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Measurement of the $1s2p2p'\ ^4P^e$ Resonance in He$^-$ Photodetachment

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A large resonance in the photodetachment spectrum of metastable He$^-$, which was recently predicted theoretically, has been observed experimentally. Located at 1.2344 eV with a width of 7.0 meV, the detachment cross section reaches a maximum of ~ 70 Å^2. Its size results from the large oscillator strength associated with the first allowed transition in He$^-$, $(1s2s2p)^4P^o \rightarrow (1s2p2p')^4P^e$.

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Although He$^-$ has been known for a number of years to exist in the metastable $(1s2s2p)^4P$ state, and is the simplest negative ion next to H$^-$, little attention has been given to its photodetachment properties until recently and very little theoretical work has been done on higher-lying He$^-$ states in the quartet system (as compared to the doublet resonances seen in electron scattering). Recently, we made a preliminary survey[1] of the He$^-$ photodetachment spectrum, using a variety of lasers to make measurements at 15 photon energies between 0.12 and 4 eV. Independently, and by a different measurement technique, Compton, Alton, and Pegg[2] obtained cross sections between 1.77 and 2.75 eV using a flash-lamp-pumped dye laser. Agreement between these two experiments was quite good, considering their preliminary nature.

Although there were large energy gaps between some of our data, they nevertheless showed an interesting skeletal profile, and suggested[1] that the cross section increases significantly at or near the energy threshold for leaving the neutral He in the excited 2^3P state. Subsequently, Hazi and Reed[3] carried out the first photodetachment calculations on He$^-$. They obtained quite good agreement with our results, and found a large peak just above the 2^3P threshold, where the cross section increased by two orders of magnitude to about 25 Å^2. From a separate scattering phase-shift analysis, they found the peak to be a shape resonance located at about 1.233 eV, associated with the $(1s2p2p')^4P^e$ state. Other recent calculations[4,5] have also found this He$^-$ state to lie just above He(2^3P).

In Fig. 1 we compare a logarithmic plot of our original data with the results of Hazi and Reed. Although our results are on the average 30%–40% higher than the calculations, the relative agreement is excellent. The obvious prominence of the predicted resonance stimulated our desire to measure it experimentally.

In this Letter we report the successful observation of this resonance, which occurs in an experimentally difficult wavelength region between 900 and 1012 nm. To our knowledge, these measurements make the first experimental use of a tunable cw dye laser at these wavelengths. The results confirm the essential aspects of the Hazi and Reed calculations, while finding the peak cross section even higher than predicted. This feature dominates the He$^-$ photodetachment spectrum far more than the corresponding 2^1P resonance in H$^-$, which reaches only 1 Å^2 with a width of 16 meV.[6,7]

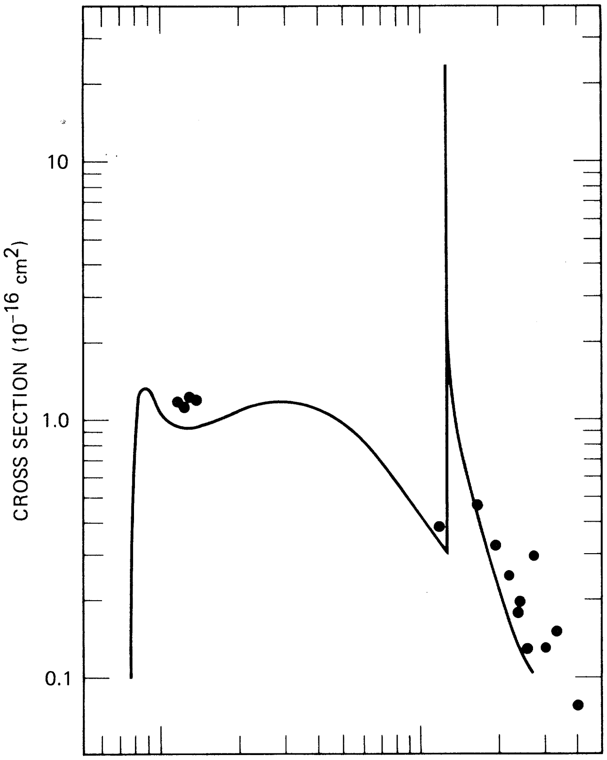

FIG. 1. He$^-$ photodetachment cross section as a function of photon energy. Solid curve: calculations of Hazi and Reed (Ref. 3); data points: previous results (Ref. 1).

The experimental apparatus and method described earlier[1] were used here with only minor modifications. The He$^-$ ions are formed from a 1300-eV He$^+$ beam by two successive electron-capture collisions in an alkali-vapor oven (we used both Na and K). After passing into a separately pumped interaction chamber the He$^-$ component was electrically deflected through 15° and directed along a 15-cm field-free drift path, where it was intersected by the laser beam at an angle of ~60°. The drift path terminated in an ion-beam–defining aperture (2.4 mm diam), behind which the ions were swept away and into a Faraday cup by a second electric field. Neutral atoms formed along the drift path were detected and counted with a Channeltron electron multiplier which followed the second deflection field. The laser beam was mechanically chopped and counts with laser on and off were stored in a PAR model-1112 processor.

Relative cross sections were obtained by normalizing the difference in counts accumulated with the laser on and off to the background (laser off) caused by autodetachment. A small component of the background caused by collisional detachment was determined at several pressures and was subtracted from the background. Absolute cross sections were obtained[1] by using the measured autodetachment lifetimes[8] of 16 ± 4, 10 ± 2, and 500 ± 200 μs for the $J=\frac{1}{2}$, $\frac{3}{2}$, and $\frac{5}{2}$ fine-structure states, and the established initial statistical population[8] to determine the negative-ion flux.

Considerable attention was given to the overlap of the laser and ion beams. Variable apertures at both entrance and exit windows in the apparatus enabled accurate laser beam alignment as well as a determination of the photon flux as a function of aperture diameter, which was fitted by a Gaussian distribution. The ion beam diameter was determined by the circular aperture that terminated the ion drift path; the ion beam was assumed to have constant density over this diameter. The beam overlap integral was solved numerically as a function of the ratio of the laser beam diameter at the $\frac{2}{3}$ power point to the diameter of the ion beam. The laser profile was determined for each measurement. The laser power was measured with a calibrated Spectra Physics model-404 power meter.

Leduc and Weisbuch[9] had found that IR-140 dye could be made to lase in the range 860–1013 nm when pumped by the ir lines of a Kr$^+$ laser.

Using a CR3000K laser with about 2.3–2.4 W output in these lines at 752 and 799 nm to pump a Coherent model-590 dye laser, we were able to cover the 930–990 nm range without great difficulty using the normal dye-laser ir optics. With considerable time and effort we were able to extend the measurements to 1008 nm.[10] Finally, with the use of a parabolic pump mirror and short-focus folding mirror as recommended by Leduc and Weisbuch, we were able to reach 1012 nm on the low-energy side of the peak. In most measurements beyond 995 nm, the laser power was less than 20 mW, and the typical signal/background ratio was 10^{-3}. With a total count rate limited to $\sim 10^4\ \mathrm{s}^{-1}$ to assure linearity, long runs (typically 1 h) were required to reduce the uncertainties due to counting statistics and beam fluctuations. The monochromator used to measure the photon wavelength was calibrated against four NeI lines between 944 and 966 nm, and we estimate an absolute uncertainty of ≤ 0.15 nm, or 0.18 meV near the resonance peak. The Doppler shift arising from the 60° angle of intersection was accounted for.

The measurements reported here cover the sharp portion of the resonance that rises above $1\times10^{-16}\ \mathrm{cm}^2$ in Fig. 1. The results are shown in a semilog plot in Fig. 2. The error bars represent probable errors based on variations from the mean of a number of short (10–15 min) runs made at each wavelength.

The ^4P partial cross sections of Hazi and Reed[3] for $\mathrm{He}^- + h\nu \rightarrow \mathrm{He}(2^3P) + e(kp)$ are also shown.

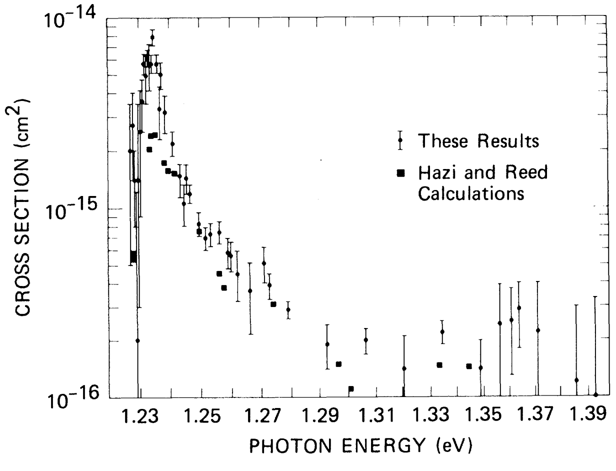

FIG. 2. Semilog plot of He$^-$ photodetachment cross sections obtained in this work, and the calculated values of Hazi and Reed for $h\nu + \mathrm{He}^-(^4P^o) \rightarrow \mathrm{He}(2^3P) + e(kp)$, $^4P^e$.

They find an additional 10% contribution to the summed oscillator strength in this interval due to the $^4D^e$ channel.[11] Figure 2 shows excellent agreement between experiment and calculations in the general shape and location of the peak. A linear plot of the data between 1.22 and 1.27 eV in Fig. 3 shows that the measured values are actually considerably higher than the calculations in the immediate vicinity of the peak, reaching about 70 Å^2, compared with the calculated maximum of 24 Å^2. On the other hand, we find that the peak is located at 1.2344 ± 0.0004 eV, with a full width at half maximum of 7.0 ± 0.4 meV, in excellent agreement with the values of 1.233 eV and 7 meV which Hazi and Reed determined from the scattering phase-shift analysis.

We determined the oscillator strength responsible for the cross section $\sigma(\nu)$ from the relation $f = mc(he^2)^{-1}\int\sigma(\nu)d\nu$, finding $f = 0.49$ for energies between 1.22 and 1.25 eV, close to the resonance, and $f = 0.72$ for the more extended range 1.22–1.42 eV. Hazi and Reed obtain[11] a summed oscillator strength of 0.54 including the 10% ^4D contribution, between 1.22 and 1.42 eV,[10] or 72% of our value. These are large values but comparable to $f = 0.54$ for the similar $(1s2s)2^3S - (1s2p)2^3P$ transition in He. The transition $(1s2s2p)^4P^o$

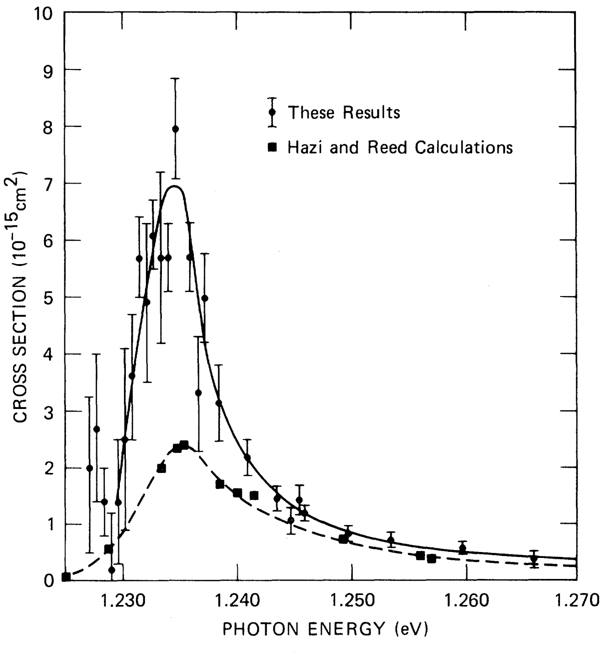

FIG. 3. Linear plot of the present results in the immediate vicinity of the resonance. Squares are the $^4P^e$ cross sections of Hazi and Reed. The smooth curves are visual fits to the data.

→ $(1s2p2p')^4P^e$ is the first fully allowed transition in He⁻, and as an open channel shape resonance above 2^3P it accounts for the extreme magnitude of the detachment cross section. We find an increase from 0.39 Å^2 at 1.117 eV[1] to 70 Å^2 at 1.23 eV, a factor of 180.

Our absolute values are generally 20%–40% higher than the calculations[3] at all energies, perhaps partly because of our normalization using the measured autodetachment lifetimes. If the actual autodetachment rate is slower than that obtained from the measured lifetimes, then our absolute cross sections are correspondingly too high. As measurements of slow decay rates are apt to err on the fast side as a result of other losses, our cross sections could be uniformly high. However, the relative energy dependence would be unaffected.

These results confirm the location and width of the resonance determined from the scattering phase-shift calculations of Hazi and Reed,[3] but find the peak higher and narrower than the cross-section calculations. The close agreement in location and width also lends confirmation to the value 77.4 ± 0.2 meV for the electron affinity of He 2^3S calculated by Bunge and Bunge[12] and also obtained by Hazi and Reed.[3]

Finally, we mention the possibility of additional structure in the 1.2–1.4 eV region. In Fig. 2 there is an apparent increase near 1.35 eV, and Fig. 3 shows some evidence of structure below 1.23 eV. The latter is suggestive of an interference profile, and the former could suggest the possibility of ^4S or ^4D structure in this area. We plan to investigate these regions in detail and will attempt to reach the 2^3P threshold using a much more sensitive experimental configuration.

We thank A. V. Hazi for pursuing the first photodetachment calculations on He⁻ and for communicating the results. We are grateful to T. J. Johnston (Coherent Radiation) for valuable help with the infrared dye-laser operation, and to T. F. Johnston (Coherent Radiation) for the use of optical components. We also thank Phil Cosby and Dave Huestis for experimental and theoretical support. This work was supported by the National Science Foundation and the U. S. Air Force Office of Scientific Research.

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