sim 20% of the circumference.

The assignment of the two decay components in the beam is based on a determination of the initial populations, taking into account the flight time from the charge-exchange cell to the detection region as well as the timing of the dataacquisition system. In all the lifetime spectra, the relative populations of the components are within 47–53 %. This is consistent with a population of the three fine-structure levels in accordance with their statistical weights, which may be expected after production by two-electron capture, and favors the interpretation that the fast-decaying component is indeed associated with the weighted average of the $J=3/2$ and 1/2 levels, and that their lifetimes are not very different.

The measured lifetimes of the $J = 5/2$ level at eight different temperatures are shown in Fig. 3 (open circles). The values range from 304.1 μ s at 22.1 °C to 347.0 μ s at -53.9 °C. At a given temperature *T*, the decay rate induced by photodetachment due to blackbody radiation can be calculated from

$$
\Gamma_{BB} = \int_{BE/\hbar}^{\infty} \sigma(\omega) \Phi(\omega) d\omega, \tag{1}
$$

where *BE* is the binding energy, $\sigma(\omega)$ is the photodetachment cross section, and $\Phi(\omega)$ denotes the distribution of thermal photons described by the Planck radiation law $[25]$:

$$
\Phi(\omega)d\omega = \frac{1}{\pi^2 c^2} \frac{\omega^2 d\omega}{e^{\hbar \omega/\mathbf{k} \mathbf{T}} - 1}.
$$
 (2)

FIG. 4. Decay rate of He^- due to blackbody-radiation photodetachment as a function of temperature calculated from Eq. (1) .

Values of $\Phi(\omega)$ are readily obtained from Eq. (2), whereas for $\sigma(\omega)$, accurate theoretical photodetachment cross sections up to photon energies of 0.6–0.7 eV above the He $(1s2s³S)$ threshold are needed to evaluate Eq. (1) numerically. Several theoretical studies were recently conducted on this subject (see Ref. $[26]$, and references therein), and Fig. 4 shows the calculation of Γ_{BB} in a temperature range from -100 to 100° C utilizing the total He⁻ photodetachment cross section of Liu and Starace [26]. The decay rate decreases from 570 to 126 s^{-1} in the measured temperature range. The result of a similar calculation using the cross section of Xi and Froese Fischer $\lceil 27 \rceil$ agrees with the data in Fig. 4 within a few thousandths, whereas the cross section of Saha and Compton $[28]$, used in the previous ASTRID experiment $[5]$, yielded a decay rate 5% lower.

Correcting the eight measured lifetimes of the ${}^{4}P_{5/2}$ level for the blackbody radiation-induced photodetachment, the zero-temperature values shown by filled circles in Fig. 3 are obtained. The main contribution to the uncertainty stems from the fitting procedure described earlier. Including an estimate of potential systematic uncertainties, a determination of the mean weighted value of all the independent zerotemperature values results in a final lifetime of $\tau_{5/2}$ =365 \pm 3 μ s, which is indicated by the solid line in Fig. 3. Finally, by adding Γ_{BB} to this value, it is possible to determine the expected measured lifetime as a function of temperature, which is given by the dashed line in Fig. 3. As opposed to this behavior of $\tau_{5/2}$, the short-lived component stemming from the decay of the weighted average of the $J = 3/2$ and $1/2$ fine-structure levels has been found to be practically independent of photodetachment from blackbody radiation. Taking the average of the eight measurements, the value of the lifetime is determined to $\tau_{\langle 3/2,1/2\rangle}=11.1\pm0.3\,\mu s.$

III. DISCUSSION AND CONCLUSION

The present results from the study of He⁻(1*s*2*s*2*p*⁴*P_I*) lifetimes with the electrostatic storage ring ELISA are listed in Table I, together with the values from previous experimental and theoretical investigations. For the $J=5/2$ level, the value of 365 ± 3 μ s is higher than, but in agreement with, the earlier magnetic storage ring experiment (350 ± 15 μ s [5]), and represents an improved accuracy by a factor of 5. This improvement can be accounted for by (i) the elimination of the mixing between the $J = 5/2$ and $3/2$ finestructure components due to the presence of magnetic fields; (ii) the performance of the lifetime measurements far below room temperature, which has reduced the correction of the lifetime due to blackbody radiation-induced photodetachment from approximately 20% to below 5% ; and (iii) an improved storage of the negative-ion beam in the storage ring.

The present lifetime, however, is 6% longer than the recently reported electrostatic ion trap value of Wolf *et al.* $(343\pm10 \ \mu s)$ [18]), and the deviation cannot be explained by the 5% lower Γ_{BB} used by Wolf *et al.*, since applying our room-temperature value of 570 s^{-1} to their data only increases the lifetime \sim 4 μ s. The deviation seems most likely to be due to a less perfect storage of the He^- ions or an unidentified loss mechanism in the ion trap experiment. We have been able to reproduce the ion trap result at room temperature by using worse storage conditions for the negative ions (equivalent to large beam oscillations in the present study) than those applied in the final experiments. A poor storage of the negative ions will always result in a shorter lifetime than the true one, since a $(s$ mall) fraction of the ions may be removed from the circulating beam due to interaction with parts of the apparatus.

With respect to the most recently calculated value (345 ± 10 μ s [23]), the present lifetime is also longer, but it should be noted that the calculation was performed just after the lifetime from the magnetic storage ring experiment became available. The theoretical value is very sensitive to the electron correlation used to describe the initial wave function for the ${}^{4}P_{5/2}$ state, and a minor modification of the wave function will bring the calculated lifetime into agreement with the experimental one.

A comparison of the weighted average lifetime for the *J* $=3/2$ and $1/2$ fine-structure components obtained in the present experiment and the ion trap experiment also exhibits a deviation, with the present one 25% longer than the trap value (11.1 \pm 0.3 μ s versus 8.9 \pm 0.2 μ s), and with both experiments claiming an accuracy of only 3%. Even though the data analysis of this average lifetime will be influenced by the lifetime value of the longer-lived $J = 5/2$ fine-structure component, a change of this value from 343 to 365 μ s is not sufficient to explain the discrepancy. The deviation is most likely due to the same reasons, or to one of these, discussed above in connection with $\tau_{5/2}$. The present average lifetime is in good agreement with both the $J=3/2$ lifetime reported from the ASTRID experiment (12 ± 2 μ s [5]) and with the only calculation available (11.4 μ s [12]).

The present study has shown that the electrostatic storage ring ELISA offers improved possibilities for studies of negative ions, e.g., with respect to lifetime studies of atomic and molecular ions $\lvert 8 \rvert$. Supplied with an electron target it will provide a good alternative to the larger magnetic rings. The mass independence allows for the storage of very heavy systems of biophysical interest, e.g., proteins, which is presently in progress. The cooling scheme applied here to reduce the blackbody radiation-induced photodetachment will be further optimized in the future with respect to nitrogen consumption and the lowest possible temperature. Another important consequence of the cooling is that the residual-gas pressure can be reduced to around 10^{-12} mbar, resulting in storage lifetimes of several minutes for beams consisting of stable ions. Finally, with its ''table-top'' size, it may also be possible to perform experiments combining synchrotron radiation from the magnetic storage ring ASTRID with stored heavy ions in ELISA.

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