Observation of the Metastable Negative Argon Ion Ar^-

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The predicted metastable $^4S^o_{3/2}$ Ar^- ion has been observed for the first time. It was produced from an Ar^+ beam by two-step electron capture in Cs vapor. Decay-rate measurements favor the existence of only one state with a lifetime of 350 ± 150 ns.

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In this Letter we report the observation of the metastable negative ion Ar^- and the measurement of its autodetachment lifetime of 350 ± 150 ns. It is now the fourth verified example of a negative ion that is formed from a metastable excited state of neutral species whose ground state cannot bind an electron. Rapid autodetachment of these ions is forbidden by their spin configurations; decay into the continua can occur only through spin-orbit or spin-spin interactions. The $^4P^o$ state of He^- is well known and has been studied, but only recently has research on other ions been undertaken. The spin configurations that render these ions metastable also strongly inhibit their production in conventional negative-ion sources, but this difficulty is overcome using the process of two-step electron capture by the parent positive ions in alkali vapors, as was demonstrated originally by Donnally and Thoeming[1] for He^- production. In this process, for example

$$He^+ + Cs \rightarrow He^*(1s2s\,^3S) + Cs^+ \quad (1a)$$

followed by

$$He^* + Cs \rightarrow He^-(1s2s2p\,^4P^o) + Cs^+, \quad (1b)$$

the parent metastable neutral is efficiently produced by near-resonant electron capture (1a), and the low ionization potential of Cs facilitates the second electron capture (1b) to form the negative ion. The cross section at He^+ beam energies of 1–5 keV for reaction (1a) is very large, $\sim 10^{-14}$ cm^2, while that for (1b) is much smaller ($\sim 2\times 10^{-16}$ cm^2),[2] and is the production-limiting step.

Using this two-step electron-capture technique, Bae and Peterson have recently discovered[3] the previously unsuspected metastable ion He_2^- and also have shown[4] the metastability of Be^-. On the other hand, we found strong evidence against the existence of the previously reported H_2^- and H_3^- ions, and obtained null results in producing Mg^-, N^-, and N_2^-.[5] During that search for other negative ions, we also looked for Ne^- and Ar^- without success. However, it was recognized[5] that the production of the more massive ions would be inhibited in our apparatus because its small mass-analyzing magnet restricted the maximum velocity of the beams to quite low values, which would both suppress the second step [reaction 1(b)] in the formation process and increase the loss by autodetachment. These problems were overcome in the present collaboration by the use of the LEAPA II accelerator at the Lawrence Berkeley Laboratory, which permitted a search for these metastable negative ions up to mass 100 amu at energies up to 18 keV.

Recently, using configuration interaction calculations, Bunge *et al.*[6] concluded that the $3s^23p^54s4p\,^4S$ state of Ar^- lies more than 135 meV below the $3s^23p^54s\,^3P^o$ state of Ar and is metastable. They also concluded that there is no similar state of Ne, whose negative-ion states are either slightly unbound or decay via $E1$ radiative transitions to a lower unbound state. Not only do we confirm here the existence of Ar^-, we also report the inability to find any trace of the metastable Ne^-, in agreement with the calculations of Bunge *et al.*[6]

A schematic diagram of the experimental apparatus is shown in Fig. 1. The positive-ion beam was extracted from a duoplasmatron ion source at full acceleration potential. The beam was focused, momentum selected by a 15° magnetic analyzer, collimated by two 2.5-mm-diam apertures separated by 10 cm, and directed through a cesium-vapor heat pipe[7] to form the negative ions. The Cs target thickness was typically $\sim 10^{15}$ cm^{-2}. The beam was then defined by a 5-mm-diam aperture $A1$ and separated into the positive, zero, and negative charge-state components by an electrostatic deflector $D1$ in the analysis chamber. The operating pressure in the analysis chamber was typically less than 2×10^{-6} Torr. The positive beam was directed to a Faraday cup FC1 for monitoring the beam current, and the strong undeflected neutral-beam current was collected in the Faraday cup FC3 after traversing a long (50 cm) pipe section installed to reduce interference from secondary neutral particles and uv photons produced at the collector surface.

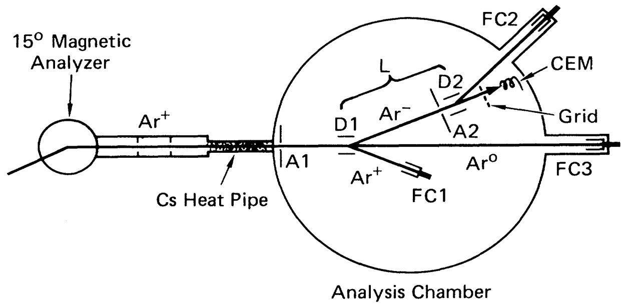

FIG. 1. Schematic diagram of the main experimental arrangement. Deflection angles at $D1$ and $D2$ are both 22.5°.

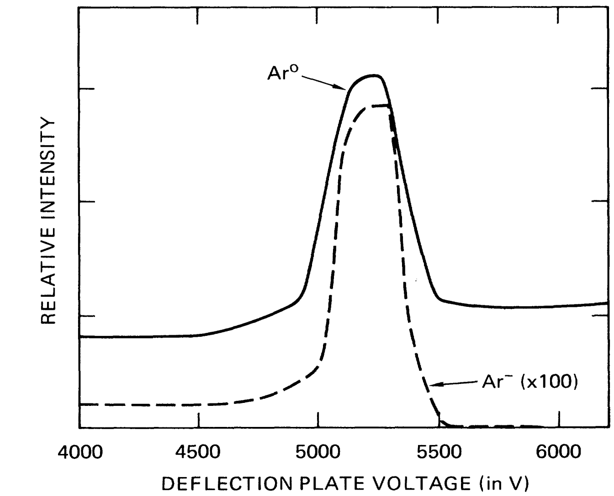

FIG. 2. Relative intensities of the autodetached Ar^0 neutrals measured at the CEM and the Ar^- current to FC2, as functions of the deflection voltage on $D1$.

The negative beam was directed along a 15-cm field-free flight path L to allow observation of its autodetachment, and through a 6.4-mm-diam aperture $A2$. It then could either be directed to a second Faraday cup FC2 by deflector $D2$, or pass undeflected to a channel electron multiplier (CEM). The CEM was preceded by an electrostatic grid which was used to control secondary electrons. All neutral atoms created along L by either autodetachment or collisional detachment of the negative ions were also monitored by the CEM. Several grounded stainless-steel plates, which also terminated the fringe fields of $D1$ and $D2$, and magnets were placed to block and deflect secondary particles. With this arrangement we were able to reduce the background current measured by FC2 to less than 2×10^{-15} A for a parent positive beam of 5×10^{-7} A.

Analogously to He^- production by reactions (1a) and (1b), Ar^- can be produced by the successive electron-capture reactions

$$Ar^+ + Cs \rightarrow Ar^*(3s^2 3p^5 4s\,^3P_2^\circ) + Cs^+ \qquad (2a)$$

and

$$Ar^* + Cs \rightarrow Ar^-(3s^2 3p^5 4s 4p\,^4S_{3/2}^e) + Cs^+. \qquad (2b)$$

The long-lived $Ar^*(^3P_2)$ metastable effectively does not decay in this experiment. On the other hand, Ar^- is expected to decay through autodetachment,

$$Ar^- \rightarrow Ar^0(3s^2 3p^6\,^1S_0) + e, \qquad (3)$$

faster than the (11 μs) lifetime[8] of $He^-(^4P_{3/2})$, because this transition occurs primarily through spin-orbit coupling to autodetaching doublet states,[9] which is much stronger in Ar than He.

A series of measurements was performed to establish the existence of the metastable Ar^- ion. First, the negative ion was confirmed to have the same momentum and kinetic energy as the initial parent Ar^+ ions. Next, its autodetachment was established by monitoring the neutral products at the CEM. In fact, it was found that the neutral Ar^0 beam generated along L was so intense that the CEM could be used as a neutral-current monitor (instead of a counter) when the grid was positively biased to extract secondary electrons from the CEM. Collisional detachment was examined by measuring the neutral currents at several different pressures in the analysis chamber. They were found to be essentially constant, indicating that collisional detachment was negligible. It was also important to test whether the apparent Ar^0 signals came from the autodetachment of Ar^- ions or from some other source, such as secondaries produced by the beam striking some surface. To do this we monitored simultaneously the secondary emission current from the CEM and the actual negative ion current at FC2, with $D2$ set at the proper voltage, and scanned the deflector voltage of $D1$. It can be seen in Fig. 2 that these currents track identically, except for some background in the CEM current mainly due to secondary particles (high-energy neutral atoms or uv photons) produced on other surfaces in the chamber.

As a further test, we obtained some ^{36}Ar and used it as the ion-source gas. Figure 3 shows the mass spectrum of the positive ions that yielded negative ions of the same energy, measured at FC2. The typical Ar^- current at FC2, at a beam energy of 18 keV, was $\sim10^{-12}$ A. When ^{40}Ar was used as the source gas, negative ions of mass 12, 16, and 40 amu were observed (indicated by a dashed line). The $^{12}C^-$ and $^{16}O^-$ ions came from an unidentified ion-source contaminant, and no noticable neutral (autodetachment) current was observed at the CEM from these common stable ions. When the source gas was changed to ^{36}Ar, the 40-amu peak disappeared and a 36-amu peak appeared (indicated by a solid line), while the mass 12- and 16-amu peaks from the contaminant remained.

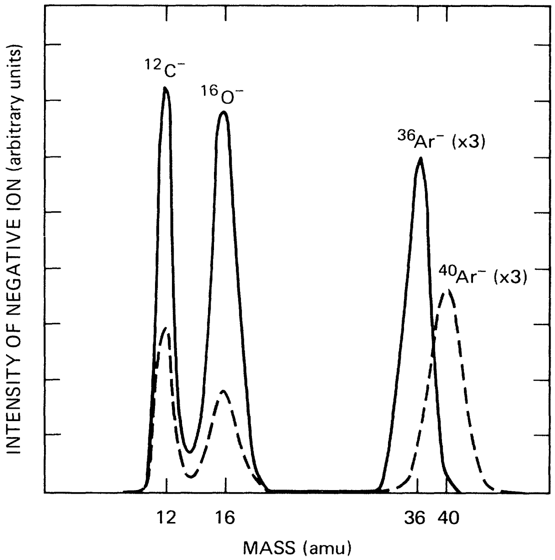

FIG. 3. Mass spectrum of negative ions measured at FC2. Solid line, ^{36}Ar as ion-source gas. Dashed line, ^{40}Ar gas.

The Ar^- currents seen in Fig. 3 have been reduced a factor of $\sim 10^{-2}$ by autodetachment along the flight path (15 cm along L and 20 cm between $D2$ and FC2, in Fig. 1), so they were originally much larger than the O^- and C^- contaminant ion currents.

To estimate the Ar^- autodetachment lifetime, we used the CEM as a particle counter to measure the average effective decay rate along L, as defined by

$$\Gamma_{\text{eff}} = v \ln[I_T/(I_T - I^0)]/L, \quad (4)$$

where v is the beam velocity, L is the field-free path length between $D1$ and $D2$, and I_T and I^0 are the CEM count rates of the total (negative plus neutral) and neutral beams. At the beam energies used in the experiment ($\geqslant 8$ keV) it is reasonable to assume that the secondary-electron coefficients of Ar^- and Ar^0 are both greater than 1 and thus that the count rates can represent the corresponding currents. For these decay-rate measurements, it was necessary to reduce the parent Ar^+ beam considerably in order to avoid saturation of the count rates of the CEM ($I_T \sim 10^4$ s^{-1}), and thus to maintain linearity in counts per current. As mentioned above, the collisional detachment contribution to I^0 was negligible compared to autodetachment.

Decay rates were measured at several energies between 8 keV ($v = 2\times 10^7$ cm/s) and 18 keV ($v = 3\times 10^7$ cm/s), and within the experimental errors they were constant at $\Gamma_{\text{eff}} \simeq 2.9\times 10^6$ s^{-1}. These results support the prediction of Bunge *et al.*[6] that metastable Ar^- is a ^4S state, and thus has only one fine-structure level and a unique decay rate. In this case the lifetime of $Ar^-(^4S)$, as determined by this work, is 350 ± 150 ns. The relatively large uncertainty in the lifetime is largely due to the combined effects of (a) the fact that the high decay rate made the difference between I_T and I^0 very small ($< 10\%$ relative to each) and thus subject to large statistical uncertainties, (b) the need to subtract a substantial background, and (c) fluctuations in the ion-beam intensity. This uncertainty could be reduced greatly in an improved experiment.

We also looked for metastable Ne^-, Kr^-, and Xe^- and could not find any detectable amounts. The results indicate that either Ne^- does not exist or that it has a lifetime much less than 50 ns. Considering the lifetimes of He^- (10–500 μs)[8] and Ar^- (350 ns) it is unlikely that a metastable Ne^- would have a lifetime much less than 50 ns; therefore, we conclude that it does not exist. On the other hand, our null results on the existence of Kr^- and Xe^- are somewhat less conclusive, because these ions are slower than Ar^- and probably have lower production efficiencies and greater autodetachment losses at these energies. Further experiments using another accelerator with higher energies should clarify the existence of these heavier ions.

In summary, we have observed the metastable Ar^- ion and performed preliminary lifetime measurements. The results indicate that Ar^- probably has only one state with a lifetime of 350 ± 150 ns. We have also found that a metastable state of Ne^- apparently does not exist. Both of these results support the theoretical results of Bunge *et al.*[6]

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