# Single and Double Electron Loss Cross Section for 2-50-keV H<sub>1</sub><sup>-</sup> Ions Incident upon Hydrogen and the Inert Gases

J. F. WILLIAMS\*

Physics Department, Australian National University, Canberra, Australia (Received 6 September 1966)

Direct determinations of the cross sections for single and double electron loss in single collisions by 2-50-keV  $H_1^-$  ions incident upon hydrogen molecules and inert-gas atoms are made by the method of measuring the rate of growth of the fast-collision products, H10 and H1+, with increasing target-gas number density. Previous measurements of  $\sigma_{-1,0}$  have included contributions from  $\sigma_{-1,1}$ . It is shown that allowance for such contributions is only partially successful in improving the agreement between previously published values.

#### INTRODUCTION

HE cross section for the single-electron loss (detachment) from fast negative hydrogen ions,  $\sigma_{-1,0}$ , has been determined previously only by methods which use the assumption that the simultaneous doubleelectron loss cross section  $\sigma_{-1,1}$  is a most unlikely event. In the experiments of Hasted<sup>1,2</sup> with the inert gases, the loss cross section was determined from the current of detached electrons which gives the sum  $(\sigma_{-1,0}+2\sigma_{-1,1})$ . In the later experiments of Stier and Barnett,<sup>3</sup>  $\sigma_{-1,0}$  was determined indirectly by the attenuation of a beam of atoms in a gas of sufficient pressure either to equilibrate the charge distribution or to allow only single collisions to occur. Although these workers measured changes in the composition of the primary beam and not in the currents of slow ions or electrons formed in the target gas, their experiments also measure a sum, namely  $(\sigma_{-1,0} + \sigma_{-1,1})$ . As Stier and Barnett's values are generally lower than Hasted's values, one may readily expect that  $\sigma_{-1,1}$  is not negligible compared with  $\sigma_{-1,0}$ . Further, in argon (see Fig. 4), the differences between the two sets of data suggest that  $\sigma_{-1,1}$  increases quite quickly with increasing energy in the vicinity of 4 keV.

In the present investigation direct measurements of both  $\sigma_{-1,0}$  and  $\sigma_{-1,1}$  are made simultaneously by determining the rate of growth of the fast-collision products with increase in target-gas number density. Small projectile-energy increments of 0.5 keV are used in the vicinity of 4 keV where a large increase in  $\sigma_{-1,1}$  is anticipated.4

The double-electron loss cross section  $\sigma_{-1,1}$  has been measured previously by Tisone and Branscomb,<sup>5</sup> whose relative values agree well with the absolute values determined by Fogel<sup>6</sup> when their data are normalized at 4 keV.

- <sup>8</sup> P. M. Stier and C. F. Barnett, Phys. Rev. 103, 896 (1956). <sup>4</sup> L. M. Branscomb, Advan. Electron. Electron Phys. 9, 43,
- (1957). <sup>6</sup> G. C. Tisone and L. M. Branscomb, Joint Institute for Laboratory Astrophysics, Boulder, Colorado, Report No. 20, (1964) (unpublished).

<sup>6</sup>Y. M. Fogel, V. A. Amkudinov, and R. E. Slabospitskii, Zh. Eksperim. i Teor. Fiz. **32**, 453 (1957) [English transl.: Soviet Phys.—JETP **5**, 382 (1957)].

#### EXPERIMENTAL METHOD

The apparatus and method used for the cross-section determinations are identical to those previously discussed<sup>7,8</sup> in detail.

The negative hydrogen ions were obtained from electron-capture collisions of the protons emergent from the electrodeless discharge source in the free-drift region between the accelerating lens and the momentumanalyzing magnetic field.7 This region was isolated from the subsequent vacuum system by a differentially pumped canal (2-mm diam ×15-mm length) which permitted the gas pressure along the drift path to be raised to  $10^{-4}$ -mm Hg in order to obtain  $H_1^-$  beam currents of the order of 10<sup>-9</sup> A.

Relative cross-section values were determined from the slope of the linear portion of the graph of the growth of the fast-collision products versus collision-cell gas number density. Typical curves are shown in Fig. 1. Both  $\sigma_{-1,0}$  and  $\sigma_{-1,1}$  were measured simultaneously at various energies in the range 2-50 keV. These relative cross-section values were standardized<sup>7</sup> in the manner of Fite9 and McClure10 against the well-known singleelectron-capture cross section  $\sigma_{1,0}$  for proton energies of 10 keV in each of the target gases, namely,  $8.2 \times 10^{-16}$ cm<sup>2</sup>/molecule in H<sub>2</sub>,  $9.5 \times 10^{-17}$  cm<sup>2</sup>/atom in He, 2.9×10<sup>-16</sup> cm<sup>2</sup>/atom in Ne, 9.9×10<sup>-16</sup> cm<sup>2</sup>/atom in Ar, 1.45×10<sup>-15</sup> cm<sup>2</sup>/atom in Kr, and 1.8×10<sup>-15</sup> cm<sup>2</sup>/ atom in Xe.

#### RESULTS

Figures 2 and 3 show the results obtained for the cross sections  $\sigma_{-1,1}$  for 2–50-keV H<sub>1</sub><sup>-</sup> ions incident upon H<sub>2</sub>, He, Ne, Ar gas targets. There is fair agreement between the energy dependence of the two sets of values, but Fogel's values are mostly well outside the present experimental accuracy of  $\pm 8\%$ .

Recently Tisone and Branscomb<sup>5</sup> have made relative measurements of  $\sigma_{-1,1}$  in hydrogen over the energy range from 0.5 to 4 keV. If their values are normalized

<sup>\*</sup> Present address: Université Laval, Québec, P.Q., Canada. <sup>1</sup> J. B. Hasted, Proc. Roy. Soc. (London) **A212**, 235 (1952). <sup>2</sup> J. B. Hasted and J. B. H. Stedeford, Proc. Roy. Soc. (London) **A227**, 466 (1955).

<sup>&</sup>lt;sup>7</sup> J. F. Williams and D. N. F. Dunbar, Phys. Rev. **149**, 62 (1966). <sup>8</sup> J. F. Williams, Rev. Sci. Instr. **37**, 1205 (1966). <sup>9</sup> W. L. Fite, D. G. Hummer, R. F. Stebbings, and L. M. Branscomb, Phys. Rev. **119**, 668 (1960).

<sup>&</sup>lt;sup>10</sup> G. W. McClure, Phys. Rev. **130**, 1852 (1963).



F1G. 1. The linear growth of the fastcollision products  $H_1^o$  with increasing collision cell gas number density n' is seen for (a) 15-keV  $H_1^-$  incident upon neon and for (b) 45-keV  $H_1^-$  incident upon krypton gas.

to the present values at 4 keV then there is good agreement between all the common points over the common energy range of 2 to 4 keV.

While the two electrons detached from a  $H_1^-$  ion may not be captured by the inert-gas target atoms to form stable negative ions and so must be ejected into the continuum, the remaining target atom may be left in an excited state and so may complicate the interpretation of the measured cross section  $\sigma_{-1,1}$ .

Figure 4 shows the cross section  $\sigma_{-1,0}$  for 2–50-keV  $H_1^-$  ions incident upon the five inert gases. In all five gases earlier data<sup>1,3</sup> are about 10% higher than the present values. When either  $2\sigma_{-1,1}$  or  $\sigma_{-1,1}$ , as appropriate, is subtracted from those data, then agreement with the present values is considerably improved. The present values of  $(\sigma_{-1,0}+2\sigma_{-1,1})$  become increasingly smaller than Hasted's values<sup>2</sup> as the atomic number of the target gas increases. Figures 2 and 3 show that



FIG. 2. The double-electron loss cross section  $\sigma_{-11}$  for  $H_1^-$  ions incident upon hydrogen molecules and helium atoms is shown as a function of  $H_1^-$  energy. Open circles—present results; solid line—Fogel (Ref. 6); cross with circle—relative values of Tisone and Branscomb (Ref. 5) normalized to the present value at 4 keV.

while  $\sigma_{-1,1}$  does increase quickly in the energy region around 8 keV, the values of  $\sigma_{-1,1}$  are not large enough and do not increase quickly enough with increasing projectile energy to account satisfactorily for the shape of Hasted's results.

The Born-approximation calculations of  $\sigma_{-1,0}$  by Sida<sup>11</sup> for a helium target gas show an energy dependence similar to that shown by the experimental values. It could be expected<sup>12</sup> that the Born approximation would be accurate down to an energy of about 50 keV, when the calculated values would start to considerably



FIG. 3. The double-electron loss cross section  $\sigma_{-11}$  for  $H_1^-$  ions incident upon neon and argon atoms is shown as a function of  $H_1^-$  energy. Open circle—present results in neon; solid circle—present results in argon; solid line—Fogel (Ref. 6).

<sup>&</sup>lt;sup>11</sup> D. W. Sida, Proc. Phys. Soc. (London) **A68**, 240 (1955). <sup>12</sup> M. R. C. McDowell and G. Peach, Proc. Phys. Soc. (London) **74**, 463 (1959).

overestimate the true cross-section values. However, the experimental evidence presented in Fig. 4 shows that the predicted cross section is, at all energies, less than the experimental values. While the magnitude of the predicted cross sections depends strongly upon the



FIG. 4. The present values of the single-electron loss cross section  $\sigma_{-10}$  for H<sub>1</sub><sup>-</sup> ions incidents upon He, Ne, Ar, Kr, and Xe atoms, is shown as a function of the H<sub>1</sub><sup>-</sup> energy and is compared with the composite values of other workers. Open circle—present values of  $\sigma_{-10}$ ; dashed line—present values of  $(\sigma_{-10}+2\sigma_{-11})$ ; dotted line—Hasted  $(\sigma_{-10}+2\sigma_{-11})$  (Ref. 1); line made up of three dashes followed by three dots—Stier and Barnett  $(\sigma_{-10}+\sigma_{-11})$  (Ref. 3); dashed and dotted line—Sida (Ref. 11); solid line with error bars—Hasted and Stedeford  $(\sigma_{-10}+2\sigma_{-11})$  (Ref. 2).



FIG. 5. The present values of the single electron loss cross section  $\sigma_{-10}$  for H<sub>1</sub><sup>-</sup> ions incident upon hydrogen molecules is shown as a function of H<sub>1</sub><sup>-</sup> ion energy and compared with the composite values of other workers. Open circles—present values; dotted line—Hasted and Smith ( $\sigma_{-10}+2\sigma_{-11}$ ) [J. B. Hasted and R. A. Smith, Proc. Roy. Soc. (London) A235, 349 (1956)]; line composed of a dash followed by two dots—Whittier ( $\sigma_{-10}+\sigma_{-11}$ ) [A. C. Whittier, Can. J. Phys. 32, 275 (1954)]; line composed of dot followed by two dashes—Stier and Barnett ( $\sigma_{-10}+\sigma_{-11}$ ) (Ref. 3); solid line—first Born-approximation calculations by McDowell (Ref. 12) for atomic hydrogen; dashed line—values of the total electron-production cross section for H<sub>1</sub><sup>-</sup> ions incident upon atomic hydrogen by Hummer (Ref. 9). Note that both McDowell's and Hummer's values have been multiplied by a factor of 2 in this figure. This procedure is not meant to imply any equivalence of two hydrogen atoms to one hydrogen molecule for the cross section  $\sigma_{-10}$ .

actual wave function used, the energy dependence of the cross section is insensitive to the choice of wave function, so it appears that, for the cross section  $\sigma_{-1,0}$ , the Born approximation may be valid down to an energy as low as several keV.

Figure 5 shows (a) the present values of the cross section  $\sigma_{-1,0}$  for 2-50 keV  $H_1^-$  ions incident upon molecular-hydrogen gas, (b) the experimental values of the total electron-production cross section for  $H_1^$ ions incident upon atomic hydrogen measured by Hummer et al.,<sup>9</sup> and (c) the first Born-approximation prediction by McDowell and Peach<sup>12</sup> for an atomichydrogen target. Both sets of values in atomic hydrogen have been multiplied by two in Fig. 5 only in order that the data may be shown in one figure. It appears that a hydrogen atom may be equally as effective as a hydrogen molecule in stripping the outer electron from an  $H_1^-$  ion. The comparison of the experimental values in atomic hydrogen with the predictions by McDowell and Peach<sup>12</sup> suggests that the Born approximation may be valid for the simple electron-detachment process for energies as low as several keV in agreement with the data in helium.

The experimental data in hydrogen and the inert gases do not show any subsidiary maxima which might indicate a possible large cross section for the production of excited states in either the projectile or target atoms. McDowell's predictions indicate that the main contribution to  $\sigma_{-1,0}$  should come from single transitions

cross section.

of the  $H_{1-k}$  ion, that is, from electron detachment from  $H_1^-$  with the remaining  $H_1^0$  atom in its ground state while the target atom is left in its ground state. The predicted peak for this process is in good agreement with the maximum at 10 keV in the data of Hummer et al.<sup>5</sup> It is only at the upper end of the present energy range that one may have expected to find any appre-

PHYSICAL REVIEW

VOLUME 154, NUMBER 1

5 FEBRUARY 1967

## Statistical Treatment of Ionization in Atomic Collisions\*

M. H. MITTLEMAN<sup>†</sup>

Lawrence Radiation Laboratory, University of California, Livermore, California

AND

L. WILETS Department of Physics, University of Washington, Seattle, Washington (Received 11 May 1966)

A previously derived statistical model of atomic collisions is discussed and applied to the problem of ionization in the collision of two slow heavy atoms. The diffusion equation (in energy of excitation) resulting from the model is solved for a simplified nontrivial model. A universal curve is obtained for the cross section for single ionization. The experimental data of Lee and Gilbody are fitted remarkably well.

### I. INTRODUCTION

CLOW collisions between atoms are characterized by  $\mathbf{O}$  the formation of a quasimolecule. For sufficiently slow collisions, the system remains in the same adiabatic state in which it was formed. For finite velocities, the adiabatic levels are no longer eigenstates of the system; in the language of perturbation theory, transitions occur among the adiabatic states.

The case of simple systems, in which a pair of levels may undergo a near crossing, has been analyzed by Landau<sup>1</sup> and Zener.<sup>2</sup> Transitions are most likely to occur in the vicinity of the near crossing. If only one near crossing is present, the Landau-Zener model yields a three-parameter (two of which are scale parameters) excitation function. These parameters depend upon fine details of molecular level curves and are very difficult to obtain theoretically, although in principle they can be obtained from experimental excitation functions. If several near crossings occur, the number of parameters increases and the analysis becomes prohibitively complicated.

As the complexity of the quasimolecular system increases, the Landau-Zener model breaks down in at least two, closely related, ways: (1) A transition does not occur at a point, but over an appreciable distance.

(2) If another crossing occurs before the first is completed, the single-crossing formula fails. Not only does such an effect become more likely as the level density increases, but transitions can also be made simultaneously to other nearby levels. A different approach is then required.

ciable contribution from excited states to the measured

ACKNOWLEDGMENT

for his comments and assistance during the course of

this work and on the preparation of this paper.

The author is indebted to Professor D. N. F. Dunbar

A theory which attacks the problem from the region of greatest complexity was proposed by one of the authors.3 We will not rederive the equations here, but state the assumptions and conclusions.

## II. THE STATISTICAL MODEL

It is assumed that the level density is high and that near crossings are frequent. By high density we mean that the level spacing is comparable to or smaller than the "residual" interelectron energies, the actual separation energy which would occur for an isolated crossing, characteristically a fraction of an electron volt. Frequency of crossings is to be measured in terms of the characteristic distance during which a single-crossing transition would occur.

$$\Delta R \sim \left| \frac{2hR}{d\epsilon_{12}/dR} \right|^{1/2},\tag{1}$$

where  $d\epsilon_{12}/dR$  is the rate of approach of the pair of levels. The conditions do not appear difficult to satisfy

<sup>\*</sup>Work performed under the auspices of the U.S. Atomic Energy Commission.

<sup>&</sup>lt;sup>1</sup> Present address: Space Science Laboratory, University of California, Berkeley, California.
<sup>1</sup> L. D. Landau, Phys. Sowjetunion 2, 46 (1932).
<sup>2</sup> C. Zener, Proc. Roy. Soc. (London) A137, 696 (1932).

<sup>&</sup>lt;sup>3</sup> L. Wilets, in Proceedings of the Second International Conference on the Physics of Electronic and Atomic Collisions (W. A. Ben-jamin, Inc., New York, 1961), p. 47; Phys. Rev. 116, 372 (1959).