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Storage-ring experiments with 10–100-keV Ca^- beams: Role of blackbody radiation

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The lifetime of 10–100-keV Ca^- beams in a heavy-ion storage ring was measured to be $\sim$490 μs. A combination of experimental and theoretical evidence strongly suggests that the observed decay is associated with blackbody-radiation-induced photodetachment of $Ca^-(4s^2 4p\,^2P)$ ground-state ions at room temperature. Blackbody-radiation effects may also explain the previously reported existence of a long-lived (290 ± 100 μs) metastable $Ca^-(4s4p^2\,^4P)$ ion [D. Hanstorp *et al.*, Phys. Rev. Lett. **63**, 368 (1989)].

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Studies of the structure and dynamics of negative ions are of fundamental relevance for atomic physics (for recent reviews, see Ref. [1]). A system which has turned out to be particularly interesting is the negative calcium ion (Fig. 1), which has provided a significant challenge to both experimentalists and theorists. The history of Ca^- studies is complicated. Because of the firm belief that closed-subshell alkaline-earth atoms would not bind an extra electron, all Ca^- beams were originally assumed to be composed of metastable ions. Thus, the photodetachment work of Heinicke *et al.* [2] using an arc-based continuum light source almost two decades ago, reported on relative cross sections for a metastable species. Some years later, the view that Ca^- beams are composed of metastable species was further supported when Bunge *et al.* [3] calculated that $Ca^-(4s4p^2\,^4P)$ was quite strongly bound with respect to $Ca(4s4p\,^3P)$, by $\sim$550 meV. Our understanding changed abruptly in 1987 with the prediction [4] and observation [5] of a *stable* $Ca^-(4s^2 4p\,^2P)$ ion. Since then, a significant number of calculations dealing with the electron affinity [6–14], fine structure [15], and photodetachment cross sections [11,16] of $Ca^-(^2P)$ have appeared. The discovery of the stable Ca^- ion naturally led to a renewed interest in the properties of the metastable species. By utilizing a production technique which was believed to produce almost entirely metastable species, Hanstorp *et al.* [17] measured a decay rate for a Ca^- beam which led to a lifetime for a long-lived component of 290 ± 100 μs. They attributed this lifetime to $Ca^-(4s4p^2\,^4P_{5/2})$

More recently two difficulties have arisen: the first concerning the electron affinity of the Ca^- *ground state* and the second related to the lifetime of a long-lived Ca^- *metastable state*. Several theoretical calculations on the ground-state ion indicated that the electron affinity was in the range of 45–82 [6–8,10,11,13,14] meV, while in two other theoretical works values of 0 meV [12] and 22 meV [9] were obtained. Thus the observations of Pegg *et al.* [5], who had utilized electron spectroscopy and fast-ion beam techniques to obtain an electron affinity of 43 ± 7 meV, were in good agreement with the majority of theoretical works. However, the very recent tunable-laser photodetachment work of Walter and Peterson [18] reported that the Ca^- ground state was much more weakly bound, with an electron affinity of 18.4 ± 2.5 meV. Moreover, this latter result is now strongly supported by independent electric-field dissociation measurements [19]. With regard to the metastable state, calculations of the autodetachment rates for $Ca^-(4s4p^2\,^4P_{5/2}$ and $^4P_{3/2})$ yielded values exceeding $10^9\ s^{-1}$, invalidating the original assignment of Hanstorp *et al.* [17], but leaving $^4P_{1/2}$ as a possible long-lived state [20]. However, very recent calcu-

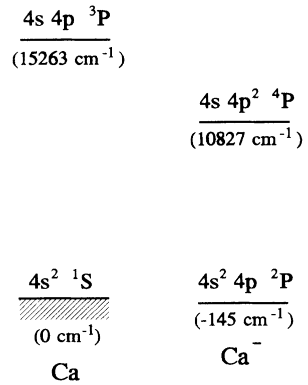

FIG. 1. Simplified energy-level diagram of the calcium atom and the corresponding negative ion. The ground-state $Ca^-(4s^2 4p\,^2P)$ ion is bound with respect to $Ca(4s^2\,^1S)$, while the $Ca^-(4s4p^2\,^4P)$ ion is bound with respect to $Ca(4s4p\,^3P)$, and is predicted (theoretically) to autodetach on a submicrosecond time sale. The binding energies of the ions have been taken from the most recent accepted values, as described in the text.

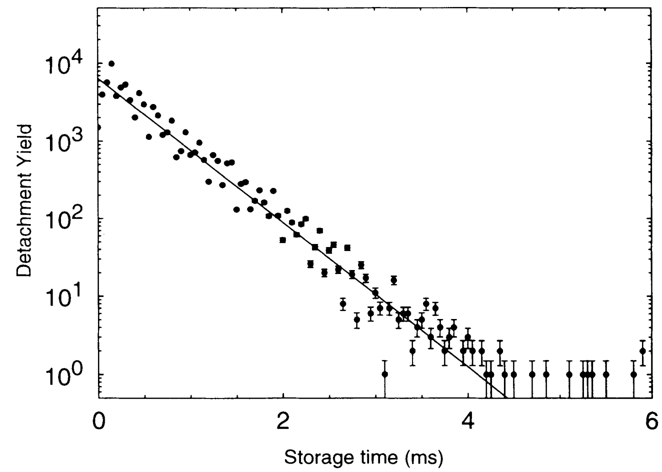

FIG. 2. Semilogarithmic plot of the detachment yield vs time following injection of a weak (pA) 100-keV Ca^- beam. The straight line represents the exponential fit. The rapid fluctuation of the detachment yield is due to betatron oscillations in the storage ring, as described in the text.

lations by Brage, Miecznin, and Fischer [21] on the $J=\frac{1}{2}$ component have led to a lifetime of only ~0.1 μs, and considerable doubt as to the precise value of the lifetime remains. The large discrepancy between theoretical calculations and experiment on the $Ca^-(4s4p^2\,^4P)$ lifetime prompted us to initiate a series of measurements aimed at the observation of the metastable ion lifetime utilizing a heavy-ion storage ring.

We have utilized the Aarhus Storage Ring Denmark (ASTRID) which facilitates ion-lifetime measurements in the ~10 μs–100 ms range [22]. Very recently, this experimental approach has yielded accurate experimental lifetimes for the homologous $Be^-(2s2p^2\,^4P_J)$ ion [23]. A detailed description of the experimental facility can be found in recent publications [22,23]. Mass- and charge-state-analyzed $^{40}Ca^-$ ions, with an energy in the range of 10–100 keV, were injected into the storage ring. A tandem-channel-plate detector positioned at one corner detected fast neutral atoms which were produced along a 10-m straight section of the ring. This approach allowed monitoring of the decay for several lifetimes of the beam with a good signal-to-noise ratio. With injection of nanoampere beams, the detector was often partially saturated at early times, but to avoid such effects in the search for a short-lived metastable component, picoampere beams were used. By using different charge-exchange materials (Na or K) in the vapor cell, one could attempt to vary the relative fraction of stable and metastable Ca^- ions. Electron capture from Na vapor favors the production of the ^1S ground state of Ca, and the Ca^- ion which is subsequently formed is most likely in the $4s^2 4p\,^2P$ ground state. On the other hand, charge exchange in K or Cs [17,18] is almost in resonance with the formation of $Ca(4s4p\,^3P)$ and may favor production of the metastable $Ca^-(4s4p^2\,^4P)$ ions, as claimed by Hanstorp *et al.* [17].

Figure 2 shows a typical neutral-atom signal versus time for Ca^-. It was obtained with Na in the charge-exchange cell, which should strongly favor production of ground-state Ca^- ions as pointed out above. The data can be described by means of a single exponential-decay process. As discussed in detail elsewhere [22,23], the oscillatory behavior (Fig. 2) is due to betatron oscillations in the ring. Since the beam cannot be stored perfectly on axis, a varying detection efficiency from one revolution to the next is observed due to the finite size of the detector. Nevertheless, the ability to follow the decay out to several beam lifetimes (~50–100 revolutions) effectively averages betatron oscillation effects. It is also important to note that the injected beam decays totally. The few counts at times greater than 4 ms are consistent with noise in the tandem-channel-plate detection system. With the injection of even more intense beams than is the case in Fig. 2, the ratio of the initial signal to the background was $\sim 10^5$:1. This should be contrasted with the ratio which would be expected *if* approximately equal fractions of the metastable and stable species were present, and *if* the stable ion lifetime were dictated entirely by collisional detachment via rest gas collisions. Assuming a metastable ion lifetime ~0.5 ms and a stable ion lifetime ~0.5 s, the ratio would be $\sim 10^3$:1. The studies were also conducted at three energies (10, 25, and 100 keV) but no energy dependence of the beam lifetime was observed. Finally, a search was performed for a short-lived (metastable) component by changing to potassium metal in the charge-exchange cell and by injecting weak beams to avoid detector saturation, but the data always exhibited a single exponential decay with a lifetime of 490 ± 20 μs.

The decay of the entire beam within a few milliseconds necessitated the investigation of various destruction mechanisms for ground-state ions. In light of the new electron affinity of only ~18 meV [18,19], four mechanisms in particular were considered: collisional destruction on rest gas, intrabeam scattering, electric-field stripping, and blackbody-radiation-induced photodetachment. The rest gas in the ring is primarily H_2. In order to test for an unexpectedly high collisional detachment cross section on a H_2 target, the average background pressure was degraded by a factor of ~ 100 by introducing hydrogen gas. However, this led to a change in lifetime of only $\leq 10\%$. Intrabeam scattering was eliminated as a relevant destruction mechanism, since the results were found to be independent of the Ca^- beam current. Electric field ($\mathbf{E} \sim \mathbf{v} \times \mathbf{B}$) stripping can also be neglected since, for example, at a beam energy of 25 keV, $|\mathbf{E}| \sim 410$ V/cm, which is much too small to induce motional field stripping [24]. This is also consistent with the lack of an observed energy-dependent lifetime. Our beam decay rate is, however, consistent with blackbody-radiation-induced photodetachment. Absorption of photons with wavelengths less than ~ 67 μm will detach $Ca^-(4s^2 4p\,^2P)$ ions. By folding the Planck radiation distribution with theoretical wavelength-dependent photodetachment cross sections [11,16] in a computer model, we predict a lifetime for $Ca^-(4s^2 4p\,^2P)$ of 520 μs (Ref. [11]; "length" formalism) and 960 μs (Ref. [16]; accurate initial state and correlated final state, length formalism) on the basis of this mechanism alone. The "velocity" formalism of Ref. [11] leads to a doubling of the predicted lifetime (1150 μs), while the same formalism and accurate initial state and correlated final state from Ref. [16] leads to a slight reduction in lifetime (870 μs). The theories [11,16], however, utilized binding energies which were a factor of 3–4 too large (58 meV [11] and 69 meV [16]), and thus the cross sections can only be considered approximate values. Therefore we consider the agreement between theory and experiment to be quite acceptable. Furthermore, in our calculations, only one fine-structure component was assumed, with a binding energy of 18.4 meV. Calculations of the $^2P_{1/2}$-$^2P_{3/2}$ fine-structure splitting [15] by Vosko, Chevary, and Mayer and Dzuba *et al.* have yielded 5.4 and 6.9 meV, respectively, but no evidence of a two-component blackbody-radiation-induced decay is evident from our data.

In order to investigate further blackbody-radiation effects on $Ca^-(4s^2 4p\,^2P)$, the temperature variation of the lifetime was tested. By heating one half of the storage ring to 383 K, the lifetime was decreased to 416 ± 15 μs. The temperature rise degrades the vacuum by a factor ~ 15, which would only increase the decay rate by $\leq 1\%$. The observed fractional change in decay rate was, however, much less than we predicted from calculations based on Refs. [11,16], and the effect of a much lower electron affinity on the magnitude of the cross sections and their wavelength dependence needs to be evaluated before rigorous comparisons with experiment will be possible. It was, however, feasible to make rigorous comparisons of theoretical and experimental data for blackbody-radiation effects in the case of the negative helium ion, which is bound by only 77 meV [25]. For this light ion, accurate theoretical results are available [26,27]. For $He^-(1s2s2p\,^4P_{5/2})$, our calculations indicate that blackbody-radiation-induced photodetachment represents a $\sim 20\%$ effect at room temperature [28]. Moreover, in the He^- case, the observed decrease in lifetime upon heating one-half of the ring to 383 K was in very good agreement with our calculations [28].

Our lifetime is in good agreement with that of Ref. [17], when various different aspects of the respective experiments are taken into account. The present data, however, do not exclude the existence of a metastable component, as claimed by Hanstorp *et al.* In particular, an observed photodetachment threshold in the tunable laser work of Peterson and co-workers [17,18] suggests that a metastable ion with a lifetime on the short microsecond time scale may indeed exist. Their data appear to be quite consistent with detachment of the metastable $Ca^-(^4P)$ ion, assuming that the electron affinity is accurately given by Bunge *et al.* [3]. Our data indicate that either the lifetime of such a component is shorter than ~ 30 μs or our production intensity ratio, $I[Ca^-(^4P)]/I[Ca^-(^2P)]$, is rather low. It also appears that the very high production efficiency claimed for the metastable species in Ref. [17] was an overestimate [18,20]. If their decay [17] could have been entirely attributed to a metastable species, the associated lifetime would have to be renormalized to a shorter value. The obvious role of blackbody radiation, however, renders the earlier data inconclusive.

Blackbody-radiation effects are well known from experiments with Rydberg atoms, and have even been recently utilized to calibrate the relative density of highly excited sodium atoms via the photon-induced ionization rate [29]. Corresponding examples for negative ions are rare. One recent case comes from experiments on the storage of H^- at the low-energy antiproton ring (LEAR) at CERN [30], where a very significant effect from light emission by ionization gauges was observed. Since for H^- the electron affinity is 754 meV [1], it is unperturbed by an emitter at room temperature, but is sensitive to near-infrared and visible photons from hot filaments. The effect was particularly noticeable in the LEAR experiments due to the low collisional detachment rate at megaelectron volt energies. Finally, it is interesting to note that blackbody-radiation effects in the storage ring might be used to advantage in specific cases to discriminate strongly versus weakly bound ions. This could be realized for ions of the appropriate electron affinities through the introduction of a "hot pipe" at, say, ~ 600 K. Midinfrared lasers and nonlinear optical techniques could also be applied in future storage-ring-based investigations. For example, a laser pulse of wavelength greater than ~ 2.3 μm (and less than the ^2P threshold detachment wavelength) can saturate the photodetachment of ground-state $Ca^-(4s^2 4p\,^2P)$ ions, while the metastable $Ca^-(4s4p^2\,^4P)$ species would be unaffected.

In summary, we conclude that ground state $Ca^-(4s^2 4p\,^2P)$ ions were effectively photodetached on a submillisecond time scale by blackbody radiation at room temperature. The analytical strengths of storage-ring-based lifetime studies have also been clearly demonstrated. The total extinction of the injected beam, even under conditions of ion production where ground-state $Ca^-(4s^2 4p\,^2P)$ should have been formed exclusively, led convincingly to the blackbody-radiation mechanism. Nevertheless, a single photodetachment-threshold feature seen by Peterson and co-workers [17,18], which appears to be associated with $Ca^-(^4P)$, still suggests the existence of a metastable ion with a lifetime $\gtrsim 1$ μs.

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[1] H. Hotop and W. C. Lineberger, J. Phys. Chem. Ref. Data **14**, 731 (1985); V. Esaulov, Ann. Phys. (Paris) **11**, 493 (1986); T. Andersen, Phys. Scr. **T34**, 23 (1991); D. R. Bates, Adv. At. Mol. Phys. **27**, 1 (1991).

[2] E. Heinicke, H. Kaiser, R. Rackwitz, and D. Feldman, Phys. Lett. **50A**, 265 (1974).

[3] C. F. Bunge, M. Galán, R. Jauregui, and A. V. Bunge, Nucl. Instrum. Methods **202**, 299 (1982).

[4] C. Froese Fischer, J. B. Lagowski, and S. H. Vosko, Phys. Rev. Lett. **59**, 2263 (1987).

[5] D. J. Pegg, J. S. Thomson, R. N. Compton, and G. D. Alton, Phys. Rev. Lett. **59**, 2267 (1987).

[6] C. Froese Fischer, Phys. Rev. A **39**, 963 (1989).

[7] S. H. Vosko, J. B. Lagowski, and I. L. Mayer, Phys. Rev. A **39**, 446 (1989).

[8] L. Kim and C. H. Greene, J. Phys. B **22**, L175 (1989).

[9] C. W. Bauschlicher, S. R. Langhoff, and P. R. Taylor, Chem. Phys. Lett. **158**, 245 (1989).

[10] W. R. Johnson, J. Sapirstein, and S. A. Blundell, J. Phys. B **22**, 2341 (1989).

[11] G. F. Gribakin, B. V. Gul'tsev, V. K. Ivanov, and M. Yu. Kuchiev, J. Phys. B **23**, 4505 (1990).

[12] P. Fuentealba, A. Savin, H. Stoll, and H. Preuss, Phys. Rev. A **41**, 1238 (1990).

[13] R. D. Cowan and M. Wilson, Phys. Scr. **43**, 244 (1991).

[14] T. Brage and C. Froese Fischer, Phys. Rev. A **44**, 72 (1991).

[15] S. H. Vosko, J. A. Chevary, and I. L. Mayer, J. Phys. B **24**, L225 (1991); V. A. Dzuba, V. V. Flambaum, G. F. Gribakin, and D. P. Sushkov, Phys. Rev. A **44**, 2823 (1991).

[16] C. Froese Fischer and J. E. Hansen, Phys. Rev. A **44**, 1559 (1991).

[17] D. Hanstorp, P. Devynck, W. G. Graham, and J. R. Peterson, Phys. Rev. Lett. **63**, 368 (1989).

[18] C. W. Walter and J. R. Peterson, Phys. Rev. Lett. **68**, 2281 (1992).

[19] M. J. Nadeau, X. L. Zhao, M. A. Garwan, and A. E. Litherland (unpublished).

[20] J. R. Peterson, Aust. J. Phys. (to be published).

[21] T. Brage, G. Miecznin, and C. Froese Fischer (unpublished); T. Brage (private communication).

[22] L. H. Andersen, T. Andersen, H. K. Haugen, N. Hertel, P. Hvelplund, S. P. Møller, and W. W. Smith, Phys. Lett. A **162**, 336 (1992).

[23] P. Balling, L. H. Andersen, T. Andersen, H. K. Haugen, P. Hvelplund, and K. Taulbjerg (unpublished).

[24] M. J. Nadeau and A. E. Litherland, Nucl. Instrum. Methods Phys. Res., Sect. B **52**, 387 (1990).

[25] A. V. Bunge and C. F. Bunge, Phys. Rev. A **19**, 452 (1979).

[26] A. U. Hazi and K. Reed, Phys. Rev. Lett. **24**, 2269 (1981).

[27] H. P. Saha and R. N. Compton, Phys. Rev. A **64**, 1510 (1990).

[28] T. Andersen *et al.* (unpublished).

[29] K. Harth, M. W. Ruf, and H. Hotop, Z. Phys. D **14**, 149 (1989).

[30] D. Manglunki, in *Physics at LEAR with Low Energy Antiprotons,* Fourth LEAR Workshop, edited by C. Amsler *et al.* (Harwood Academic, Chur, Switzerland, 1988).