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ELECTRON AFFINITY OF HELIUM VIA LASER PHOTODETACHMENT OF ITS NEGATIVE ION*

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In this Letter we report the first direct measurement of the electron affinity of helium (2^3S), by the technique of energy analysis of electrons photodetached from a beam of negative helium ions by monochromatic (laser) light.

Although the He^- ion was first identified experimentally in 1939 by Hiby,[1] it was not until 1955 that Holøien and Midtdal[2] showed theoretically that the $1s2s2p\,^4P$ state would be sufficiently metastable against autoionization to account for Hiby's observation. Holøien and Midtdal employed a variational technique to show that the $1s2s2p\,^4P$ state of negative helium lies below the $1s2s\,^3S$ state of neutral helium. Pietenpol[3] has shown that the $J=\frac{5}{2}$ component of this state is strongly metastable, with a calculated lifetime against autoionization of about 1 msec. A reliable theoretical value for the affinity is difficult to obtain because the calculated variational upper bound to the negative-ion energy level must be subtracted from the large 2^3S term value (19.8 eV) of excited, neutral helium. In their recent paper Holøien and Geltman,[4] using a 30-parameter wave function, find an affinity $E_A \geq 0.033$ eV.[5] They also quote the unpublished result $E_A \geq 0.069$ eV obtained by Weiss using the method of superposition of configurations.[6] This latter result indicates that the barrier penetration analysis used by Smirnov[7] to interpret the electric field quenching data of Riviere and Sweetman[8] is too uncertain in this case to yield a precise value for the electron affinity.

In the present experiment we analyze the energy of the electrons photodetached from a beam of negative helium ions passing through the high-power intracavity electromagnetic field of a continuous-duty argon-ion laser. With this monochromatic source one expects the electron energy spectrum to show a sharp peak at the appropriate energy-conserving kinetic energy. Transitions from excited ion states and/or residual excitation left in the neutral atom (or molecule) will consequently appear directly in the electron energy spectrum.

Our apparatus (illustrated in Fig. 1) consists basically of five parts: ion source, Wien (mass) filter, laser light source, electron energy analyzer, and pulse-counting and control electronics. Figure 2 shows the interaction region in more detail.

Positive ions of helium and deuterium (and sometimes hydrogen) are extracted from a hot-cathode arc-discharge source. This beam of about 0.1 μA is accelerated to 2.5 keV and focused through an oven containing potassium vapor at a few microns pressure. Here some 1 % of the He^+ (and perhaps 10 % of the H^+ and D^+) double-charge exchange with the potassium, producing a remarkably monoenergetic well-defined beam of the desired negative ions.[9] These ions are then decelerated to 700 V, elec-

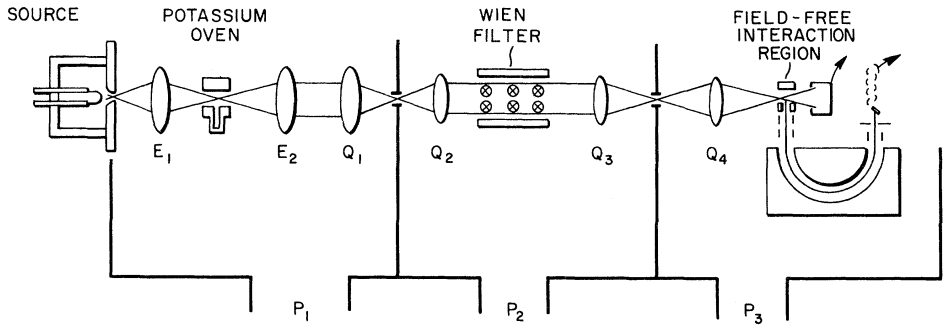

FIG. 1. Block diagram of the ion-beam apparatus. E_1 and E_2 are "Einzel" lenses. The symbol Q represents a 12-element symmetrical quadrupole lens. The operating pressure in the interaction chamber is held to 10^{-8} Torr or less by the 200-liter/sec ion pump, P_3.

trostatically deflected through 10°, and focused through a differential pumping iris. This early separation of the negative beam from the positive and neutral beams was found essential to minimize background. The recollimated negative-ion beam next may be mass analyzed by the Wien filter, a high-transmission, variable-resolution velocity filter utilizing balanced magnetic and electric deflection.[10] Behind the exit slit of the analyzer, which also serves as a differential pumping iris, the negative-ion beam is brought to a focus in the interaction chamber. We estimate the diameter of the beam to be somewhat less than 1 mm at the focus, with $f/6$ convergence. The ions

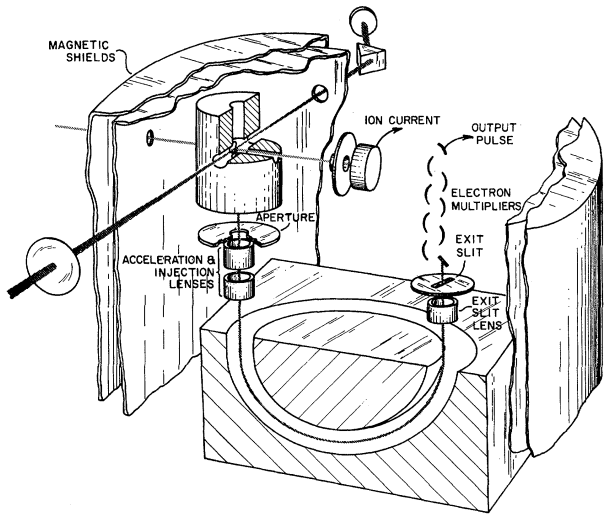

FIG. 2. Detail of the interaction chamber and electron analyzer. To increase the useful solid angle, and to minimize the effect of patch fields, the electrons are accelerated to 11.2 eV for analysis. The nominal trajectory has about 5.7 cm radius. All critical surfaces are copper.

are then collected in a guarded Faraday cup.

The argon ion laser uses 5 kW average input to the hot-cathode discharge through a water-cooled quartz capillary tube, 2.5 mm i.d. and 50 cm long. A lens of 32-cm focal length between the discharge tube and the interaction chamber produces a focal spot of approximately $\frac{1}{4}$ mm diam. The light passes at Brewster's angle through a prism for wavelength selection. The laser cavity is completed by a mirror of 32-cm radius of curvature. High-quality fused quartz windows are mounted at Brewster's angle in the vacuum envelopes of both the laser and the photodetachment chamber. Because of the inevitable optical losses, the presently available circulating power is only about $\frac{1}{2}$ W (each way) on either of the two strongest argon lines, 4880 and 5145 Å. This value of light flux results in an integrated photodetachment probability of something in excess of 10^{-5} for a typical negative ion.

Only those electrons emitted into the 10^{-2} sr accepted by our electron-injection optics can travel out of the field-free interaction chamber into the hemispherical electron analyzer,[11] where they have an energy of 11.2 eV. The electrons are further accelerated after the analyzer to 112 eV before encountering the exit slit, to minimize the effect of patch fields on resolution. Those electrons passing through the 0.012-in. (50-mV equivalent full width at half maximum) exit slit are accelerated and finally impinge on the cathode of an electron multiplier. To discriminate against background (arising from collisional detachment, multiplier dark pulses, etc.) we use a digital form of synchronous detection. The laser is triggered at a 120-cps rate (with about a 30 to 40% duty factor) and the multiplier pulses are accumulated in the "signal plus background" or

the "background only" channels of a dual scaler, as appropriate. The scaler gate times are digitally derived from a quartz oscillator to allow extended counting of weak signals without base-line errors. For convenience, the output of a photocell monitoring the laser is fed to a precision integrator whose output pulse, upon reaching a preselected integral level, terminates the counting. The electron analyzer voltages are obtained from a high-stability power supply programed by a precision decade resistor box, thus obviating the need for large numbers of precise voltage measurements.

In the analysis of experiments on the lighter negative ions, it is essential to recognize the non-negligible velocity of the negative-ion center of mass: The electrons must be emitted slightly in the backward direction in the center-of-mass system to fall within our collection angle (90° laboratory angle with respect to the beam axis). This transformation into laboratory coordinates carries with it a reduction of kinetic energy, amounting to 190 mV for a 700-V deuterium beam. Since this kinematic correction can be very accurately calculated if the ion beam energy is well known (see later), negative-ion energy levels accurate to a few millivolts may immediately be calculated from the observed electron kinetic energy spectra. For still more precise results —such as reported here—we may use the comparison with the experimental isotope shift (hydrogen versus deuterium) to refine our knowledge of the 90° angle between the ion beam and the direction of electron collection. Optionally, the difference in the laboratory electron energies for 2^3S and 2^3P final helium-atom states may be compared with the known spectroscopic splittings to give another measure of the angle. The ion energy was determined by a differential retarding analysis by applying a bias to the Faraday cup and guard. Tests performed with 700-V positive ions showed energy widths of about $\frac{1}{2}$ V and agreement to within 1 V with the dc potential applied to the ion source. Thus, for the negative ions we can have confidence in the 11-eV typical widths and 4-eV typical energy loss from the value calculated from the applied voltages. Measurement of the electron's laboratory energy for an ion (H^- or D^-) of known affinity[12] gives the contact potential differential between the externally applied and the internal effective potentials.

To eliminate any time-dependent or beam-dependent drifts, all the data reported here have been taken with a "combined beam" technique, whereby the ion source is made to produce the desired negative ion and a reference ion, for example H^- and/or D^-. By virtue of its linear construction, the Wien filter may be operated at zero mass resolution. Typically we obtained 3 nA total beam current: 1.5 nA of He^-, 1 nA of D^-, and 0.5 nA of H^-.

To test our understanding of the systematic corrections required, we have listed in Table I the results of three separate experiments, of several runs each.[13] Experiment I used the splitting between 2^3S and 2^3P final states to determine the effective collection angle as described previously. One run of this group is shown in Fig. 3. The average of six runs with line centers taken from plots of the individual runs gives 80.5 ± 3.0 mV. Experiment II is probably our best, with all parameters such as laser wavelength (5145 Å) and beam energy (715 eV) nearly optimum. Although some of the early runs in this series were affected by oxygen contamination of the negative-ion beam, the last four runs were essentially limited only by counting statistics. A computer was used to least-squares fit these runs with a weakly skewed Lorentz function, which is a usable approximation to our analyzer response function. The 1-mV probable error appears to be mainly statistical in origin. In experiment III we deliberately changed the conditions which influence the known systematic effects. The bluer but weaker 4880-Å line gives 130 mV more, the shift of beam energy downward to 606 eV gives 29 mV less loss of energy due to kinematics, and the laser spot was repositioned to change the "90°" collection angle. Although the signal-to-background ratio was somewhat degraded, we obtain the same result again, within the statistical error. Systematic shifts due to two finite solid-angle effects have been numerically estimated and included in the results, although the largest is only 0.9 mV. Thus, we believe the affinity of $He(2^3S)$ is known to be 80.0 mV with about 2 mV probable error. We have assigned ±8 mV as the error limits at 95% confidence for systematic effects, unknown at present.

The cross section for photodetachment of He^- into helium 2^3P may be directly taken from the count rates and beam densities normalizing against the known H^- (D^-) values.[14] In these three cases an s electron goes into p-wave con-

Table I. Summary of experimental results.

Experiment	λ Å	V_{beam} (eV)	$E(He,S)-E(He,P)$ (mV)	$E(He,S)-E(D)$ (mV)	$A(He,^3S)$ (mV)
I					
He^S, He^P, D	5145	723.5	1131.2 ± 1.6	778.3 ± 2.0	80.5 ± 3.0[a]
			$E(D)-E(H)$	$E(He,S)-E(D)$	
II					
He^S, D, H	5145[b]	715	t_1 192.4	769.0	80.9
			t_2 192.7	769.3	80.4
			t_4 192.9	770.9	79.0
			192.2 ± 0.5	769.3 ± 0.8	80.9 ± 1.0[c]
III					
He^S, D, H	4880	606	170 ± 5	762	78 ± 2[d]
Final average					80.0 ± 2[e]

[a]Average of six runs.
[b]At λ = 5145 Å, $\sigma(He\,^3P) = 7\times10^{-19}$ cm^2 ($\pm 2x$), $\sigma^*(He\,^3S) = 5\times10^{-18}$ cm^2 ($\pm 2x$) (see text).
[c]Average of four runs.
[d]Sum of six runs.
[e]Probable error; limit of error (95% confidence) is ±8 mV.

tinuum. The He^- differential cross section to photodetach into the 2^3S is about 7 times larger than the corresponding 2^3P differential cross section.[15] Because of the availability of both s-wave and d-wave channels, the angular distribution and hence the total cross section must be regarded as unknown at present.

We expect this laser photodetachment technique for the study of negative ions to yield reliable energy-level information for a number of interesting negative ions, such as O_2^-, NO^-, N^-, alkali-metal negative ions, and halogen molecular negative ions.

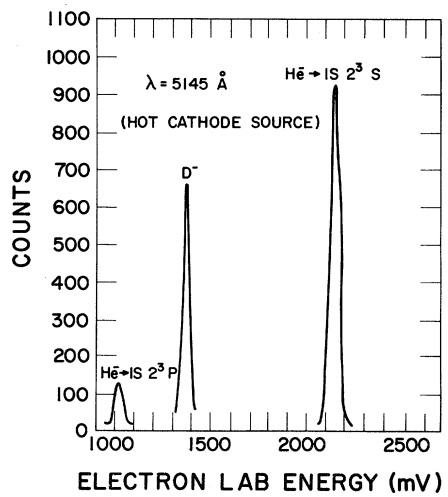

FIG. 3. Plot of a typical run from Experiment I. Total counting time here was about 20 min. The energy scale has not been corrected for the small contact-potential shift (< 50 mV). The weakest peak is truly helium, as evidenced by its observation with the mass-analyzed beam.

We are indebted to R. Ennis of Ortec for assistance in getting the double-charge-transfer apparatus set up and working and for the loan of some important components. We are indebted to C. Kuyatt and J. A. Simpson for their expert design of the electron analyzer and for their helpful comments. We are particularly pleased to recognize the coupling between the presence of Dr. E. Holøien as a Visiting Fellow at Joint Institute of Laboratory Astrophysics in 1965-1966 and our interest in helium negative ions. We have profited greatly from discussions with L. M. Branscomb about negative ions and negative-ion sources. His generous help and enthusiasm have made this work "faster and livelier."

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[1]J. W. Hiby, Ann. Physik 34, 473 (1939).
[2]E. Holøien and J. Midtdal, Proc. Phys. Soc. (London) A68, 815 (1955).
[3]J. L. Pietenpol, Phys. Rev. Letters 7, 64 (1961).
[4]E. Holøien and S. Geltman, Phys. Rev. 153, 81 (1967).
[5]These authors comment that when an error in Ref. 2 is corrected, the original four-parameter calcula-

tion actually leads to an unbound state.

[6]Footnote f in Table I of Ref. 4.

[7]B. M. Smirnov, Teplofiz. Vysokikh Temperatur, Akad. Nauk SSSR 3, 775 (1965) [translation: High Temp. 3, 716 (1965)].

[8]A. C. Riviere and D. R. Sweetman, Phys. Rev. Letters 5, 560 (1960).

[9]B. Donnally, to be published.

[10]The Wien filter construction is based in part on the more sophisticated design by L. Wählin, used by the University of Colorado cyclotron group.

[11]C. E. Kuyatt and J. A. Simpson, Rev. Sci. Instr. 38, 103 (1967).

[12]We have used 755.1 mV as the affinity for hydrogen, taken from the calculation by C. L. Pekeris, Phys. Rev. 112, 1649 (1958).

[13]An additional experiment was done with an rf ion source. Unfortunately, no direct measurement of the ion energy was made; the value of 920 eV determined from the H-D splitting gives 78 mV for the helium affinity.

[14]S. Geltman, Astrophys. J. 136, 935 (1962).

[15]These differential cross sections are measured essentially in the direction of the optical electric vector.

ELECTRIC DIPOLE MOMENT OF THE CESIUM ATOM. A NEW UPPER LIMIT TO THE ELECTRIC DIPOLE MOMENT OF THE FREE ELECTRON*

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A few years ago, experiments were begun to search for an electric dipole moment (EDM) of the cesium atom.[1,2] The importance of these experiments lies in the fact that the observation of an EDM in an atomic system of well-defined angular momentum would be direct evidence for a violation of both space-inversion and time-reversal invariance.[3]

The experiments were performed on an atomic beam magnetic resonance apparatus. The transition between the levels (4, −4) and (4, −3) of cesium was induced in a Ramsey double hairpin in a weak uniform magnetic field $\vec{H}$. A set of parallel electric field plates was situated between the rf loops. A small resonance shift, linear in the applied electric field $\vec{E}$, was observed. This effect could possibly have been interpreted as due to the interaction of an EDM of the cesium atom with this field. Stated as an upper limit on the existence of an EDM of the cesium atom, the result of these experiments was $d_{Cs} = (2.2 \pm 0.1) \times 10^{-19}$ cm$\times e$.

As was pointed out at the same time, there was an alternative explanation for this effect. As the atom moves with velocity $\vec{v}$ through the electric field it experiences a motional magnetic field $\vec{v} \times \vec{E}$. A resonance shift, linear in $\vec{E}$, due to the interaction of the atom's magnetic dipole moment with this motional magnetic field results if $\vec{E}$ is not parallel to $\vec{H}$. The experiments are very sensitive to this alignment as a misalignment of 0.01 rad would explain the result above.

The possibility that a $\vec{v} \times \vec{E}$ effect was causing the linear shift was indirectly taken into account in later work by comparing shifts in various alkalies. This led to a new upper limit for the cesium atom of $d_{Cs} = (5.1 \pm 4.4) \times 10^{-20}$ cm$\times e$.[4,5] King,[6] using a square-wave phase-modulation technique to eliminate $\vec{v} \times \vec{E}$, found $d_{Cs} = (0.8 \pm 8.0) \times 10^{-20}$ cm$\times e$. Sandars,[7] in a recent experiment where $\vec{v} \times \vec{E}$ was taken into account by comparing results from beams in opposite directions, found as an upper limit $d_{Cs} = (9 \pm 10) \times 10^{-21}$ cm$\times e$.

In this Letter we wish to report the results of experiments in which $\vec{v} \times \vec{E}$ is measured directly on the Zeeman transitions $(F, m_F = -I \pm \frac{1}{2}) \leftrightarrow (F, -I - \frac{1}{2})$ of several of the alkali atoms. The approach differs from that used in the previous work in the following ways:

(1) A new, longer resonance region is used in which the $\vec{H}$ field (horizontal field) is produced by rectangular Helmholtz coils mounted on a cylindrical form enveloping the electric field plates and rf loops.[8] An additional pair of coils was mounted at right angles to the $\vec{H}$-field coils in order to produce a magnetic field (vertical field) perpendicular to $\vec{H}$. Adjustment of the current in this latter pair of coils allows us to vary the alignment of $\vec{E}$ and $\vec{H}$ electronically, and thus manipulate the $\vec{v} \times \vec{E}$ effect. The entire resonance region is magnetically shielded by three concentric Hipernom cylinders to minimize noise due to fluctuations in the ambient field.

(2) Digital signal-processing techniques are used. The direction of the $\vec{E}$ field is reversed periodically and the resonance signal is accumulated in two counters gated synchronously