K-Shell Photodetachment of He^-: Experiment and Theory

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High-resolution K-shell photodetachment measurements of He^- giving rise to He^+ ions have been performed using a merged synchrotron vacuum ultraviolet photon-ion beam technique. The measurements on this fundamental negative ion display dramatic structure differing substantially, qualitatively and quantitatively, from the corresponding process in neutral atoms and positive ions, owing to the dominance of correlation in both initial and final states of He^-. In addition, this experimental investigation provides an unambiguous test of two independent theoretical calculations that report serious discrepancies and shows excellent agreement with one of them.

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The investigation of innershell photodetachment processes in negative ions offers a new perspective for understanding strongly correlated systems. Recent theoretical calculations have indicated that the phenomenology of these processes differs substantially from the corresponding innershell ionization processes in atoms and positive ions [1–3]. The difference is a consequence of the dominant effects of correlation in both initial and final states of the innershell photodetachment process; the emerging photoelectron interacts with the wave functions of the outershell electrons on its way out of the negative ion, thereby providing a unique probe of the dynamics of these highly correlated systems. Thus, these studies allow us to focus specifically upon the effects of electron-electron correlation and permit us to transfer the understanding gained to systems where these effects are also important, but much more difficult to disentangle, such as in neutral atoms, positive ions, molecules, clusters, and solids. In addition to their fundamental interest, the properties of these ions are of applied importance since the production and destruction of negative ions in systems such as dilute plasmas appearing in the outer atmospheres of stars are strongly affected by the characteristics of these ions [4].

From an experimental point of view, unlike outershell studies [5–9], innershell photodetachment is largely unexplored territory; only Li^-, the simplest multishell negative ion has been investigated, and that only this year [3,10]. These experimental studies have confirmed that the response of innershell electrons to ionizing radiation does indeed differ both qualitatively and quantitatively from neutral atoms and positive ions, at least for the case of Li^-. How general is this finding? To begin to answer this question, innershell photodetachment measurements on He^-, a negative ion that is very markedly different from Li^-, and significantly more complicated, are of great importance. To begin with, He^- does not form a stable ground state; the lowest extant state is $1s2s2p\ ^4P$, bound by only 77 meV [11] relative to the excited $1s2s\ ^3S$ He state. Thus, we are probing excited state photodetachment. Also, this state is metastable, making the experiment much more difficult than with a negative ion in a stable ground state. The situation is also much more complicated because the ^4P symmetry of the initial state of He^- allows ^4S, ^4P, and ^4D final states, while the initial ^1S state of Li^- allows only ^1P final states. In addition, the photoexcitation of the $1s$ electron in He^- leads primarily to a "hollow" ion, i.e., in the $1s$ photoexcitation region "hollow" resonances dominate the cross section, thereby allowing detailed study of these highly unstable states. Finally, since He^- contains only three electrons, innershell photodetachment calculations nearing exactness are, in principle, possible. Two such calculations have been performed, and they disagree markedly, despite the fact that we are dealing only with a three electron system [1,2]. Experimental results were therefore necessary to settle the issue. For these reasons, then, we have undertaken experimental investigations of innershell ($1s$) photodetachment of the metastable $1s2s2p\ ^4P$ state of He^-.

In this Letter, we report on the first experimental investigation which confirms that the photodetachment cross section, leading to He^+ does indeed differ qualitatively and quantitatively from the corresponding small, structureless, monotone decreasing cross sections in neutral He or He^+ in the threshold region. In addition, our experimental results indicate a clear preference for one of the previous many-body calculations [1,2] of He^-.

The experiment was performed at the Advanced Light Source on the high flux and high resolution atomic and molecular undulator beam line 10.0.1. Two sets of data were recorded with low (100 meV) and high (70 meV) photon resolution, in the 38–44 eV photon energy range. The photon energy step size was varied in order to define narrow structures in the spectrum. We used 35 meV steps between 38.5–40.5 eV, 50 meV steps between 40.5 and 42.8 eV and 15–30 meV steps between 42.8–44 eV. The smaller step size was used to scan the structures while the larger one was used in between the structures. He^-

ions, produced using an rf charge exchange ion source (Alphatross) [12], were used in a photon-ion experimental apparatus described elsewhere [13]. The He^- ion beam was accelerated to 13 keV, and a beam of about 20–150 nA reached the interaction region. The ions were merged collinearly with a counterpropagating photon beam along a 29 cm long interaction region producing photodetached neutral He atoms and He^+ ions from subsequent Auger decay of the excited neutral He atoms. He^+ ions, doubly photodetached, were detected with a microchannel plate detector as a function of photon energy. The resulting signal was normalized to the primary He^- ion beam as well as the photon intensity. In the case of negative ions, these merged experiments are a serious challenge since even if one achieves perfect overlap between the photon and ion beam, the signal is very easily swamped by background noise due to stripping of the negative ions with the residual gas (even though the background pressure in the interaction region was 10^{-10} torr) or with apertures in the ion beam line. In order to reduce collisional noise, and correct for the background ionization, the photon beam was chopped at 1 Hz and the photodetachment signal was determined by subtracting the light-off signal from the light-on signal. The statistical error in the data was decreased by summing multiple sweeps of the photon energy range of interest. The photon energy scale was separately calibrated using known resonances positions for neutral gases and corrected for the Doppler shift which amount to about 106 meV for 40 eV photons.

Negative ion photodetachment provides an extreme theoretical challenge because the wave functions of both initial and final states are so very sensitive to electron correlation effects. Two calculations [1,2] based upon two different methodologies, but each of which introduced the same correlation effects have been performed. One calculation [1] is based on an enhanced version of the R-matrix code, with an upgraded asymptotic part to handle the negative ion system; the other [2] employs a spline basis and a multiconfiguration Hartree-Fock (MCHF) methodology. Nevertheless, these two calculations have yielded significantly different results.

In order to compare our experimental results with the calculations, it is necessary to extract the He^+ production cross sections from the calculated total photodetachement cross sections. This has been done by subtracting the channels from the total photodetatchment cross section that do not result in He^+ production. These are the $1snl$ channels, whose cross sections are extremely small in the experimental energy range. In addition, there is the $2p^2\ ^3P$ channel, which is metastable against autoionization and decays primarily by radiation to the $1s2p\ ^3P$ state. The other channels are autoionizing states of He that decay almost exclusively via autoionization to He^+.

Our low resolution experimental results, along with the He^+ production cross sections of both calculations are shown in Fig. 1. Since the cross-section measurements are relative, a meaningful comparison requires normalization. In addition, both theoretical calculations have been shifted in photon energy to align their thresholds with the experimental thresholds; the calculations of Refs. [1] and [2] have been shifted by +106 and 185 meV, respectively. Then, the experimental results were normalized to the theory of Ref. [1] at 42 eV and the theory of Ref. [2] was shifted vertically to 20 Mb for the sake of clarity. Also, the intensity of the first peak of Ref. [2], which reaches 120 Mb, has been truncated to 60 Mb. We choose to normalize at 42 eV because this energy is in a broad, flat, nonresonant region of the spectrum. Furthermore, this normalization results in excellent agreement between experiment and theory in the nonresonant regions over the entire energy range considered. Finally, since theory is in accord with the Thomas-Reiche-Kuhn sum rule [1], if normalized significantly differently, the experimental results would not be in accord with the sum rule.

The experimental data show a peak of about 15 Mb at 38.88 eV, just above the first (He $2s2p\ ^3P$) $1s$ detachment threshold. Note that below the first $1s$ threshold, the experimental cross section is, within errors bars, zero as expected. Above the peak is a long tail and a broad flat region, up to about 42.8 eV. Then we move into a region of structures, starting at the second $1s$ threshold (the first photodetachment plus excitation threshold He $2s3s\ ^3S$) up to 44 eV. As discussed above, the He $2p^2\ ^3P$ photodetachment channel, which opens at lower energy than $2s3s\ ^3S$ [1,2] does not lead to He^+ production and is, therefore, not expected in the present experiment. Comparison of the experimental results with the two theoretical spectra shows a clear preference for the R-matrix results [1]. The agreement here is excellent qualitatively and quite good quantitatively as well.

A more detailed comparison with the higher resolution data is shown in Fig. 2, along with the theoretical R-matrix cross section [1]. It is clear that the agreement in this region is excellent. Unlike in the case of neutral atoms and positive ions, here the structures are of varying origin, both resonances and threshold maxima; the latter arise because the photodetachment cross section for each channel starts from zero at threshold. The strong resonance just above 43.4 eV is clearly seen. This resonance has been characterized as [1,2] $(2s3p^2 + 2p3s3p)\ ^4P$ and lies just below the opening of the $(2s3p + 2p3s)\ ^3P$ channel of He which is so strongly mixed that a single-particle designation is completely inadequate. This He state is often designated $(23sp+)\ ^3P$. Experimental resolution appears to lower and broaden this resonance relative to theory. In addition, a *very* narrow $2s3s4s\ ^4S$ resonance is predicted just below 43 eV. It's narrowness ($\sim$0.1 meV) [1,2], would require even higher photon resolution to measure it in order to confirm its prediction and the slight hint of its presence in our data. As a matter of fact, this resonance may well be somewhat stronger than predicted by theory [1,2] owing to the omission of the $1s4s\ ^3S$ target state of He which

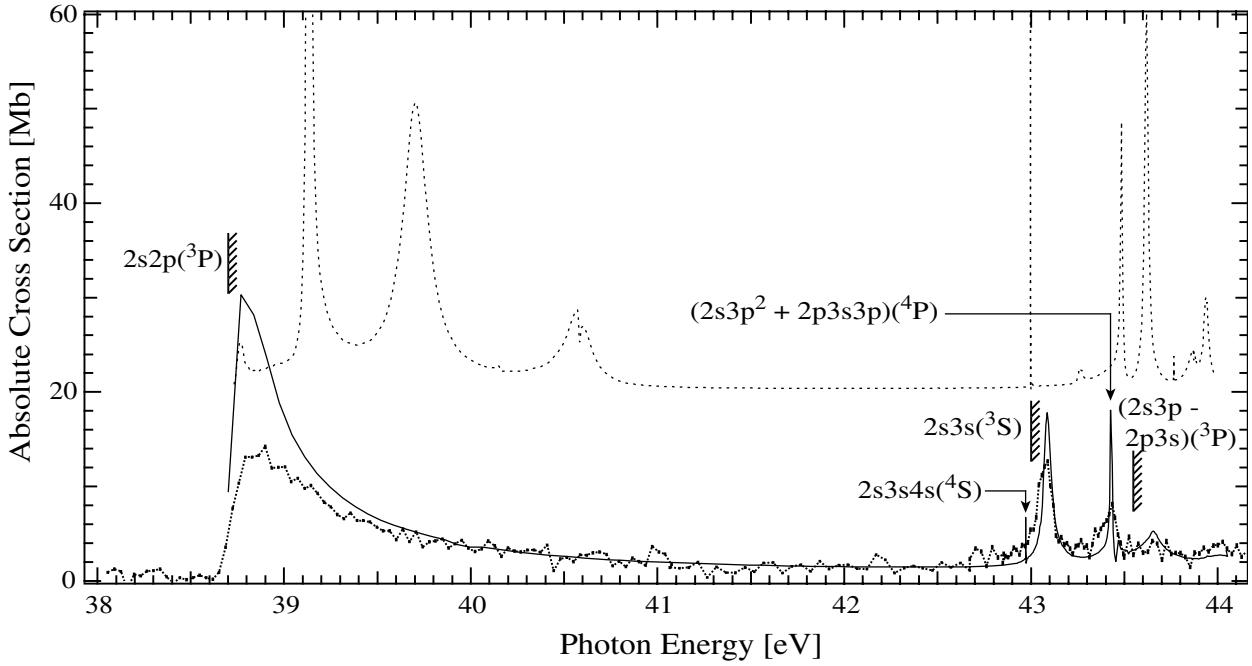

FIG. 1. Photodetachment cross section of the $1s2s2p\ ^4\mathrm{P}$ state of He^- in the $1s$ photoabsorption region producing He^+. The low-resolution experimental results are shown along with the theoretical results of Refs. [1] and [2] shown with dotted lines, respectively. The He thresholds and He^- resonances are indicated by the hatched lines and arrows, respectively.

precludes the possibility of the $2s3s \rightarrow 1s\epsilon s$ Auger decay which is likely to be one of the strongest decay channels of the $2s3s4s\ ^4\mathrm{S}$ resonance.

The rest of the structures seen are nonresonant threshold maxima. The threshold maximum above the first $1s$ detachment threshold, $2s2p\ ^3\mathrm{P}$, seen in Fig. 1 at 38.88 eV, provides poorest agreement of experiment with Ref. [1]; the latter is a factor of 2 larger than experiment here. This

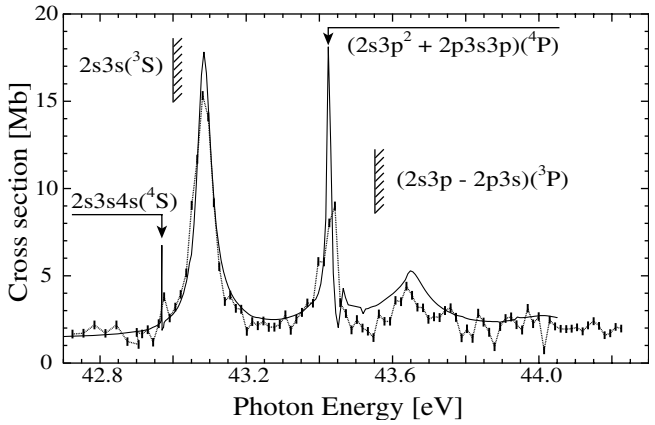

FIG. 2. Photodetachment cross section of the $1s2s2p\ ^4\mathrm{P}$ state of He^- in the $1s$ photoabsorption region producing He^+. The vertical bars are the high-resolution experimental results, and the solid line is the theoretical result of Ref. [1]. The height of the vertical bars at each data point represents the estimated uncertainties. The predicted He thresholds and He^- resonances are indicated by the hatched lines and arrows, respectively.

is reminiscent of an even larger discrepancy between theory and experiment at the first $1s$ threshold in Li^- photodetachment [3,10,14]. The basic cause of this discrepancy in He^- and Li^- is not yet understood. But it does underscore the fact that in the application of the R-matrix formalism to $1s$ photoabsorption of even three- and four-electron systems, some important aspect(s) of the physics of the process are still not treated correctly.

However, the threshold peak above the $2s3s\ ^3\mathrm{S}$ threshold, at about 43.1 eV, is seen in Fig. 2 to be in excellent agreement with theory. Additionally, the threshold maximum just above the opening of the $(2s3p - 2p3s)\ ^3\mathrm{P}$ channel, also designated $(23sp-)\ ^3\mathrm{P}$, just above 43.6 eV, shows quite good agreement as well. Thus it is clear that, except for the maximum above the first $1s$ threshold, excellent *quantitative* agreement with R-matrix theory [1] is found. It is equally clear that a number of structures predicted by the calculation of Ref. [2] are not observed.

The factor of 2 disagreement between our experimental cross section and R-matrix theory [1] just above the first $1s$ photodetachment threshold, is somewhat troubling, especially since the higher energy range displays excellent agreement. However, a similar discrepancy was observed for Li^- [3,10]. The only important approximation in the calculations, for both cases, is the omission of the higher members of the set of singly excited and doubly excited final states of the neutral atom, along with the continua associated with each of them. One would expect this omission to affect the higher energy photodetachment cross section, which probes directly the more highly excited states of the

neutral atom, more than the lower energy. Evidently this is not the case. We hope our experimental results will, however, stimulate further theoretical studies and provide a resolution to this discrepancy.

In summary, the first experimental K-shell photodetachment of an open-shell negative ion, He^-, leading to He^+, has been performed. It reveals a wealth of structure, qualitatively and quantitatively unlike photoabsorption of atoms and *positive* ions; structure that arises from the dominance of electron correlation in *both* initial and final states of the He^- ion. The measurements also provide an unambiguous test of two independent calculations [1,2] that showed major discrepancies, even though both in principle included similar correlation effects, and clearly demonstrated which calculation is correct.

Finally, although the initial $1s2s2p\ ^4P$ state of He^- is highly correlated, this correlation is in the outer shell; the $1s$ wave function is almost unaffected by the outer electrons and is virtually the same as in He^+. Thus, the study of K-shell photodetachment is essentially a study of outer shell correlation of initial and final states probed from within. The fact that $1s$ photoabsorption of He^- is so different from He^+ is a clear signature of the dominance of correlation effects. In our judgment, investigations of innershell photodetachment of other systems are likely to yield new surprises; we hope that this work will stimulate such experimental and theoretical studies.

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