Partial Cross Sections for the Photodetachment of Metastable He^-

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Partial cross sections for the photodetachment of a 2s or 2p electron from metastable $(1s2s2p)^4P$ He^- ions have been determined using crossed-beam photoelectron spectroscopy. Kinematic effects associated with detachment from a fast beam source have been exploited, in a novel way, to enhance the precision of the measurements. Calculated cross sections for the photodetachment of H^- were used to establish a scale for the He^- measurements. Radiative attachment cross sections are derived from the photodetachment results using the principle of detailed balance.

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The three-electron He^- ion has a relatively simple structure. As such, it has received a great deal of experimental and theoretical attention since its discovery by Hiby.[1] It is well known that stable He^- ions are nonexistent due to the fact that an electron is unable to attach itself to a He atom in its ground state. The He^- ion is instead the prototype of an unusual class of unstable, yet long-lived, negative ions that are metastable against decay via electron (autodetachment) and photon (electric dipole) emission. The ion is formed in the core-excited and spin-aligned $(1s2s2p)^4P$ state when an electron attaches itself to a He atom in the metastable $(1s2s)^3S$ excited state. Autodetachment via the strong electrostatic interactions is spin forbidden. The process proceeds, however, at a slower rate via the weaker magnetic interactions among the electrons. There exist a differential metastability among the fine-structure levels due to their different coupling strengths to the continuum. The lifetimes of the three levels and their relative spacings have been measured by Novick and co-workers.[2] The He^- ion lives long enough (tens to hundreds of microseconds) that a beam of ions can be formed in an accelerator and subsequently used as an essentially stable source for experiments. The structure of He^- is now well established. The electron affinity of $He(2^3S)$ has been calculated by Bunge and Bunge[3] to be 77.51 ± 0.04 meV. This quantity was first measured by Brehm, Gusinow, and Hall[4] using crossed laser-ion-beam photoelectron spectroscopy and later by Peterson, Bae, and Huestis[5] using a merged-beam threshold photodetachment technique. The latter value is in excellent agreement with theory.

The detailed manner in which metastable ions such as the He^- ion interact with radiation is not as well known, however, although the subject is of fundamental interest. In particular, experimental data on absolute partial cross sections, which are the subject of this paper, are sparse. The simplest stable negative ion, H^-, has a single open photodetachment channel, $h\nu + H^-(1\,^1S) \rightarrow H(1\,^2S) + e^-(kp)$, in the visible ($h\nu < 10.96$ eV). In contrast, photodetachment of He^- can lead to five possible final (continuum) states in the visible ($h\nu < 4.85$ eV); i.e., ^4S and ^4D states associated with the process $h\nu + He^-(2\,^4P) \rightarrow He(2\,^3S) + e^-(ks,d)$ and ^4S, ^4P, and ^4D states associated with the process $h\nu + He^-(2\,^4P) \rightarrow He(2\,^3P) + e^-(kp)$. The technique of energy and angle-resolved photoelectron spectroscopy has allowed us to quantitatively investigate, as a function of photon energy, the competition between the two resolved photodetachment processes that leave the He atom in the $2\,^3S$ and $2\,^3P$ states, respectively. The partial cross sections, $\sigma(^3S)$ and $\sigma(^3P)$, for the two processes have each been measured relative to the known cross section, $\sigma(^2S)$, for photodetaching a reference D^- ion. As far as it is known, the present data represent the only published measurement of branching ratios for competing photodetachment channels. At the present time there are no published calculations of the partial cross sections for He^- photodetachment, although work is currently in progress.[6]

The cross-section ratios were determined from measurements of the yields and angular distributions of photoelectrons ejected from the ions at the intersection of crossed laser- and negative-ion beams. The fast moving (~ 30 keV) and tenuous beam of negative ions was produced by double charge exchange between a beam of positive ions and atoms in a Li vapor cell. The negative-ion beam was intersected orthogonally by a linearly polarized beam of photons from a flashlamp-pumped pulsed dye laser. Sets of apertures were used to ensure that the overlap of the two beams remained unaltered during the relative measurement process. Following photodetachment events, electrons ejected from the He^- ions, in the direction of motion of the ion beam (forward-directed electrons), were collected and energy analyzed using a spherical-sector electron spectrometer.

The angular distributions of the detached electrons were determined by measuring their yields as a function of the angle between the fixed collection direction and the direction defined by the polarization vector of the laser beam. The polarization vector could be rotated in a plane perpendicular to the propagation direction of the laser by the use of a $\lambda/2$ phase retarder (double Fresnel rhomb). The output power of the laser was carefully chosen to avoid saturation on each of the photodetachment processes studied. The instantaneous intensities of the photon and ion beams were monitored for normalization purposes. A synchronous detection scheme, based on the time structure of the pulsed laser, was used to discriminate against the rather large electron background. Further details of this crossed-beams apparatus can be found in a previous publication.[7]

The relative cross sections (angular integral) were obtained by comparing, under identical geometrical conditions, the yields of electrons, $Y(\mathrm{He})$ and $Y(\mathrm{D})$, produced in the photodetachment of beams of He^- and D^- ions, respectively. The cross-section ratio for these two processes can be expressed by the following relation:

$$\frac{\sigma(\mathrm{He})}{\sigma(\mathrm{D})}=\frac{Y(\mathrm{He})\phi(\mathrm{D})\rho(\mathrm{D})g(\mathrm{D})[1+\beta(\mathrm{D})]}{y(\mathrm{D})\phi(\mathrm{He})\rho(\mathrm{He})g(\mathrm{He})[1+\beta(\mathrm{He})]}\,. \quad (1)$$

In this expression the yield ratio is multiplied by several measured factors that take account of the different photon fluxes, ϕ, ion densities, ρ, frame-transformed solid-angle factors, g, and asymmetry parameters, β, associated with photodetaching electrons from the two beams. All geometric factors are adjusted to be equal and therefore cancel out. A novel technique, which is illustrated in Fig. 1, has been used to cancel out the efficiency factors associated with the collection and detection of electrons from the two beams. The three photoelectron spectral peaks shown in Fig. 1, which have different energies in the ion frame, can be kinematically shifted to the same energy in the laboratory frame by an appropriate choice of the ion-beam energies, E_i. The efficiencies for collecting and detecting electrons of the same energy from the He^- and D^- beams then become equal and cancel out. The ion-beam energies, which are needed to determine both the ion densities and solid-angle transformation factors shown in Eq. (1), can be determined precisely ($\sim 0.1\%$) from an *in situ* analysis of the separation of the peaks in the photoelectron spectra. For example, the He^- photodetachment spectrum consists of a pair of peaks (one for each exit channel) whose separation in the ion frame, $E(^3S\text{-}^3P)$, is well known from photon spectroscopy. A determination of their separation in the laboratory frame can then be used to measure the ion-beam energy. This technique, which depends on either kinematic shifting or doubling of the spectral lines, is described in more detail by Pegg *et al.*[7] Angle-resolved yield measurements were made on each spectral line to determine the asymmetry parameters β that characterize the shape of the photoelectron emission patterns. The

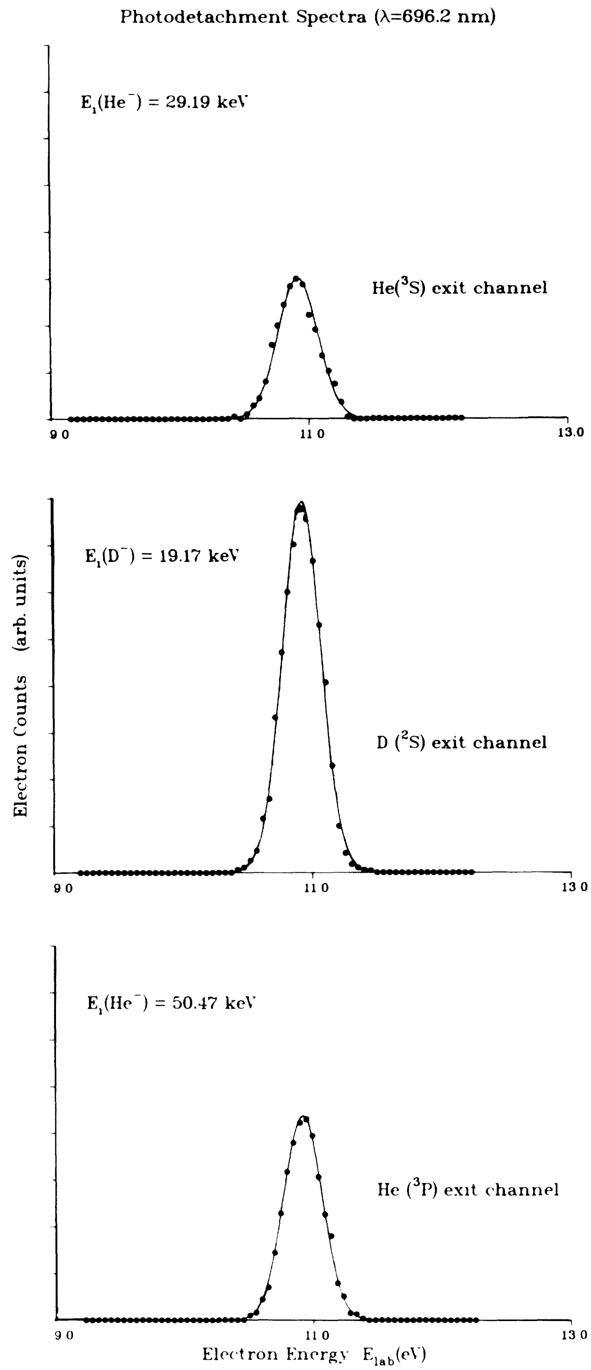

FIG. 1. Three spectral peaks kinematically shifted to the same laboratory-frame energy by an appropriate choice of ion-beam energies. The peak yields were used in the measurement of relative cross sections for the photodetachment of He^- and D^- ions.

measurement technique was first tested by photodetaching electrons from beams of D^- and Li^- ions. The predicted $\cos^2\Theta$ ($\beta=2$) distribution was obtained in all cases. The ^3P exit channel in He^- photodetachment involves the ejection of an s orbital electron, similar to photodetachment in the stable D^- and Li^- ions. The measured values of β, however, are considerably smaller ($\beta \sim 1.5$) than the $\beta = 2$ predicted by an independent-electron model. It is suspected that the presence of the $(1s2p^2)^4P$ shape resonance[5] lying just above the threshold for the opening of the ^3P channel is perturbing the angular distribution of the photoelectrons in this case. The spectral dependence of the asymmetry parameters characterizing the angular distribution of electrons ejected in the photodetachment of He^- are the subject of a paper by Thompson *et al.*[8] Calculations have been made by Saha and Compton.[6]

The partial cross-section ratios, $\sigma(^3S)/\sigma(^2S)$ and $\sigma(^3P)/\sigma(^2S)$, have been measured at photon energies of 1.781, 1.946, and 2.091 eV. The results are $\sigma(^3S)/\sigma(^2S) = 0.60 \pm 0.02$ (1.781 eV), 0.57 ± 0.04 (1.946 eV), and 0.49 ± 0.04 (2.091 eV); $\sigma(^3P)/\sigma(^2S) = 0.28 \pm 0.02$ (1.781 eV), 0.18 ± 0.02 (1.946 eV), and 0.12 ± 0.01 (2.091 eV). The uncertainties quoted on these values represent 2 standard deviations of the weighted mean of several data sets each comprising a number of individual spectra added together. The major source of statistical error in each data set originates in the measurement of yield ratios ($\sim 4\%$) and asymmetry-parameter ratios ($\sim 3\%$). Uncertainties in other measured quantities are considered to be negligible. Systematic error sources have been investigated and are estimated to be within the quoted 2σ limits. A scale for the relative cross-section measurements can be readily established by assuming theoretical values for the $\sigma(^2S)$ cross section for photodetaching the D^- ion. Several calculations of H^- photodetachment cross sections have been made and can be used for this purpose. The perturbation-variation results of Stewart[9] have been arbitrarily chosen for the purpose of normalization in the present work. The results of Broad and Reinhardt[10] fall within 2% of those of Stewart over the range of the present experiments. Combining the theoretical values (assuming a 3% uncertainty) with the measured cross-section ratios produces the partial cross sections (in Mb) shown in Table I. The sum of the partial cross sections, σ_{total}, and the measured asymmetry parameters, β, are also tabulated. The branching ratios for the ^3S channel are 0.70 ± 0.06 (1.781 eV), 0.75 ± 0.10 (1.946 eV), and 0.80 ± 0.18 (2.091 eV). The measured partial and total cross sections are also shown in Fig. 2, along with the results of the total cross-section calculation of Hazi and Reed.[11] The calculation and the sum of the present partial cross-section measurements are seen to be in good agreement at all the photon energies used in the experiment. The only other partial cross-section measurement for He^- is that of Brehm, Gusinow, and Hall.[4] Their estimates, at a photon energy of 2.410 eV, are limited in precision to a factor of 2 since no angular distribution information was obtained in the experiment. The less precise total cross-section measurements of Compton, Alton, and Pegg[12] and Hodges, Coggiola, and Peterson[13] are in essential agreement with the sum of the partial cross sections measured here.

TABLE I. Photodetachment of He^-: cross sections (Mb) and asymmetry parameters.

	Photon energy (eV)		
	1.781	1.946	2.091
$\sigma(^3S)$	22.9 ± 1.0	20.5 ± 1.5	16.6 ± 1.8
$\beta(^3S)$	1.15 ± 0.02	1.28 ± 0.02	1.19 ± 0.07
$\sigma(^3P)$	10.0 ± 0.6	6.7 ± 0.6	4.1 ± 0.4
$\beta(^3P)$	1.52 ± 0.04	1.59 ± 0.03	1.53 ± 0.05
σ_{total}	32.9 ± 2.4	27.2 ± 3.1	20.7 ± 3.0

The measured cross sections for photodetaching an electron from the He^- ion can be used to indirectly determine, using detailed balance arguments,[14] the cross sections, σ_a, for the far less probable inverse process of radiative attachment. The derived radiative attachment cross sections (in barns) are $\sigma_a(^3S) = 166 \pm 7$ (1.781 eV), 162 ± 12 (1.946 eV), and 120 ± 13 (2.091 eV); $\sigma(^3P) = 74 \pm 5$ (1.781 eV), 45 ± 4 (1.946 eV), and 30 ± 3 (2.091 eV).

The relatively high precision ($\sim 5\%$ in favorable cases) of the present cross-section ratio measurements reflects our ability to exploit, in a novel way, certain kinematic effects associated with a fast moving source of ions and the simultaneous collection of electrons in the forward direction. These features of the present apparatus have not been used in previous photodetachment

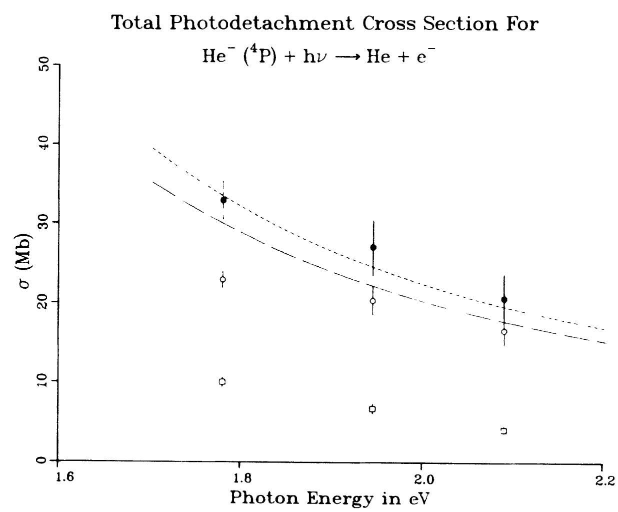

FIG. 2. The open squares and circles represent the partial cross sections for photodetaching He^- via the $He(^3P)$ and $He(^3S)$ exit channels, respectively. The solid circles, which are the sum of the partial cross sections, are compared with the results of the total cross-section calculation of Ref. 11 (shown as continuous curves). The length and velocity forms are represented by the upper and lower curves, respectively.

studies. A scale for the relative measurements has been established by the use of accurate theoretical values for the cross sections for the photodetachment of the reference ion, D^-. As a result we have been able to determine partial cross sections for the photodetachment of He^- to $< 10\%$.

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