Precision Lifetime Measurements of He^- in a Cryogenic Electrostatic Ion-Beam Trap

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We have developed a small purely electrostatic ion-beam trap which may be operated in thermal equilibrium at precisely controlled temperatures down to 10 K. Thus, we avoid magnetic field induced mixing of quantum states and may effectively eliminate any influence from absorption of photons from blackbody radiation. We report the first correction-free measurement of the lifetime of the $1s2s2p\ ^4P^o_{5/2}$ level of $^4He^-$ yielding the high-precision result $359.0 \pm 0.7\ \mu$s. This result is an essential proof-of-principle for cryogenic electrostatic storage rings and traps for atomic and molecular physics.

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The He ground state cannot bind an additional electron to form a stable bound state. However, the He^- ion has been reported in beam experiments since 1925 [1]. In 1955, Holøien and Midtdal [2] established theoretically that the metastable $1s2s2p\ ^4P^o_J$ state in He^- is bound with respect to the lowest excited state of neutral He, $1s2s\ ^3S^e$ (cf. Fig. 1). Since then, results have been reported from both experimental [3–13] and theoretical studies [11,14–22] aimed at a more refined description of the energetics and metastabilities of this intriguing, highly correlated, He^- state. In general, doubly excited three-electron systems attract significant attention as precise measurements of their various properties are particularly well suited to test theoretical descriptions of electron-electron interactions. In this context, He^- takes a special place as the noncentral parts of the interaction potential for the electrons are much more important on a relative scale than in the corresponding neutral and positively charged atomic systems. Furthermore, due to the weak interaction with the nucleus, the wave functions of He^- extend to large distances, and as subtle details in atomic wave functions may control the corresponding decays, calculations of the relaxation of excited states in He^- are particularly demanding and important.

In Coulomb autodetachment processes, the total angular momentum quantum number, J, the parity, and the total energy of the system are conserved. In addition, as spin-orbit interactions are weak, the total orbital angular momentum, L, and the total spin, S, are also fairly good quantum numbers. For energetic reasons, the $1s2s2p\ ^4P^o_J$ state in He^- can only autodetach to the neutral He ground state $1s^2\ ^1S^e$, and thus the only available continua are $1s^2\epsilon l\ ^2L$ with parities $(-1)^l$ and total angular momenta $J = l \pm 1/2$. Thus, neglecting weak radiative processes, the $1s2s2p\ ^4P^o_{1/2}$ level can only decay to the $1s^2\epsilon p\ ^2P^o_{1/2}$ continuum, $1s2s2p\ ^4P^o_{3/2}$ only to $1s^2\epsilon p\ ^2P^o_{3/2}$, and $1s2s2p\ ^4P^o_{5/2}$ only to $1s^2\epsilon f\ ^2F^o_{5/2}$. In the former two cases, autodetachment is governed mainly by small admixtures of the rapidly autodetaching $1s2s2p\ ^2P^o_{1/2}$ and $^2P^o_{3/2}$ levels in the respective $J = 1/2$ and $J = 3/2$ levels of the quartet state (this process is often referred to as relativistically induced Coulomb autodetachment). For the $^4P^o_{5/2}$ level, autodetachment leads to a final state in which neither L nor S is conserved. Also, here Coulomb autodetachment may be relativistically induced through the very small admixtures of the $1s2p(^1P)3d\ ^2F^o_{5/2}$ and the $1s2s(^1S)4f\ ^2F^o_{5/2}$ [20] levels, which have the same J and parity as $^4P^o_{5/2}$ and the $1s^2\epsilon f\ ^2F^o_{5/2}$ continuum. However, the main contribution to the decay is due to spin-spin and spin-other-orbit interactions (i.e., to *direct* relativistic couplings) between the $^4P^o_{5/2}$ level and the $^2F^o_{5/2}$ continuum. It has been known for quite some time that the $J = 1/2$ and $J = 3/2$ levels have lifetimes of the order of 10 μs, and that the lifetime of the $^4P^o_{5/2}$ level is 30–40 times longer. However, all experimental studies up to this point have been performed under conditions requiring substantial corrections for instrumental effects and direct, high-precision measurements have until now not been possible.

Here, we present the first direct high-precision measurement of the $^4P^o_{5/2}$ lifetime, where we have effectively eliminated all instrumental influences on the result by using a cryogenic, compact, and purely electrostatic ion-

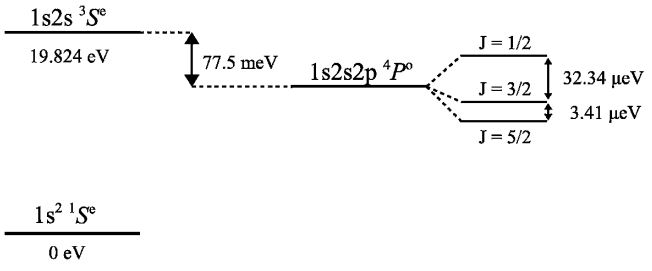

FIG. 1. Schematic energy diagrams of the ground state and the lowest excited state of ^4He, and the $1s2s2p\ ^4P^o$ state of $^4He^-$ [7,9,11].

beam trap. In [23] (and references therein), the design of larger, cryogenically operated, ion storage rings are described. Cryogenic traps and rings are very useful for studies of stabilities of, and reactions with, loosely bound atomic and molecular systems. Such methods are essential when photodetachment by blackbody radiation influences ion stabilities under normal (room temperature) laboratory conditions.

The electrostatic ion-beam trap, ConeTrap [24], consists of two cone-shaped electrodes, where the cone openings face each other from opposite sides of a centered, grounded, box-shaped electrode (cf. Fig. 2). The trap, of total length 175 mm, is mounted inside a cryogenically cooled aluminum vacuum chamber, surrounded by a copper heat shield and an outer stainless steel vacuum chamber. This arrangement and a resistive heater allow us to regulate the common trap and inner chamber temperature to within ± 1 K down to 10 K and, furthermore, to keep the system in thermal equilibrium. When ions are injected in the trap, the entrance cone electrode is kept on ground potential, and the exit cone on a potential which is slightly higher than the 2.5 kV beam acceleration voltage. The ion beam is reflected, *and focused*, by the electric field between the center electrode and the exit cone. During this reflection, the entrance cone is switched (rise time $\approx$100 ns) to the same value as the exit cone, thus trapping the 0.8 μs long He^- ion pulse. The trap is operated in the storage mode [24] giving trajectories that fill the volume indicated in the lower part of Fig. 2 (calculated by means of the SIMION package [25] for a 4 mm diameter ion beam). In order to shield the stored He^- ions from thermal radiation

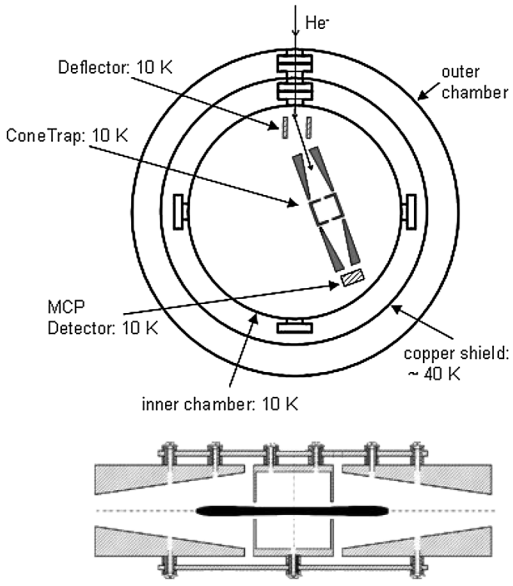

FIG. 2. Upper: A schematic of the chamber with ConeTrap. The inner chamber, ConeTrap, the deflector (and a 4 mm diameter tube in front of it), and the detector are thermally anchored to the cold stage of the cryogenerator and may be cooled down to 10 K. Lower: Expanded view of ConeTrap illustrating simulated ion trajectories (cf. text).

from the beam line, the trap is mounted at an angle of 20° with respect to the incident beam (Fig. 2). Neutral He atoms leaving the trap through the hole in the exit cone are detected by a microchannel plate (MCP) detector, and we obtain decay curves through the rate of detected neutrals as a function of time. Note that the recordings of the decay curves are relative measurements, and hence knowledge of the exact value of the detection efficiency (about 50% for 2.5 keV He) is not needed for the extraction of the lifetimes. Recently, MCP detectors have been successfully tested under cryogenic conditions [26].

The $^4He^-$ ions are produced in double collisions ($^4He^+ + Cs \rightarrow {}^4He^*$, $^4He^* + Cs \rightarrow {}^4He^-$) of 2.5 keV $^4He^+$ in a cesium charge exchange cell, which is out of sight from the ConeTrap interior. Approximately 0.1% of the He^+ ions are converted to He^- yielding typical currents of 50 pA. The period for the He^- ion motion in the trap is 1.6 μs, and injections are made at a repetition rate of 280 Hz, allowing 3.5 ms long measuring cycles with few He^- ions in the trap.

The measured decay rate for the population of a given J level is, in general,

$$\Gamma_{\text{measured}} = \Gamma(^4P_J^o) + \Gamma_{\text{BB}} + \Gamma_{\text{coll}} + \Gamma_{\text{ion-ion}} + \Gamma_{\text{fields}} + \Gamma_{\text{instr}} \quad (1)$$

where $\Gamma(^4P_J^o)$ is the true autodetachment rate, and Γ_{BB} is the rate due to photodetachment by blackbody (BB) radiation. Γ_{coll}, $\Gamma_{\text{ion-ion}}$, Γ_{fields}, and Γ_{instr} refer to the depletions due to collisions with the residual-gas, ion-ion scattering, electric and magnetic fields, and any other unknown instrumental effects, respectively. Here, we will argue that all terms on the right hand side of Eq. (1), except $\Gamma(^4P_J^o)$, are negligibly small for temperatures below 100 K.

The $1s2s2p\ ^4P_J^o$ state of He^- is bound by only 77.5 meV relative to the $1s2s\ ^3S^e$ state of He [9,11] (Fig. 1). Thus, any room temperature lifetime measurement needs to be

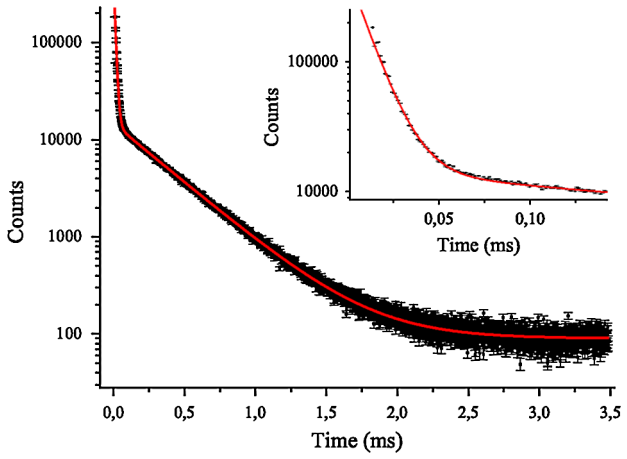

FIG. 3 (color online). The decay curve of $^4He^-$ measured at 10 K. The solid curve is a fit to the data. The inset shows a time region in which the decay of the short lived $J = 1/2$ and $J = 3/2$ levels dominate the intensity.

corrected for photodetachment by BB radiation photons. Such corrections rely on calculated cross sections for photodetachment and accurate knowledge of the photon energy distribution—i.e., of the temperature (or distribution of temperatures)—that the He^- ions experience throughout the measurements.

Here, we have measured the depletion of the three fine-structure levels of the $1s2s2p\ ^4P^o$ state of He^- at 10, 50, and 296 K. At 10 K (Fig. 3) and 50 K, we measured decay rates for the $J = 5/2$ level corresponding to lifetimes of $358.8 \pm 0.7\ \mu$s and $362 \pm 2\ \mu$s, respectively (statistical errors, 1 standard deviation). As the BB radiation driven photodetachment rate is negligible at both 10 and 50 K (see below), our low-temperature result for $J = 5/2$ is the weighted average: $\tau = 359.0 \pm 0.7\ \mu$s. Using this result and the photodetachment cross sections [22], we calculate the BB contributions to the total decay rate as a function of temperature and obtain the curve shown in Fig. 4. This curve yields an expected 296 K lifetime of 297.3 μs, in agreement with our room temperature measurement of $296 \pm 4\ \mu$s. In Fig. 4, we show the present measured $J = 5/2$ lifetimes for 10, 50, and 296 K, and results by Pedersen *et al.* [13] and Wolf *et al.* [12] at intermediate and high temperatures. The most recent theoretical result, by Miecznik *et al.* [20], is shown at 0 K. Note that the (measured) lifetime is independent of limited temperature variations at 10 and 50 K as, in fact, the thermal photodetachment rate is negligible for all temperatures below 100 K. In contrast, the only previous measurements below room temperature [13] required careful measurements of the full temperature distribution in order to deduce a value for the 0 K lifetime.

We measured the pressure in our cold chamber in a room temperature volume (directly connected to the cold region) to be 1.5×10^{-10} mbar at 10 K, 1.7×10^{-10} mbar at 50 K, and 4.0×10^{-10} mbar at 296 K. In addition, we made a room temperature measurement at a hundred times higher pressure of 4.0×10^{-8} mbar yielding a measured lifetime

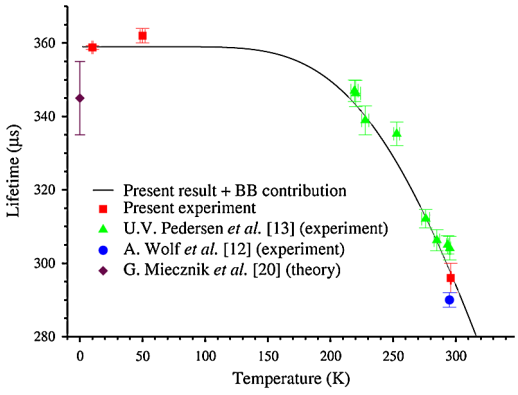

FIG. 4 (color online). Temperature dependence of the measured lifetime of the $1s2s2p\ ^4P^o_{5/2}$ level of $^4He^-$. The effect on the decay rate from photodetachment by blackbody radiation can readily be seen as a decrease in the measured lifetime above 100 K.

of $290 \pm 10\ \mu$s for $J = 5/2$. Ascribing the full difference between the low ($296 \pm 4\ \mu$s) and the high ($290 \pm 10\ \mu$s) pressure results to residual-gas collisions, we determine $\Gamma_{\text{coll}} = 0.26 \pm 0.48\ \text{s}^{-1}$ at 1.5×10^{-10} mbar, which is negligible compared to the measured decay rate, Γ_{measured}, for $J = 5/2$ of $2784 \pm 5\ \text{s}^{-1}$. Having shown that a residual-gas density of $10^8\ \text{cm}^{-3}$ does not significantly affect the measured lifetime, we may safely ignore losses due to interactions with other stored ions as, on average, only 3–4 ions are trapped per injection cycle.

As the trap is electrostatic, there is no Zeeman-mixing between the J levels. With magnetic storage devices, such mixings may contribute substantially to the depletion and have to be corrected for [10]. In principle, electric fields may also induce decays. This may be through electric field induced tunneling, which is negligible at the present trap field strengths below 200 kV/m [27], and/or through Stark mixing of the J levels. However, the field induced shifts of the $^4P^o$ J levels are only a few neV [28,29], which is negligible in relation to the fine-structure splittings (Fig. 1). In order to determine any remaining limitations in the storage time, 2.5 keV He^+ ions were stored in ConeTrap. At 4.0×10^{-10} mbar, the storage lifetime is 6 ± 2 s, corresponding to an upper limit for contributions from other instrumental effects of $\Gamma_{\text{instr}} \approx 0.17 \pm 0.06\ \text{s}^{-1}$. Thus, all terms of the sum in Eq. (1) are negligibly small except for the spontaneous autodetachment rate, and we conclude that our low-temperature measurement for the $1s2s2p\ ^4P^o_{5/2}$ He^- level represents the true intrinsic lifetime $\tau_{5/2} = 359.0 \pm 0.7\ \mu$s.

Pedersen *et al.* [13] used the electrostatic heavy-ion storage ring ELISA to measure the lifetime of the $1s2s2p\ ^4P^o_{5/2}$ level of He^- at temperatures down to below $-50\,^\circ$C. From the average of ten temperature measurements at different positions in ELISA and the photodetachment cross sections [21,22], they determined the contribution to the measured decay rate from BB radiation, and indirectly deduced a 0 K result of $365 \pm 3\ \mu$s. This is significantly longer than what we report here from a direct measurement at low, well defined, temperatures and at thermal equilibrium. Most likely, the ELISA result [13] was somewhat limited in precision due to the uncertainties in their estimate of the temperature distribution. Wolf *et al.* [12] used an electrostatic ion-beam trap at room temperature and corrected for BB radiation by means of the photodetachment cross sections [18], and indirectly deduced a 0 K result of $343 \pm 10\ \mu$s for $J = 5/2$. Miecznik *et al.* [20] used the multiconfiguration Hartree Fock approximation to calculate a lifetime of $345 \pm 10\ \mu$s.

Although the statistical quality of the present data is high, we cannot directly, in a three-component fit (with seven free parameters, including background), extract separate values for the two short lifetimes. With a double-exponential fit to the data in Fig. 3, the average lifetime for $J = 1/2$ and $J = 3/2$ is $10.8 \pm 0.8\ \mu$s (cf. Table I). Notably, this fit does yield results consistent with

TABLE I. Experimental and theoretical lifetimes of the $1s2s2p\ ^4P^o_J$ levels of $^4He^-$. The errors given for "This work" and "This work[a]" are 1 standard deviation.

Reference	Lifetime (μs) $J = 1/2$	Average	$J = 3/2$	$J = 5/2$
Experiment				
Nicholas 1968 [3]		18.2 ± 2.7		
Novick 1970 [4]	16 ± 4		10 ± 2	500 ± 200
Blau 1970 [5]		11 ± 5		345 ± 90
Simpson 1971 [6]		9^{+5}_{-3}		
Alton 1983 [8]		16.7 ± 2.5		
Andersen 1993 [10]			12 ± 2	350 ± 15
Wolf 1999 [12]		8.9 ± 0.2		343 ± 10
Pedersen 2001 [13]		11.1 ± 0.3		365 ± 3
This work		10.8 ± 0.8		359.0 ± 0.7
This work[a]	7.8 ± 1.0	10.8 ± 0.1	12.3 ± 0.5	
Theory				
Estberg 1968 [14]				266
Laughlin 1968 [15]				550
Estberg 1970 [16]				455
Davis 1987 [17]				497
Brage 1991 [19]	10.7		11.8	405
Miecznik 1993 [20]				345 ± 10

[a]Result when the relative initial populations of the $J = 1/2$, $J = 3/2$, and $J = 5/2$ levels are assumed to be 1/6, 1/3, and 1/2, respectively, (cf. text).

a 1:1 statistical population, at the time of production of the ions, between on one side the $J = 1/2$ and $J = 3/2$ levels together and, on the other side, the $J = 5/2$ level. Assuming that *all three* fine-structure levels are populated in proportion to their statistical weights (1:2:3 for $J = 1/2$, $3/2$, and $5/2$), a new two-component fit yields an average lifetime of 10.8 ± 0.1 μs. An analysis based on different starting points for three-component fits yields separate $J = 3/2$ and $J = 1/2$ lifetimes of 12.3 ± 0.5 μs and 7.8 ± 1.0 μs under the rather well-founded assumption of a strict statistical population of the three fine-structure levels at the time of production in the cesium cell (cf. the row marked by an a in Table I).

In this work, we have used a novel technique based on a purely electrostatic ion-beam trap which may be operated at room temperature and at cryogenic temperatures down to 10 K to perform the first correction-free measurements of the lifetime of the $1s2s2p\ ^4P^o$ $J = 5/2$ fine-structure level in He^-. All previous experimental studies of this lifetime have been hampered by the need to accurately measure the temperature distribution in the apparatus, and by the need to make theory based, substantial, corrections of the results. Our new, direct, high-precision measurement of the $^4P^o$ $J = 5/2$ lifetime of 359.0 ± 0.7 μs presents a serious challenge for advanced theories describing, in detail, the properties of the spatially extended wave functions of He^- (dipole polarizability 8169.1 a_0^3 [29])—the lightest atomic three-electron system.

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