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Helium Negative Ions from Metastable Helium Atoms

BAILEY L. DONNALLY AND GEORGE THOEMING
Lake Forest College, Lake Forest, Illinois

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Helium negative ions were produced from He^+ by a two-step process in which He $(1s2s)^3S_1$ was first produced by a nearly resonant charge transfer and then He^- was produced by capture of a $2p$ electron. Both collisions were with cesium atoms. The maximum yield of He^- was 1.2% of the incident He^+ when the energy of the He^+ was 3 keV. The cross sections relevant to these processes were obtained.

I. INTRODUCTION

THREE-ELECTRON systems in excited-core states which are metastable against radiation and auto-ionization have been discussed theoretically by a number of authors.[1-3] The first of these systems to attract general interest was He^-. Following its experimental discovery by Hiby[4] in a Thompson-type mass spectrometer and later by Dukel'skii *et al.*[5] in a charge transfer experiment, Holøien and Mitdal[2] calculated that the $(1s2s2p)$ $^4P_{5/2}$ state of He^- should be metastable against auto-ionization. A recent calculation by Holøien and Geltman[3] indicates that the electron affinity is >0.033 eV. Pietenpol[6] estimated the lifetime of this state to be 1.7×10^{-3} sec which is a generous length of time for it to be observed and studied in beam experiments. A similar system, Li $(1s2s2p)$ $^4P_{5/2}$, has been experimentally studied by Feldman and Novick,[7] but the only experiments on the atomic properties of He^- have been those of Sweetman[8] who showed that the lifetime is $>10^{-5}$ sec and of Riviere and Sweetman[9] who studied dissociation of He^- by an electric field. Using the results of Riviere and Sweetman, Demkov, and Drukarev[10] estimated the electron affinity to be 0.08 eV.

Helium negative ions for the experiments mentioned above and for use with tandem electrostatic accelerators have been produced by collisions between He^+ and various gases[5,11,12] including He, Ne, Ar, Kr, O_2, N_2, and H_2. The cross sections for these collisions[5,11] are of the order of 10^{-19} or 10^{-20} cm² and the maximum fraction of He^+ that can be converted to He^- is 2.4×10^{-4} for He^+–H_2 collisions[12] and less for other collisions. The He^+ energy at which this maximum yield occurs is 140 keV for H_2 and is within a factor of 2 of this energy for the other gases except Ar.

The high energy of the He^- obtained from the collisions mentioned above is a serious disadvantage in atomic physics studies and the low yield makes it difficult to get adequate intensities for either nuclear physics or atomic physics experiments. This paper reports a new process for producing helium negative ions from helium positive ions for which the maximum yield is about two orders of magnitude higher than that for processes reported in the past and which will produce He^- in abundance at much lower energy.

II. A TWO-STEP PROCESS FOR PRODUCING He^-

Since He^- is probably in the metastable $(1s2s2p)$ $^4P_{5/2}$ state,[1,2] it seems reasonable to try to produce He^- by the following two-step process[13]: (1) Make $He(1s2s)$ $^3S_1$ from He^+ by a nearly resonant charge transfer with an alkali atom, and (2) make He^- from $He^-(1s2s)$ $^3S_1$ by pickup of a $2p$ electron.

This is an attractive possibility if we carefully choose both collisions because the reaction in the first step should have a large cross section in the keV energy range. Lorents and Peterson[14] found the total-charge-transfer cross section for He^+–Cs collisions to be 1.66×10^{-14} cm² at a He^+ energy of 2 keV. They point out that this very large cross section is probably due to the nearly resonant charge transfer to excited states of He. All triplet states of He produced in these collisions decay quickly to the $(1s2s)$ $^3S_1$ state, so if we assume, in the absence of more detailed knowledge, that the

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[1] Ta-You Wu, Phil. Mag. **22**, 837 (1936); Ta-You Wu and S. T. Shen, Chinese J. Phys. **5**, 150 (1944).

[2] E. Holøien and J. Mitdal, Proc. Phys. Soc. (London) **A68**, 815 (1955).

[3] E. Holøien and S. Geltman, Phys. Rev. **153**, 81 (1967).

[4] J. W. Hiby, Ann. Physik **34**, 473 (1939).

[5] V. M. Dukel'skii, V. V. Asfrosimov, and N. V. Fedorenko, Zh. Eksperim. i Teor. Fiz. **30**, 792 (1956) [English transl.: Soviet Phys.—JETP **3**, 764 (1956)].

[6] J. Pietenpol, Phys. Rev. Letters **7**, 64 (1961).

[7] P. Feldman and R. Novick, Phys. Rev. Letters **11**, 278 (1963).

[8] D. R. Sweetman, Proc. Phys. Soc. (London) **76**, 998 (1960).

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

[10] Y. N. Demkov and G. F. Drukarev, Zh. Eksperim. i Teor. Fiz. **47**, 918 (1964) [English transl.: Soviet Phys.—JETP **20**, 614 (1965)].

[11] P. M. Windham, P. J. Joseph, and J. A. Weinman, Phys. Rev. **109**, 1193 (1958).

[12] T. Jorgensen, Jr., C. E. Kuyatt, W. W. Lang, D. C. Lorents, and C. A. Sautter, Phys. Rev. **140**, A1481 (1965).

[13] V. M. Dukel'skii *et al.* (Ref. 5) have shown that when He^+ collides with Ne, Ar, and Kr, the He^+ picks up two electrons in a single collision to produce He^-. On the other hand, Jorgensen *et al.* (Ref. 12) suggest that metastable helium atoms play a role in producing He^- when He^+ collides with H_2, N_2, O_2, and Ar. Further, Holøien and Mitdal (Ref. 2) surmise that He^- observed by Hiby (Ref. 4) was formed from a $He(1s2s)^3S_1$ intermediate state.

[14] D. C. Lorents and J. R. Peterson, in *Abstracts of Papers of the 4th International Conference on the Physics of Atomic and Electronic Collisions* (Science Bookcrafters, Inc., Hastings-on-Hudson, New York, 1965), p. 328. See also H. G. Dehmelt and F. G. Major, Phys. Rev. Letters **8**, 213 (1962); P. Mahadevan, C. E. Carlston, and G. D. Magnuson, Bull. Am. Phys. Soc. **10**, 692 (1965).

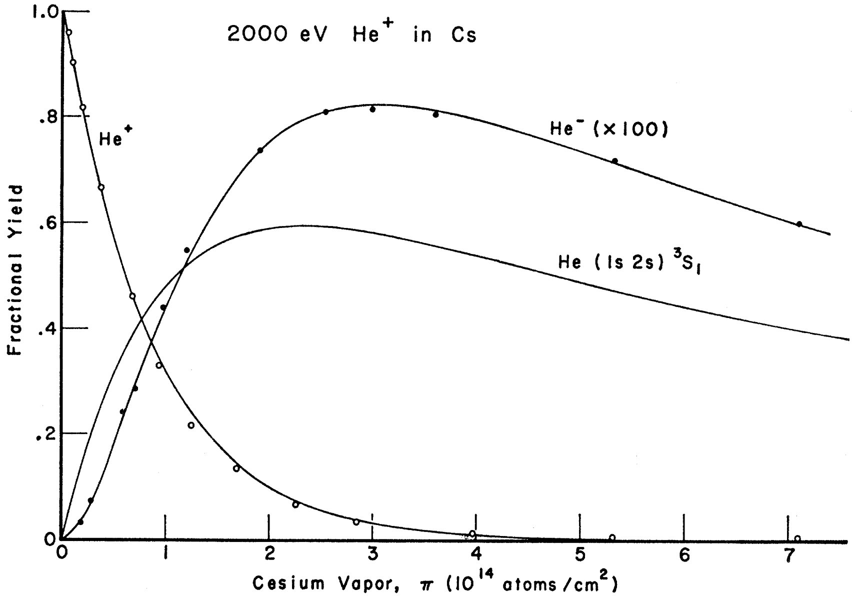

FIG. 1. The fraction of He$^+$, He$(1s2s)^3S_1$, and He$^-$ emerging from the exchange cell as a function of cesium vapor thickness. The circles represent measured values. The solid lines are theoretical curves calculated as described in the text.

excited He atoms are statistically distributed between singlet and triplet states, then $\frac{3}{4}$ of the collisions will produce triplet states, and the cross section for production of He$(1s2s)$ $^3S_1$ or states that decay to He$(1s2s)$ $^3S_1$ will be about 1.2×10^{-14} cm^2. Singlet states of He resulting from He$^+$–Cs collisions probably do not participate to an important extent in the second step of this process for producing He$^-$ because the metastable He$^-$ is a quartet state, and the spin-orbit interaction would have to produce a spin flip in the helium singlet states at the same time that a $2p$ electron is captured. The collision time is too short to allow this in a significant fraction of the collisions. Spin exchange with the core electrons of Cs is unlikely since this would require that the Cs$^+$ be left in an excited state, making the energy defect for the collision about 17.2 eV. Of course, auto-ionizing, short-lived, doublet states of He$^-$ may be produced from singlet states but they will not be observed in the experiment to be described here.

For the second step in the process to produce He$^-$, we need a reaction of the type He$(1s2s)$ $^3S_1+X\rightarrow$ He$^-+X^+$. If we choose the atom X such that for large internuclear separations the potential-energy curve for He$^-+X^+$ lies a few electron volts above the potential curve for He$(1s2s)$ $^3S_1+X$, the Coulomb interaction will tend to bring these two curves together as the internuclear separation decreases. If He$^-+X^+$ lies so far above He$(1s2s)$ $^3S_1+X$ that the molecular potential-energy curves are not close together at internuclear separations of a few angstroms, then the Massey criterion[15] suggests that the cross section for the reaction He$(1s2s)$ $^3S_1+X\rightarrow$He$^-+X^+$ will be small in the energy range studied. This argument implies that X should have a low ionization energy, so one of the alkali atoms is a reasonable choice. This is fortunate, since the He$(1s2s)$ $^3S_1$ is produced in the first step by a He$^+$–alkali-atom collision, and therefore both collisions can take place in a single gas cell containing alkali atoms.

III. APPARATUS

To experimentally test the notions outlined above, a He$^+$ beam was sent through a cell containing alkali metal in equilibrium with its vapor, the He$^-$ formed by collisions in this cell was separated from the neutral and positive component of the beam by an electrostatic analyzer, and then the He$^-$ was collected in a Faraday cup connected to an electrometer.[16] The alkali atom density was varied by changing the temperature of the cell containing the alkali metal.

IV. RESULTS AND DISCUSSION

The fraction of the incident 2-keV He$^+$ ions converted to He$^-$ in a cesium-vapor cell as a function of the gas thickness π expressed in atoms per square centimeter is shown in Fig. 1.

To describe the processes quantitatively we write:

$$dF^+/d\pi=-\sigma_{+0}F^+, \tag{1}$$

$$dF^t/d\pi=\sigma_{+t}F^+-(\sigma_{t0}+\sigma_{t-})F^t, \tag{2}$$

$$dF^-/d\pi=\sigma_{t-}F^t-\sigma_{-0}F^-. \tag{3}$$

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[15] H. S. W. Massey, Rept. Progr. Phys. 12, 248 (1949).

[16] The apparatus used was similar to that described by Donnally and Sawyer, Phys. Rev. Letters 15, 439 (1965), except that the gas cell was not used.

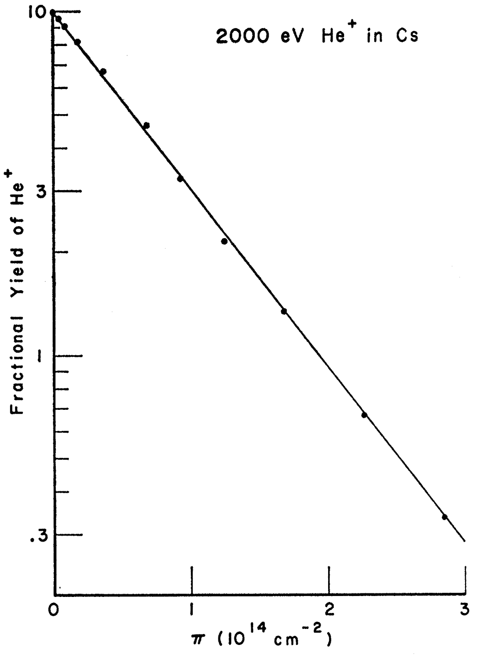

FIG. 2. The fraction of He$^+$ emerging from the exchange cell as a function of cesium vapor thickness.

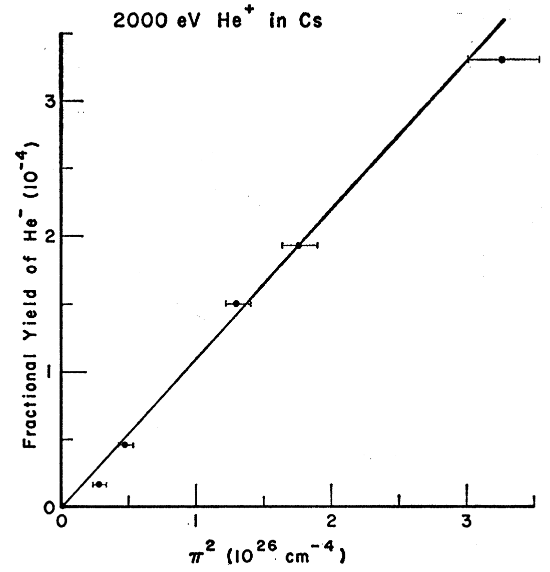

FIG. 3. The fraction of He$^-$ emerging from the exchange cell as a function of cesium vapor thickness for small values of cesium vapor thickness. Since we are trying to show the functional form in this graph, the error bars shown here include only the error in π due to uncertainty in the cesium cell temperature and not uncertainties that might change the slope but not the functional form. Errors in F^- are small in this data.

In these equations F^+, F^t, and F^- are the fractions of the total beam emerging from the cesium cell as He$^+$, He$(1s2s)\ ^3S_1$, and He$^-$, respectively,[17] σ_{+0} is the total-charge-transfer cross section for He$^+$ in cesium, σ_{+t} is the cross section for the process[18] $\mathrm{He}^+ + \mathrm{Cs} \rightarrow \mathrm{He}(1s2s)\ ^3S_1 + \mathrm{Cs}^+$, σ_{t-} is the cross section for the process $\mathrm{He}(1s2s)\ ^3S_1 + \mathrm{Cs} \rightarrow \mathrm{He}^- + \mathrm{Cs}^+$, σ_{t0} is the cross section for destruction of He$(1s2s)\ ^3S_1$ in Cs by processes other than $\mathrm{He}(1s2s)\ ^3S_1 + \mathrm{Cs} \rightarrow \mathrm{He}^- + \mathrm{Cs}^+$, and σ_{-0} is the stripping cross section for He$^-$ in Cs. In writing these equations we assumed that the stripping cross section for helium atoms, both ground state and excited, in Cs is negligible. This approximation is justified by the data on F$^+$ as a function of π plotted in Fig. 2. It is clear that $\ln F^+$ is a linear function of π as is implied by Eq. (1). If stripping of neutral helium were not negligible, the experimental values of $\ln F^+$ would lie above the straight line for large values of π. All processes involving loss or capture of two electrons are also ignored. Further, we ignored in these equations triplet helium atoms produced by stripping of He$^-$. This last assumption is questionable and is discussed later in this paper. The processes contributing to σ_{t0} are: (a) spin exchange in which triplet atoms are converted to singlet atoms; (b) Penning ionization of Cs by He$(1s2s)\ ^3S_1$; (c) formation of those double He$^-$ states which promptly autoionize leaving singlet helium atoms. The solutions of Eqs. (1), (2), and (3) are:

$$F^+ = \exp(-\sigma_{+0}\pi), \tag{4}$$

$$F^t = \frac{\sigma_{+t}}{\sigma_{t0}+\sigma_{t-}-\sigma_{+0}}\left[\exp(-\sigma_{+0}\pi) - \exp(-\sigma_{t0}\pi - \sigma_{t-}\pi)\right], \tag{5}$$

$$F^- = \frac{\sigma_{+t}\sigma_{t-}}{\sigma_{t0}+\sigma_{t-}-\sigma_{+0}}\left[\frac{\exp(-\sigma_{+0}\pi) - \exp(-\sigma_{-0}\pi)}{\sigma_{-0}-\sigma_{+0}} - \frac{\exp(-\sigma_{t0}\pi - \sigma_{t-}\pi) - \exp(-\sigma_{-0}\pi)}{\sigma_{-0}-\sigma_{t0}-\sigma_{t-}}\right]. \tag{6}$$

For small values of π, Eq. (6) reduces to $F^- = \sigma_{+t}\sigma_{t-}\pi^2/2$. Fig. 3 shows the experimental values of F^- for very small values of π plotted against π^2. Since this is a straight line within experimental error, it shows that He$^-$ is formed in a two-step process, and from the slope of the straight line, we get $\sigma_{+t}\sigma_{t-} = 2.43\times10^{-30}$ cm^4. Using the slope of the line in Fig. 2 along with Eq. (4) we get $\sigma_{+0} = 1.14\times10^{-14}$ cm^2. This value of σ_{+0} differs from the value 1.66×10^{-14} cm^2 measured by Lorents and Peterson[14] but we are not able to explain the discrepancy. Since $\sigma_{+0} \geq \sigma_{+t}$ and $\sigma_{+t}\sigma_{t-} = 2.43\times10^{-30}$ cm^4, we can say that $\sigma_{t-} \geq 2.13\times10^{-16}$ cm^2.

It is possible to determine σ_{-0} and $(\sigma_{t0}+\sigma_{t-})$ by fitting the theoretical curve obtained from Eq. (6) to the measured F^--versus-π curve. The solid lines in Fig. 1 are theoretical curves with $\sigma_{-0} = 2.0\times10^{-14}$ cm^2 and

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[17] When we write He$^-$ we mean the He$^-(1s2s2p)\,^4P_{5/2}$ state and not the states of He$^-$ which immediately auto-ionize.

[18] Actually this should be interpreted as the cross section for production of He$(1s2s)\,^3S_1$ and any state decaying to He$(1s2s)\,^3S_1$.

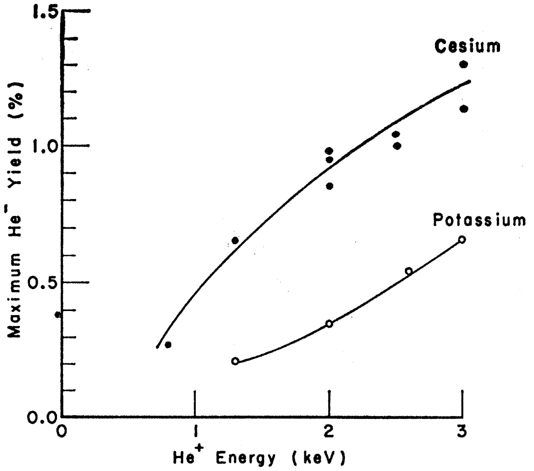

FIG. 4. The maximum fraction of incident He^+ converted to He^- in the cesium exchange cell as a function of energy of the incident He^+. The maximum yield from a potassium cell is also shown.

$(\sigma_{t0}+\sigma_{t-})=1.0\times10^{-15}$ cm². It is possible to get unambiguous values for these cross sections because the maximum yield of He^- is a very sensitive function of the cross section for destruction of He^-, σ_{-0}, while the value of π at which this maximum occurs and the slope of the F^- versus π curve are determined predominately by $(\sigma_{t0}+\sigma_{t-})$ once σ_{-0} is approximately fixed. In calculating the theoretical F^t-versus-π curve shown in Fig. 1, we *assumed*, for reasons discussed earlier, that $\sigma_{+t}=\frac{3}{4}\sigma_{+0}$. If this assumption is incorrect, the magnitude of F^t is in error but the shape of the curve is correct. If we insert a term $+\sigma_{-0}F^-$ on the right side of Eq. (2), implying that all He^- ions that are destroyed produce triplet He atoms, the value of $(\sigma_{t0}+\sigma_{t-})$ needed to make the theoretical curve fit the experimental curve is 1.25×10^{-15}. The values of $\sigma_{+t}\sigma_{t-}$, σ_{-0} and σ_{+0} are not affected by this change. The large value of σ_{-0} is probably due to the nearly resonant process $He^-+Cs\rightarrow He(1s2s)\ {}^3S_1+Cs^-$ and therefore we probably are more correct in including the term $\sigma_{-0}F^-$ in Eq. (2) than in eliminating it. On the other hand we cannot rule out a process like $He^-+Cs\rightarrow He(1s^2)+Cs+e$. The experimental error in the data of Figs. 1–3 arise mainly from uncertainties in (a) the temperature of the cesium cell, (b) the effective length of the cesium cell, and (c) the incident He^+ beam intensity which could not be monitored during a complete run. Considering these uncertainties, we estimate that the accuracy of σ_{+0} is about ±15%, the accuracy of the product $\sigma_{+t}\sigma_{t-}$ is about ±20%, and the accuracy of σ_{-0} is about ±20%. Because of the uncertainty of whether or not to include the term $+\sigma_{-0}F^-$ on the right side of Eq. (2), $(\sigma_{t0}+\sigma_{t-})$ is estimated to be accurate to ±50%.

Figure 4 shows the maximum yield of He^- as a function of He^+ energy. The value of π at which this maximum occurs is roughly independent of He^+ energy and is about 3×10^{14} atoms/cm². It should be noted that the maximum yield of He^- at 3 keV is about two orders of magnitude higher than the yields reported previously even at much higher He^+ energy.

By moving the cesium oven away from the electrostatic analyzer and using suitable beam-defining apertures, we have found that the emittance of the He^+ beam is changed by less than 0.1 cm rad $eV^{1/2}$ by the two-step electron-capture process.

If the He^+ picked up one or both electrons from alkali atoms having electronic polarization (produced, say, by optical pumping or by selection by deflection in a strong inhomogenous magnetic field), the resulting He^- would be polarized. If He^{3+} picked up the electrons, the electronic angular momentum would couple to the nuclear angular momentum to produce nuclear polarization of the helium-negative ion. This scheme, however, does not seem as attractive for polarizing He^- as the scheme proposed by Feldman and Novick[19] involving the different metastabilities of the substates of He^-. The slow He^- ions made possible by the processes reported here make the inviting Feldman and Novick proposal even more attractive.

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The authors wish to express their appreciation to Dr. J. L. McKibben for a conversation which directed their attention to the problem of producing helium negative ions, and to Roger Becker for help in taking data and making some calculations.

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[19] P. Feldman and R. Novick, in *Proceedings of the Congrès International de Physique Nucléaire, II,* edited by P. Gunginberger (Centre National de Rechierche Scientifique, Paris, 1964), p. 285.