Cross Sections for Double Electron Capture by 2–50-keV Protons Incident upon Hydrogen and the Inert Gases

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The cross section for capture of two electrons by protons in single collisions with hydrogen and inert gas targets has been measured over the energy range 2 to 50 keV. Cross-section values are given relative to the well-known single-electron-capture cross section by protons in each target gas. In hydrogen, a peak value of 1.0×10^{-17} cm^2/molecule at 17 keV is obtained for the double-capture cross section $\sigma_{1,-1}$. The double-peaked cross section versus velocity function, which is probably attributable to $10/-12$ and $10/-13e$ processes, is confirmed. In He, the cross section $\sigma_{1,-1}$ reaches a maximum of 7.10^{-19} cm^2/atom at 30 keV and steadily decreases to 2.10^{-20} cm^2/atom at 2 keV. In Ne, the present values show a peak cross section of 1.2×10^{-18} cm^2/atom being reached at 14 keV. The present values when extrapolated above 50 keV agree well with Mittleman's corrected Born approximation calculations for H_2.

INTRODUCTION

THE double electron capture by protons in single collisions with hydrogen molecules and helium atoms is uncomplicated by the presence of excited states and may be represented symbolically as

$$H_1^+ + H_2 \rightarrow H_1^- + 2H_1^+ + E, \tag{1}$$

$$H_1^+ + He \rightarrow H_1^- + He^{++} + E. \tag{2}$$

The two electrons of the target gas are in their ground states prior to collision; after collision they are both captured by the incident proton to form the negative hydrogen ion H_1^- which is unlikely to have any bound excited states.[1] There are no free electrons produced in the collision. For these reasons the collision is attractive for both theoretical and experimental study.

The cross sections $\sigma_{1,-1}$ for these collisions have been measured over the energy range 2–50 keV by the method of passing a monoenergetic beam of protons through a differentially pumped target gas cell and observing the emergent fast H_1^- ions as a function of the target gas number density. Previous measurements by Fogel[2–5] and McClure[6] in hydrogen were made by this method.

The only measurement of $\sigma_{1,-1}$ in helium is that of Fogel.[5]

In the other inert gases Fogel[5] has shown that the cross section has two maxima in the energy range 2–50 keV, the lower energy maximum being attributed to a $10/-12$ collision and the higher energy maximum to a $10/-13e$ collision. (A charge—exchange collision of the type $A^{a+}+B^{b+}\rightarrow A^{c+}+B^{d+}$ may be simply referred to as an ab/cd collision.)

Since it seemed improbable that a $10/-13e$ collision would have a larger cross section than a $10/-12$ collision, the cross section $\sigma_{1,-1}$ has been remeasured in the other inert gases.

EXPERIMENTAL METHOD

The apparatus was identical to that described previously.[7] Momentum-analyzed proton beam currents of about 1 μA were focused through a 1.0-mm beam defining aperture into the collision cell. The large exit aperture of the collision cell made possible the collection of all fast secondary collision products which may be produced up to 3.2° from the primary beam. The negatively and positively charged hydrogen ions were collected in Faraday cups, while the uncharged particles were detected by a secondary electron-emitting-type detector, similar to that described by Gardon.[8]

The method of measurement, which has already been discussed fully,[7] determines the relative cross sections from the slope of the linear portion of a graph of the number of fast collision products versus collision cell gas number density. The single electron capture cross section, σ_{10} and the double electron capture cross section, $\sigma_{1,-1}$, were measured simultaneously at various energies in the range 2–50 keV. These relative cross section values were standardized in the manner of Fite[9] and McClure[6] against the well known single electron capture cross section, σ_{10}, at 10 keV in each of the target gases. This standardization procedure avoided both the difficulties in determining the pressure profile in the collision cell and the inaccuracy, which may be

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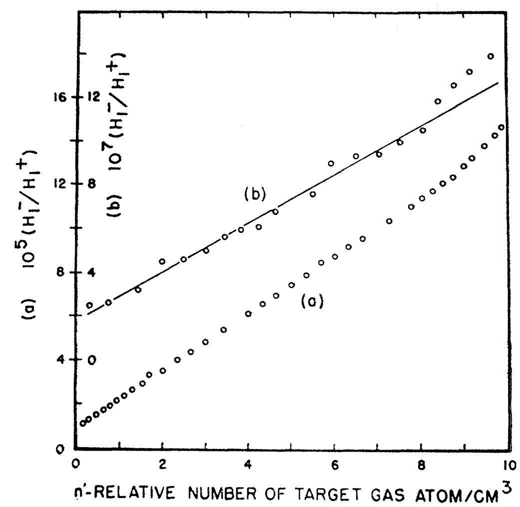

FIG. 1. The growth with relative target gas number density, n', of the ratio of the current of collision product ions H_1^- to the current of primary beam ions, H_1^+ for (a) 10-keV H_1^+ incident upon a molecular hydrogen target gas and (b) 2-keV H_1^+ incident upon helium target gas.

as large as 25%, in an absolute pressure measurement by a liquid-nitrogen-trapped McLeod gauge.[10]

Figure 1 shows two examples of the growth of collision product ions H_1^- with target gas number density. The slopes of the linear sections are used to determine the cross sections. Both graphs show well-defined linear regions.

RESULTS

Figure 2 shows $\sigma_{1,-1}$ over the energy range 2–50 keV in hydrogen. Above 10 keV there is good agreement between the present data and those of McClure,[6] which are considerably larger than those of Fogel.[5] Below 10 keV there is a further disagreement, which is much greater than the experimental inaccuracy, between the present values and those of Fogel and McClure. Fogel's earlier results[2,3,4] are quite different from the later values.

Recent Born approximation calculations by Mittleman[11] show little agreement with the experimental values in the energy region below 50 keV where the Born approximation is expected to be poor. However, an extrapolation of the experimental values to higher energies shows better agreement with the Born approximation when it is modified to correct for the nonorthogonality of initial- and final-state wave functions rather than with the unmodified approximation.

For the present apparatus it has been shown[7] that the diameters of the entry and exit canals are sufficiently large that $\sigma_{1,-1}$ is insensitive to any increase in their values. While most of the earlier values[2–5] have been obtained with the use of cylindrical canals to cause

TABLE I. Composition of charged and neutral ion beams produced by a 10 keV proton beam in hydrogen at typical gas pressures.

Voltage between deflection plates (V)	Target gas pressure (10^{-5} mmHg)	Detected current H_1^+ (A)	H_1^0 (10^{-10} A)	H_1^- (10^{-13} A)
0	0.25	9.0×10^{-7}	1.9	17.0
250		2.9×10^{-14}	0.65	0.02
0	8.2	8.93×10^{-7}	61	313
250		56×10^{-14}	0.65	1.6

large pressure differentials at each end of the collision cell, Kozlov[12] has sought to eliminate any error by separating the fast secondary H_1^- ions from the fast primary protons using a retarding electric field within the target gas. His values in the energy region 0.2–5 keV show a similar energy dependence to the present values but are a factor of three larger in magnitude.

It is readily seen that very large errors may arise from the presence in the primary proton beam of small percentages of neutral atoms which may originate from neutralization of the primary beam upon collision with a beam upon collision with a beam defining aperture, the collision-cell entry canal wall or residual gas atoms along the beam path before the collision cell. From the review paper by Allison[13] it is seen that such neutral atoms are lost from the primary beam by electron capture rather than by electron loss processes, since

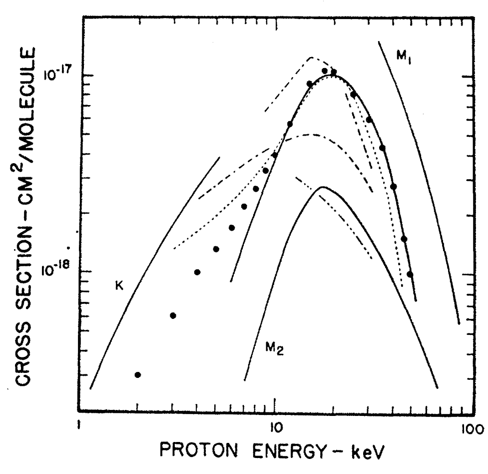

FIG. 2. The cross section σ_{1-1} as a function of primary proton energy for molecular hydrogen target gas. ••• Present experimental values; —— G. W. McClure (Ref. 6); –·····–··· Y. M. Fogel (Ref. 2); –·–·–· Y. M. Fogel (Ref. 3); – – · – – · Y. M. Fogel (Ref. 4); · · · · · · Y. M. Fogel (Ref. 5); ——K V. G. Kozlov (Ref. 12); ——M_1 M. H. Mittleman (Ref. 11), Born approximation; ——M_2 M. H. Mittleman, (Ref. 11), modified Born approximation (see text).

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TABLE II. Values of the individual terms of Eq. (3) used to determine the excitation energy, E_{ex}, of the doubly ionized target atoms of Ar, Kr, and Xe.

Target atom	E_{max} (keV)	$\|\Delta E\|$ (eV)	$-E_C$ (eV)	$-E_\infty$ (eV)	E_{ex} (eV)	V^i (doubly charged ion) (eV)
Ar	28	40.8	30.2	29.07	41.9	40.9
Kr	22	36.3	24.0	24.27	36.0	36.9
Xe	16	30.9	20.85	19.04	32.7	32.1

$\sigma_{0,-1} \gg \sigma_{01}$ particularly at low energies. Then it is readily shown that the contribution to $\sigma_{1,-1}$, for a percentage N of neutral atoms in the primary beam, is approximately $N\sigma_{0,-1}/\sigma_{1,-1}$. For example, in He at 3 keV proton energy $\sigma_{0,-1}/\sigma_{1,-1} = 1.1\times10^{-18}/0.025\times10^{-18} = 44$ while at 30 keV, $\sigma_{0,-1}/\sigma_{1,-1} = 6.3\times10^{-18}/0.65\times10^{-18} = 9.7$. Therefore, if at 3 keV N equals 1% the impurity neutral atoms contribute almost half as many, H_1^- ions as do the primary protons. This possible source of inaccuracy has been measured by the use of a pair of electrostatic deflection plates placed immediately before the collision-cell entry canal and has been allowed for at every $\sigma_{1,-1}$ cross section measurement. As shown in Table I for a determination at 10 keV the error is generally quite small.

Figure 3 shows $\sigma_{1,-1}$ for protons incident upon helium gas. The agreement with Fogel's data[5] is similar to that in hydrogen, the present values being smaller than Fogel's values at low energies.

There is no agreement between the theoretical and experimental values. The Born approximation calculations of Gerasimenko[14] for proton energies above 100 keV are clearly orders of magnitude too large and of quite a different energy dependence from that shown by the experimental data. Rosentsveig's perturbed stationary state calculation gives values[15] from 2 to 10 keV which show little similarity in either energy dependence or magnitude with the experimental data.

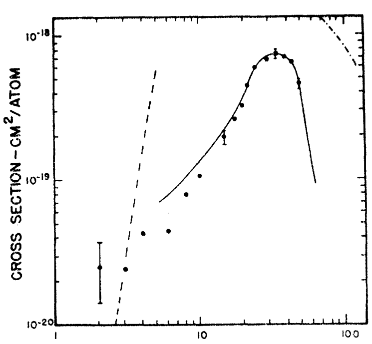

FIG. 3. The cross section σ_{1-1} as a function of primary proton energy for helium target gas. • • • Present experimental values; —— Y. M. Fogel (Ref. 5); - - - L. Rosentsveig (Ref. 15); -·-·- V. Garasimenko (Ref. 14).

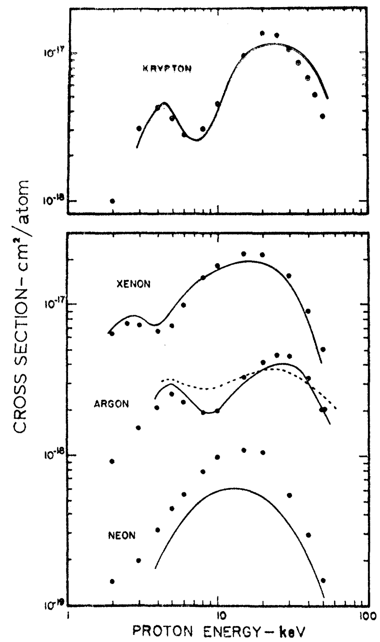

FIG. 4. The cross section σ_{1-1} as a function of primary proton energy for neon, argon, krypton, and xenon target gases. • • • Present experimental values; —— Y. M. Fogel (Ref. 5); - - - - V. V. Afrosimov (Ref. 17).

Figure 4, shows σ_{1-1} for protons incident upon Ne, Ar, Kr and Xe. It is readily shown by application of the Massey adiabatic hypothesis[16] that the lower energy peak is due to a maximum in the collision process $10/-12$, and the second peak occurs for the collision process in which the excitation energy of the target atom is its third ionization potential, i.e., a $10/-13e$ collision, within 4% (see Table II). In applying the adiabatic criterion the product, ma, of the interaction distance, a, and the number of electrons transferred in the collision, m, is assumed equal to 7 Å. Then the energy defect $|\Delta E|$ which corresponds to the experimentally observed maximum cross section value at energy E_{max} is calculated and the excitation energy E_{ex} of the doubly ionized target atom is found from the relationship.

$$\begin{aligned}|\Delta E| &= E_\infty - E_{ex} - E_C \\ &= V^i(\mathrm{H}) + S(\mathrm{H}) - V^i(\mathrm{He}) \\ &\qquad - V^i(\mathrm{He}^+) - E_{ex} - E_C, \quad (3)\end{aligned}$$

where V^i is the ionization energy of a given atom and S is the electron affinity; E_C is the Coulomb attraction energy between the collision products and is calculate in the manner of Hasted.[16]

Even though this explanation is readily made by involving only the ground states of the doubly- and triply-charged target ions, the experimental cross section is, in fact, the superposition of all the cross sections for double electron capture by protons in which the target atoms are left in their ground doubly- and triply-ionized

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states and all higher excited states. The adiabatic criterion then indicates that the probability of the target atom being left in any of its excited states is small compared with that for the ground ionized states.

Two facts indicate why a second peak is not observed in the cross section $\sigma_{1,-1}$ in Ne. First an adiabatic hypothesis argument indicates that the maximum cross section for a $10/-13e$ collision in Ne should occur at a proton energy of about 95 keV which is above the present range. Secondly, Fig. 4 shows that, as the atomic number of the target decreases, the height of the higher velocity peak relative to that of the lower velocity peak decreases while the height of the lower velocity peak itself decreases, i.e., the maximum values of the cross sections for $10/-12$ and $10/-13e$ processes have a different dependence upon atomic number. Thus in Ne it is probable that the $10/-13e$ peak is too small to be seen above the $10/-12$ peak.

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Positron Annihilation in Helium

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The modified adiabatic scattering function previously used to compute phase shifts for positron-helium scattering below the positronium threshold is applied to the problem of annihilation in helium. A large enhancement factor relative to the Dirac rate is found, varying with energy from 2.30 to 3.16. The probability of finding the spectator electron in states of the He^+ ion other than the ground state is computed and found to be small but probably observable.

I. POSITRON SCATTERING FUNCTION

We have previously discussed the adiabatic approximation[1] and applied it to the problem of low-energy positron-helium scattering.[2] Let us now consider its application to the computation of positron annihilation in helium.

The scattering wave function for a positron of momentum $\mathbf{k}$ has the form

$$\Psi_{\mathbf{k}}(\mathbf{r}_1,\mathbf{r}_2,\mathbf{x})=\chi_{\mathbf{k}}(\mathbf{x})[1+G(\mathbf{r}_1,\mathbf{x})+G(\mathbf{r}_2,\mathbf{x})]\phi(\mathbf{r}_1,\mathbf{r}_2). \quad (1)$$

Here $\mathbf{x}$ is the positron coordinate, and $\mathbf{r}_1$, $\mathbf{r}_2$ are the coordinates of the two electrons, all measured from the helium nucleus. The function ϕ describes the ground state of the helium atom. The asymptotic form of the function Ψ will be correct if $\chi_{\mathbf{k}}\rightarrow e^{i\mathbf{k}\cdot\mathbf{x}}$ since the correlation function G vanishes for large values of x, and the two electrons are assumed to be independently polarized.

Taking the positron interaction as perturbation

$$V=2\sum_{i=1,2}\left[\frac{1}{x}-\frac{1}{|\mathbf{x}-\mathbf{r}_i|}\right], \quad (2)$$

one can evaluate $G(\mathbf{r},\mathbf{x})$ correct to first order in V by the method of Dalgarno and Lewis.[3] If we make the shielding approximation employed previously[2]

$$\phi(\mathbf{r}_1,\mathbf{r}_2)=\pi^{-1}\beta^3\exp[-\beta(r_1+r_2)], \quad (3)$$

the equation for $G(\mathbf{r},\mathbf{x})$ becomes

$$\nabla^2G-2\beta\frac{dG}{dr}=2\left[\frac{1}{x}-\frac{1}{|\mathbf{x}-\mathbf{r}|}\right]-2\beta e^{-2\beta x}\left[1+\frac{1}{\beta x}\right]. \quad (4)$$

This is similar to the equation solved by Dalgarno and Lynn[4] for the case of hydrogen, and can be related to that result directly by making the change of variables $\mathbf{y}=\beta\mathbf{r}$, $\mathbf{z}=\beta\mathbf{x}$. Then one finds that $\beta G(\mathbf{y},\mathbf{z})$ satisfies the same equation as does the function derived byDalgarno and Lynn,[4] and can be taken over from their work. They derived an expression, in elliptical coordinates, which contains implicitly all terms in the Legendre polynomial expansion,

$$G(\mathbf{y},\mathbf{z})=\sum_{l=0}^{\infty}g_l(y,z)P_l(\cos\alpha), \quad [\cos\alpha=\hat{y}\cdot\hat{z}]. \quad (5)$$

Our previous experience[1] indicated that the monopole term in the expansion gives excessive short-range attraction (correlation), and in the following it will be completely suppressed. Then, in elliptical coordinates

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