Charge Changing Processes in Hydrogen Beams

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Charge changing processes of hydrogen beams in gases (H_2, He, N_2, O_2, Ne, Ar, Kr, Xe), atomic hydrogen, alkali metal vapors (Li, Na, Mg, K, Cs), and gaseous carbon are reviewed primarily from an experimental point of view. Following a simple description of charge changing phenomena and typical techniques of measurement, problems associated with the experiments are discussed. Experimental cross-section results for the various charge changing processes are presented in figures with critical comments. A brief review of the theoretical developments relevant to the charge changing processes of hydrogen beams is also presented.

CONTENTS

1. INTRODUCTION

When a beam of fast ions or atoms passes through a material, not only do the projectiles excite or ionize the atoms in the material, the incident beam particles themselves undergo capture or loss of electrons. These processes are called attachment and detachment (or stripping) (Massey and Burhop, 1969). As a result, positive or negative ions or neutral atoms may emerge after passage through the material.

Recently, these charge changing phenomena have aroused increased interest in connection with the design of radiation detectors, with radiation damage, with studies of astrophysics, with controlled thermonuclear fusion, with the acceleration of multiply charged heavy ions, and with slowing down or energy loss of heavy ions in matter. The production of negative ion beams is a primary problem in the design and construction of tandem accelerators (van de Graaff, 1960). The production of negative ions also plays an important role in astrophysics, in mass spectrometry, and in gas discharges.

Moreover, the production of heavy ions with high charge states by means of charge changing processes is one of the important factors in the design or use of heavy-ion cyclotrons, linear accelerators, and tandem accelerators in order to obtain high-energy heavy ions. Presently, the multiple acceleration of heavy ions, such as uranium ions with an energy range of up to 1 GeV (Rose, 1967), has been discussed and studied with

successive (repeated) multiple electron stripping during the course of the acceleration in tandem accelerators.

This principle, suggested first by Hortig (1963, 1966) will open new possibilities in heavy-ion acceleration and depends, primarily, on the degree of the electron stripping in heavy ions Z_{eff}/Z, where Z_{eff} and Z are the effective charge and the atomic number of the heavy ion, respectively.

In addition to the interest generated in the design of accelerators, special attention has been directed to charge changing phenomena in connection with nuclear fusion studies; for example, with the neutral particle injection system (cf. Post, 1958; Sweetman, 1962; Bezbatcheko, 1964; Barnett *et al.*, 1964).

The first precise measurement on a charge changing collision was made by Henderson (1922) using alpha particles emitted from radioactive sources. Since then, much experimental work has been done on charge changing phenomena for many kinds of ions or atoms including uranium (Grodzins *et al.*, 1967).

On the theoretical side, Fowler (1924) calculated the cross section for electron capture using classical electrodynamics. After the early development of theories of the cross section for the electron capture based on quantum mechanics by Oppenheimer (1928) and Brinkman and Kramers (1930), many theoretical calculations (Bates, 1962; Bates and McCarrol, 1962) have been made. A detailed discussion of the theoretical development is presented in Sec. 7.

As shown by Nikolaev (1965) relations between the cross sections for electron capture by protons and those for heavy ions and atoms exist, so that the cross sections for electron capture by heavy ions and atoms can be estimated from those by protons. Therefore, the accumulation and survey of experimental data for hydrogen ions and atoms are thought to be basic requirements for a general study of charge changing phenomena. Moreover, as would be expected, the most comprehensive theoretical and experimental studies have been those for charge changing processes involving hydrogen. These will be described and reviewed in the present paper.

Since Bartels (1930) did the first experimental work on proton charge change, charge changing processes for protons and hydrogen atoms have been extensively studied. Massey and Burhop (1969) have reviewed the work done in this area in the 1930 decade. The Allison reviews (1958a) and (1962), following the earlier Allison and Warshaw review (1953), include works before 1958. More than ten years have passed since the publication of the second Allison review, and during that period the activity in this field has been greatly expanded. Data have been accumulated in high-energy regions up to a few MeV for various gases. The highest energy investigated up to now is 37 MeV (Acerbi *et al.*, 1967, 1969) in the measurement of electron capture by protons.

Meanwhile, charge changing processes of heavier ions and atoms, such as He, C, N, O, F, Ne, or Ar, have been investigated, mainly by groups in the United States (Northcliffe, 1963; Moak *et al.*, 1967; Rose and Gales, 1967), Great Britain (Gilbody *et al.*, 1963), and the Soviet Union (Nikolaev, 1965; Pivovar *et al.*, 1965). However, because of experimental difficulties in obtaining heavy ions with high energy, experimental data on the charge changing processes for these heavy ions are few and scattered, except for equilibrium charge compositions of certain ions. Also, theoretical calculations of electron capture and loss by heavy ions are very complex and at present represent only gross estimates (Dmitriev and Nikolaev, 1963; Nicolaev, 1967). The present situation concerning charge change studies on heavy ions and atoms have been summarized by Nikolaev (1965) and Lo and Fite (1970) and are not included here. This paper, therefore, will be limited to the experimental results on the charge changing processes of hydrogen (positive and negative) ions and atoms. *Note added in proof:* More recent results are reviewed by H. Betz (1972, Rev. Mod. Phys. **44,** 465).

Section 2 presents the basic description of charge changing phenomena, and charge change in hydrogen beams are described as a special case in Sec. 3. Section 4 describes typical experimental methods for the measurements. In Sec. 5, detection and measurement problems are discussed. Experimental data are given in the figures of Sec. 6. Section 7 describes some theoretical aspects of charge changing processes. In the last section, the prospects for future work are discussed.

2. THE BASIC RELATIONS FOR CHARGE CHANGING PROCESSES DURING PASSAGE THROUGH MATTER

During the passage of a fast beam through matter, such processes as ionization (Kieffer and Dunn, 1966; Rudge, 1968) or excitation (Moiseiwitch, 1968) and charge change may occur. In general, these charge changing processes can be represented (Nikolaev, 1965; Layton *et al.*, 1967) as:

$$A^p+B\rightarrow A^m+B^n+(m+n-p)e, \qquad (1)$$

wherein an incident fast projectile A with initial charge p collides with a target atom B, initially neutral, and undergoes the loss or capture of electrons into charge state m, while the target atom acquires charge n. Thus $(m+n-p)$ electrons are released in this process.

The cross section for the process (1) is denoted by $\sigma_{pm}{}^{0n}$, where the superscripts pertain to the initial and final charge states of the target (gas) atom, and the subscripts pertain to those of the incident fast beam. In principle, the indices p, m, and n can range from zero (or -1 if the negative ion exists) to Z, the total number of electrons in the respective atom.

The cross section $\sigma_{pm}{}^{0n}$ may include the following

terms:

1. Pure ionization without any stripping ($m=p$ and $n\geq 1$). The cross section is denoted by $\sigma_{pp}{}^{0n}$ or simply $\sigma_i{}^{0n}$.
2. Pure stripping without any ionization ($m>p$ and $n=0$). The cross section is denoted by $\sigma_{pm}{}^{00}$ or simply $\sigma_{pm}{}^{s}$.
3. Stripping with ionization ($m>p$ and $n\geq 1$). In this stripping–ionization process, the stripping of the incident fast beam and ionization of the target occur in the same collision.
4. Electron capture, or charge change ($m<p$ and $n\geq 1$). The cross section is denoted by $\sigma_{pm}{}^{0n}$, in general.

In order to completely describe the above processes, coincidence experiments (cf. Afrosimov *et al.*, 1965; Everhart and Kessel, 1965; Kessel *et al.*, 1965) are required in which the charge states of the interacting beam and atom are measured before and after the collision. However, experiments of this type are very difficult. Therefore, it is usually the case that the charge states of only the incident fast beam or of only the slow secondary ion are investigated separately.

2.1 Fast Beam Measurement

In this case, the measurements are performed independent of the charge state n of the slow target ion produced by the collision. The variation of the charge composition in the fast beam during the passage through the matter is described by the following differential equations:

$$\frac{dF_m}{d\pi}=\sum_{j=-1}^{Z}{}'(F_j\sigma_{jm}-F_m\sigma_{mj}) \qquad m=-1, 0, 1, 2, \cdots, Z, \tag{2}$$

$$\sum_{m=-1}^{Z}F_m=1. \tag{3}$$

In what follows the indices range from -1 to Z. However, if no negative ion exists, the lower limit of the indices must, of course, be replaced by zero. Here F_m is the fraction of the ions in the fast beam with charge m, and π is the number of the target gas atoms in a volume of matter of cross sectional area 1 cm^2 and length equal to the distance traversed along the beam path. Finally, σ_{ij} is the sum over all cross sections of processes in which the ion with charge i is transformed into that with the charge j, the summation being extended over all possible slow ion charge states

$$\sigma_{ij}=\sum_n \sigma_{ij}{}^{0n}. \tag{4}$$

The prime over the summation in Eq. (2) indicates that the value $j=m$ is omitted from the summation. The target thickness π is represented in the following form:

$$\pi=273L\nu Pl/(273+T)760, \tag{5}$$

where

L = Loschmit number of gas = 2.678×10^{19} molecules/cm^3
ν = number of atoms per gas molecule
P = target gas pressure in Torr
l = effective length of gas target in cm
T = temperature of target gas in degrees centigrade.

Numerically, at 20°C, for monoatomic gases ($\nu=1$), this becomes

$$\pi=3.29\times 10^{16}Pl. \tag{6}$$

In Eq. (2), the first term on the right-hand side represents the increase in the fraction of ions in charge state m through the transformation into charge state m from all other possible charge states j, while the second term represents the decrease of the ions in charge state m through transformation into all other possible charge states j from the charge state m.

If the fast beam traverses a sufficient amount of material ($\pi\rightarrow\infty$) and therefore undergoes a sufficient number of collisions, no further change in the charge state composition of the beam can occur and the equilibrium charge distribution is established in the beam.

The charge distribution in the equilibrium state is independent of the initial charge of the incident ion beam as it enters the target material; it is determined solely by the relation among the various cross sections for loss and capture of electrons. Therefore, in the equilibrium state, we have $dF_m/d\pi=0$, so that

$$\sum_{j=-1}^{Z}{}'(F_j{}^{\infty}\sigma_{jm}-F_m{}^{\infty}\sigma_{mj})=0, \qquad m=-1, 0, 1, \cdots, Z, \tag{7}$$

where $F_j{}^{\infty}$ represents the fraction of ions with the charge j in charge equilibrium.

These Eqs. (7), applied to a charge equilibrium measurement, give relations between the many cross sections for charge changes. From these equations alone, however, no direct determination of each individual cross section can be achieved, since the number of the cross sections to be determined is larger than the number of equations.

If a pure ion beam of charge m traverses a gas within an electromagnetic or electrostatic field, any beam component which has undergone charge change can be removed by the field and, therefore, the incident beam is decreased or attenuated. Assuming the gas pressure to be very low, so that transformations back into charge state m from all other possible charge states j are negligible (that is, neglecting multiple collisions), the differential equation governing such a situation is given by

$$\frac{dF_m}{d\pi}=-F_m\sum_{j=-1}^{Z}{}'\sigma_{mj}. \tag{8}$$

The solution of the above equation is represented by

$$F_m = \exp\left(-\sum_{j=-1}^{Z}{}' \sigma_{mj}\pi\right). \quad (9)$$

This describes the exponential attenuation of the incident beam through charge changing collisions with the target gas atom.

From the measurement of beam attenuation, the sum of the cross sections for charge change ($\sum_j{}'\sigma_{mj}$) into all other possible charge states is obtained, but the individual cross sections are not.

Even if beams with all possible charge states are prepared and the two types of experiments, equilibrium and beam attenuation measurements, are done, only $2(Z+1)$ or $2(Z+2)$ independent equations are obtained giving relations between possible cross sections. This must be compared with the number of the unknown cross sections $(Z+1)Z$ or $(Z+2)(Z+1)$. The latter number in each instance corresponds to species in which negative ions exist. Thus, even if these two types of experimental measurements are performed using beams with all possible incident charge states, no direct determination of each cross section can be obtained except in the case of hydrogen beams.

However, some assumptions make it possible to determine or estimate each of the charge changing cross sections. For example, it can be assumed in some cases that the cross sections for change of two or more elections in a single collision are negligible compared with that for a single electron. Lang (1963) gave such a discussion on the determination of the charge changing cross sections of oxygen ions with energies in the range of a few hundreds of keV. Using this assumption, in the equilibrium measurement the following simple equations can be obtained from the set of Eqs. (7)

$$F_j{}^{\infty}\sigma_{j,j+1} = F_{j+1}{}^{\infty}\sigma_{j+1,j}, \qquad j=-1, 0, 1, \cdots, Z-1. \quad (10)$$

Therefore, the ratio of the cross sections for charge change can be determined from the measurement of the fractions of the beam in the charge equilibrium state. On the other hand, one of the cross sections is obtained from the beam attenuation measurement; thus each cross section is determined. However, the accuracy of the values of the cross sections obtained is not generally good.

In contrast to the above, more accurate cross sections for charge change are obtained by solving the differental equations (2). Since the cross sections σ_{mj} are independent of the target gas pressure p or thickness π, the general solutions of the differential equation (2) are as follows:

$$F_m(\pi) = \left(F_m(0) + \sum_{j=-1}^{Z}{}' \sigma_{jm}\int_0^{\pi} F_j(x)\exp\left(\sum_{i=-1}^{Z}{}' \sigma_{mi}x\right) dx\right) \times \exp\left(-\sum_{i=-1}^{Z}{}' \sigma_{mi}\pi\right) \qquad m=-1, 0, 1, \cdots, Z \quad (11)$$

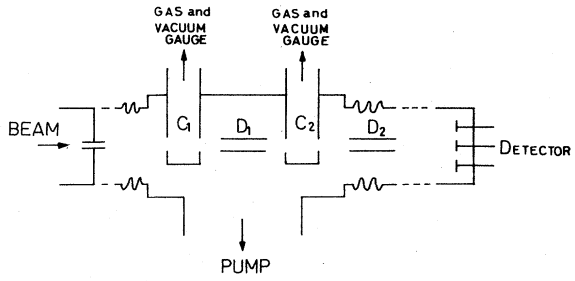

FIG. 1. An example of an experimental setup for charge change studies (Williams, 1967d). C_1 and C_2, collision chambers, are used for the production of neutral beams and for the charge changing process to be studied, respectively. D_1 and D_2, pairs of deflection electrodes, are used for sweeping out all charged beam components produced in collision chamber C_1 and for analyzing products in collision chamber C_2, respectively.

where $F_m(0)$ is the value of $F_m(\pi)$ at zero target thickness ($\pi=0$). For very small values of target thickness π, if an incident beam with charge p is introduced into the target gas, all F_j on the right-hand side are negligible except for F_p, which is approximately equal to unity. The exponential term can be expanded in a Taylor series

$$F_m(\pi) = (1-\sum_{j=-1}^{Z}{}' \sigma_{mj}\pi + \text{terms of order } \pi^2 \text{ or higher}) \times [F_m(0) + \sigma_{pm}(\pi + \text{terms of order } \pi^2 \text{ or higher})], \quad (12)$$

for $m\neq p$. For the case $m=p$, the Taylor expansion of (11) yields

$$F_p(\pi) = 1-\sum_{j=-1}^{Z}{}' \sigma_{pj}\pi + \text{terms of order } \pi^2 \text{ or higher}, \quad (13)$$

because $F_p(0)=1$. This gives the attenuation of the incident beam. A similar relation has already been obtained in Eq. (9). In the case $m\neq p$, the growth rate of the ion with charge m is given by

$$F_m(\pi) = \sigma_{pm}(\pi - \tfrac{1}{2}\sum_{j=-1}^{Z}{}' \sigma_{mj}\pi^2 + \text{terms of order } \pi^3 \text{ or higher}), \quad (14)$$

since $F_m(0)=0$.

From Eq. (13), the sum of the cross sections for all possible charge changes $\sum_j{}'\sigma_{pj}$ can be obtained (by the attenuation method). On the other hand, from Eq. (14), the cross section for the loss and capture of electrons σ_{pm} can be determined (by the growth method). This can be carried out experimentally if the incident beam is mass and charge selected by passage through a magnetic field, and if the outgoing beam is also analyzed by a suitable electromagnetic or electrostatic analyzer and the final charge fractions F_m recorded. However, neutral beams pose a special problem. These must be produced only by charge exchange in the first collision chamber which is one

of two collision chambers placed in tandem along the beam path. After passage through the first chamber, all charged beam components are removed from the beam by a deflecting field (Fig. 1). Therefore, only the neutral beam enters the second or main collision chamber where the charge changes of the neutral beam occur. Other procedures are quite the same as in the charged incident beam.

2.2 Slow Ion and Electron Measurement

Using a weak transverse electric field produced by a pair of electrodes located in the collision chamber, a slow positive ion produced in the collision process [B^{n+} in Eq. (1)] can be drawn to one plane electrode with electrons drawn to the other plane. (Slow negative ions are also drawn to this latter electrode. However, since the production of negative ions is small, these will be neglected.) This experimental technique is usually called the condenser method.

From the measurement of the slow positive ions and electrons, the total, or apparent, cross sections for positive ion production σ_+ and that for the electron production σ_- are given as follows:

$$\sigma_+ = \sum_m \sum_n n\sigma_{pm}{}^{0n}, \tag{15}$$

$$\sigma_- = \sum_m \sum_n (m+n-p)\sigma_{pm}{}^{0n}. \tag{16}$$

Therefore, the total cross section σ_t for charge changing collisions can be obtained by subtracting σ_- from σ_+ either experimentally or arithmetically. Thus, we have

$$\sigma_t = \sigma_+ - \sigma_- = \sum_m \sum_n (p-m)\sigma_{pm}{}^{0n} = \sum_m (p-m)\sigma_{pm}. \tag{17}$$

From Eq. (17), the sum of the cross sections for all possible charge changing processes can be estimated. In general, the individual cross sections cannot be obtained by this method alone. However, at very low energy, the cross sections for electron loss are usually very small, and the neutral fast beam ($m=0$) is predominant. Under such an assumption, Eq. (17) reduces to

$$\sigma_t = p\sigma_{p0}. \tag{18}$$

When the incident fast beam is singly charged ($p=1$), the cross section for single charge change σ_{10} can then be determined.

This condenser method is very attractive in the case of very low-energy incident beams, especially those below 1 keV, where the detection of the fast neutral beam produced in the collision process is very difficult. However, this method is rarely used for measurement at high energies (over a few hundreds of keV) for a proton or a neutral hydrogen beam, because at these higher energies the cross section for charge capture is itself very small and the other charge changing processes are no longer negligible by comparison.

3. THE HYDROGEN BEAM

A hydrogen beam constitutes the simplest case in charge changing collisions. It has only three components, or charge states: namely, a positive ion (proton, or H^+), a neutral atom (H^0) and a negative ion (H^-). Therefore, the differential equations (2) describing charge changing collisions become very simple (Fogel and Mitin, 1956; Fogel, 1960):

$$dF_1/d\pi = -(\sigma_{10}+\sigma_{1-1})F_1+\sigma_{01}F_0+\sigma_{-11}F_{-1}, \tag{19}$$

$$dF_0/d\pi = \sigma_{10}F_1-(\sigma_{01}+\sigma_{0-1})F_0+\sigma_{-10}F_{-1}, \tag{20}$$

$$dF_{-1}/d\pi = \sigma_{1-1}F_1+\sigma_{0-1}F_0-(\sigma_{-11}+\sigma_{-10})F_{-1}, \tag{21}$$

$$F_1+F_0+F_{-1}=1, \tag{22}$$

where F_1, F_0, and F_{-1} denote, respectively, the fractions of protons, neutral hydrogen atoms, and negative hydrogen ions in the beam. As seen in Eqs. (19)–(22), even in the simplest case of a hydrogen beam, there are six cross sections for charge change; namely, σ_{10}, σ_{1-1}; σ_{01}, σ_{0-1}; σ_{-10}, σ_{-11}.

3.1 Equilibrium Measurement

When charge equilibrium is established, we have $dF_1/d\pi = dF_0/d\pi = dF_{-1}/d\pi = 0$. Using Eqs. (19)–(22), the fractions of various charge components in the charge equilibrated beam can be calculated in terms of six cross sections by

$$F_1{}^\infty = D^{-1}[\sigma_{01}(\sigma_{-11}+\sigma_{-10})+\sigma_{0-1}\sigma_{-11}], \tag{23}$$

$$F_0{}^\infty = D^{-1}[\sigma_{10}(\sigma_{-10}+\sigma_{-11})+\sigma_{1-1}\sigma_{-10}], \tag{24}$$

$$F_{-1}{}^\infty = D^{-1}[\sigma_{0-1}(\sigma_{1-1}+\sigma_{10})+\sigma_{01}\sigma_{1-1}], \tag{25}$$

$$D = (\sigma_{-11}+\sigma_{-10})(\sigma_{10}+\sigma_{01})+\sigma_{1-1}(\sigma_{01}+\sigma_{0-1}) + (\sigma_{10}+\sigma_{-11})\sigma_{0-1}+\sigma_{1-1}\sigma_{-10}. \tag{26}$$

The ratios of various components are given by

$$\frac{F_1{}^\infty}{F_0{}^\infty} = \frac{\sigma_{01}\sigma_{-11}+\sigma_{0-1}\sigma_{-11}+\sigma_{01}\sigma_{-10}}{\sigma_{10}\sigma_{-11}+\sigma_{10}\sigma_{-10}+\sigma_{1-1}\sigma_{-10}}, \tag{27}$$

$$\frac{F_{-1}{}^\infty}{F_0{}^\infty} = \frac{\sigma_{10}\sigma_{0-1}+\sigma_{1-1}\sigma_{01}+\sigma_{1-1}\sigma_{0-1}}{\sigma_{10}\sigma_{-11}+\sigma_{10}\sigma_{-10}+\sigma_{1-1}\sigma_{-10}}, \tag{28}$$

$$\frac{F_{-1}{}^\infty}{F_1{}^\infty} = \frac{\sigma_{10}\sigma_{0-1}+\sigma_{1-1}\sigma_{01}+\sigma_{1-1}\sigma_{0-1}}{\sigma_{01}\sigma_{-11}+\sigma_{0-1}\sigma_{-11}+\sigma_{01}\sigma_{-10}}. \tag{29}$$

From these measurements, the individual cross sections cannot be determined as mentioned above. However, some simplifying assumptions are possible in special cases. For example, if the beam energy is below a few tens of kiloelectron volts, the cross section for double charge change can be neglected. Thus, σ_{1-1} and σ_{-11} are negligible, and $\sigma_{10} \gg \sigma_{01}$, so that the following

simple relations are obtained:

$$F_1^\infty/F_0^\infty=\sigma_{01}/\sigma_{10} \quad (30)$$

(low-energy beam)

$$F_{-1}^\infty/F_0^\infty=\sigma_{0-1}/\sigma_{-10}. \quad (31)$$

On the other hand, at higher energies σ_{0-1} and σ_{1-1} become negligibly small. Therefore, in this case, we have

$$F_1^\infty/F_0^\infty=\sigma_{01}/\sigma_{10} \quad (30)$$

(high-energy beam)

$$F_{-1}^\infty/F_0^\infty=(\sigma_{1-1}\sigma_{01}+\sigma_{10}\sigma_{0-1})/\sigma_{10}(\sigma_{-11}+\sigma_{-10}). \quad (32)$$

The ratios of cross sections can therefore be estimated in some cases by equilibrium measurements. If one of the cross sections can then be measured by other methods, the other cross section can be calculated from Eq. (30) or Eq. (31).

3.2 Beam Attenuation Measurement

If a pure incident beam is admitted into a collision chamber in a magnetic field of sufficient strength to deflect any charged component out of the beam before it can be neutralized by a second process, then the attenuation of the incident neutral hydrogen beam is given by

$$F_0(\pi)=\exp\left[-\pi(\sigma_{01}+\sigma_{0-1})\right], \quad (33)$$

where $F_0(0)=1$. From the attenuation measurement of the neutral hydrogen beam, the sum $(\sigma_{01}+\sigma_{0-1})$ can be obtained. Similarly, from measurements of pure proton and negative hydrogen ion beams, the sums $(\sigma_{10}+\sigma_{1-1})$ and $(\sigma_{-10}+\sigma_{-11})$ can be obtained.

In principle, each of six cross sections for the charge change of hydrogen beams can be determined from measurements of the fractions of the charge compositions by the charge equilibrium method taken together with the sums obtained from the attenuation data of the incident of positive, neutral, and negative hydrogen ion beams. However, the relations among the cross sections are rather complicated.

In some energy regions, simple relations are obtained: if the cross sections for the double charge change σ_{1-1} and σ_{-11} are negligible, σ_{10} and σ_{-10} can be obtained from the attenuation of the incident proton and negative ion beams. Therefore, from Eqs. (30) and (31), σ_{01} and σ_{0-1} are calculated.

3.3 Growth Rate Measurement

As shown above, the individual cross sections cannot be deduced from a single measurement by either of the above two methods alone; rather, a combination of the two measurements is required to determine each cross section. On the other hand, a method using Eq. (14) is feasible. This method is based on the measurement of the growth of new charge components produced in the collision chamber at very low target gas pressure. If a pure proton beam is used as incident beam, the initial conditions which obtain at $\pi=0$ are $F_{-1}=F_0=0$ and $F_1=1$. It therefore follows from Eqs. (20) and (21), that

$$\left.\begin{aligned}\sigma_{10}&=(dF_0/d\pi)_{\pi=0}, \quad (34)\\ \sigma_{1-1}&=(dF_{-1}/d\pi)_{\pi=0} \quad (35)\end{aligned}\right\}\ \text{for } H^+ \text{ incident beams.}$$

Similarly, if pure neutral beams or negative ion beams are used as incident beams, the following results are obtained:

$$\left.\begin{aligned}\sigma_{01}&=(dF_1/d\pi)_{\pi=0} \quad (36)\\ \sigma_{0-1}&=(dF_{-1}/d\pi)_{\pi=0} \quad (37)\end{aligned}\right\}\ \text{for } H^0 \text{ incident beams,}$$

and

$$\left.\begin{aligned}\sigma_{-11}&=(dF_1/d\pi)_{\pi=0} \quad (38)\\ \sigma_{-10}&=(dF_0/d\pi)_{\pi=0} \quad (39)\end{aligned}\right\}\ \text{for } H^- \text{ incident beams.}$$

Therefore, the determination of the cross sections for the various charge changing processes can be reduced to the study of the dependence of the initial growth rate of beams of different charge on the pressure of the target atom gas. Consequently, the cross section can be determined from the linear part of the growth of the new charge components observed at such low pressure that multiple collisions are negligible. However, in some cases, the above condition, often called the single collision condition, is not always satisfied. Even in such cases, however, the cross sections can be deduced from the data.

The general solutions of the differential Eqs. (19)–(21) are given by

$$F_1=a_0+a_1\exp(-\gamma_1\pi)+a_2\exp(-\gamma_2\pi), \quad (40)$$

$$F_0=b_0+b_1\exp(-\gamma_1\pi)+b_2\exp(-\gamma_2\pi), \quad (41)$$

$$F_{-1}=c_0+c_1\exp(-\gamma_1\pi)+c_2\exp(-\gamma_2\pi), \quad (42)$$

where a_0, a_1, a_2, γ_1, γ_2, etc. are constants which depend upon the cross sections σ_{10}, σ_{1-1}, etc. (Fogel and Mitin, 1956; Fogel, 1960). For sufficiently small values of $\gamma_1\pi$ and $\gamma_2\pi$, the above expressions are expanded in a Taylor series. By neglecting terms higher than π^3, while assuming the incident beam to be a pure proton beam, the growth of the negative hydrogen ion beam, for example, is given by

$$F_{-1}=\sigma_{1-1}\pi+\tfrac{1}{2}(\sigma_{10}\sigma_{0-1}+\sigma_{1-1}\sigma_{10}+\sigma_{1-1}^2-\sigma_{1-1}\sigma_{-10}-\sigma_{1-1}\sigma_{-11})\pi^2$$

$$=A\pi+B\pi^2. \quad (43)$$

By measuring the growth curve of the negative ion, therefore, the coefficient A of the term linear in π can be calculated from the least square fitting method. Then, the cross section σ_{1-1} can be determined. Similarly, from measurements of the growth curve of the neutral beam, the cross section σ_{10} can be determined. Finally, if a neutral or negative beam is used, the cross sections σ_{01} and σ_{0-1} or σ_{-10} and σ_{-11} can be obtained.

The symmetry of the differential equations (19)–(21) should be noted. If the indices 0 and 1 or 0 and -1 or 1 and -1 are interchanged, the results are identical. Therefore, the solutions of Eqs. (19)–(21) can be written in compact form. Taking $(F_i(0)=1)$, then we have

$$F_i=\sigma_{ij}\pi+\tfrac{1}{2}(\sigma_{ik}\sigma_{kj}+\sigma_{ij}\sigma_{ik}+\sigma_{ij}^2-\sigma_{ij}\sigma_{jk}-\sigma_{ij}\sigma_{ji})\pi^2$$
$$i,j,k=-1,0,1 \quad \text{and} \quad k\neq i\neq j, \tag{44}$$

where i and j indicate the charge of the incident and observed beams and k is the other one of the three charge states of hydrogen beam.

This method remains the same for ions more complicated than hydrogen. The only difference is that the resultant beam is not a three component system but a multicomponent system with all possible ionic states. The coefficient B of the quadratic term π^2 in Eq. (43) becomes a function of a larger number of the cross sections for charge change between possible charge states. The prescriptions for the more general case are given in the preceding section.

However, Eq. (43) or (44) for the determination of the cross section for charge change of any ion or atom remains valid as long as the pressure of the target gas is not too high, even though the single collision condition is not satisfied. Writing the results in terms of quantities familiar to experimentalists, the cross section for charge change in which the incident beam with charge i is transformed into that with charge j is given approximately by

$$\sigma_{ij}=1.035\times10^{-19}[(T/lP)(I_j/I_i)] \text{ in cm}^2, \tag{45}$$

where

l = effective length of the collision chamber (cm)
T = temperature of the target gas in the chamber (°K)
I_j = beam current of new charge state j produced in the collision
I_i = current of the incident beam entering the collision chamber
P = pressure of the target gas in the collision chamber (torr).

However, more exact relations are required for more accurate measurements. Nikolaev *et al.* (1961) have given the following relation, taking into consideration charge change due to the residual gases, to achieve an accuracy of 1–2 percent,

$$F_j=\delta_{ij}+g_{ij}+\tfrac{1}{2}\sum_p(g_{ip}g_{pj}+\gamma g_{ip}'g_{pj}-\gamma g_{ip}g_{pj}')$$
$$+\tfrac{1}{6}\sum_{p,q}(g_{ip}g_{pq}g_{qj}+2\gamma g_{ip'}g_{pq}g_{qj}-\gamma g_{ip}g_{pq}'g_{qj}$$
$$-\gamma g_{ip}g_{pq}g_{q'j})+(1/24)\sum_{p,q,r}g_{ip}g_{pq}g_{qr}g_{rj}$$
$$+(1/120)\sum_{p,q,r,s}g_{ip}g_{pq}g_{qr}g_{rs}g_{sj}, \tag{46}$$

where i is the charge of the incident beam, j one of the charge components of the emerging beam and

$$\delta_{ij}=\begin{cases}1 & \text{if } i=j\\ 0 & \text{if } i\neq j\end{cases},$$

$$g_{ij}=\sigma_{ij}\pi,$$
$$g_{ij}'=\sigma_{ij}'\pi',$$
$$\gamma=(\beta_1'-\beta_2')-(\beta_1-\beta_2),$$

β_1, β_2 = relative amount of gas molecules before and after the collision chamber, and β and π pertain to the admitted gas, while β' and π' pertain to the residual gases. Other expressions are the same as those used above.

3.4 Condenser Method

As mentioned in Sec. 2.2, the total cross section for charge change can be obtained from this method [see Eq. (17)]. In the case of hydrogen, Eq. (17) becomes very simple. For an incident proton beam, we have $p=1$ and $m=0$ or -1. Therefore, the total cross section $\sigma_t(H^+)$ is given by

$$\sigma_t(H^+)=\sigma_{10}+2\sigma_{1-1}. \tag{47}$$

The possible collision processes in this case are as follows:

(a) pure ionization

$$H^++B\rightarrow H^++B^{n+}+ne \qquad (n\geq1), \tag{48}$$

(b) one electron capture plus ionization

$$H^++B\rightarrow H^0+B^{n+}+(n-1)e \qquad (n\geq1), \tag{49}$$

(c) double electron capture plus ionization

$$H^++B\rightarrow H^-+B^{n+}+(n-2)e \qquad (n\geq2). \tag{50}$$

The cross sections σ_{10} and σ_{1-1} in Eq. (47) correspond to the processes (49) and (50), summed up over all possible final charge states of the target. Similarly, for pure neutral hydrogen, we have $p=0$ and $m=-1$ or 1. The total cross section is

$$\sigma_t(H^0)=\sigma_{01}-\sigma_{0-1}. \tag{51}$$

The possible collision processes are:

(a) pure ionization

$$H^0+B\rightarrow H^0+B^{n+}+ne \qquad (n\geq 1), \tag{52}$$

(b) one electron capture plus ionization

$$H^0+B\rightarrow H^-+B^{n+}+(n-1)e \qquad (n\geq 1), \tag{53}$$

(c) one electron loss plus ionization

$$H^0+B\rightarrow H^++B^{n+}+(n+1)e \qquad (n\geq 0). \tag{54}$$

The cross sections σ_{0-1} and σ_{01} in Eq. (51) correspond to the processes (53) and (54). Finally, for the pure negative ion beam, we have $p=-1$ and $m=0$ or 1. Therefore, the total cross section is

$$\sigma_t(H^-)=\sigma_{-10}+2\sigma_{-11}. \tag{55}$$

The possible collision processes are:

(a) pure ionization

$$H^-+B\rightarrow H^-+B^{n+}+ne \qquad (n\geq 1), \tag{56}$$

(b) one electron loss plus ionization

$$H^-+B\rightarrow H^0+B^{n+}+(n+1)e \qquad (n\geq 0), \tag{57}$$

(c) double electron loss plus ionization

$$H^-+B\rightarrow H^++B^{n+}+(n+2)e \qquad (n\geq 0). \tag{58}$$

The cross sections σ_{-10} and σ_{-11} in Eq. (55) correspond to the processes (57) and (58).

Therefore, from this measurement, the sum of the cross sections (47), (51), or (55) can be obtained. However, in moderate energy regions, the cross sections for double charge change are much smaller than those for single charge change. That is, $\sigma_{10}\gg\sigma_{1-1}$ and $\sigma_{10}\gg\sigma_{-11}$. Also, the cross section for the loss of electrons in the neutral hydrogen atom beam is larger than that for the capture of an electron; namely, $\sigma_{01}\gg\sigma_{0-1}$.

Thus, from the total cross section for charge change measured by the condenser method some cross sections such as σ_{10}, σ_{01} and σ_{-10} can be estimated:

$$\sigma_{10}\simeq\sigma_t(H^+), \tag{59}$$

$$\sigma_{01}\simeq\sigma_t(H^0), \tag{60}$$

$$\sigma_{-10}\simeq\sigma_t(H^-). \tag{61}$$

4. TYPICAL MEASURING METHODS

Some typical methods currently being used for the measurement of charge changing cross sections are:

4.1 Charge Equilibrium Method

When a beam passes through a sufficient thickness of the target gas or solid, equilibrium of the charge distribution in the ion beam is established. The target thickness required to achieve equilibrium is about 0.3 μg/cm^2, which corresponds to a target gas atom thickness of 10^{17}–10^{18} atoms/cm^2. The pressure of the target gas required to establish this thickness in a collision chamber of length about 10 cm is 10^{-2}–10^{-1} torr. Usually two methods are used to check the establishment of the equilibrium charge state. The first is to measure the change of final state charge distribution with variation of the target thickness and the second is to measure the change of final state charge distribution with variation of charge state of the incident beam. The fraction of the ion beam in the charge equilibrium state (F_i^∞) can be determined by electrostatic or electromagnetic analysis. From these fractions the ratios of some cross sections can be calculated as described in Sec. 3. The mean charge $\bar{\imath}$ of the ion beam in equilibrium, defined by

$$\bar{\imath}=\sum_i iF_i^\infty, \tag{62}$$

is also obtained in the charge equilibrium method. This quantity is of considerable importance in its own right.

In determining σ_{10} and σ_{01}, σ_{0-1} and σ_{-10} for hydrogen, Stier *et al.* (1956) first measured the fraction of the ion beam after passage through a target thickness of gas atoms sufficient to establish charge equilibrium, and estimated σ_{10}/σ_{01} and $\sigma_{0-1}/\sigma_{-10}$, assuming double charge change to be negligibly small. Then, by the beam attenuation method to be described in the next section, σ_{10} and σ_{-10} were determined, respectively, from measurements on a proton incident beam and a negative ion incident beam (again neglecting double charge change). Finally, this yielded σ_{01} and σ_{0-1}.

However, in this method, metastable states, as are found, for example, in a helium beam, may cause errors in determining the ratios of the charge fractions of the ion beam. Therefore, this method is rarely used for the determination of cross sections.

4.2 Beam Attenuation Method

The collision chamber, filled with target gas, is placed in a magnetic field. Since ions with different charges have different trajectories in the magnetic field, the alteration of the charge state of an ion by a charge changing collision causes an alteration in its trajectory. Therefore, ions which undergo charge change in the collision chamber cannot reach a detector which intercepts the incident beam, thereby decreasing the ion beam measured by the detector. The measurement of the beam attenuation gives the sum of the cross sections for all possible charge changing collisions from the incident beam charge state. However, in addition to the beam attenuation caused by the charge alteration, this measurement may include the attenuation of the beam by elastic scattering of the incident beam outside of the detector aperture. This type of error becomes more serious with low energy incident beams.

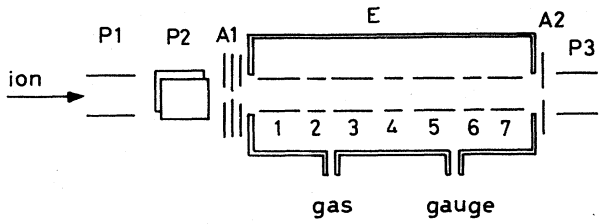

FIG. 2. Condenser method for the study of charge changing processes (Layton *et al.*, 1967). P_1 and P_2 are pairs of steering plates. A_1 and A_2 are collimation slits. P_3 are a pair of analyzing plates. E(1, 2, 6, 7) and E(3, 4, 5) are guard electrodes and current measuring electrodes, respectively.

As mentioned above, this method is not used independently for the measurement of each cross section, but is combined with other methods, such as the charge equilibrium method.

4.3 Condenser Method

The condenser method, shown schematically in Fig. 2, is often used for measurements on energy beams of less than a few keV. From such measurements, the total cross section for charge change; that is, $\sigma_{10}+2\sigma_{1-1}$ for an incident proton beam, $\sigma_{01}-\sigma_{0-1}$ for an incident neutral beam, and $\sigma_{-10}+2\sigma_{-11}$ for an incident negative ion beam is obtained.

Since double charge change is usually very small compared with single charge change, σ_{10} or σ_{-10} is obtained for incident proton or negative beams by neglecting σ_{1-1} or σ_{-11}. However, for incident neutral beam this is not always the case; σ_{01} and σ_{0-1} are not so different, but are even, in some energy regions, comparable. Therefore, special care is necessary.

Curran *et al.* (1959) used this method for the neutral incident beam. First, they measured the beam attenuation of the incident neutral beam by applying a strong electric field ($\sim$ a few kV/cm) to the condenser plates to remove all of the charged components produced in collisions with target gas atoms and determined the sum of the cross sections ($\sigma_{01}+\sigma_{0-1}$). Second, using the condenser method, the current to the condenser plates was measured, and the difference of the cross sections ($\sigma_{01}-\sigma_{0-1}$) was obtained. From these two measurements, therefore, the charge change cross sections σ_{01} and σ_{0-1} were calculated.

Similar procedures using charged incident ion beams, may be applied to determine other cross sections. However, the double charge change cross sections σ_{1-1} and σ_{-11} thus obtained may be subject to a large uncertainty, since σ_{1-1} and σ_{-11} are very small compared with σ_{10} and σ_{-10}, respectively.

Fedorenko *et al.* (1956) and Afrosimov *et al.* (1960) nevertheless used this method with some modifications (the so called wide slit mass spectrometer method) to obtain the double charge change cross sections. These authors determined the cross section σ_{1-1} for H^+ on Ar up to 180 keV. This method, however, is inferior and rarely used for double charge change, particularly in the high-energy region over a few tens of keV. On the other hand, this method is very effective for measurements on very low-energy beams. Recently, Koopman (1967, 1968) used this method to determine the cross sections for incident protons in the energy range of 400–1500 eV in atmospheric gases.

4.4 Energy Retardation Method

This method is used exclusively for the measurement of double charge changing processes of singly charged positive ions. It is especially useful for a proton beam with the relatively low energy of a few keV. Since double charge changing processes at such low energy may be accompanied by considerable scattering, the usual collision chamber system with narrow channels (to insure a good pressure drop to the outside) and the separated charge analyzer may severely limit the focusing angle of the negative ion produced in a double charge changing process and may lead to some systematic errors in the measurement of the cross section.

The principle of this method, shown schematically in Fig. 3, utilizes a retarding electric field to separate the incident positive ion beam from the secondary negative ion beam produced by a double charge changing process. The positive ion beam with energy E_0 enters a collision chamber through a small channel S. Those ions which undergo a double charge changing collision in the collision chamber are, of course, transformed into a negative ion beam. Separation of the beam components is achieved by A, B, and C, a series of gridded electrodes placed in the beam path. The first gridded electrode A is grounded. On the second gridded electrode B, a positive potential $V^b > E_0$ is applied to prevent the incident positive ion beam from reaching a Faraday cage which is placed at the end of the collision chamber. Therefore, only the negative ion beam, further accelerated by a uniform electric field between the gridded electrodes A and B and somewhat decelerated between B and C, can reach the Faraday cage. To suppress the secondary electron from the Faraday cage and grids, a magnetic field is applied. On C, a negative potential V^c is applied such that $E_0 > V^c > E_{\max}$, where $E_{\max}$ is the maximum energy of any negative ion which can be knocked out of grid A. This suppresses the effect of negative ion emission from the grid A resulting from bombardment by the incident positive or the secondary neutral beams. The distance between A and B must be quite short compared with that between the channel S and the grid

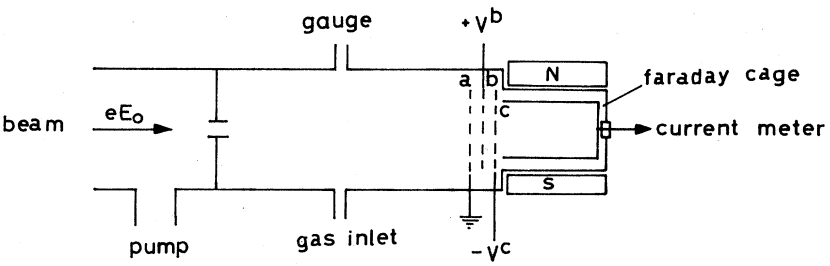

FIG. 3. Energy retardation method for charge change study (Kozlov *et al.*, 1962). See text.

A. This latter distance is to be considered as the length of the collision chamber.

This method is very attractive for measurements of double charge change in very low-energy beams which are accompanied by substantial angular scattering. It was introduced and developed by Kozlov and Roshkov (1962) for measurement of the double charge changing cross section σ_{1-1} of protons with energies of 500–5000 eV. In principle, this method may also be applied to the measurement of double charge change in a hydrogen negative ion beam (σ_{-11}), by introducing the pure negative ion beam and applying a negative potential on the grid B. The method has not yet been attempted for this case.

4.5 Growth Rate Method

Most of the cross sections for charge change have been obtained by the growth rate method. It consists basically of passing a pure incident ion or atom beam through the collision chamber and measuring the growth rate of new charge state fractions at such low pressures that the single collision regime is ensured. Then, by plotting the intensity of the beam in a given charge state against the gas pressure, the cross section for that charge changing process can be estimated from the slope of the linear portion in the I vs p curve. In the process of fitting the experimental data, great care must be taken not to include a parabolic portion of the curve [see Eq. (43)]. However, it is difficult to separate the linear and parabolic terms of the curve. In some cases, they cannot be separated, as will be mentioned later.

To avoid this difficulty, Allison (1958c) made two measurements to determine σ_{20} and σ_{21} for helium. First, the sum $(\sigma_{20}+\sigma_{21})$ was measured by the attenuation method. Then, the growth curves of neutral and singly charged helium were measured, and the ratio of the cross sections σ_{20}/σ_{21} estimated from the initial values. From this ratio and the sum, individual cross sections could be deduced without analytical fitting of the growth curve.

4.6 Least-Squares Method

In charge changing processes involving high-energy heavy ions, there are many charge states and, therefore, many different charge changing cross sections. The approximation (43) does not always hold in such cases, even for sufficiently small values of π; some charge components with small cross sections do not increase linearly with the target thickness. In such cases, a different method is necessary for the determination of the cross section. Recently, least-squares fitting methods using a digital computer have been developed (Betz, 1969; Datz *et al.*, 1970; Winter *et al.*, 1960; Betz *et al.*, 1971) by which the cross sections can be determined with good accuracy over a range of 3 to 4 orders of magnitude. The cross sections are determined so that the distribution of charge fractions computed from Eq. (2) can best reproduce the experimental data. For a given set of cross sections σ_{ij} (as parameters), Eq. (2) can be integrated numerically with initial conditions set at $\pi=0$. To obtain the best values for the cross sections, the following square sum should be minimized

$$S=\sum_i \sum_m \{F_m(\pi_i)-F_m{}^c(\pi_i)\}^2 W_m(\pi_i). \qquad (63)$$

Here $F_m(\pi_i)$ and $F_m{}^c(\pi_i)$ are the measured and calculated fractions of charge state m at a target thickness π_i, respectively. Here $W_m(\pi_i)$ is the weighting factor connected with experimental uncertainties of $F_m(\pi_i)$ and π_i, and can be given by errors of the measured fractions $\{\Delta F_m(\pi_i)\}^2$ which should include errors in the target thickness π_i. Minimization of a square sum is a well-known technique.

From this least-square analysis of all measured charge fractions, the best set of the cross sections can be obtained. This procedure can be applied to any number of measured nonequilibrium distributions. Moreover, residual gases inside and outside the target chamber will modify only the initial charge distributions $F_m(0)$.

4.7 Merging Beam Method

This is a new technique, developed independently by Trujillo *et al.* (1966) and Belyaev *et al.* (1967) and suitable for measuring the cross sections of charge change at energies of a few eV. In this method, two superposed monoenergetic beams with high laboratory energy and small energy spread move in the same direction along a common axis. Therefore, their interaction energy is quite small compared with their laboratory energy (a few tens of keV). Difficulties

often encountered in very low-energy beam study, such as repulsion due to its own space charge or strong scattering in collision processes can be avoided. However, this method is rarely used for study of high energy beam collisions.

5. ASSOCIATED TECHNIQUES

5.1 Beam Energy Measurement

Since charge change cross sections at high energy (especially those for the capture of one or two electrons) are strongly dependent on the energy of the beams, the beam energy must be accurately determined.

A. Thresholds or Resonances of Nuclear Reactions (Hund, 1953; Marion, 1961, 1966, 1968)

In the high-energy region, the energy scale over 164 keV can be calibrated with an accuracy of one part in 10^3–10^4 by means of threshold or resonance values of some nuclear reactions. [The lowest resonance energy nuclear reaction which can be used for energy calibration is the 164 keV resonance of the ^{11}B (p, γ) reaction.] To obtain these high accuracies of incident beam energy, the accelerator voltage, energy analyzing systems, and other associated systems must be highly stabilized. In most measurements of charge change, however, such highly stabilized beam energies are not required. For the present purposes, accuracy of one per cent in the knowledge of beam energy is thought to be sufficient.

B. Bleeder Current

Voltages below 164 kV can be measured by the current through high precision, high megohm bleeders or by electrostatic voltmeters. But this type of measurement sometimes causes some systematic errors in beam energy determination. For example, leakage current through the surface of the bleeders or corona current from the edges of the bleeders can make the energy of the beam appear to be larger than it really is. Usually, uncertainties associated with the measurement of the energy of the beams by this method are estmiated to be of the order of a few per cent, except in those cases in which extremely careful measurements were performed.

C. Solid State Detectors (Dearnaley and Northrop, 1966)

Recently, solid state detectors have been widely used for the determination of beam energy for energies over a few hundreds of keV. This low-energy region is limited by the noise level of the solid state detectors or preamplifiers whose energy resolution is of the order of 20 keV. Moreover, surface barrier type solid state detectors make it possible to considerably lower this low-energy limit. A linear dependence of pulse amplitude on proton energy in the range 18–250 keV was observed. More recently, this type of detector with an energy resolution of about 10 keV was developed to measure spectra of neutral particles emerging from a nuclear fusion apparatus (Bogdanov and Maksimenko, 1965), the energy resolution being limited mainly by preamplifier noise. This situation may be improved by using cooled field effect transistors for preamplifiers. Since the pulse heights from a solid state detector are proportional to the energies of the incident ions, these are analyzed by a multichannel pulse height analyzer calibrated with beams from accurately stabilized accelerators or with nuclear reaction products (e.g. alpha particles) emitted from by radioactive materials such as U, Am, or Po. The energy of the beam can thus be determined from the pulse height distributions in the pulse height analyzer, with accuracy believed to be within $\pm$ one percent. Scientillation detectors may also be used, but the accuracy of this type of detector is much lower ($\sim$10%) than that of solid state detectors.

D. Energy Ranger

At present, the operational energy of a cyclotron is estimated and changed by the so called foil or ranger, which determines the proton range in a set of thin aluminium foils (Bethe and Ashkin, 1963; Whaling, 1958; Fano, 1963). With careful measurements, the uncertainty in this method can be maintained at about $\pm$1%.

E. Energy Spread in an Ion Source

The ion beam extracted from an ion source has some inherent energy spread depending on the type of the ion source itself or of its parameters (Collins *et al.*, 1965). Usually, from the low voltage arc type of ion sources such as duoplasmatron (Von Ardenne, 1962), the energy spread is very small, generally less than a few tens of electron volts (Collins *et al.*, 1965). On the other hand, the ion beam from an rf-type ion source sometimes has a very large energy spread of up to 1 keV depending on the source parameters. The type of rf exciting circuit (Ero, 1958; Collins *et al.*, 1965; Garner and Swann, 1965; Collins *et al.*, 1966; Vorotonikov, 1966; Tawara and Sonoda, 1970) is particularly important in determining the energy spread. However, these energy spreads are usually negligibly small when compared with fluctuations of the accelerator energy.

F. Energy Loss During Collision (Fedorenko 1959)

In experiments requiring neutral or negative ion beams, these are obtained by means of a converter chamber placed just in front of the main collision chamber. The collisional energy loss of a beam as it passes through the converter must be taken into consideration in determining its energy. The results by Fogel *et al.* (1957), however, have shown the energy loss of the ion beam with an energy of, say, 5 keV is quite small. Consequently, the energy of the neutral

and negative ion beam produced in the charge changing collision can be considered to be equal to that of the incident positive ion beam.

5.2 Beam Intensity Measurement

Of primary concern in the measurement of charge changing cross sections is an accurate measurement of the intensities of the ion and atom beams.

A. Faraday Cage

The charged particles, positive or negative, can be measured with considerable accuracy by a well-designed Faraday cage with either guard electrodes or a magnetic field to suppress the secondary electrons emitted from the surface of the Faraday cage. (Pruit, 1966). In some cases, the charge collected in the Faraday cage is integrated with an integrating electrometer or alternatively with a conventional electrometer in conjunction with a recorder. The uncertainty in the intensity measurement as determined by an integrating electrometer is estimated to be usually less than 1–2%, while in the electrometer-recorder combination, the accuracy with which the average intensity can be determined may be poorer, perhaps 5–10 percent. This latter error may then be the main contribution to errors in the cross section measurements. In particular, the instabilities in intensity of the ion or atom beam cause a large uncertainty in such averaging procedures. The calibration of the electrometer itself can be made accurate to less than 1–2%.

This type of intensity measurement is, of course, unsuitable for neutral atom beams.

B. Thermal Detector

If a neutral beam has enough power, its intensity can be estimated from the measurement of the heat dissipated at the target by thermosensitive elements such as thermocouples or thermistors (Gardon, 1953; Chambers, 1964b). However, the intensity of a neutral beam produced in a charge changing process is usually very weak. Thermal detection, therefore, becomes difficult. Recently, Berkner *et al.* (1968) have developed a thermal detector using a small disk (1 in. in diameter and a few tens of mills in thickness) of barium titanate or barium titanate–lead zirconate. It can be employed for intensity measurements down to a minimum power of about a half microwatt of beam intensity. Berkner *et al.* have shown it to have good linearity, long term stability, and repeatability over a wide range (1 μW–100 mW). Independently, van de Runstraat *et al.* (1970) have measured beam powers down to about 10 nW using a glass-coated thermister, by controlling the Faraday cage temperature.

The response of the thermal detectors is independent of the charge states of the ion beams. Therefore, the calibration for the neutral beam can be made by mounting the thermal detector in a Faraday cage which is struck by the charged particle beam with a known energy (Berkner *et al.*, 1968).

C. Secondary Electron Emission-Type Detector and Charge-Equilibrated Faraday Cage

For medium-energy neutral beams, secondary electron emission-type detectors are used by many workers (Michijima, 1968). However, since the rate of secondary electron emission strongly depends on the surface condition of the electron emitters (Large and Whitlock, 1962; Chambers, 1964a; Gibbs and Commings, 1966) careful calibration is required in each measurement. The emission rate of secondary electrons is also dependent on the type of ion beam, its energy, and its charge states (Large, 1963; Daly and Powell, 1964; Morita *et al.*, 1966). This type of detector is affected by the ionization of residual gas near the detector by the secondary electrons, which often causes a spurious current signal in the detector. Thus, when the intensities of different charge components are compared, the emission rate for the beam with each charge state must be accurately known.

An alternative procedure is to charge equilibrate the beam by passage through a thin foil or gas converter before it arrives at the detectors (Jorgensen *et al.*, 1965; Gibbs and Commings, 1966; Mechbach and Nemirovsky, 1967). For example, the intensity of a neutral beam with energies over a few tens of keV can be measured by a Faraday cage with a thin foil window, such as Ni, Au or aluminized nylon or polystyrene. After passage through the foil, the beams are in charge equilibrium. Since the charge composition in the equilibrium state does not depend on the charge of the ion beam impinging on the thin foil, a single movable detector suffices for the measurement of all charge components in the beam (Mechbach and Nemirovsky, 1965).

D. Electron Multipliers

In the measurement of very low-ion beam intensity, secondary electron multipliers are sometimes used. These depend on electron emission due to ion or atom bombardment of a metal surface (conversion dynode), such as Cu–Be. The electrons are multiplied by the following dynodes, made of Cu–Be or Ag–Mg, just as in photomultipliers. Sometimes, photomultipliers with the photocathode removed and directly exposed in the chamber vacuum can be used for this purpose. The electron multipliers have a gain of 10^7–10^8. The gain of the electron multiplier depends on the ion specimen, its charges, and its energy (Barnett *et al.*, 1954). Therefore, the gain for each ion detected must be separately calibrated. Moreover, the gain may be substantially altered if the dynodes are exposed to air or humidity. Furthermore, Barnett *et al.* report that the gain is dependent on the current measured. Thus the input ion intensity must be restricted to 10^{-15} A or less.

The useful life of this type detector is generally short because of the damage to the conversion dynode surface due to bombardment by the energetic ion beams.

E. Magnetic Channel Multipliers

Recently magnetic channel multipliers (Wiley and McLarsen, 1955) with crossed electric and magnetic fields have been developed. They have a continuous dynode strip and simple and compact structure. Their characteristics are more stable than are those of electron multipliers and they have wide operating range, responding to beams of from a few tens of eV to 100 keV energies (O'Brien *et al.*, 1967).

F. Molecular Beam Detectors

In the very low-energy range, molecular beam detectors can be used for neutral beam measurement (Weiss, 1961). The neutral beams may be ionized on the surface of a filament and then detected by their reduction of the space charge limitation on a diode. The ionization of the neutral beam by electron impact is also used for its detection. However, absolute determination of the intensity of a neutral beam by these methods is very difficult.

G. Proportional Counters

McClure and Allensworth (1966) have used a specially made proportional counter with a very thin window ($\simeq$5 μg/cm^2) for the detection of a hydrogen beam at energies of a few keV. Because of the difficulty of manufacturing thin windows, this type of detector is rarely used.

H. Secondary Electron Scintillation Detectors

Instead of direct measurement of the slow neutral beam, the electrons emitted from the target, biased at a negative potential (10–20 keV), are accelerated into a scintillator and detected by a photomultiplier (Daly, 1960; Daly *et al.*, 1965; Tisone, 1965). In this method, the radiation damage of the scintillator due to the electrons is quite small and the photoconversion of the electron in the plastic scintillator is more efficient than that of heavy particles. Since the secondary emission-type detectors become less efficient and the beam power also becomes small for a very low-energy beam, intensity measurement of a low energy neutral beam is very difficult. In an absolute determination, the number of electrons emitted per incident particle (the secondary emission coefficient) must be known, except when pulse counting is used. In this method, solid state detectors may be substituted for the scintillator and photomultiplier combination, thereby making the measuring system fairly simple.

I. Nuclear Plates (Chalkin and Fremlin, 1960)

Tracks produced in nuclear emulsion by energetic bombarding particles are observed with a microscope and counted. However, this method is rarely used in the present connection.

J. Scintillator or Solid State (Semiconductor) Detector (Birks, 1964)

Scintillation detectors (which may be plastic for very high energy, or CsI for medium energy), ZnS, or semiconductor detectors are all widely used for high-energy neutral beams as well as for charged particle detection. Because of the development of fast electronic techniques, intensities of about 10^7 particles/sec (corresponding to 1 pA) can be directly counted with good accuracy and stability.

Generally speaking, in the measurement of beam intensity, counting detectors such as scintillation detectors or semiconductor detectors are usually used for intensities of 10^{-16} A or less, while intensities greater than 10^{-14} A can be measured by a Faraday cage or secondary emission detector with aid of an electrometer.

5.3 Pressure Measurement

The major contribution to errors in the measurement of charge changing cross sections is that of the pressure determination. An absolute determination of the pressure in the collision chamber can be usually obtained through a McLeod gauge. However, the pumping effect by a liquid-nitrogen-trapped McLeod gauge must be taken into consideration (Ishii and Nakayama, 1961; Meike and Reich, 1963). The mercury vapor streaming from the McLeod to the cold trap causes the same pumping action as does oil vapor in a diffusion pump. Moreover, the McLeod gauge cannot be used for continuous measurement or monitoring of the pressure.

Therefore, ionization gauges, Philips gauges, or Alphatrons, etc., calibrated with the McLeod gauge, are used for the monitoring. However, the characteristics of these devices are dependent on the gases to be measured and, therefore, calibrations are required for each gas.

On the other hand, the characteristics of a capacitance manometer are independent of the type of the target gas, and it is also suitable for continuous monitoring of pressure over a wide range. It is reported to have excellent linearity over a wide pressure range (10^{-3}–10 torr).[1]

Recently Acerbi *et al.* (1970) have developed a unique device for precise measurement of the absolute pressure of the gas target–an electromagnetic balance method. In their device, the pressure of the collision chamber is estimated from a force exerted on a small disk fitted into a hole drilled through a wall of the collision chamber. This device can be calibrated with an accurately known weight. Thus, Acerbi claims that a precision of better than $\pm$1.5% can be obtained.

[1] MKS Instruments Inc., Baratron capacitance manometer.

Along with the absolute pressure measurement, the precise determination of the effective length of the collision chamber is both very important and very difficult. The difficulty is due to diffusion of the target gases into high vacuum regions through the entrance and exit channels of the collision chamber. For this purpose, [cf. Welsh, 1967; Welsh *et al.* (1967)] it is usually assumed that the pressure in a long channel with small diameter falls off linearly; thus the effective length of the collision chamber is approximately equal to the center-to-center distance of the channels. However, this assumption is generally not accurate.

Toburen *et al.* (1968a, b; 1969) have tried an alternative approach. A simple estimate of the difusion region has been made assuming that (1) the pressure attenuation outside the collision chamber is proportional to the inverse square of the distance from the channels (isotropic propagation condition), and that (2) the pressure at a distance equal to the radius of the aperture of the channels along the beam path is the same as that in the collision chamber (molecular flow condition).

The absolute pressure and the effective length do not affect the cross section independently, but the product of the pressure (P) and the effective length of the collision chamber (l) is the important parameter. Under the assumptions outlined above, the total effective value of this product (Pl) is given by

$$Pl=P_0l_0+P_0(r_1+r_2)+P_0r_1^2\int_{r_1}^{\infty}\frac{dr}{r^2}+P_0r_2^2\int_{r_2}^{\infty}\frac{dr}{r^2}$$

$$=P_0l_0+2P_0(r_1+r_2)=P_0[l_0+2(r_1+r_2)], \qquad (64)$$

where

P = effective pressure
l = effective length of the collision chamber
P_0 = real pressure in the collision chamber
l_0 = geometrical length of the collision chamber
r_1 = radius of the entrance aperture
r_2 = radius of the exit aperture.

According to the above calculation, the *increase* of the effective length of the collision chamber depends only upon the diameters of the apertures of the channels, and is independent of the geometrcial length of the collision chamber.

The percentage increase in the length of the collision chamber is found to be less than 0.5% for a relatively long collision chamber (l_0) = 17.5±0.06 in., $2r_1$ = 0.020 in., $2r_2$ = 0.062 in.). This is not a serious error. On the other hand, it is 2.7% for a short chamber (l_0 = 3.06± 0.06 in.).

Since the cross sections for charge change can usually be determined from the linear portion in the plot of the beam intensity versus the pressure in the growth rate method (See Sec. 4.5), the effect of uncertainty in the measurement of the absolute pressure in the collision chamber is minimized. For example, an error in the absolute pressure measurement as high as 10% may cause about 1% error in the cross sections so determined. (See Sec. 4.4.)

On the contrary, no absolute determinations are required in the relative measurement of the cross sections (Fite *et al.*, 1958, 60; McClure, 1963a, b; Williams and Dunbar, 1966). In a first approximation, the cross section can be given in the form

$$\sigma_{ij}=(I_j/I_i)(1/Nl). \qquad [\text{See Eq. (45)}] \qquad (65)$$

Assuming the molecular flow condition to be satisfied in the collision chamber, then we have $Nl=Cn'$, where n' is the relative value of the pressure in the collision chamber, and C is the calibration constant, which is fixed for a given target and a given geometry of the collision chamber. The pressure may then be measured by the usual ionization gauge, which does not require calibration for each gas. The calibration constant C is determined by assuming the cross section value for a certain charge changing process at a given energy already measured with sufficiently high accuracy in each target gas and measuring the conversion ratio (I_j/I_i) of the incident ion beam to the corresponding charge state beam. In many cases, the cross section for single electron capture (σ_{10}) is used as a standard process because there are a large number of experimental measurements of this cross section and theoretical calculations are also available. During data accumulation, the fluctuations and drifts of the pressure in the collision chamber must be kept as small as possible [for example, 0.5% as by Welsh *et al.* (1967)].

As will be mentioned later, alkali or alkali-earth metal vapors have large charge changing cross sections. Therefore, many investigations on these metal vapors are currently in progress at various laboratories. However, the determination of the target density of such vapors is quite different from that of gases. Usually, the pressure of such metal vapors is estimated from the temperature of the metal oven using the known vapor pressure at that temperature. Also, surface ionization detectors (Ramsey, 1956) are often used. However, in both cases, the determination of the absolute vapor pressure is very difficult and it is thought that an uncertainty of about ±20% may be assigned to it. A calorimetric manometer (Butusov *et al.*, 1968) is sometimes used for measuring the supersonic flow of the metal vapor. This consists of a small disk and a thermocouple to measure the temperature rise of the disk. The temperature increases linearly with the flow density of the metal vapor. From the temperature rise of the disk, the density and pressure of the metal vapor can be estimated.

5.4 Effects Caused by Beam Impurities

The ion beam incident into the collision chamber will always contain, as impurities, some ions in other charge states originating from collisions (a) with the

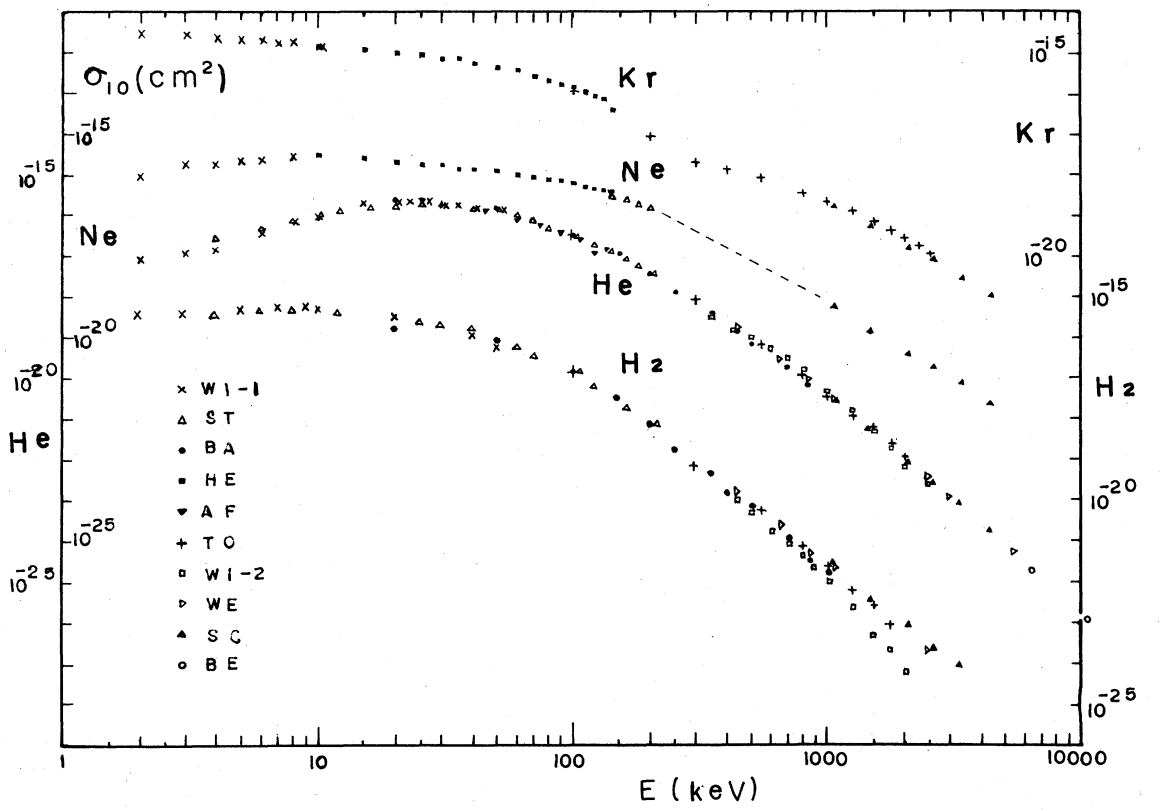

FIG. 4. Single electron capture by a proton in H_2, He, Ne, and Kr. WI-1; Williams *et al.* (1966). ST: Stier and Barrett (1956). BA: Barnett and Reynolds (1958). HE: de Heer *et al.* (1966). AF: Afrosimov *et al.* (1958). TO: Toburen *et al.* (1968a). WI-2: Williams (1967c). WE: Welsh (1967), Welsh *et al.* (1967). SC: Schryber (1967). BE: Berkner *et al.* (1965).

edges of the beam defining aperture, (b) with the walls of the entry and exit channels of the collision chamber, and (c) with the residual gas atoms along the beam path between a mass-analyzing magnet and the entrance channel of the collision chamber.

Since products from the processes (a) and (b) are almost independent of the gas pressure of the collision chamber and are constant in a given system, they do not influence the slope of the growth rate of the main collision products, from which the cross sections for the charge change are determined. Their effects can be minimized by optically aligning the knife-edge beam defining slits and choosing the diameter of the entrance channel larger than that of the incident ion beam. However, an inevitable instability of the beam position due to energy fluctuations sometimes makes their effects large. In such a case, the impurity products due to effects (a) and (b) are not always constant.

In methods such as the condenser method or the equilibrium method for the determination of charge changing cross sections, the impurities due to the processes (a) and (b) are indistinguishable from the incident beam. Therefore, cross sections obtained by these methods may contain some systematic errors due to impurity contamination of the incident beam.

The effect due to process (c) can be estimated by measuring relative amounts of ion beam components with different charge before the target gas is admitted into the collision chamber. However, the increase of the target gas pressure also increases the effective length of the collision chamber due to the diffusion of the target gas from the channels into the high vacuum regions. Such an end effect can be estimated from a measurement using two collision chambers with different length, but the same geometry of entrance and exit channels. If the same end geometry of the channels is used, the increase of the effective length of the collision chamber should be the same.

Careful measurements show that the cross sections obtained with a short collision chamber are greater than those obtained with a long collision chamber. This difference is in close agreement with the calculation given by Eq. (64) for the increase of the collision chamber length (Toburen *et al.*, 1968a).

On the other hand, in the determination of relative cross sections, the increase of the collision chamber length causes no errors. Some discussion of errors

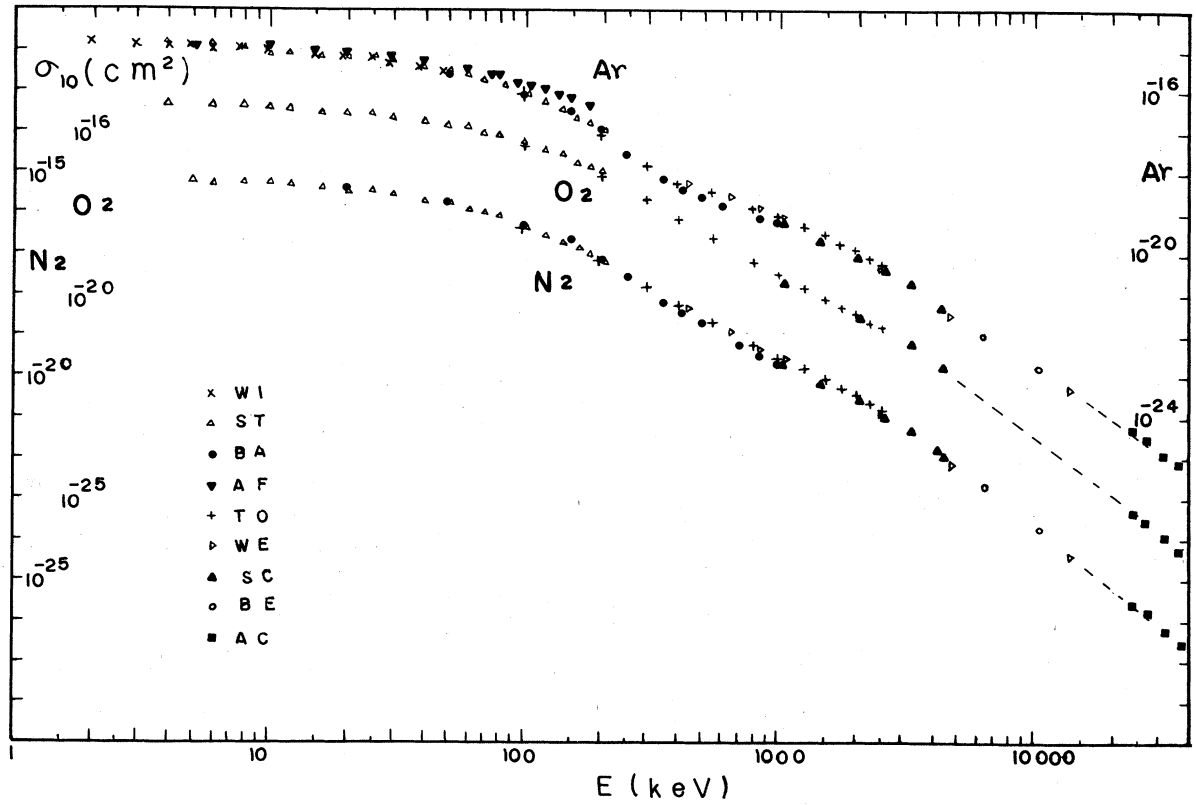

FIG. 5. Single electron capture by a proton in N_2, O_2, and Ar. WI: Williams *et al.* (1966). ST: Stier and Barnett (1956). BA: Barnett and Reynolds (1958). AF: Afrosimov *et al.* (1960). TO: Toburen *et al.* (1968a). WE: Welsh (1967), Welsh *et al.* (1967). SC: Schryber (1967). BE: Berkner *et al.* (1965). AC: Acerbi *et al.* (1967, 1969). The result of Szostak *et al.* (10^{-24} cm^2/atom at 4 MeV) seems to be too small, and is therefore omitted in this figure.

arising from beam impurities produced by the processes described above are given by Williams and Dunbar (1966) for high-energy proton charge changing experiments.

5.5 Effect of Metastable Beams

An effect due to metastable states in the beam on charge changing cross sections was first observed by Barnett and Stier (1958). The cross sections σ_{01} of He and Ne neutral beams, which were prepared by the conversion of He^+ and Ne^+ ions in hydrogen target gas, were independent of the pressure in the conversion chamber if the pressure was high ($\simeq$10 μ). However, when the pressure was decreased, the cross section σ_{01} increased. This effect means that neutral beams so converted contain a higher percentage of metastable atoms at low pressure than at higher pressure of the conversion chamber, since the destruction probability of the metastable atoms increases with the gas pressure of the conversion chamber. At higher pressures, there is no significant fraction of metastable atoms in the neutral beam emergent from the conversion chamber. Recently this effect in He beams has been observed by many workers (Allison, 1958b; Wittkower *et al.*, 1967a, b; Gilbody *et al.*, 1968, 70; Miers and Anderson, 1970a, b; Tawara, 1971a, b). Also, this effect is observed in the production of negative helium ions from positive helium ions incident on a hydrogen target gas (Collins and Stroud, 1967). The effects of metastable ions are also observed in Li^+, B^+, O^+, F^+, Na^+, and K^+ ion beams (Fogel *et al.* 1959, 61; Fogel, 1960) and also in Ar^+, Kr^+ and Xe^+ (Lockwood, 1970).

In the case of hydrogen atoms, the atomic beam must be considered to contain some fraction of the beam in excited states, although the majority of excited states decay before entering the main collision chamber. For example, the $2p$ state will decay to the ground state with a lifetime of the order of 10^{-9} sec. The metastable $2s$ state, whose lifetime is considerably longer ($\simeq$150 millisec), can, however, be quenched by a deflection electric field greater than 30 V/cm usually placed just behind the conversion chamber to deflect away all charged particles.

Other highly excited states may have long lifetimes so that the beam in such states may reach the main collision chamber. However, since the excited state population is proportional to n^{-3} (where n is the principal quantum number), the fraction of the neutral

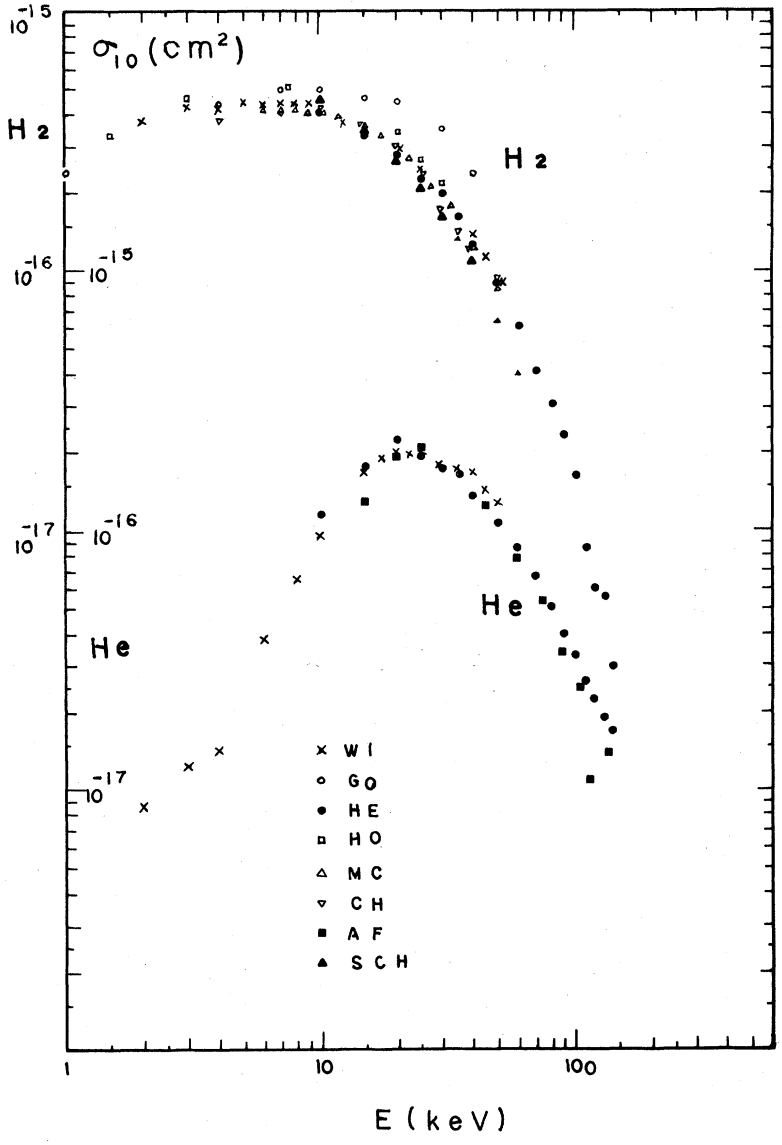

FIG. 6. Single electron capture by a proton in H_2 and He (low energy). WI Williams *et al.* (1966). GO: Gordeev *et al.* (1964). HE: de Heer *et al.* (1966). HO: Hollricher (1965). MC: McClure (1963b). CH: Chambers (1963). AF: Afrosimov *et al.* (1958). SCH: Schwirzke (1960).

beam in highly excited states is negligibly small. Therefore, in many investigations of neutral hydrogen atom beams, including the work of Barnett *et al.* (1958) no effects of the beam in such highly excited states are observed.

5.6 Impurities in the Target Gas

Many kinds of high purity gas are available from commercial sources. They usually contain less than 0.1%–0.5% of impurities. However, since the cross sections for some gases, say, hydrogen atom or molecule targets are usually very small compared with residual gases such as oxygen or nitrogen, a small percentage of impurities present in hydrogen gas has a strong effect (Berkner *et al.*, 1965). Therefore, the hydrogen gas is usually introduced into the collision chamber through heated palladium or nickel pipes in order to prevent other gases from being admitted into the chamber. Condensable impurities can be greatly reduced by using cold traps in or near the collision chamber.

In many cases, errors due to impurities in the target gases caused by using commercially available gases are estimated to be less than one per cent. It is, however, necessary to insure that no impurities are contained in the connecting pipe lines from the gas reservoir to the collision chamber (including variable leak valves and other valves). These pipe lines must be heated to remove any adsorbed gas. Further, cold traps should be inserted between the gas reservoir and the collision chamber except when the target gas itself is condensable.

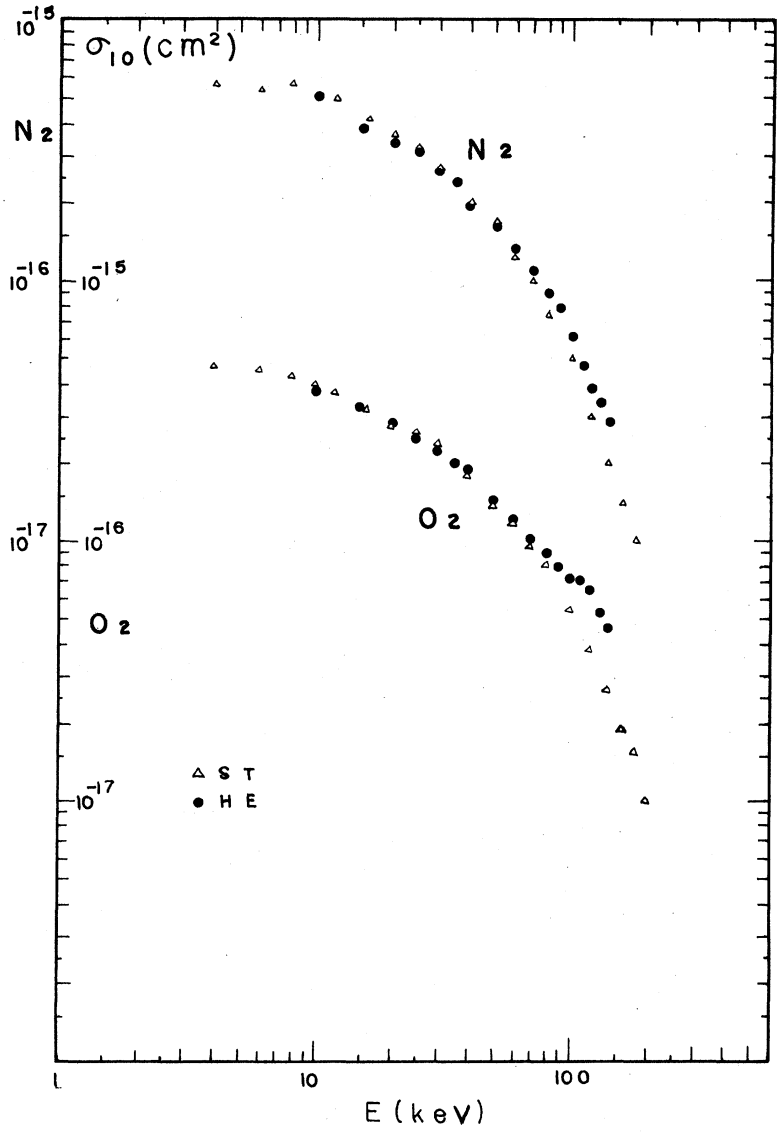

FIG. 7. Single electron capture by a proton in N_2 and O_2 (low energy). ST: Stier and Barnett (1956). HE: de Heer *et al.* (1966).

5.7 Effect of External Fields

Other possible causes of error may be found in the separation of the ion beam into different charge components by external electrostatic or magnetic fields. If the external fields are strong enough to separate the different charge state components of the ion beam, then some ion beam component may strike the aperture edge of the exit channel. This effect becomes pronounced in a very low-energy beam. External electric fields can be shielded by grounded cylindrical shields, so that unwanted electrostatic deflection can be reduced to a negligible level.

Stray magnetic fields (e.g., from mass-analyzing magnets) are generally of the order of a few gauss. Therefore, extreme care must be taken in a long collision chamber. These stray magnetic fields can be reduced by shielding the ion beam path with soft iron or μ-metal sheets.

5.8 Errors in Fitting a Straight Line to Experimental Data

The relation between the growth rate of the fraction I_j/I_i and the gas pressure of the collision chamber is marked by two different pressure regimes (Fogel *et al.*, 1957). At first, the growth is linear in pressure up to a pressure of the order of 10^{-4}–10^{-3} torr. Above this, a remarkable deviation from linearity is observed. In the linear region, multiple collisions are negligible. That is, we have $B\pi \ll A$ in Eq. (43). The relation in this case is given in an apprixomate form

$$I_j/I_i = A\pi, \tag{66}$$

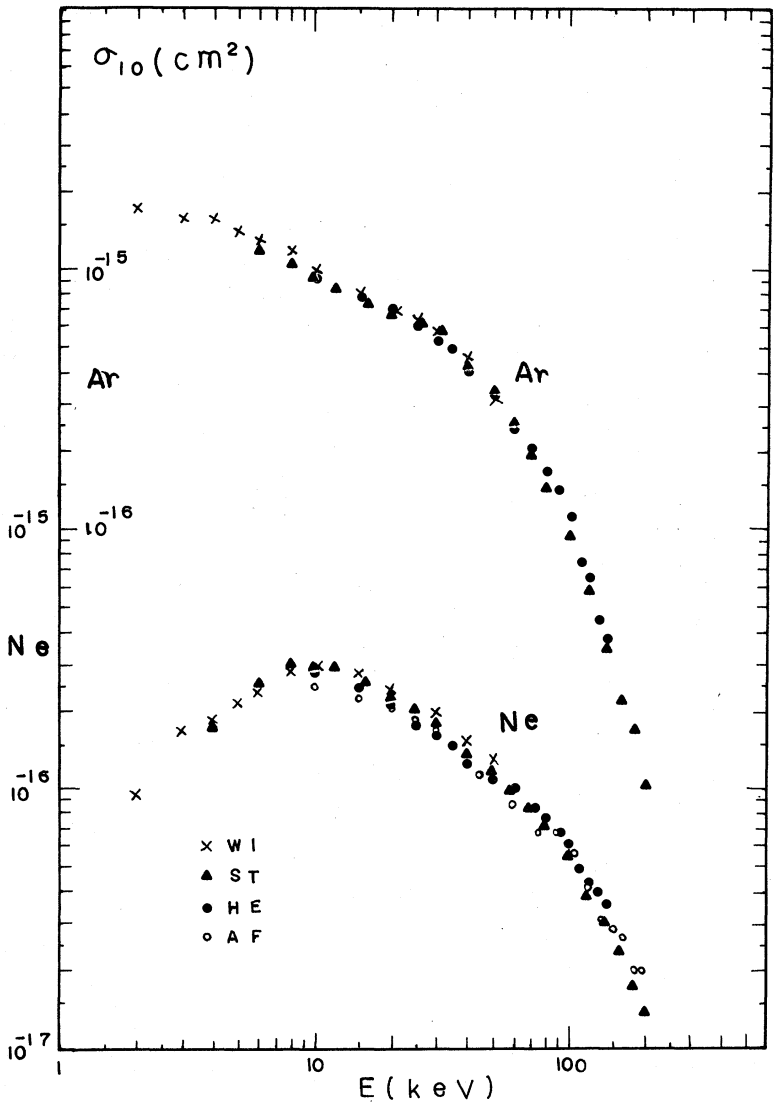

FIG. 8. Single electron capture by a proton in Ne and Ar (low energy). WI: Williams *et al.* (1966). ST: Stier and Barnett (1956). HE: de Heer *et al.* (1966). AF: Afrosimov *et al.* (1960).

and the cross section can be determined from the linear portion of the experimental curve by the method of least squares. If deviations from linearity are apparent even at pressures close to the residual gas pressure in the collision chamber, it is then convenient to express the relation (43) in the following form

$$\pi^{-1}(I_j/I_i) = A + B\pi. \tag{67}$$

The curve of $(I_j/I_i)/\pi$ vs pressure π has a linear portion, and the cross sections can be calculated by the least-square method.

In plotting these relations, collisions with the residual gases, both in the collision chamber and outside, must be taken into consideration. That is, $(I_j/I_i)-(I_j/I_i)_r$ must be plotted against $\pi-\pi_r$, where $(I_j/I_i)_r$ is the value of (I_j/I_i) at the residual gas pressure π_r. This method, however, has some inherent systematic errors associated with (a) the effect of the pressure and composition of the residual gas, (b) unequal scattering of the incident beam and product beam in the collision chamber, and (c) the unequal attenuation of these beams during the passage from the collision chamber to the Faraday cage.

The effect (a) can be determined by measuring the dependence of the cross sections on $(I_j/I_i)_r$; usually the associated errors are small. This effect can be reduced an order of magnitude by inserting cold traps in the collision chamber. For example, $(I_j/I_i)_r$ decreased from 10^{-3} to 10^{-4} with the use of a cold trap in an experiment by Fogel and Mitin (1956). Since the vapors of various organic substances such as diffusion pump oils are condensed on the surface of the cold trap, the pressure of the residual gas is greatly reduced. As a consequence, the associated systematic errors

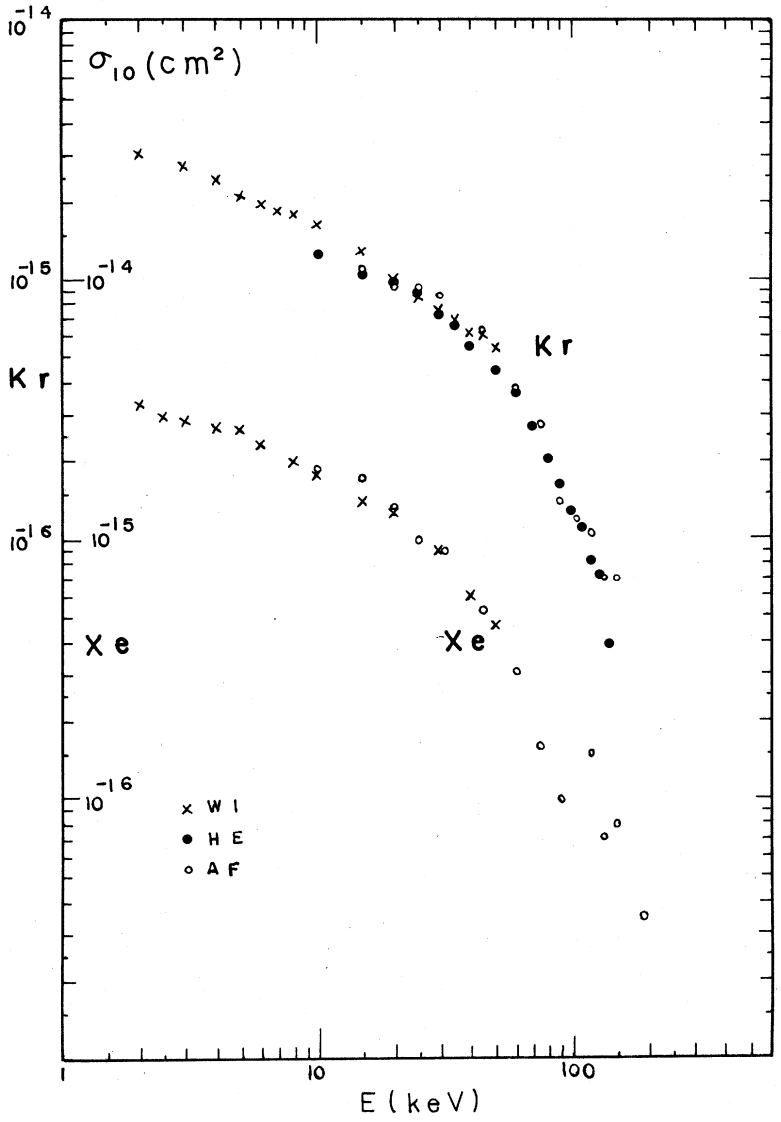

FIG. 9. Single electron capture by a proton in Xe and Kr (low energy). WI: Williams *et al.* (1966). HE: de Heer *et al.* (1966). AF: Afrosimov *et al.* (1960).

can be reduced and the accuracy of the measurement is improved. Another method of reducing the effect of the residual gases is the use of a collision chamber which can be baked at a temperature of about 200–300°C together with an ultrahigh vacuum system. The lowest practicable residual gas pressure may be of the order of 10^{-9}–10^{-10} torr, attained by using ion pumps or diffusion pumps with special oil. (This is to be compared with the residual gas pressures of the order of 10^{-6} torr obtainable with ordinary diffusion pump oil.)

The effect (b) can be examined by changing the aperture of the exit channel of the collision chamber. If the ion beam is sufficiently deflected in the charge change collision processes, the ion beam which has undergone charge change may be limited by the aperture of the exit channel. A measurement by Afrosimov *et al.* (1960) shows the angular distribution of the charge changed ion beam to be broader than that of the incident ion beam. The negative ion produced by double charge change has the broadest angular distribution. However, careful design of the exit channel can limit this effect to less than one percent.

In order to estimate effect (c), it is necessary to know some other cross sections. In most cases, however, the associated errors are small and within the limit of other experimental errors.

6. SUMMARY OF EXPERIMENTAL RESULTS

This paper is limited mainly to data on rare gases (He, Ne, A, Kr, and Xe), molecular gases (H_2, N_2,

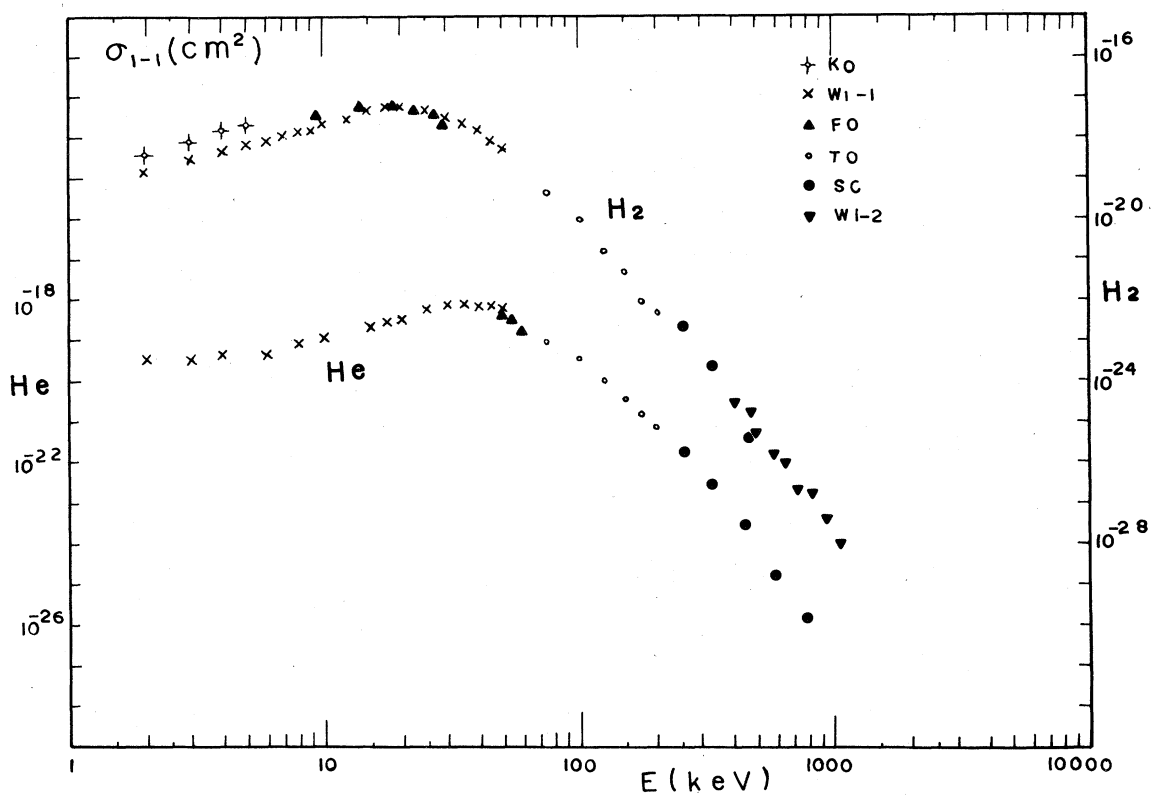

FIG. 10. Double electron capture by a proton in H_2 and He. KO: Kozlov *et al.* (1963). WI-1: Williams (1966). FO: Fogel *et al.* (1959). TO: Toburen *et al.* (1968a). SC: Schryber (1967). WI-2: Williams (1967c).

and O_2) and atomic hydrogen. Therefore, work on other gases such as CO_2 or organic gases are listed in the Tables but not shown in the following figures. However, some results on alkali vapors are briefly described.

As did many workers, Williams and Dunbar (1966) and McClure (1963a) determined the relative values of the cross sections by normalizing them to values accurately measured at some energy for each of the target gases. For an atomic hydrogen target, Fite *et al.* (1960), Hummer *et al.* (1960), Gilbody and Ryding (1966), Wittkower *et al.* (1966) and McClure (1966) determined the relative values of the cross sections and used the known values for a molecular hydrogen target. The experimental results are shown in Figs. 4–35 (except for Fig. 29). In all these figures the cross sections for the charge change are expressed in cm²/atom and the energy of the incident ion or atom beam is in keV.

Some comments on work done so far and on the results shown in figures follow:

6.1 Single-Electron Capture by a Proton

Work done so far on single electron capture by a proton is listed in Table I. Among the six charge changing cross sections of hydrogen beams, the cross section σ_{10} has been the most widely investigated for many kinds of gases and for a wide energy range. In particular, the cross sections σ_{10} for N_2, O_2, and Ar were investigated up to 38-MeV proton energy, which was the highest in the measurement of the charge change of hydrogen beams. The cross sections σ_{10} are shown in Figs. 4–9. The first two of these figures include results in an energy range up to a few tens of MeV and show the trend of the dependence of the cross section σ_{10} on the proton energy. The others show results in the energy range below 100 keV where many measurements have been carried out. Recently, Afrosimov *et al.* (1958), Schryber (1966, 67), and Toburen *et al.* (1968a, b) measured the cross section σ_{10} in the energy range of 100 keV–4.5 MeV with a fine step in energy. Their results agree well with earlier works.

Except for H_2 and He, however, data are scattered in the high-energy region above a few MeV. Szostak *et al.* (1961) measured the cross section for N_2 at 4 MeV, but their result (10^{-24} cm²/atom) seems to be much smaller than the one shown in Fig. 5 (10^{-22} cm²/atom), and is therefore not included in Fig. 5. The curves for some gases (for example, for N_2 as seen in Fig. 5) show a structure around 1-MeV incident proton energy. This structure is due to the capture of

TABLE I. Single electron capture by a proton.

Authors	Reference	Energy (keV)	Target gases	Methods
Stier and Barnett	(1956)	3–200	H_2, N_2, O_2, He, Ne, Ar	Equilibrium+growth
Barnett and Reynolds	(1958)	250–1000	H_2, N_2, He, Ar	Equilibrium+growth
Afrosimov *et al.*	(1958)	5–180	H_2	Condenser
Il'in *et al.*	(1958)	5–180	air	Condenser
Schwirzke	(1960)	9–60	H_2	Condenser
Szostak *et al.*	(1961)	4000	N_2	Growth
McClure	(1963b)	6–50	H_2	Growth
Chambers	(1963)	4–50	H_2	
Gordeev and Panov	(1964)	1–40	H_2, N_2, Ar	Condenser
Hollricher	(1965)	1.5–30	H_2, D_2	Condenser
Berkner *et al.*	(1965)	6450, 10 000	N_2, He, Ar	Thin target
Afrosimov *et al.*	(1960)	10–180	He, Ne, Ar, Kr, Xe	Condenser
de Heer *et al.*	(1966)	10–140	H_2, N_2, O_2, He, Ne, Ar, Kr	Condenser
Williams and Dunbar	(1966)	2–50	H_2, He, Ne, Ar, Kr, Xe	Growth
Williams	(1967c)	250–2500	H_2, He	Growth
Welsh *et al.*	(1967)	440–13 800	H_2, N_2, He, Ar	Thin target
Schryber	(1967)	1000–4400	H_2, N_2, O_2, He, Ne, Ar, Kr	Growth
Toburen *et al.*	(1968a, b)	100–2500	H_2, He, Ar, Kr, O_2, N_2, CO, CO_2, H_2O, CH_4, C_2H_2, C_2H_6, C_4H_{10}	Growth
Acerbi *et al.*	(1969)	32 500, 37 700	N_2, O_2, Ar	Thin target

different orbital electrons of the traget atom. Experimentally, the capture into particular states cannot be distinguished by the measuring methods mentioned above, but the total cross section for the capture into all possible states of atomic hydrogen is obtained. The structure due to this aspect of the electronic shell effect is shown in the theoretical calculations by Mapleton (1967), Bates and Mapleton (1967), and Vinogradov and Shevelkov (1971) have recently discussed the role of inner shell electrons in charge changing processes. Data on the cross section for Xe is limited below 200 keV. Therefore, this effect is not clear in Xe.

6.2 Double Electron Capture by a Proton

Work on the double electron capture of a proton is listed in Table II. The cross sections for the process σ_{1-1} are shown in Figs. 10–14. Until recently, the results of the cross section σ_{1-1} had been limited to below 60 keV, except for H^+ on A by Afrosimov *et al.* (1960) (up to 180 keV). Williams (1967c) (H_2, He), Schryber (1966, 1967) (H_2, He, Ne, A), and Toburen *et al.* (1968a, b) (H_2, He, N_2, A, Kr, H_2O, and some organic gases) have measured the cross section σ_{1-1} at energies up to around 1 MeV. Even at low energy, however, data are scarce, except for a H_2 target. The measurements for targets N_2, Kr, and Xe have been made only by Williams (1966) and Fogel *et al.* (1959). Their results, however, differ by a factor of 2. The data of Fogel and Mitin (1956) for N_2 and O_2 seem to be incorrect because of probable errors in the pressure measurements. Two maxima in the curves for A, Kr, and Xe are due to the following two processes:

$$H^+ + B \rightarrow H^- + B^{2+}$$
$$H^+ + B \rightarrow H^- + B^{3+} + e, \qquad (68)$$

where B denotes the target gas atom. The energies corresponding to these maxima agree with those which follow from Massey's (1949) adiabatic criterion.

This cross section σ_{1-1}, which determines the production of negative ions, has an important technological significance, since negative ions are required by tandem accelerators. [Note, however, that other methods for production of negative ions[2] have been devised (Lawrence *et al.* 1965).] Therefore, more accurate measurements would seem to be needed if only to find more efficient converters of protons (or more generally, of positive ions) to negative ions.

On the other hand, since Donnally and Becker (1967) investigated charge change in alkali vapors (the cross sections for double electron capture by a proton are considerably larger in these than in ordinary gases), much attention has been paid to alkali vapors as charge changers (Donnally and Thoeming, 1966, 1967). Data on alkali atoms, though as yet incomplete, will be described in a later section.

6.3 Single Electron Loss by a Neutral Hydrogen Atom

Table III lists work on single electron loss by a neutral hydrogen atom. The cross sections for this process (σ_{01}) are shown in Figs. 15–17.

[2] For example, an efficient method for direct extraction of negative ions from a duoplasmatron with an off-centered intermediate electrode has been developed.

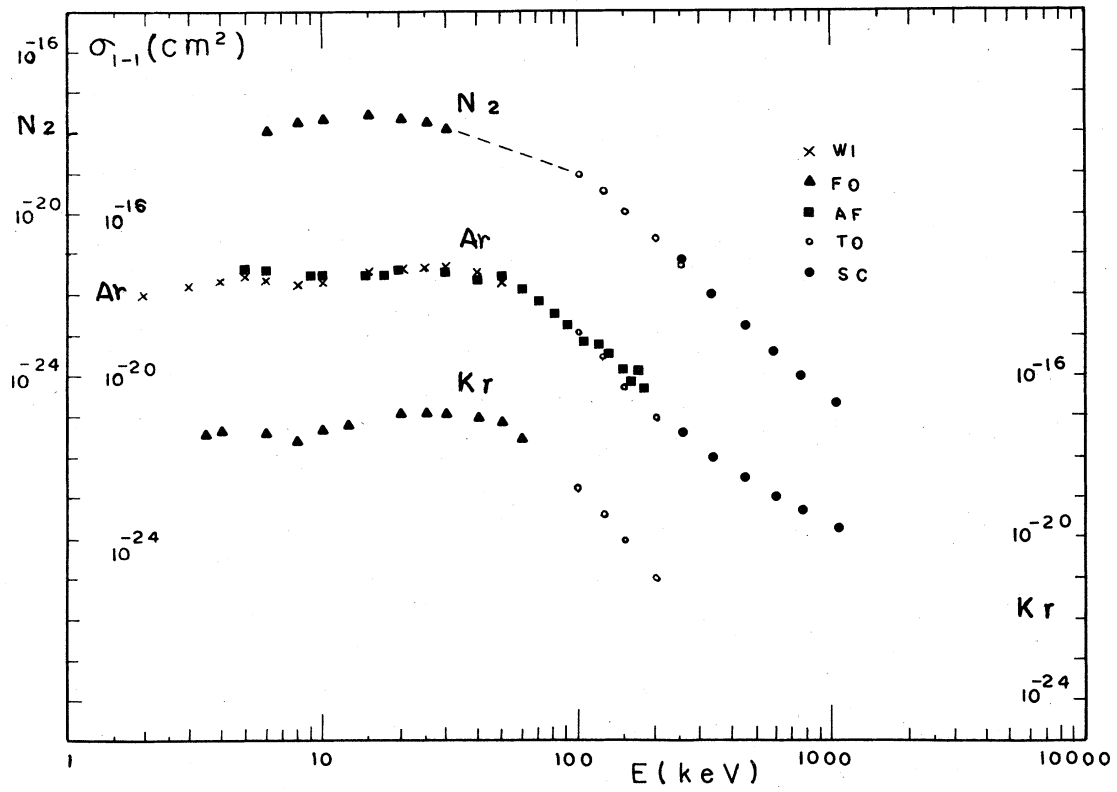

FIG. 11. Double electron capture by a proton in N_2, Ar, and Kr. WI: Williams (1966). FO: Fogel *et al.* (1959, 1960). AF: Afrosimov *et al.* (1960). TO: Toburen *et al.* (1968a). SC: Schryber (1967).

Beams of neutral hydrogen atoms are usually produced by a charge changing process in beams of protons or negative hydrogen ions accelerated to the desired energy. Data on the cross section σ_{01} above a few hundred of keV are very scarce except for H_2, He, N_2, and Ar targets. Moreover, there is considerable scatter in the available data. This may be partly because the production rate of the neutral hydrogen atoms from protons becomes very small at high energy so that it is therefore difficult to obtain sufficiently intense beams of neutral atoms. Therefore, above a few MeV the negative hydrogen ion is used as the source beam, and converted into the neutral hydrogen atom by the stripping reaction whose cross sections σ_{-10} are considerably larger than those for the proton (see Sec. 6.5 and Figs. 22–24). However, facilities for the acceleration of negative ions to such high energies (Rickey and Smythe, 1962; Burgerjon, 1966) are available at

TABLE II. Double electron capture by a proton.

Authors	Reference	Energy (keV)	Target gases	Methods
Fogel and Mitin	(1956)	9.5–29	H_2, N_2, O_2, He, Ne, Ar	Growth
Fogel *et al.*	(1959)	4–60	H_2, N_2, He, Ne, Ar, Kr, Xe	Growth
Afrosimov *et al.*	(1960)	10–180	Ar	Condenser
Szostak *et al.*	(1961)	4000	N_2	Growth
Kozlov *et al.*	(1963)	0.5–5	H_2, Ar, Kr	Retard
McClure	(1963b)	6–50	H_2	Growth
Farrokhi	(1966)	10–50	CH_4, C_2H_6, C_4H_{10}, C_3H_8	Growth
Williams	(1966)	2–50	H_2, He, Ne, Ar, Kr, Xe	Growth
Williams	(1967c)	400–2500	H_2	Growth
Schryber	(1967)	253–1025	H_2, N_2, He, Ar	Growth
Toburen *et al.*	(1968a, 1969)	75–250	H_2, He, Ar, Kr, N_2, H_2O, CH_4, C_2H_2, C_2H_6, C_4H_{10}	Growth

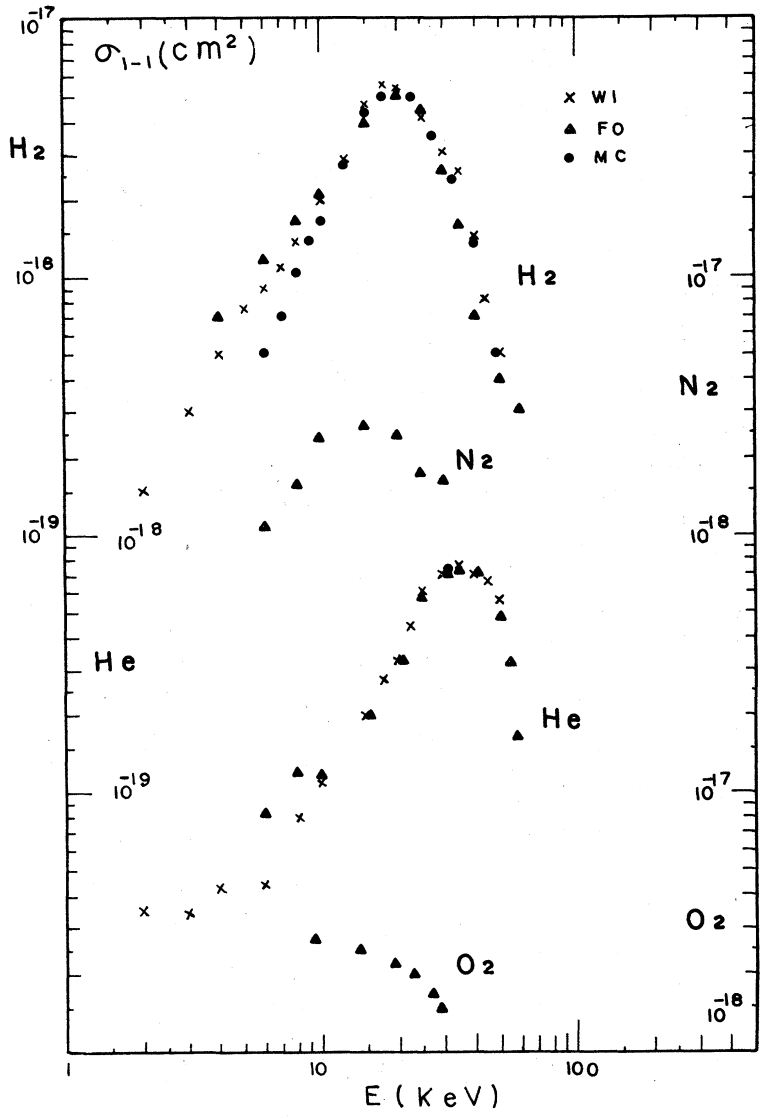

FIG. 12. Double electron capture by a proton in H_2, N_2, He, and O_2 (low energy). WI: Williams (1966). FO: Fogel *et al.* (1959), 1960). MC: McClure (1963b).

only a few laboratories (Berkner *et al.*, 1964; Smythe and Toers, 1965). In addition, data for Xe targets are also scarce and limited below 50 keV.

Fogel's (1958) results seem to be considerably smaller in all of the gases listed, compared to other results. Reasons for this difference are not clear, but seem to be due to the effect of an admixture of excited metastable atoms in the neutral atom beam entering the collision chamber. The relative amounts of metastable atoms in the neutral beam may differ depending on experimental conditions. For example, Fogel *et al.* (1958) used mercury vapor for neutralizing the proton, while other workers used ordinary gases as a neutralizer. Also, the distance between the neutralizer and the collision chamber of Fogel *et al.* was much longer than that of others.

6.4 Single Electron Capture by a Neutral Hydrogen Atom

Work on the single electron capture by a neutral hydrogen atom is listed in Table IV. The cross sections for this process (σ_{0-1}) are shown in Figs. 18–21. Most of data lie below 60 keV. The cross section σ_{0-1} for Kr and Xe up to 60 keV was measured only by Fogel *et al.* (1958) and Williams (1967a). Data in high-energy regions have only recently been obtained by Schryber (1966, 1967) for H_2, He, N_2, and Ar (250 keV–1 MeV). However, they are scarce above 100 keV. The reasons for the paucity of data in this energy region are the same as for those in the cross section σ_{01} (See Sec. 6.3.)

From a practical point of view the size of this cross

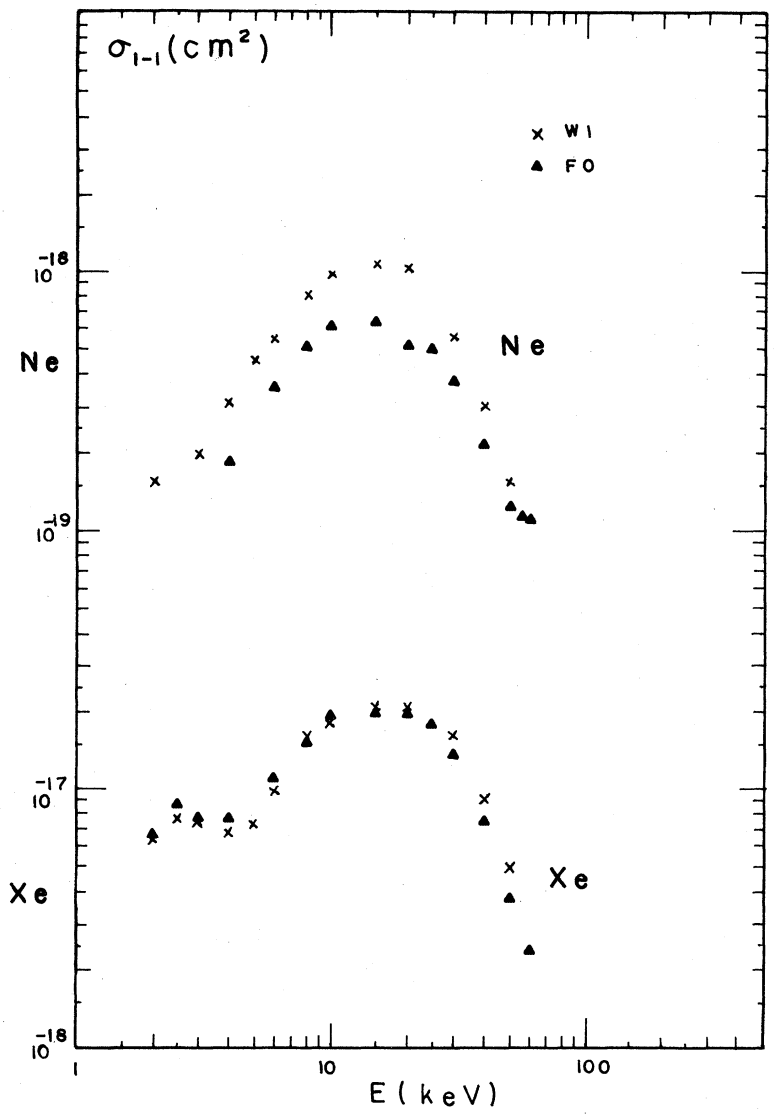

FIG. 13. Double electron capture by a proton in Ne and Xe (low energy). WI: Williams (1966). FO: Fogel *et al.* (1959).

section σ_{0-1} is an important design factor for the three stage tandem machine with a neutral beam injection system. Therefore, more measurements and a more thorough accumulation of data are necessary.

6.5 Single Electron Loss by a Negative Hydrogen Ion

Work on single electron loss by a negative hydrogen ion is listed in Table V. The cross sections for this process σ_{-10} are shown in Figs. 22–24. The difficulties in obtaining a high-energy negative hydrogen ion have limited experimental work to date. Rose *et al.* (1958) measured the cross section using a Van de Graaff accelerator. Also using a Van de Graaff type accelerator, Dimov and Dudnikov (1967) and Budker *et al.*, (1967) investigated the charge change of a negative hydrogen ion during the course of studies on high-energy particle injection into a storage ring. They measured the cross section σ_{-10} for SF_4 and some organic gases as well as ordinary gases. The former gases have considerably larger cross sections than do the latter.

Negative ions with energies over a few MeV are obtained by the negative ion accelerating AVF cyclotron (Rickey and Smythe, 1962; Burgerjon, 1966). Verba *et al.* (1963) measured the cross section in air for the electron loss of negative hydrogen ions with energies between 25 and 50 MeV which were obtained from an AVF.

Recently Snow *et al.* (1969) measured the charge changing cross section σ_{-10} of 0.5–4-keV negative hydrogen ion beams in O_2, NO_2, and atomic oxygen gases. Data in high-energy regions are scattered both in energy range and in the cross section itself. Even in

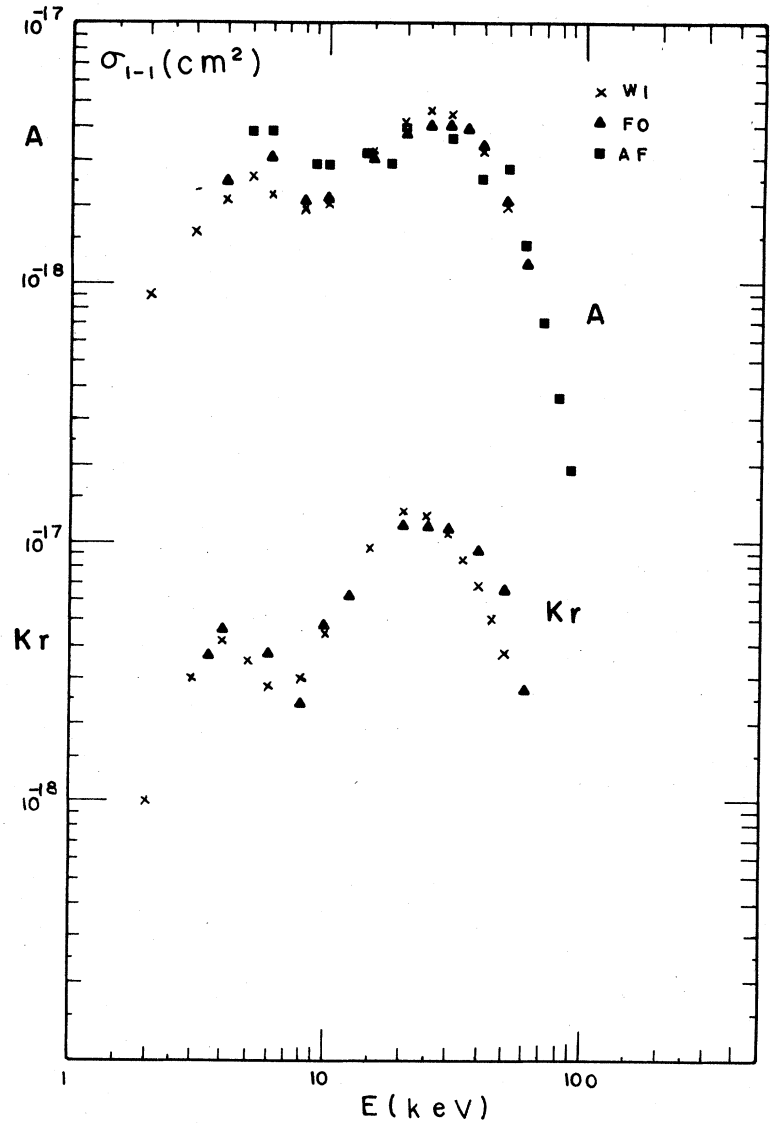

FIG. 14. Double electron capture by a proton in Ar and Kr (low energy). WI: Williams (1966). FO: Fogel *et al.* (1959). AF: Afrosimov *et al.* (1960).

low-energy regions, data are scarce compared with other data. In all measurements done so far, the negative hydrogen ion was produced by the charge change of a proton. However, such charge changing processes are accompanied with the scattering of the product beam, which becomes large in low-energy beams. Some parts of the charge changed (negative ion) beam may strike the walls of the exit channel of the collision chamber, which may introduce systematic errors in the measurement. Therefore, measurements are preferably made with the negative ion beam directly extracted from an ion source such as a Duoplasmatron (Lawrence *et al.*, 1965; Heinicke *et al.*, 1968) the qualities of which are far better than those of a beam produced by charge changing a proton beam.

6.6 Double Electron Loss by a Negative Hydrogen Ion

Work on the double electron loss of negative hydrogen is listed in Table VI. The cross sections for this process σ_{-11} are shown in Figs. 25–28. The same difficulties of acceleration of the negative ion beam described above are also enountered in this case. Data over 50 keV are completely scattered and only a few data are available, e.g., Dimov and Dudnikov (1967) (around 1 MeV), Symthe and Toevs (1965) and Berkner *et al.* (1964) (around 10 MeV).

Considerable data are available for N_2, but it is difficult to draw a smooth curve through the measured cross sections (see Fig. 28). Therefore, no conclusions

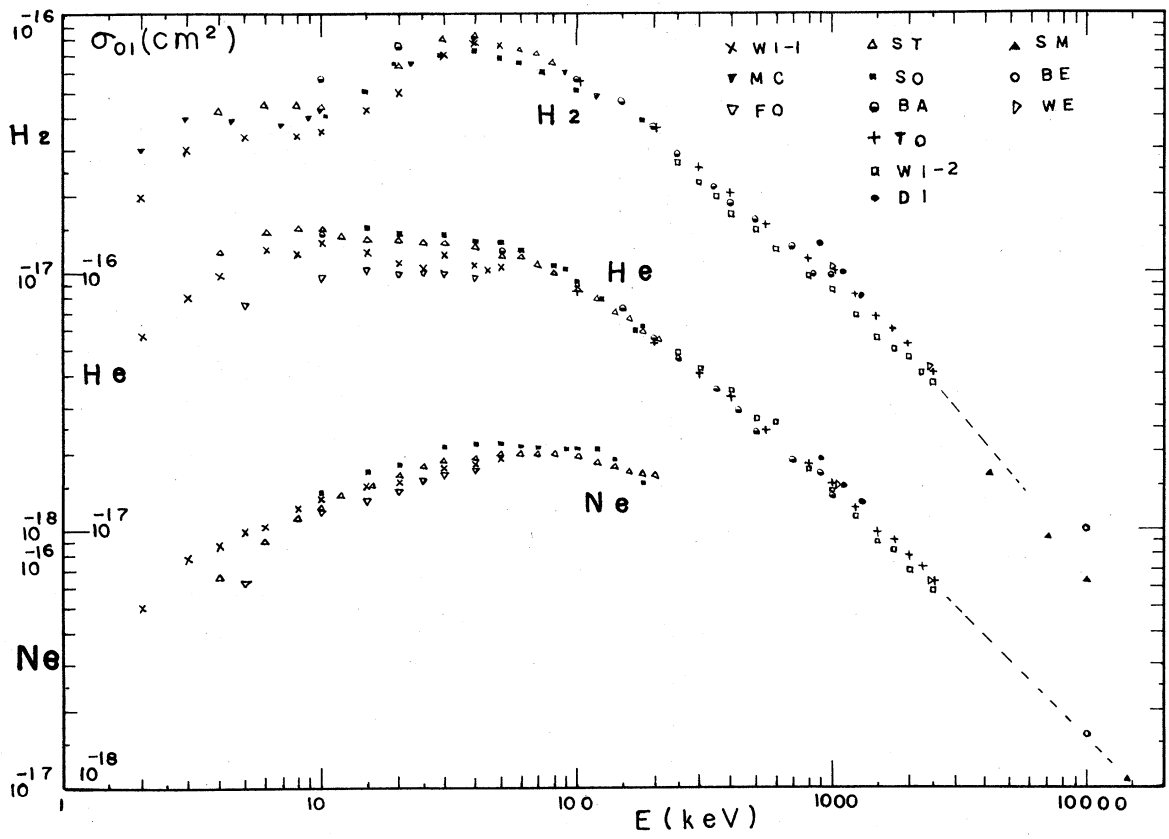

FIG. 15. Single electron loss by a hydrogen atom in H_2, He, and Ne. WI-1: Williams (1967a). MC: McClure (1964). FO: Fogel *et al.* (1958). ST: Stier and Barnett (1956). SO: Solovev *et al.* (1962). BA: Barnett and Reynolds (1958). TO: Toburen *et al.* (1968a). WI-2: Williams (1967c). DI: Dimov and Dudnikov (1967). SM: Smythe *et al.* (1965). BE: Berkner *et al.* (1964). WE: Welsh (1967), Welsh *et al.* (1967). The result of Berkner *et al.* is obtained with 20-MeV deuterons.

TABLE III. Single electron loss by a hydrogen atom.

Authors	Reference	Energy (keV)	Target gases	Methods
Stier and Barnett	(1956)	3–200	H_2, N_2, O_2, He, Ne, Ar	Equilibrium + attenuation
Barnett and Reynolds	(1958)	250–1000	H_2, N_2, He, Ar	Equilibrium + attenuation
Fogel *et al.*	(1958)	5–40	H_2, N_2, O_2, He, Ne, Ar, Kr, Xe	Growth
Curran and Donahue	(1960)	4.5–40	H_2	Condenser
Schwirzke	(1960)	10–60	H_2	Condenser
Donahue and Hushfar	(1961)	8–40	Ar, CO	Condenser
Szostak *et al.*	(1961)	4000	N_2	Growth
Solovev *et al.*	(1962)	10–180	H_2, N_2, He, Ne, Ar, Kr	Condenser
Pilipenko and Fogel	(1962)	8.5–40	H_2, N_2, CO, O_2	Growth
Pilipenko and Fogel	(1963)	15–35	CO, NO	Growth
Berkner *et al.*	(1964)	1000	H_2, N_2, He, Ar	Thin target
McClure	(1964)	2–120	H_2	Growth
Smythe and Toevs	(1965)	4000–18 000	H_2, N_2, He, Ar	Growth
Dimov and Dudnikov	(1966)	900–1300	H_2, He, CO_2, SF_4, C_3H_8, CCl_2F_2	Growth
Williams	(1967c)	250–2500	H_2, He	Growth
Williams	(1967a)	2–50	H_2, He, Ne, Ar, Kr, Xe	Growth
Welsh *et al.*	(1967)	440–13 800	H_2, N_2, He, Ar	Thin target
Schryber	(1967)	1000–4500	N_2, Ar	Growth
Toburen *et al.*	(1967)	75–250	H_2, He, Ar, Kr, N_2, H_2O	Growth

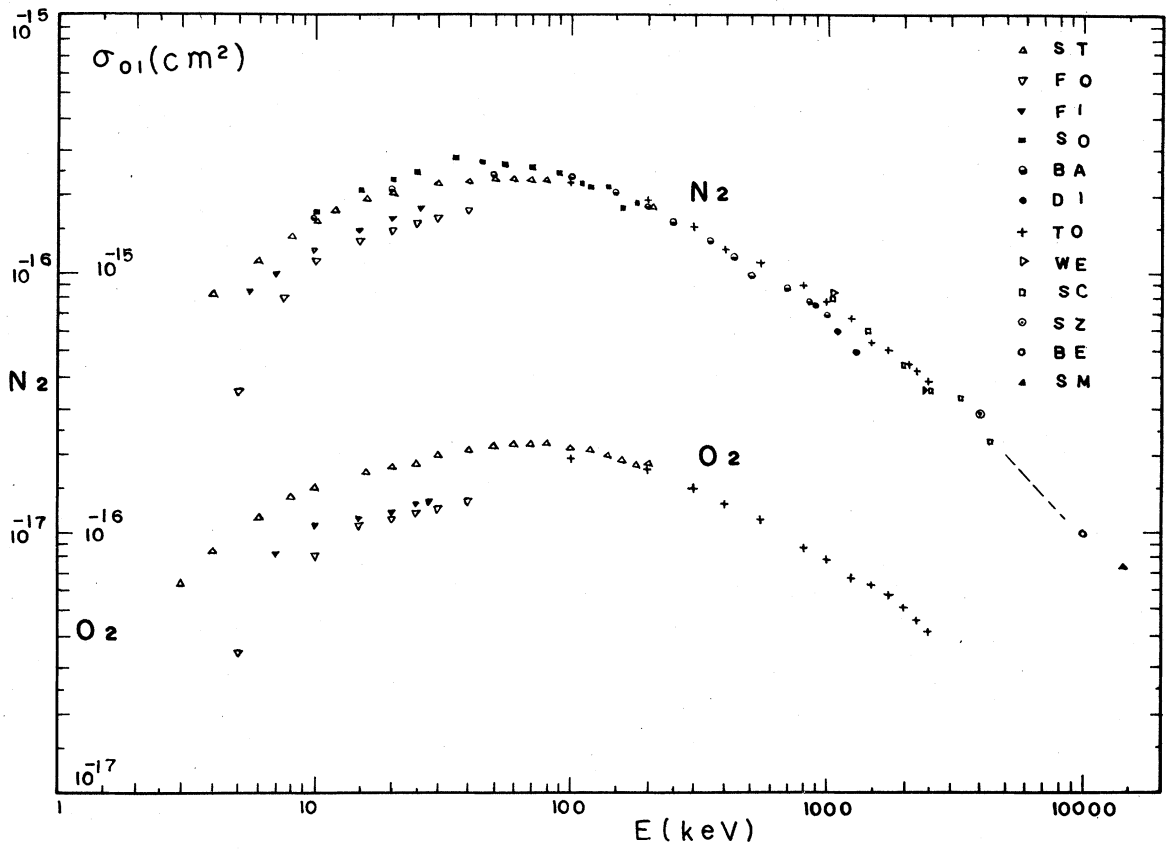

FIG. 16. Single electron loss by a hydrogen atom in N_2 and O_2. ST: Stier and Barnett (1956). FO: Fogel *et al.* (1958). PI: Pilipenko and Fogel (1962). SO: Solovev *et al.* (1962). BA: Barnett and Reynolds (1958). DI: Dimov and Dudnikov (1967). TO: Toburen *et al.* (1968a). WE: Welsh (1967), Welsh *et al.* (1967). SC: Schryber (1967). SZ: Szostak *et al.* (1961). BE: Berkner *et al.* (1964). SM: Smythe *et al.* (1965). The result of Berkner *et al.* is obtained with 20-MeV deuterons.

can be drawn on the trend of the variation of the cross section σ_{-11} with energy. Even in the low-energy region, data are very scarce. The main contributors to the measurement of this process are Fogel *et al.* (1957) and Williams (1967b). No comparisons are possible for Kr, Xe, O_2, and N_2 because only Fogel's data are available for these targets. Dimov and Dudnikov also measured the cross section σ_{-11} for SF_4 and for some organic gases around 1 MeV, but these are not reported in this paper.

In tandem machines a knowledge of this cross section σ_{-11} is very important in order to find efficient converters of the negative ion into a positive ion at the high voltage terminal of the tandem machine.

TABLE IV. Single electron capture by a hydrogen atom.

Authors	Reference	Energy (keV)	Target gases	Methods
Stier and Barnett	(1956)	4–30	H_2, N_2, O_2, He, Ne, Ar	Equilibrium+attenuation
Fogel *et al.*	(1958)	5–40	H_2, N_2, O_2, He, Ne, Ar, Kr, Xe	Growth
Curran and Donahue	(1960)	4.5–50	H_2	Condenser
Donahue and Hushfar	(1961)	8–40	Ar, CO	Condenser
Pilipenko and Fogel	(1962)	5–25	O_2, N_2, CO	Growth
Pilipenko and Fogel	(1963)	15–35	NO, CO	Growth
McClure	(1964)	2–90	H_2	Growth
Jorgensen *et al.*	(1965)	40–100	H_2	Growth+equilibrium
Schryber	(1967)	250–700	H_2, N_2, He, Ar	Growth
Williams	(1967a)	2–50	H_2, He, Ne, Ar, Kr, Xe	Growth

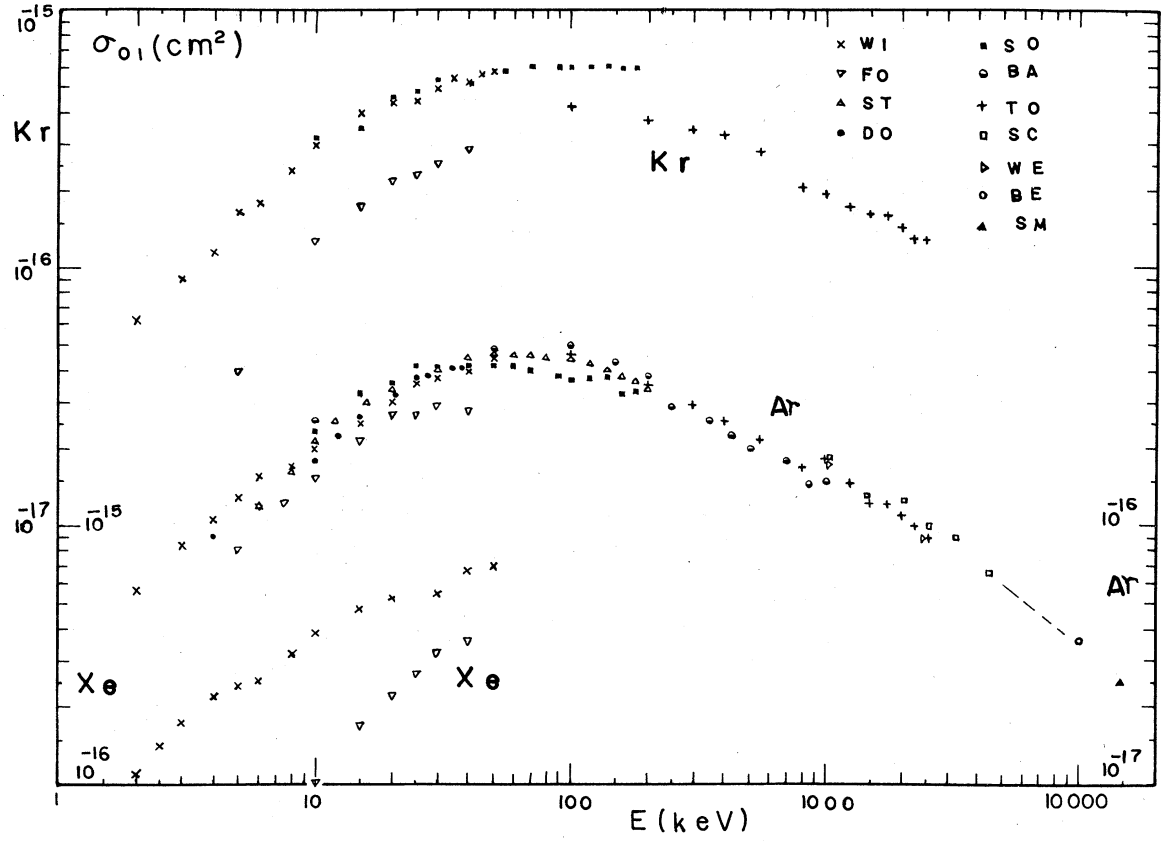

FIG. 17. Single electron loss by a hydrogen atom in Ar, Xe, and Kr. WI: Williams (1967a). FO: Fogel *et al.* (1958). ST: Stier and Barnett (1956). DO: Donahue and Hushfar (1961). SO: Solovev *et al.* (1962). BA: Barnett and Reynolds (1958). TO: Toburen *et al.* (1968a). SC: Schryber (1967). WE: Welsh (1967), Welsh *et al.* (1967). BE: Berkner *et al.* (1964). SM: Smythe *et al.* (1965). The result of Berkner *et al.* is obtained with 20-MeV deuterons.

Future measurements of this cross section σ_{-11} are required over wider energy regions.

6.7 Atomic Hydrogen

Charge changing processes between free (slow) hydrogen atoms and a high-energy hydrogen beam in a given charge state (proton, neutral atom, or negative ion), being in theory the simplest processes, have for many years been a problem of great theoretical interest. However, experiments are difficult because of the problems of atomic hydrogen target preparation. Instead, therefore, molecular hydrogen is usually used and half the value of the cross section obtained experimentally for molecular hydrogen compared with that calculated theoretically for atomic

TABLE V. Single electron loss by a negative hydrogen ion.

Authors	Reference	Energy (keV)	Target gases	Methods
Hasted and Stedeford	(1955)	3–40	He, Ne, Ar, Kr, Xe	Condenser
Stier and Barnett	(1956)	4–30	H_2, N_2, O_2	Equilibrium + attenuation
Rose *et al.*	(1958)	400–1750	H_2, O_2, CO_2, Ar	
Verba *et al.*	(1963)	25 000–50 000	air	
Berkner *et al.*	(1964)	10 000	H_2, N_2, He, Ar	Thin target
Smythe and Toevs	(1965)	4200–14 600	H_2	Growth
Dimov and Dudnikov	(1967)	900–1300	H_2, He, N_2, CO_2, C_3H_8, CCl_2F_2, SF_4	Growth
Kovacs	(1967)	200–500	N_2, CO_2	Growth
Williams	(1967b)	2–50	H_2, He, Ne, Ar, Kr, Xe	Growth
Snow *et al.*	(1969)	0.5–4	O_2, N_2, O	Crossed beam

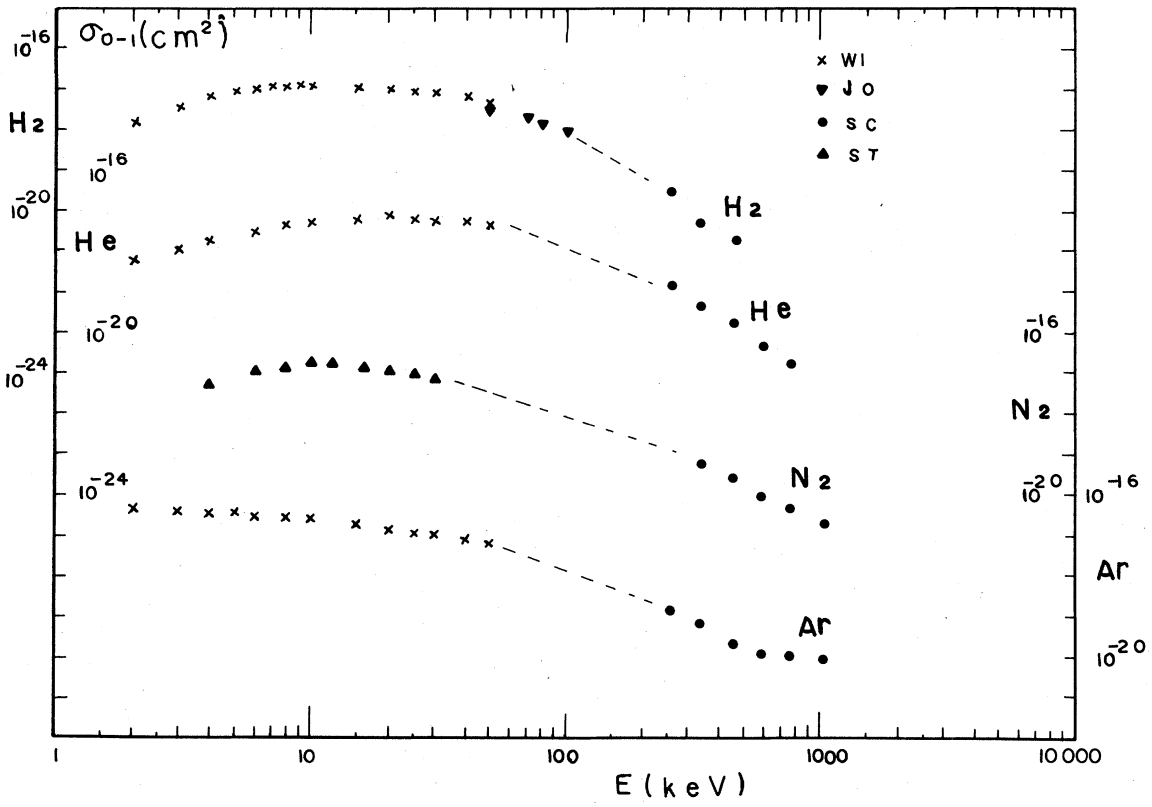

FIG. 18. Single electron capture by a hydrogen atom in H_2, He, N_2, and Ar. WI: Williams (1967a). JO: Jorgensen *et al.* (1965). SC: Schryber (1967). ST: Stier and Barnett (1956).

hydrogen. Experimental studies on the charge change for actual atomic hydrogen have only recently begun.

The first measurement using atomic hydrogen as a target was made by Fite *et al.* (1958), to determine the cross section for $H^+ + H^0 \rightarrow H^0 + H^+$ in the energy range 100 eV–40 keV. Gilbody and Ryding (1966) and Wittkower *et al.* (1966) extended the measurements up to 250 keV. Further, Ryding *et al.* (1968) have recently measured the cross section for charge change of a proton on atomic hydrogen above 1 MeV. However, their results are not yet published in detail.

Atomic hydrogen targets are usually produced by a tungsten furnace heated to 3000°K. While passing through the furnace, some of the molecular hydrogen dissociates into atomic hydrogen. It has been shown that hydrogen is practically entirely in the atomic state at target temperature above 2400°K, provided the hydrogen pressure is sufficiently low (Lockwood and Everhart, 1962). One example of such a tungsten furnace is shown in Fig. 29. In this type, the furnace made of tungsten foils is heated by conduction of an electric current applied at the terminals *E*. The molecular hydrogen gas is fed through a capillary *G* and dissociates during passage through the heated tungsten foils. The incident beam of fast protons or neutral hydrogen atoms passes directly through the tungsten

TABLE VI. Double electron loss by a negative hydrogen ion.

Authors	Reference	Energy (keV)	Target gases	Methods
Fogel *et al.*	(1957)	5–40	H_2, N_2, O_2, He, Ne, Ar, Kr, Xe	Growth
Tisone *et al.*	(1964)	0.5–4	H_2	Growth
Berkner *et al.*	(1964)	10 000	H_2, N_2, He, Ne	Thin target
Smythe and Toevs	(1965)	4200–14 600	H_2, He, N_2, O_2, Ar	Growth
Kovacs	(1967)	200–500	N_2, CO_2	Growth
Dimov and Dudnikov	(1967)	900–1300	H_2, He, N_2, CO_2, C_3H_8, CCl_2F_2, SF_4	Growth
Williams	(1967b)	2–50	H_2, He, Ne, Ar	Growth

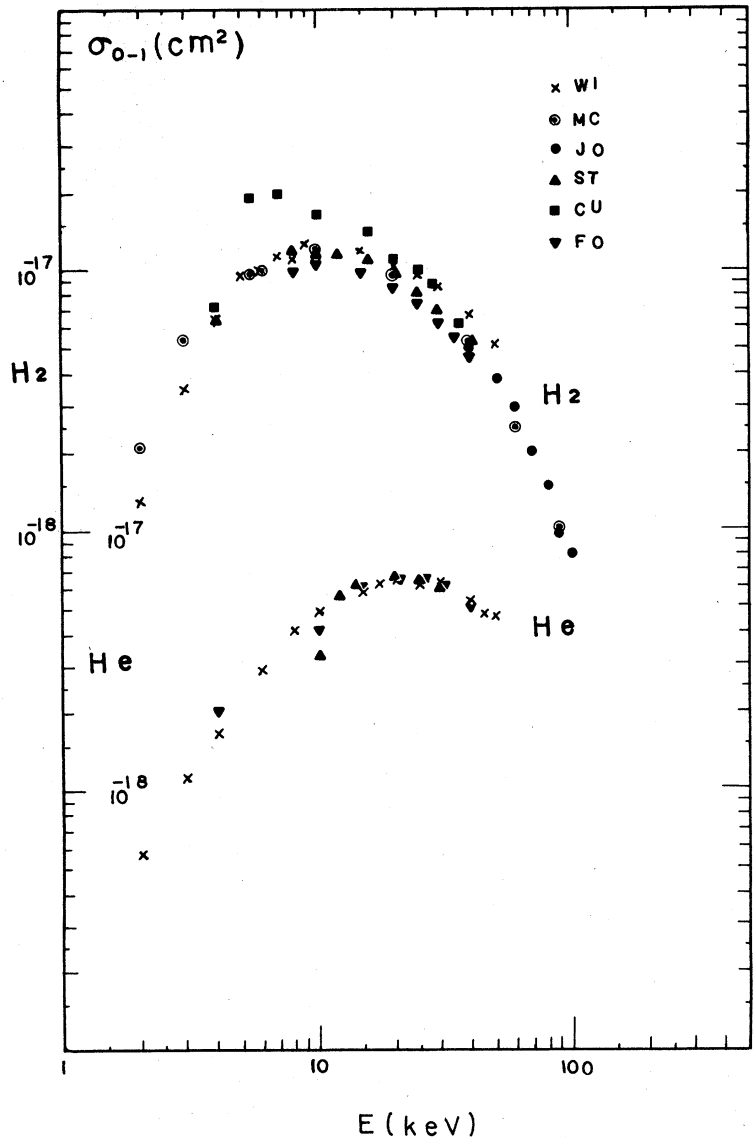

FIG. 19. Single electron capture by a hydrogen atom in H_2 and He (low energy). WI: Williams (1967a). MC: McClure (1964). JO: Jorgensen *et al.* (1965). ST: Stier and Barnett (1956). CU: Curran and Donahue (1960). FO: Fogel *et al.* (1958).

TABLE VII. Charge changing processes of a hydrogen beam in atomic hydrogen.

Authors	Reference	Energy (keV)	Measured	Methods
Fite *et al.*	(1958)	0.2–40	σ_{10}	Crossed beam
Fite *et al.*	(1960)	0.4–40	σ_{10}	Crossed beam
Hummer *et al.*	(1960)	1.25–40	σ_{-10}	Crossed beam
Fite *et al.*	(1962)	0.4–40	σ_{10}	Crossed beam
McClure	(1966)	2–117	σ_{10}	Direct
Gilbody and Ryding	(1966)	40–130	σ_{10}	Crossed beam
Wittkower *et al.*	(1966)	40–250	σ_{10}	Direct
Wittkower *et al.*	(1967b)	40–220	σ_{01}	Direct
McClure	(1968a, b)	1.25–117	σ_{01}	Direct
McClure	(1968b)	3.15–63	σ_{0-1}	Direct
Ryding *et al.*	(1968)	1000	σ_{10}	Direct

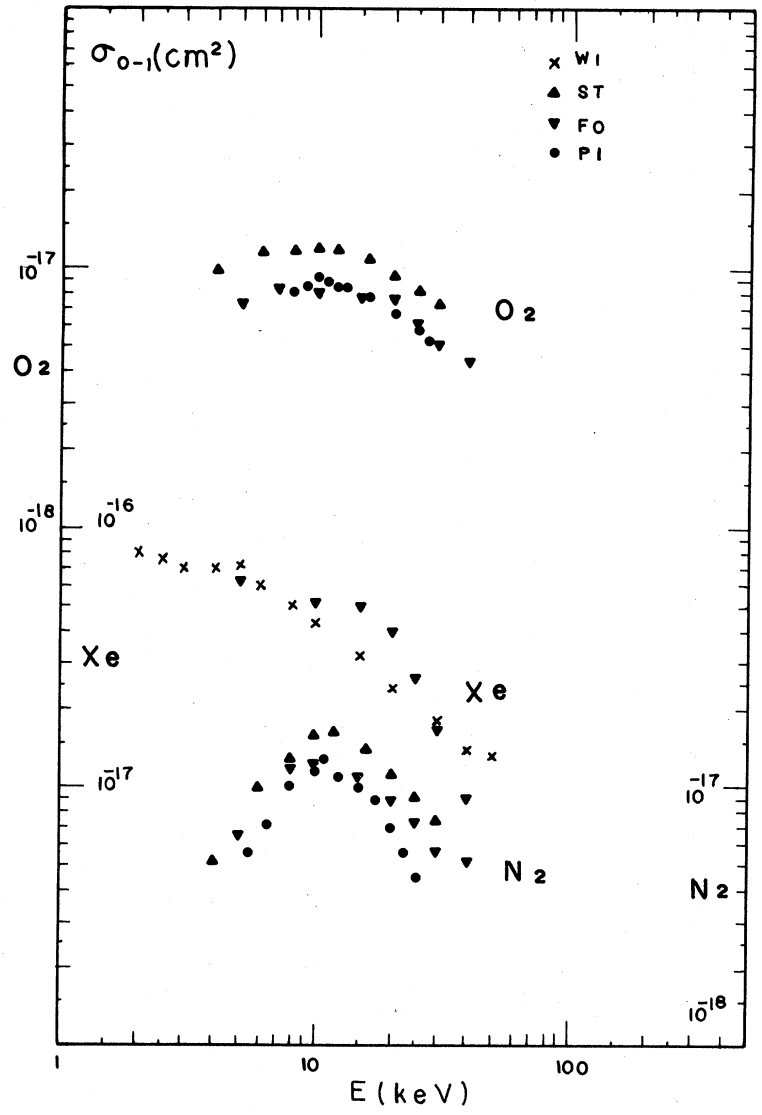

FIG. 20. Single electron capture by a hydrogen atom in N_2, O_2, and Xe (low energy). WI: Williams (1967a) ST: Stier and Barnett (1956). FO: Fogel *et al.* (1958). PI: Pilipenko and Fogel (1962).

furnace. This method provides a considerably thicker target than that available from crossed beam techniques. The charge change between the incident beam and the atomic hydrogen target may occur during passage through the atomic hydrogen furnace (through-the-furnace method) or at the intersection with the atomic hydrogen emergent from the furnace (crossed beam technique). However, it must be pointed out that accurate determination of the thickness of the atomic hydrogen target or the exact fraction dissociated is very difficult.

The ratio of the cross section for atomic hydrogen Q_1 to that for molecular hydrogen Q_2 can be obtained by comparing the measurements for nearly pure atomic hydrogen (highly dissociated molecule) from a high temperature tungsten furnace with those for entirely molecular hydrogen at room temperature. From these ratios Q_1/Q_2 absolute values of Q_1 are determined using the known values of Q_2. The ratio Q_1/Q_2 at high energy over 100 keV, measured by Wittkower *et al.* (1966), is constant at 0.42 ± 0.03, which is in good agreement with the calculated value of 0.41 in the high-energy limit by Tuan and Gerjuoy (1960).

Work on charge changing processes of atomic hydrogen is listed in Table VII. The results are shown in Figs. 30 and 31. Data presently available consist mainly of the cross sections for single electron capture by a proton and single electron loss by neutral hydrogen. The cross sections for single electron capture by neutral hydrogen have been measured only by McClure (1968a, b), and in a limited energy range, at that. The processes of single and double electron loss of the negative hydrogen ion and double electron capture of

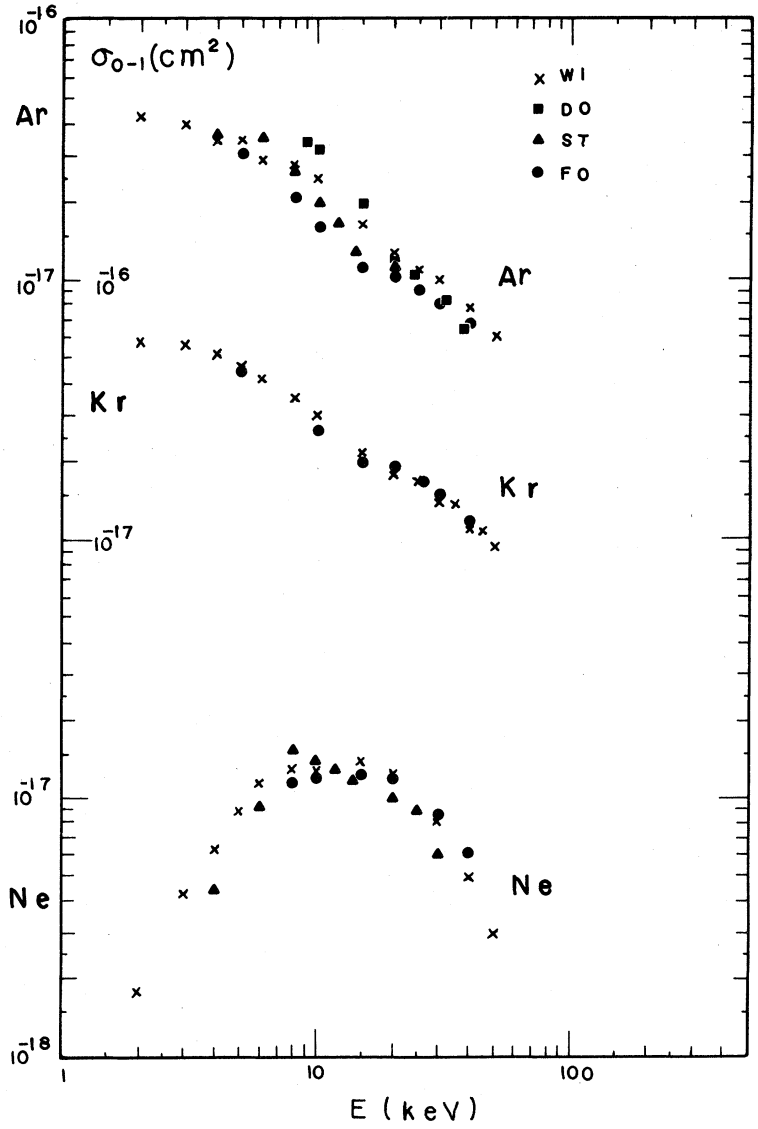

FIG. 21. Single electron capture by a hydrogen atom in Ne, Ar, and Kr (low energy). WI: Williams (1967a). DO: Donahue and Hushfar (1961). ST: Stier and Barnett (1956). FO: Fogel *et al.* (1958).

a proton incident on an atomic hydrogen target have not been investigated. Wittkower *et al.* (1967b) concluded from their results that at energies above 100 keV, half of the cross section of the neutral molecular hydrogen beam is appreciably less than the cross section for atomic hydrogen. Thus, in fast particle collisions involving electron loss, molecular hydrogen cannot be considered to be two free hydrogen atoms. However, this conclusion must be tested in other charge changing processes. In the atomic hydrogen experiments, especially in the crossed beam method, the density of atomic hydrogen is quite small compared with that of the residual gases. Therefore, the separation of that portion of the signal due to atomic hydrogen from that due to the residual gases is difficult. This is usually overcome by modulating the admission of the atomic hydrogen and using phase sensitive detectors.

6.8 Alkali and Alkaline Earth Metal Vapors

Studies on the charge change of ion beams in alkali or alkaline earth metal vapors have begun recently.

According to theoretical and experimental studies by Massey (1949) and Hasted (1960) the cross sections for charge change have shown the following trends:

(1) In such charge changing processes as single electron capture or double electron capture, the velocity of the indicent beam corresponding to the maximum cross section is proportional to the energy defect ΔE of the process.

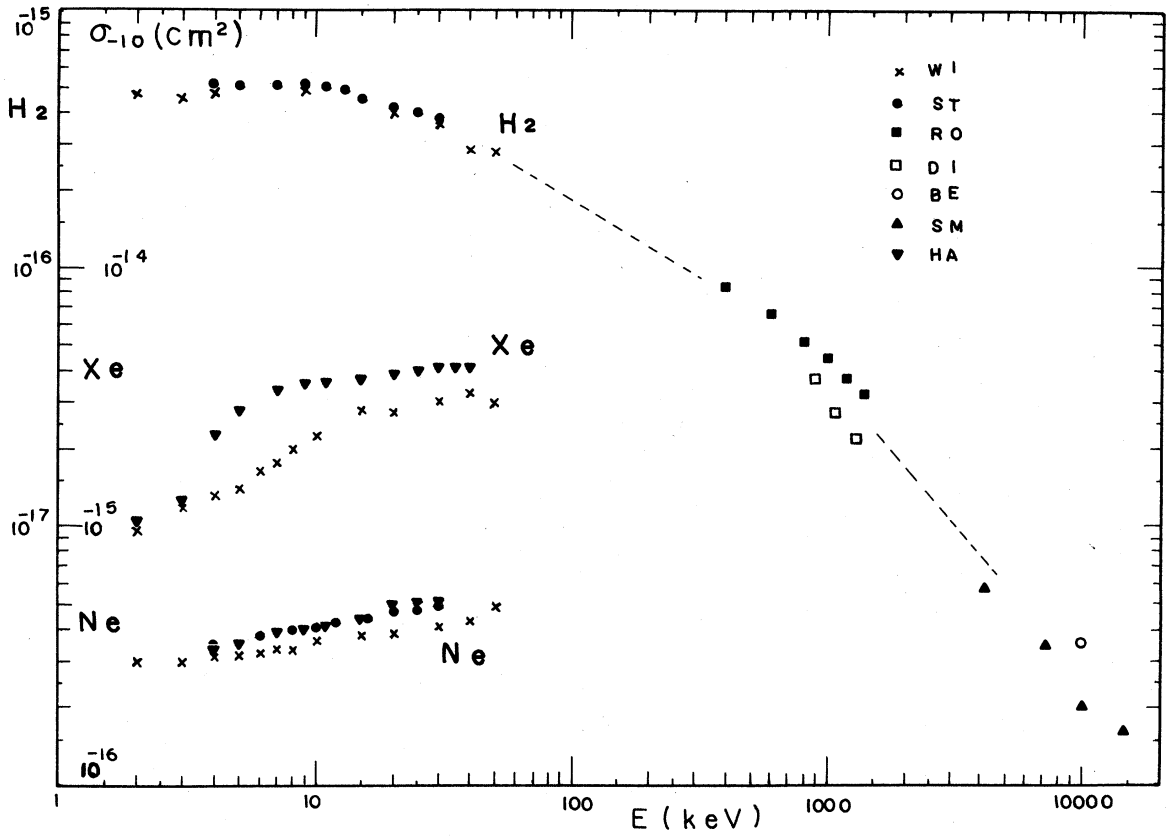

FIG. 22. Single electron loss by a negative hydrogen ion in H_2, Ne, and Xe. WI: Williams (1967b). ST: Stier and Barnett (1956). RO: Rose *et al.* (1958). DI: Dimov and Dudnikov (1967). BE: Berkner *et al.* (1964). SM: Smythe and Toevs (1965). HA: Hasted and Stedeford (1955). The result of Berkner *et al.* is obtained with 20-MeV deuterons.

TABLE VIII. Charge changing processes of a hydrogen beam in alkali vapors.

Authors	Reference	Energy (keV)	Vapors	Measured
Futch and Moses	(1967)	4–45	Mg	σ_{10}, σ_{01}
Nieman *et al.*	(1967)	30–50	K	σ_{10}, σ_{01}
Donnally and Becker	(1967)	1	K	σ_{10}
Il'in *et al.*	(1965, 1967)	10–180	Li, Na, K, Mg, Cs	$\sigma_{10}+\sigma_{1-1}$
Sellin and Granoff	(1968)	10–50	K, Cs, Rb	...
Bohlen *et al.*	(1968)	0.5–2	K, Cs	...
Schmelzbach *et al.*	(1968a)	2.5–22	K	σ_{10}, σ_{1-1}
Dyachkov and Zinenko	1968	5–70	Li, Na, Mg, Zn	...
Schlachter *et al.*	(1969)	0.5–15	Cs	σ_{10}, σ_{1-1} σ_{01}, σ_{0-1}
Berkner *et al.*	(1969)	5–70	Mg	σ_{10}, σ_{01} σ_{0-1}, σ_{-10}
Dyachkev	(1969)	10–400	Li	σ_{10}, σ_{01}
Grüebler *et al.*	(1970)	1–25	Li, Na, K, Cs	σ_{10}, σ_{1-1}

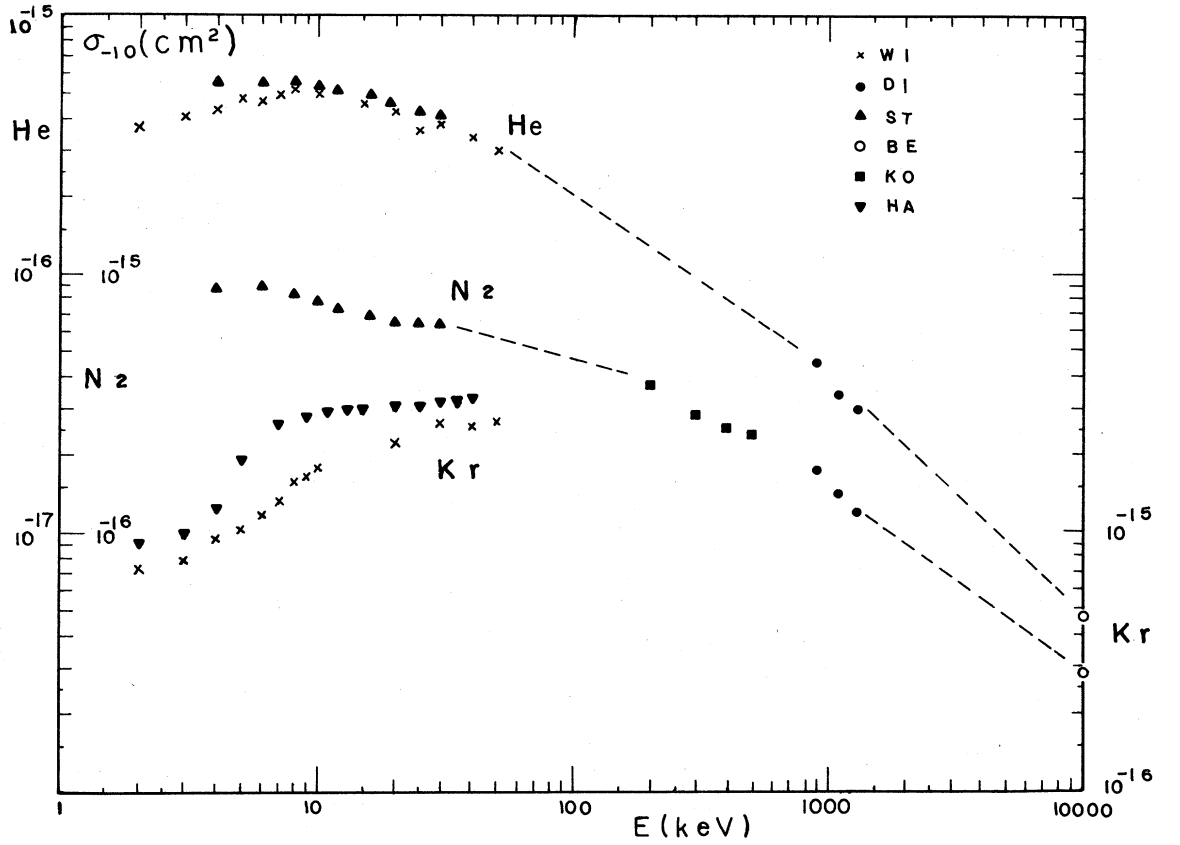

FIG. 23. Single electron loss by a negative hydrogen ion in He, N_2, and Kr. WI: Williams (1967b). DI: Dimov and Dudnikov (1967). KO: Kovacs (1967). BE: Berkner *et al.* (1964). ST: Stier and Barnett (1956). HA: Hasted and Stedeford (1955). The result of Berkner *et al.* is obtained with 20-MeV deuterons.

2. The maximum cross section for charge change is large when the energy defect ΔE is small.

(3) The maximum cross section is also large when the ionization potential of the target atom is low.

From these trends, alkali metal vapors are thought to be better choices as the targets in charge change processes. Donnally *et al.* (1964) first proved these facts in studies of the production of metastable hydrogen atoms by charge change using Cs vapor. One of the most attractive experiments on charge change in alkali vapors was done by Donnally and Becker (1967) who obtained an intense negative hydrogen ion beam corresponding to 10 percent of the protons incident, which is larger than that for ordinary gases by a factor of 10.

Since then, experiments on alkali-metal vapors have been carried out at various laboratories. However, many of these studies are made for such practical purposes as the production of intense negative ions for tandem accelerators. Therefore, basic cross section data for charge change by alkali atoms are rather scanty. Cross sections have been published by Schmelzbach *et al.* (1968a, b) (σ_{10} and σ_{1-1} for K vapor), Il'in *et al.* (1965, 1967) ($\sigma_{10}+2\sigma_{1-1}$ for Li, Na, K, Cs, and Mg vapors), Schlachter *et al.* (1969) (σ_{10}, σ_{01}, σ_{0-1}, σ_{1-1} in Cs), Dyachkov (1969) (σ_{10} and σ_{01} in Li) and Grüebler *et al.* (1970) (σ_{10} and σ_{1-1} in Li, Na, K, and Cs). However, some of these results differ by about one order of magnitude (see Fig. 35), although the results on Li by Il'in *et al.* are in reasonably good agreement with those calculated by Nikolaev (1967). Reasons for the experimental discrepancies are not clear. They may be partly due to errors in the measurement of the density of the alkali vapors. For example, Il'in *et al.* estimated the target vapor density from the chamber temperature using the known vapor pressure of the alkali metal at the temperature. In other cases, surface ionization detectors were used. Data on charge change of K vapor by Nieman *et al.* (1967) (σ_{10} and σ_{01}) have not yet been published in detail.

Bohlen *et al.* (1968) measured the ratio of negative hydrogen ion to proton beam at the optimum target thickness in K and Cs vapors in the energy range 0.5–2 keV. They found K vapor preferable to Cs for

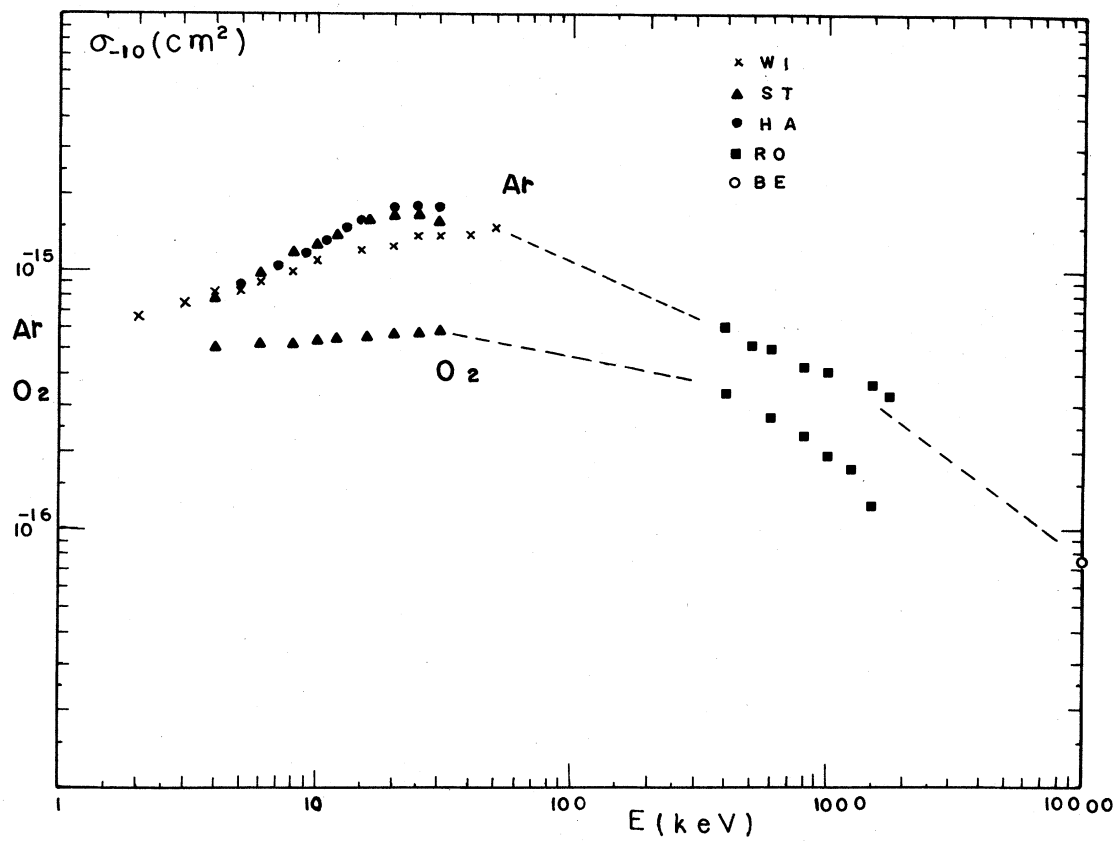

FIG. 24. Single electron loss by a negative hydrogen ion in O_2 and Ar. WI: Williams (1967b). ST: Stier and Barnett (1956). HA: Hasted and Stedeford (1955). RO: Rose *et al.* (1958). BE: Berkner *et al.* (1964). The result of Berkner *et al.* is obtained with 20-MeV deuterons.

the production of negative hydrogen ions, because the cross sections for the production of negative ions for H_2^+ and H_3^+ in K vapor were smaller than those in Cs vapor. They found this to be true despite the fact that the negative hydrogen ion–proton ratio in K was nearly equal to or slightly smaller than that for Cs in the energy range investigated.

In the experiments of both Bohlen *et al.* (1968) and Schmelzbach *et al.* (1968a) it was found that the maximum of the negative ion component is not identical with that in the charge equilibrium. This may be due to the effect of some beam components in metastable states, as has been found in He and Ne beams. As shown in Figs. 32–35, the charge change cross sections for protons are relatively high at low energies and decrease very rapidly with increasing energy. Above 20–50 keV, the cross section decreases slowly with increasing energy. In these respects, data for both alkali vapors and inert gases show similar trends.

Charge change at low energies involves mainly the weakly bound outer electron of the alkali atom and capture into the excited states is fairly probable; thus the capture probability of an outer electron of an alkali atom decreases with energy. On the other hand, it is apparent that electrons in the inner shell of an alkali atom (corresponding to the outer shell of an inert gas) play a great role in the charge change at high energy.

Recently Sellin and Granoff (1969) published results on proton–alkali vapor (Cs, K, and Ru) collisions in the energy range up to 50 keV.

Dyachkov and Zinenko (1968) investigated production ratios of neutral and negative hydrogen beams in Li, K, Mg, and Zn metal vapors from 5–40-keV protons. Zinc vapor could produce stronger neutral and negative ions than did other targets. Also Berkner *et al.* (1969) measured the charge changing cross sections (σ_{10}, σ_{01}, σ_{0-1}, and σ_{-10}) of 5–70-keV hydrogen beams in Mg vapor (Fig. 36). They cited results by Futch and Moses on the charge change of 4–45-keV protons in Mg vapor. In these measurements, the estimate of the vapor pressure of target metal in the collision region is a most difficult point.

Data on charge change for alkali or alkaline-earth metal vapors must be accumulated for practical reasons

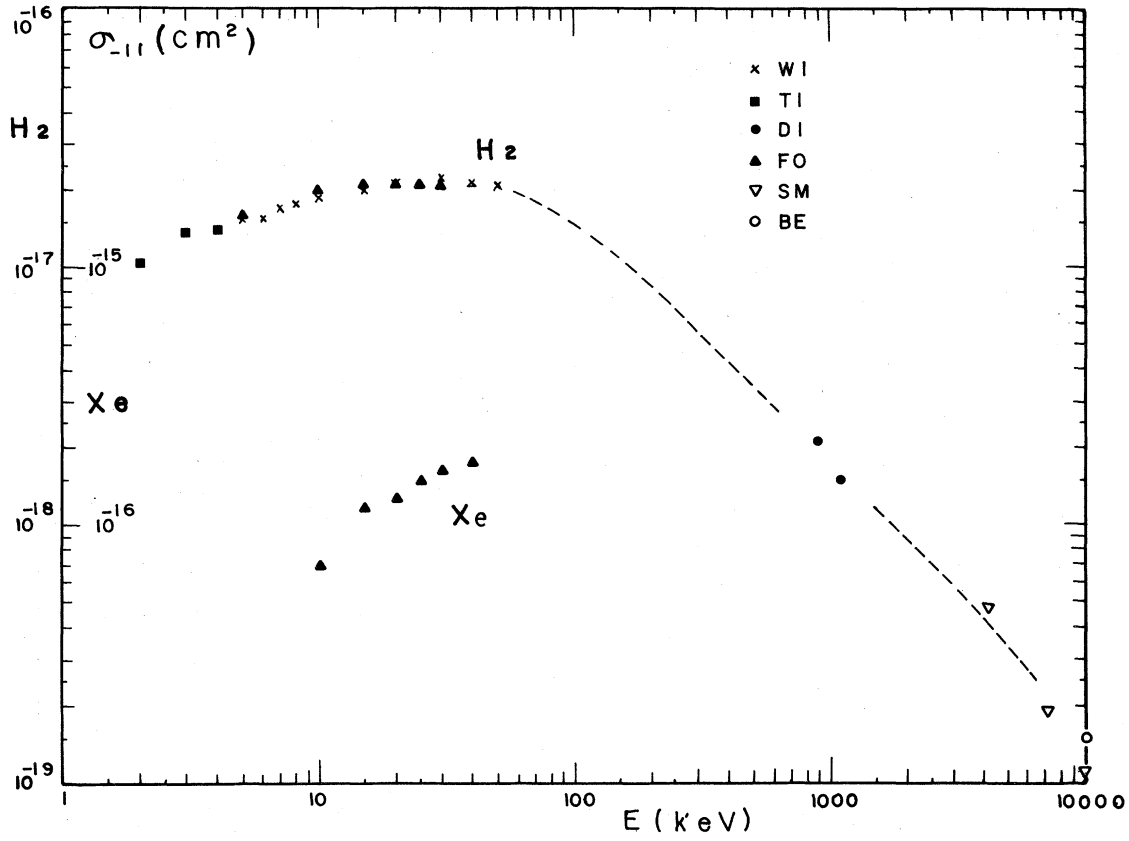

FIG. 25. Double electron loss by a negative hydrogen ion in H_2 and Xe. WI: Williams (1967b). TI: Tisone (1964), Tisone and Branscomb (1964). DI: Dimov and Dudnikov (1967). FO: Fogel *et al.* (1957). SM: Smythe and Toevs (1965). BE: Berkner *et al.* (1964). The result of Berkner *et al.* is obtained with 20-MeV deuterons.

such as the production of negative ion beams for tandem machines and for basic studies of the charge changing process.

7. THEORY

7.1 General Comments

Inasmuch as this review is intended to be primarily experimental in outlook, much of the theoretical section will be organized along the lines of the individual processes, such as, for example, charge exchange in protons on hydrogen. This first part of the section, however, is devoted to a general discussion of those theoretical concepts needed for discussing the individual processes taken up in the remainder.

In discussing the theoretical aspects of charge changing collisions, it is important distinguish between ionization and charge exchange processes. Charge exchange is an example of a rearrangement collision (i.e., one in which the colliding systems transfer at least one component particle from one system to the other). Such processes exhibit theoretical difficulties, particularly in the Born approximation, not found in simple excitation or ionization events. This difficulty will be discussed in greater detail in Sec. 7–2 below. In treating ionization from collisions involving many electron systems, it is important to include not only direct ionization, but also excitation to multiply excited (also called autoionizing) states which later ionize by an autoionization transition. These will then show up experimentally as ions just as if they had been ionized directly. The only way direct ionization and excitation to autoionizing states can be differentiated experimentally is by inelastic energy loss measurements and the energy spectrum of emitted electrons.

In general, progress has been made along three lines of approach: the Born approximation, the quantal impulse approximation, and the binary encounter approximation. These approaches will be outlined briefly, in order to provide a framework for the survey to follow:

A. The Born Approximation

The Born approximation is a formulation of a collision process which takes advantage of the fact that

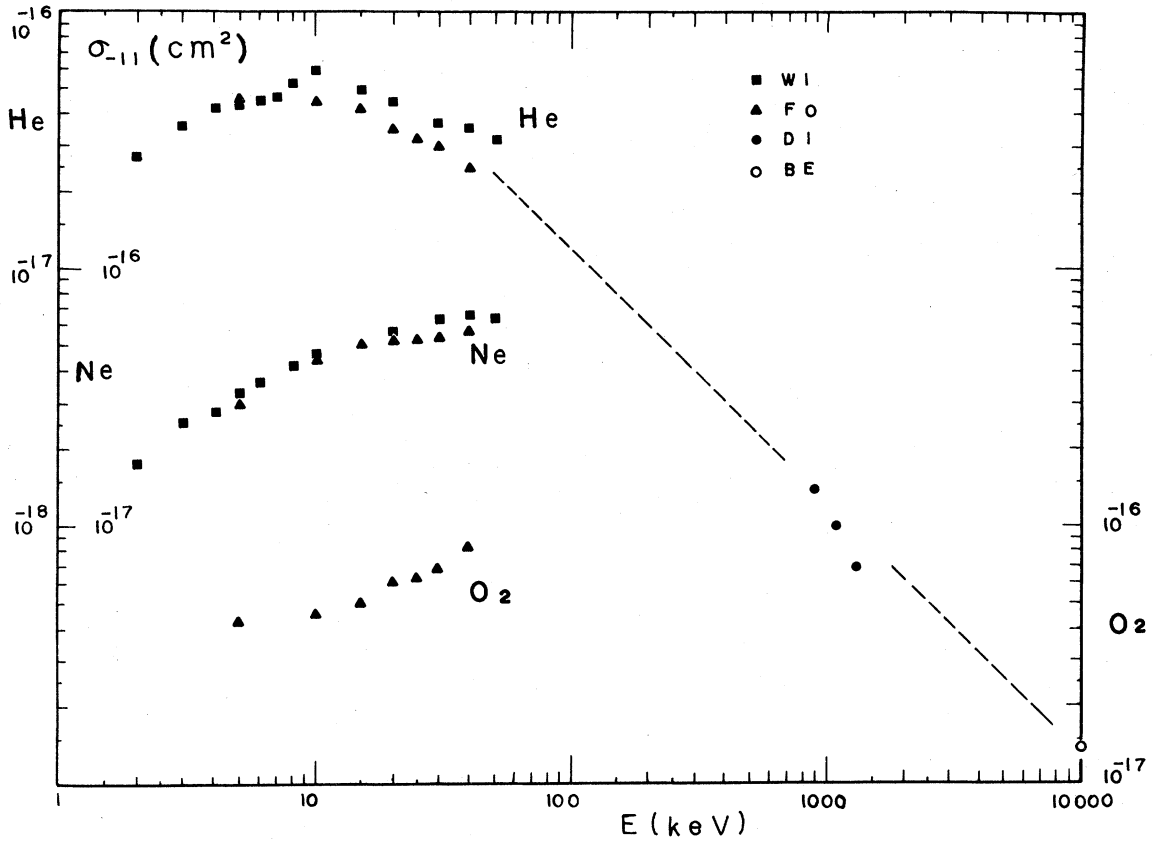

FIG. 26. Double electron loss by a negative hydrogen ion in He, O_2, and Ne. WI: Williams (1967b). FO: Fogel *et al.* (1957). DI: Dimov and Dudinkov (1967). BE: Berkner *et al.* (1964). The result of Berkner *et al.* is obtained with 20-MeV deuterons.

in a collision between two systems at high energy, the systems do not have sufficient time to interact strongly with each other. Labeling the two systems A and B, the Hamiltonian can be written in the form

$$H=H_0+V, \tag{69}$$

where

$$H_0=T_R+H_A+H_B, \tag{70}$$

$$T_R=-(\hbar^2/2\mu)\nabla_R^2. \tag{71}$$

Here, H_A is the Hamiltonian for the isolated system A in a reference frame at rest with respect to the center-of-mass of system A; similarly, H_B is the Hamiltonian describing system B in a frame at rest with respect to the center-of-mass of system B. The term T_R describes the kinetic energy of translation of one system relative to the other; $\mathbf{R}$ is the relative separation between the two centers of mass, and μ is the reduced mass. Finally, V includes those terms in the over-all Hamiltonian thus far omitted; the potential energies of components of system A interacting with components of system B. It can then be shown that the differential cross section for the scattering of a system A by system B accompanied by a transition of the internal structures of A and B from initial state Ψ_I to final state Ψ_F is given by (Mott and Massey, 1949)

$$d\sigma/d\Omega=(\mu/2\pi\hbar^2)^2(K_F/K_I)\mid M_{FI}\mid^2, \tag{72}$$

where $\hbar\mathbf{K}_I$ and $\hbar\mathbf{K}_F$ are the initial and final relative momenta of the two systems, and M_{FI} is the matrix element

$$M_{FI}=\langle\exp(i\mathbf{K}_F\cdot\mathbf{R})\Psi_F\mid V\mid\exp(i\mathbf{K}_I\cdot\mathbf{R})\Psi_I\rangle. \tag{73}$$

So long as no rearrangement occurs between any components of A or B, the application of the Born formulation to high-energy collisions is straightforward. Thus, for example, in such processes as excitation or ionization, the Born approximation presents no difficulties. It is to be noted that simple ionization is not a rearrangement process. The ionized electron is generally moving slowly with respect to its original nucleus and continues to be treated in its original reference frame.

On the other hand, the charge exchange process is

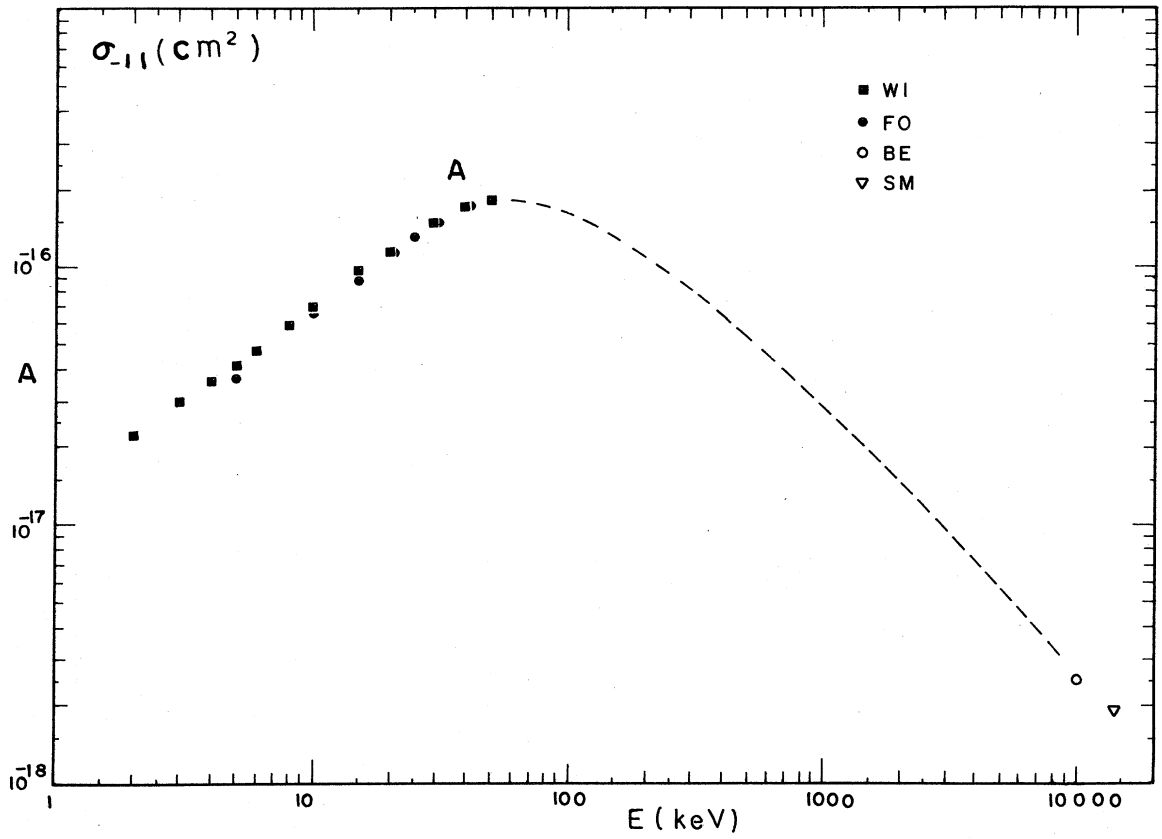

FIG. 27. Double electron loss by a negative hydrogen ion in Ar. WI: Williams (1967b). FO: Fogel *et al.* (1957). BE: Berkner *et al.* (1964). SM: Smythe and Toevs (1965). The result of Berkner *et al.* is obtained with 20-MeV deuterons.

a rearrangement collision. An electron originally in one system ends up in the other, in violation of the conditions under which the Born expression for the cross section was derived. Nevertheless, the Born approach, slightly modified, has been used without rigorous justification. In these rearrangement processes, the Hamiltonian is decomposed in two different ways:

$$H=T_I+H_A+H_B+V_I \tag{74}$$

$$H=T_F+H_C+H_D+V_F. \tag{75}$$

The initial state Ψ_I is taken to be an eigenstate of H_A+H_B while Ψ_F is an eigenstate of H_C+H_D. Clearly, there is an ambiguity as to whether to use V_I or V_F in the Born matrix element (73). This is called the post-prior discrepancy.

B. The Quantal Impulse Approximation

The Born approximation is the lowest order approximation to a rigorous quantum mechanical formulation of the scattering problem. In the present connection, this rigorous formulation has been applied to the electron exchange process in order to take into account multiple collisions of the exchanged electron which, it is believed, are important in the transfer process. A rigorous expression for the differential electron exchange cross section is given by (Goldberger and Watson, 1964)

$$d\sigma/d\Omega=(\mu/2\pi\hbar^2)^2(K_F/K_I)\mid R_{FI}^{+}\mid^2. \tag{76}$$

This agrees with expression (72), except for the matrix element which has been shown to be

$$R_{FI}^{+}=\langle\exp(i\mathbf{K}_F\cdot\mathbf{R})\Psi_F\mid V_F\mid\exp(i\mathbf{K}_I\cdot\mathbf{R})\Psi_I\rangle + \lim_{\epsilon\to 0+}\langle\exp(i\mathbf{K}_F\cdot\mathbf{R})\Psi_F\mid V_F\mid G_{\epsilon+}V_I\exp(i\mathbf{K}_I\cdot\mathbf{R})\Psi_I\rangle. \tag{77}$$

Here $G_{\epsilon+}$ is the Green's Function

$$G_{\epsilon+}=(E-H+i\epsilon)^{-1}. \tag{78}$$

It is seen that without the second term on the right-hand side, Eq. (77) reduces to the Born matrix element (73). A complete evaluation of the second term in the matrix element (77) proves to be neither tractable nor desirable. Only a selected subset of all possible

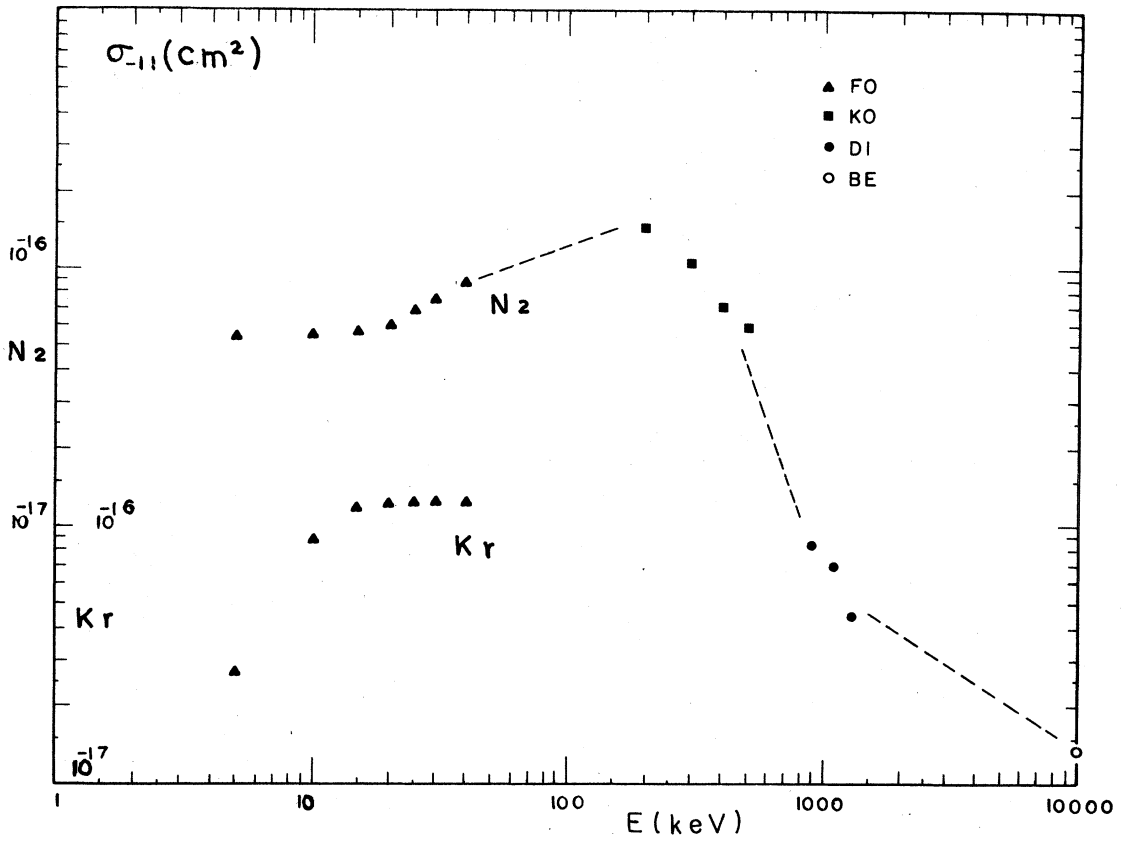

FIG. 28. Double electron loss by a negative hydrogen ion in N_2 and Kr. FO: Fogel *et al.* (1957). KO: Kovacs (1967). DI: Dimov and Dudnikov (1967). BE: Berkner *et al.* (1964). The result of Berkner *et al.* is obtained with 20-MeV deuterons.

multiple interactions are believed to be consequential to the cross section. Thus, the quantal impulse approximation neglects all those terms responsible for binding during the collision; hence the term "impulse approximation."

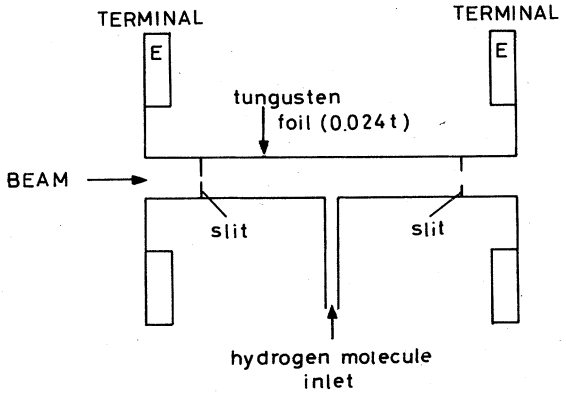

FIG. 29. A collision chamber for atomic hydrogen with a directly heated tungsten furnace. McClure (1966).

C. The Binary Encounter Approximation

In the binary encounter approximation, the atomic electrons and the nucleus of each system are all considered to act as independent scattering centers. Vriens (1969) presents an excellent summary of the binary encounter model. Each constituent particle of system A is assumed to scatter each constituent particle of system B via their mutual Coulomb interaction exactly as if the other components were not present. That is, the mutual interaction between components in the same system are neglected during the collision. This assumption would seem to be justifiable only if the effective interaction between each colliding pair of particles takes place in a volume small compared to that of the atom. This, of course, implies a momentum transfer to a component large compared to its initial momentum, which, in turn, implies that the energy transferred to the component is large compared with its binding energy. The only effect of the mutual interaction between components in the same system is to produce the initial velocity distribution of those components at the instant *before* the collision.

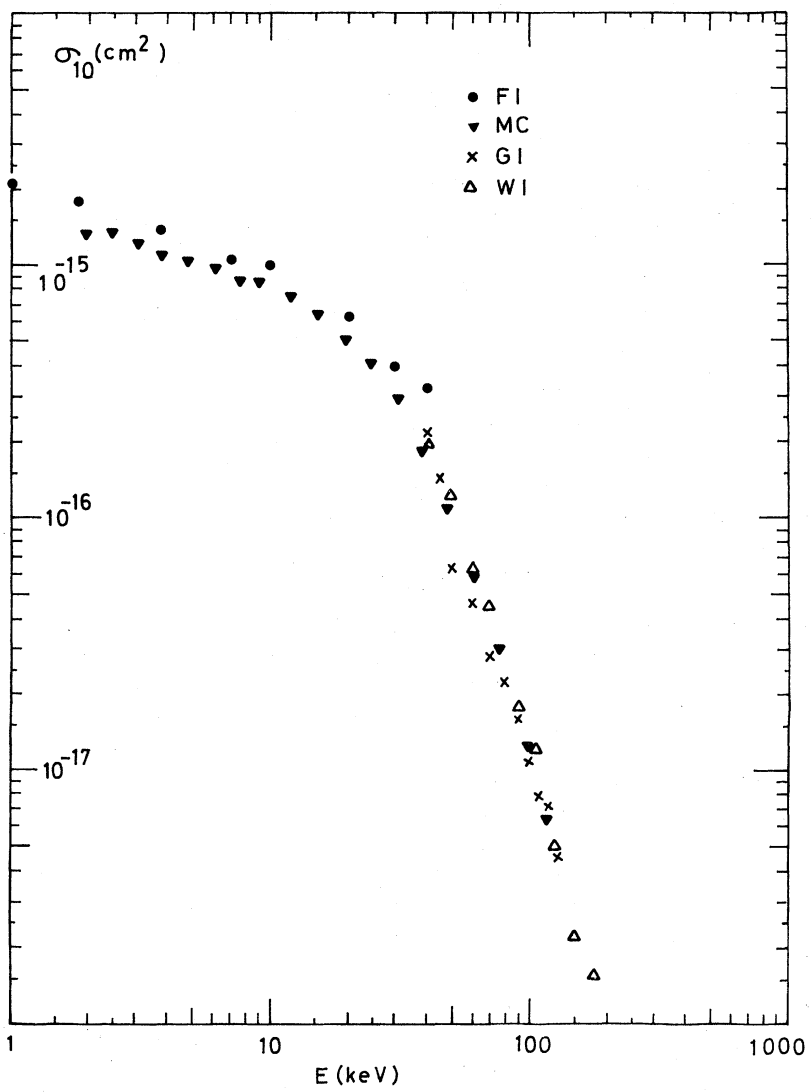

FIG. 30. Single electron capture by a proton in atomic hydrogen. FI: Fite *et al.* (1962). MC: McClure (1966). GI: Gilbody and Ryding (1966). WI: Wittkower *et al.* (1966).

What is needed to execute the binary encounter approximation for the cross section for a given process is as follows:

1. The momentum transfer differential cross section for two particles under the influence of a Coulomb force interaction. This is well known, but a little too complicated to be given here in the form in which it is needed, inasmuch as both particles have initial velocities and the relative velocity of each colliding pair is not the same as the relative velocity of the center-of-mass of system A relative to that of system B. (See Vriens, 1969.)

2. The initial velocity distribution in both system A and system B.

3. A model which attempts to describe the desired process in terms of some subset of two-body collisions. For example, the ionization cross section would be determined from the two-body cross sections which transfer to any electron an energy greater than its ionization energy.

D. Comparison of the Various Approximations

Having taken up separately the Born, the quantal impulse, and the binary encounter approximations, it is worthwhile to compare the merits of these approaches.

For any process in which the binary encounter approach is applicable, it is considerably the easiest to carry out. On the other hand, it must be remembered that for excitation and ionization processes at least, the Born is a rigorous quantum mechanical approximation, whereas the binary encounter approach is a semiquantum mechanical model. Nevertheless, when computing a given cross section for a comparison with experiment, the quantum mechanical rigor inherent in the Born approximation can be lost if not all the

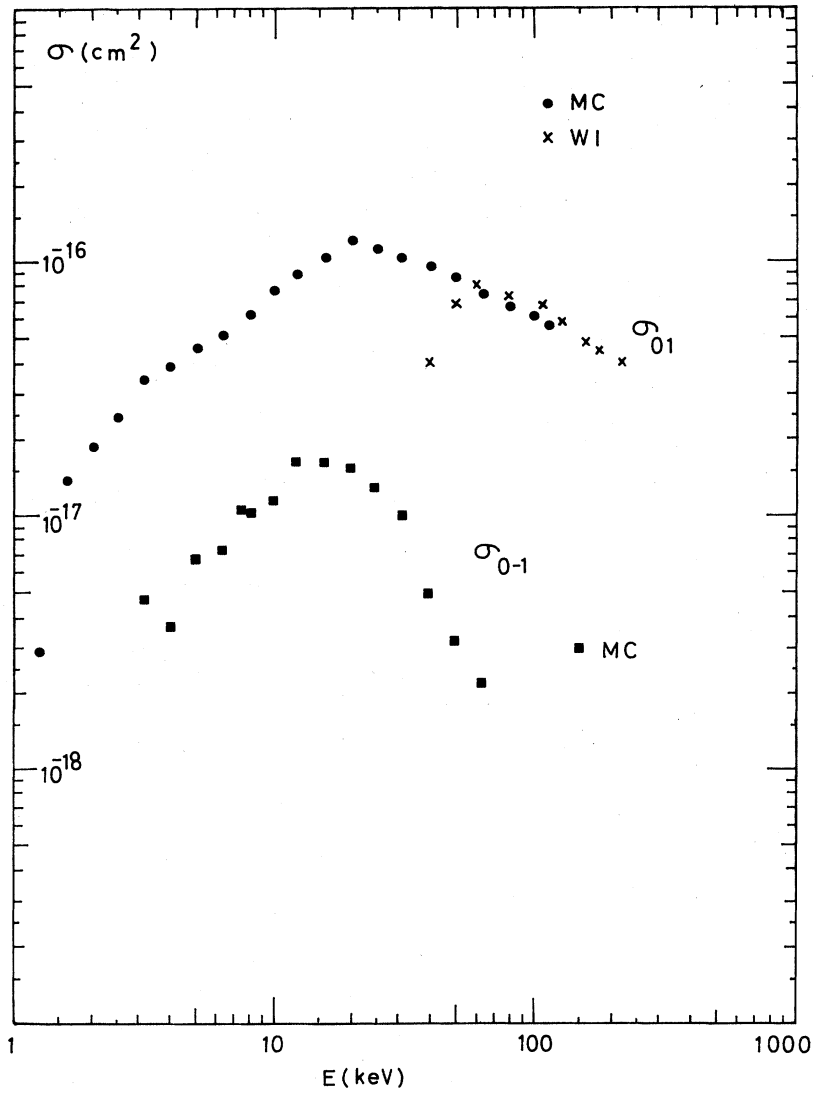

FIG. 31. Single electron capture and loss by a hydrogen atom in atomic hydrogen. MC: McClure (1968a, b). WI: Wittkower *et al.* (1967b).

final states which make up the measured cross section are taken into account in the theoretical calculation. Thus, for example, when considering the ionization cross section for the target atom, all possible final states of the projectile must be taken into account. Sum rules can often be a great help in this connection. Insofar as the binary encounter approach is concerned, the main insight required in the model to describe the process has already been pointed out to be the identification of the subset of two-body collisions which most nearly describes the final state.

None of the methods take into account any distortion of the two colliding systems when they are in close proximity. Thus, all molecular effects are ignored. These molecular effects are important in low-energy collisions, but negligible in high-energy collisions, so that all the approaches are suitable only for high-energy collisions. Beyond this, in order to be valid, the binary encounter model also requires a large momentum transfer, because, in effect, it integrates over intensities rather than amplitudes and thereby loses the phase interference inherent in the Born approximation. These phase interference effects are believed to be small for close (i.e., high momentum transfer) binary impacts.

When it comes to charge exchange phenomena, the Born approximation loses its rigorous justification, so that both approaches must be used with caution. Indeed, the binary enounter model may well be better for such processes if, as suspected (See Sec. 7.2 below), three-body interactions are involved in the charge exchange process. In this connection, the quantal impulse approximation has been employed to take into account quantum mechanically those three-body interactions which have been used in the binary encounter model for the charge exchange process.

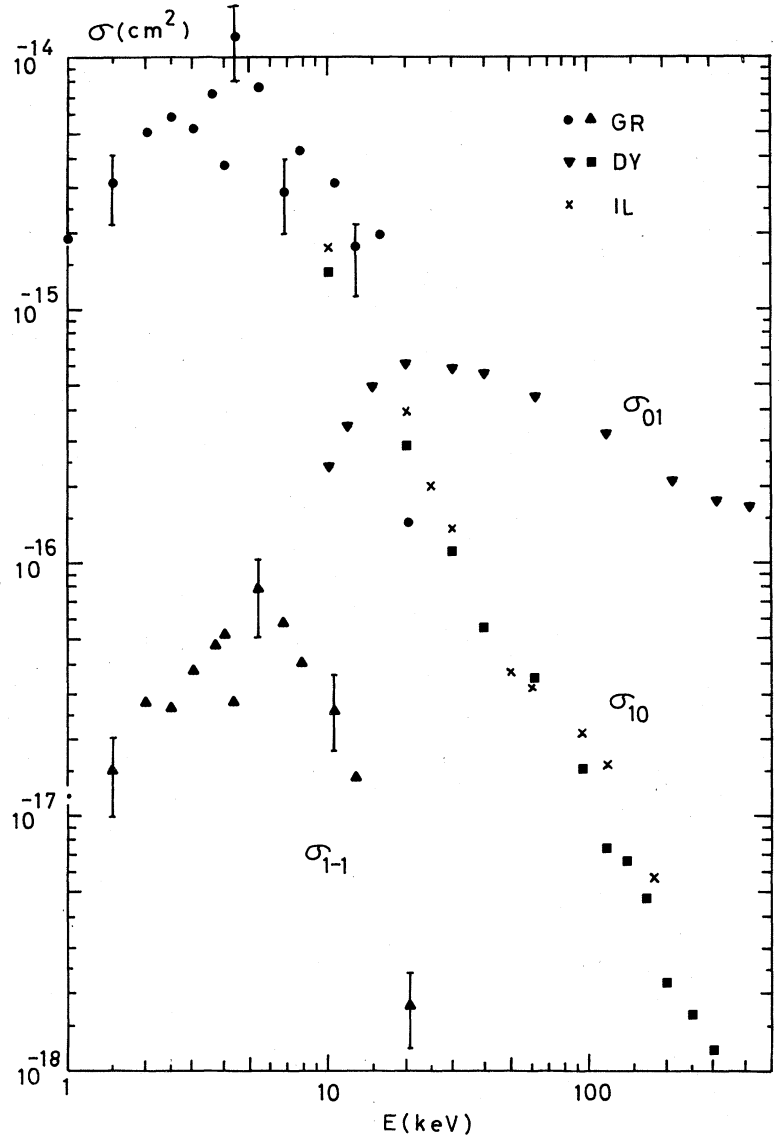

FIG. 32. Single and double electron capture by a proton in Li and single electron loss by a hydrogen atom in Li. GR: Grüebler *et al.* (1970). DY: Dyachkov (1969). IL: Il'in *et al.* (1965, 1967).

7.2 Electron Capture by a Proton

A. The Quantum Theoretic Approach

This is a true charge exchange process. As such, the application of the Born formalism is complicated by the effects of the rearrangement, often referred to as the post-prior discrepancy. The simplest case to consider theoretically is that of electron capture by a proton incident on a hydrogen atom and, understandably, this has received the most detailed and intensive treatment. Early theoretical work applying the Born approximation to this prototype problem was undertaken by Oppenheimer (1928), Brinkman and Kramers (1930), and Massey and Smith (1933). More recent work was done by Bates and Dalgarno (1952, 1953) and Jackson and Schiff (1953). Later work bringing the more powerful tools of formal scattering theory to bear on the problem was done by Pradhan (1957), Bassel and Gerjuoy (1960), McDowell (1961), and Cheshire (1962). Some aspects of the energy dependence of the cross section were re-examined by Mapleton (1964) and Coleman and McDowell (1964).

Oppenheimer demonstrated that at high incident velocity capture occurs almost entirely to s states and that the probability of capture into a state of principal quantum number n varies as n^{-3}

$$\sigma_n = \sigma_1/n^3, \tag{79}$$

where σ_n is the partial cross section for capture into a state of principal quantum number n.

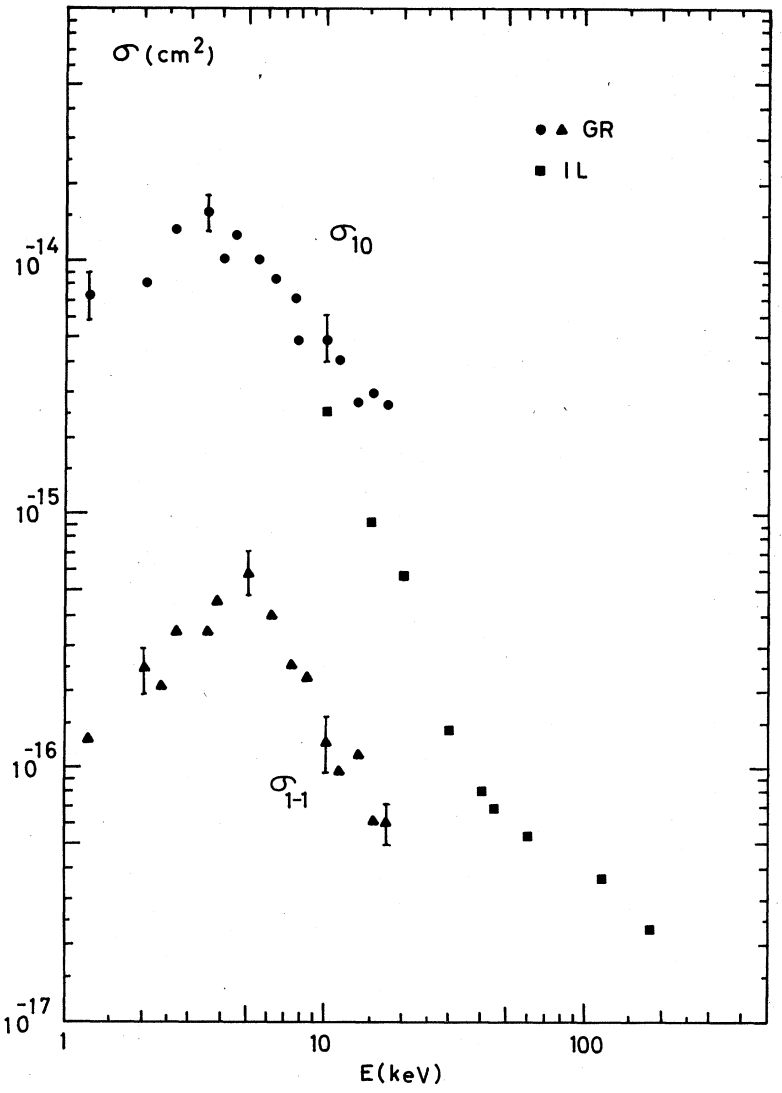

FIG. 33. Single and double electron capture by a proton in Na. GR: Grüebler *et al.* (1970). IL: Il'in *et al.* (1965, 1967).

The early work of Oppenheimer (1928), Brinkman and Kramers (1930), and Massey and Smith (1933) considered only the electron-incident proton interaction as being responsible for the capture transition in the Born approximation, neglecting the proton–proton term. This latter term would, on intuitive grounds, not be expected to be involved in the transition of the electron. However, as Bohr (1948) pointed out, the capture process is a true three body problem (as opposed to excitation and ionization processes which are essentially two body problems). Taking up this suggestion, Jackson and Schiff (1953) included the proton–proton as well as electron–proton interactions in the Born cross section calculation. They found that inclusion of the previously neglected term has a remarkable effect on the value obtained for the cross section, reducing it by a factor of 5 in the high-energy limit from the Brinkman–Kramers result. The Jackson–Schiff value is in excellent agreement with the experimental results derived from protons in molecular hydrogen in the moderate energy region below 200 keV, when due account is taken of electron capture into higher Rydberg states ($n \geq 2$). Working concurrently, Bates and Dalgarno (1952, 1953) achieved identical results and, more correctly pointed out the reason for the substantial reduction in cross section from that obtained by Brinkman and Kramers due to the proton–proton interaction term. It has, in reality, to do with the fact that the approximate initial and final wave functions used in the calculation are not completely orthogonal as required in the derivation of the formula for the Born cross section. Thus, the Jackson and Schiff-Bates and Dalgarno result does not in fact provide a three-body mechanism for the

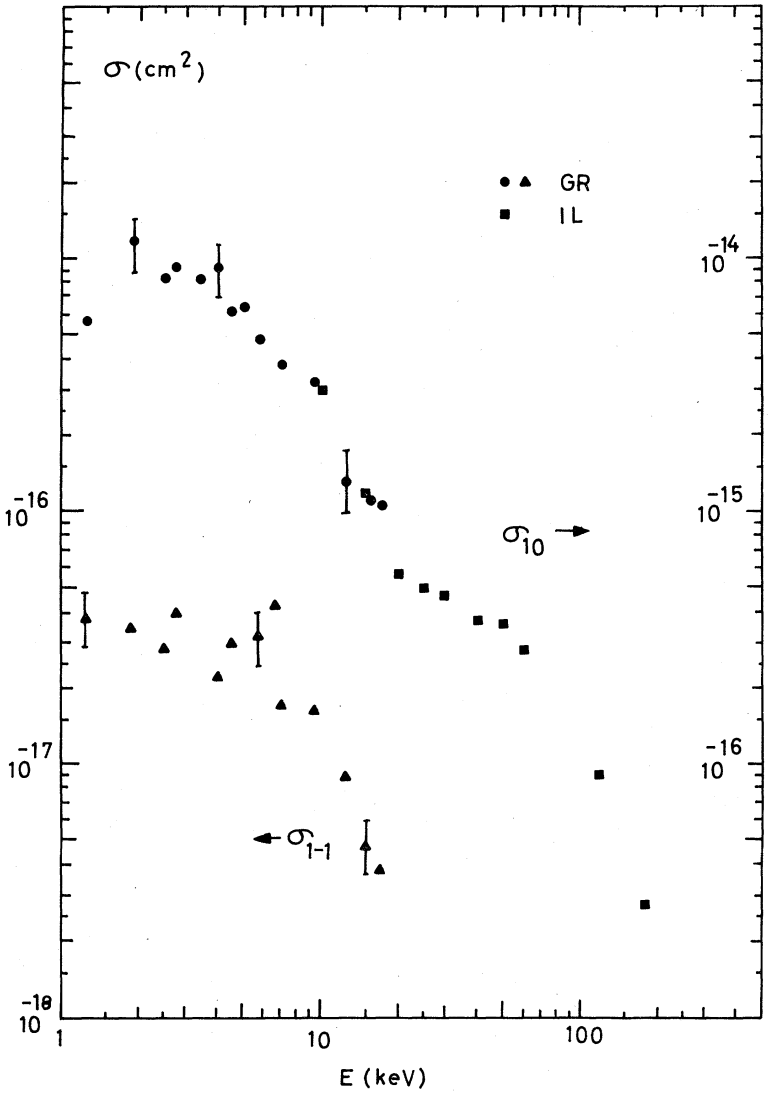

FIG. 34. Single and double electron capture by a proton in K. GR: Grüebler *et al.* (1970). IL: Il'in *et al.* (1965, 1967).

capture process. Parenthetically it might be mentioned that in the course of their calculation, Jackson and Schiff showed that for the case of the electron transfer from one proton to another, there is no ambiguity in the final result whether the post or prior interactions are used in the formulation of Born cross section calculation.

The more recent quantal impulse calculations (McDowell, 1961, Cheshire, 1962), which were designed to incorporate three-body effects in the collision, do not significantly alter agreement with experiment in the energy region below 200 keV, but show a substantial effect above 200 keV.

All of the theoretical results described above, which were calculated for protons incident on hydrogen, are, unfortunately, compared with experimental results for protons incident on molecular hydrogen, the assumption being that a hydrogen molecule acts as two isolated hydrogen atoms. Tuan and Gerjuoy (1960), however, have shown that this is not the case; the differences between molecular hydrogen and two isolated hydrogen atoms can well have as great an effect on the cross section as the differences between the various theoretical calculations.

A subsequent re-examination of problem in the very high-energy limit (above 100 MeV) by Mapleton (1964) showed that a previously overlooked back scattering contribution to the cross section becomes dominant in the high-energy limit, making the cross section depend on energy as E^{-3} instead of the E^{-6} dependence obtained by Bates and Dalgarno and Jackson and Schiff. This contribution was confirmed in the impulse approximation by Coleman and McDowell (1964).

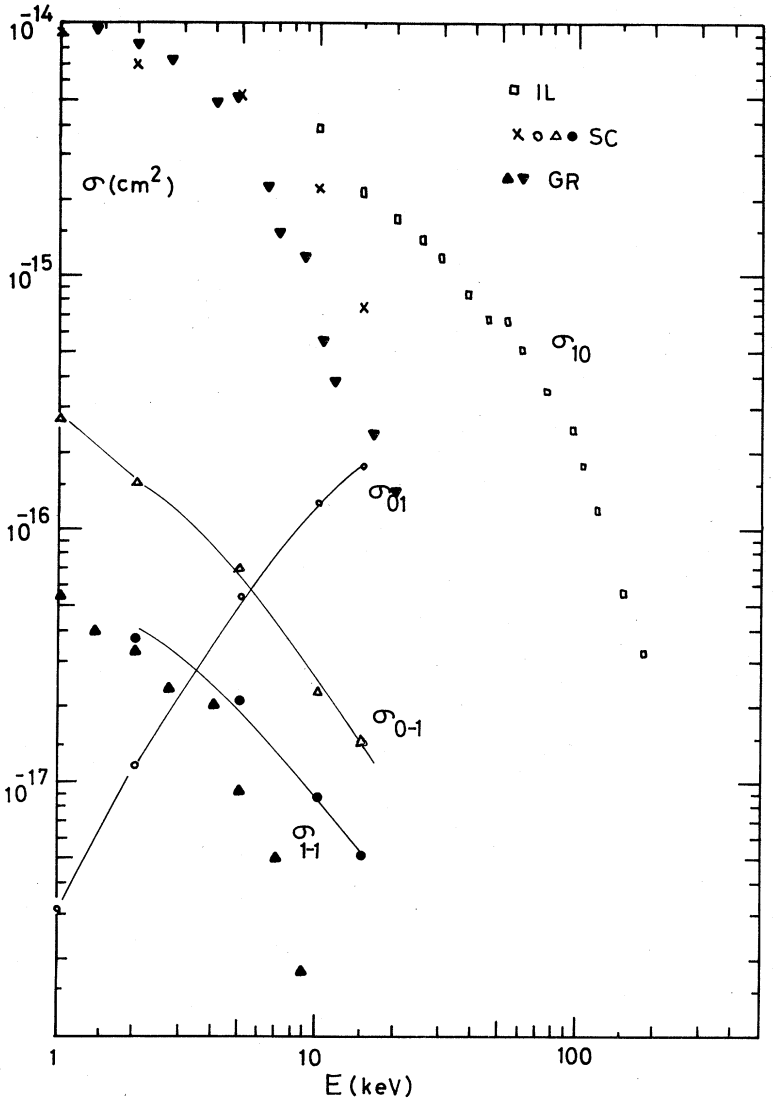

FIG. 35. Single and double electron capture by a proton, single electron loss and single electron capture by a neutral hydrogen atom in Cs vapor. IL: Il'in *et al.* (1965, 1967). (total electron capture cross section). SC: Schlachter *et al.* (1969). GR: Grübler *et al.* (1970). Data at 1 keV were deduced from a 2-keV deuteron beam.

It must be realized, however, that this back-scattering component in the cross section will *not* be seen in an experimental arrangement. Experiment looks only at charge exchange which occurs to those protons scattered in a finite angular window about scattering angle $\theta=0°$. The theoretical quantity which should be compared with experiment is the forward-scattered charge exchange component plus the backward-scattered non-charge exchange component. (Remember that the amplitudes of the two matrix elements must be added before squaring and integrating.) This combination was not presented because the authors were concerned only with the cross section as mathematically defined. It is clear, however, that a theoretical calculation of the experimentally measured quantity is needed if a meaningful comparison with future experiments in the high-energy limit is to be attempted.

B. The Classical Approach

Classical calculations on the charge exchange process date back to the work of Thomas (1927a, b, c). The model he used consisted of two binary encounters. The first collision, that between the incoming proton (proton A) and the electron bound to proton B, scatters the electron. The second collision is between the scattered electron and proton B. If this second collision is just right the electron will end up with a final velocity relative to the incoming proton A which corresponds to an energy less than the ionization energy. It will therefore be bound to proton A. It is important to note that in the strict classical approach, the first collision, by itself, could not capture. A double collision is absolutely necessary.

After Thomas, the classical approach remained

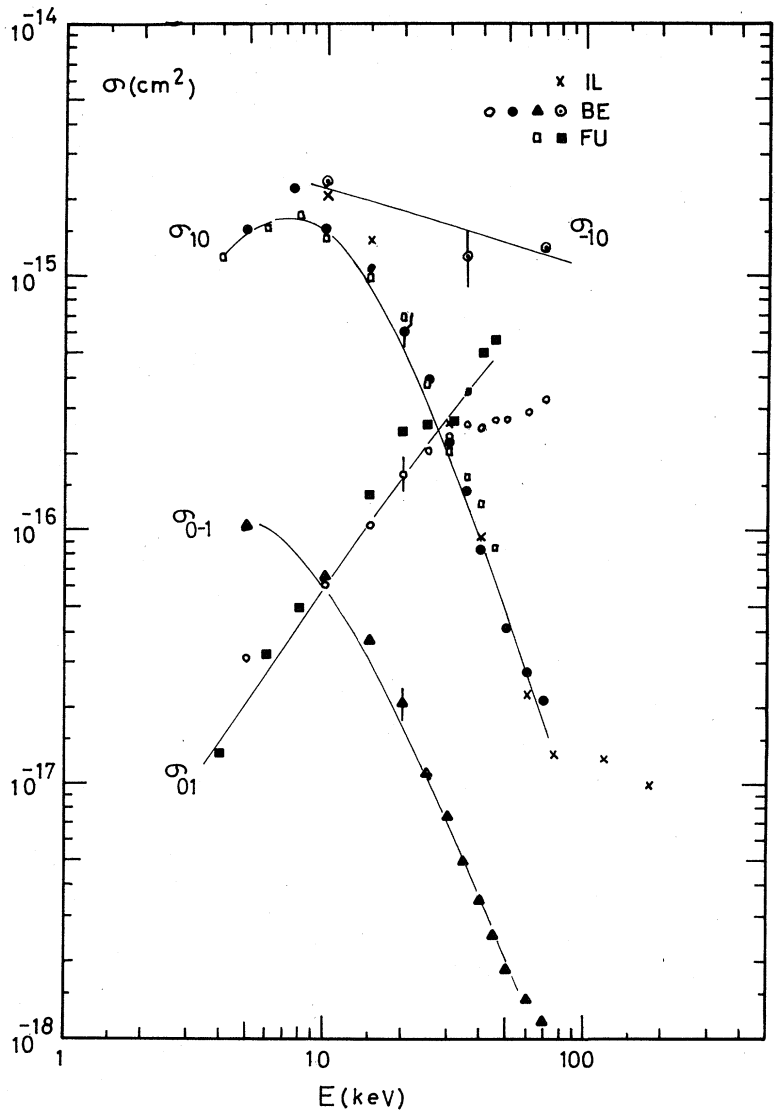

FIG. 36. Charge change by a hydrogen atom in Mg vapor. IL: Il'in *et al.* (1965, 1967). BE: Berkner *et al.* (1969). FU: Futch and Moses (cf. Berkner *et al.*, 1969).

dormant until re-introduced by Gryzinski (1957). In a series of papers, Gryzinski (1965a, b, c) considered, among many other processes, that of charge exchange. Gryzinski's model for charge exchange consists of a single collision (in violation of the precept noted above) in which the incoming proton A, with velocity v_A, transfers an amount of energy ΔE to the electron within the limits

$$\tfrac{1}{2}mv_A^2 \leq \Delta E \leq \tfrac{1}{2}mv_A^2 + 2I_H, \tag{80}$$

where m is the electron mass, and I_H is the ionization energy.

Subsequently Bates and Mapleton (1965) considered charge transfer in the high-energy limit using the classical approach of Thomas. They found that the cross section for charge exchange in this approach also follows the E^{-3} dependence on incident energy found in the quantum calculations of Mapleton (1964) and Coleman and McDowell (1964).

A different classical approach was followed in a series of papers by Abrines and Percival (1964, 1966a, b). These authors used the classical approach, but not the binary encounter approximation. They included all interactions in a Monte Carlo calculation. Agreement with experiment for this approach proved to be about as good as that obtained by Jackson and Schiff.

C. Discussion

The theoretical situation, insofar as charge exchange is concerned, is far from satisfactory. Claims have been made that the capture process is truly a three-body interaction, yet at least one classical two-body model has been suggested and the Born approximation continues to be used. Three-body interactions

incorporated in the quantal impulse approximation do not make a really substantial difference from the Born approximation. The various authors do not confront the contradictions that appear in the published literature; instead they ignore these contradictions and merely compare the predictions which follow from their models with experimental data. The data consists of protons incident on molecular hydrogen, and the applicability of these results to the case of protons incident on atomic hydrogen is itself highly suspect.

7.3 Ionization Processes

A. General Comments

Unlike charge exchange processes, ionization processes present no particular difficulties. The Born approximation is valid at sufficiently high incident energies and the classical models, which are quite easy to formulate in the binary encounter approach, should yield close approximations to the Born results.

B. Ionization of Atomic Hydrogen

Born approximation calculations were carried out by Bates and Griffing (1953) for the following processes:

$$H^+ + H(1s) \rightarrow H^+ + H^+ + e \quad (81)$$

$$H(1s) + H(1s) \rightarrow H^+ + H(1s) + e. \quad (82)$$

Classical calculations on the process (81) were carried out by Gryzinski (1965c) with the binary encounter approach and by Abrines and Percival (1966b) in the Monte Carlo approach. Subsequently, Abrines, Percival, and Valentine (1966) did a binary encounter calculation, comparing the results thus obtained with those following from the Monte Carlo calculation.

C. Collisional Detachment of the Negative Hydrogen Ion

Collision processes which remove the second electron from H^- to form neutral hydrogen could hardly be called ionization processes and are therefore termed electron detachment processes. A classical impulse approximation treatment of the processes

$$H^- + (H, He, Ne, Ar, Kr, Xe) \rightarrow H + (H, He, Ne, Ar, Kr, Xe) + e \quad (83)$$

was carried out by Bates and Walker (1967). Exploratory quantal calculations on this subject were done by Sida (1955) and McDowell and Peach (1959).

8. DISCUSSION AND CONCLUSION

Although the accumulation of data on charge changing processes for hydrogen beams incident on inert gases has not yet been as comprehensive as we would like, many experiments on hydrogen beams have been done.

Agreement between theoretical calculations and experimental results has been reported to be good in some cases (Mapleton, 1959). However, data on negative hydrogen ion beams are scarce, mainly because of limited facilities available for acceleration of the negative ion to high energy. Also, scatter in the values of the cross sections sometimes are beyond experimental uncertainties.

Looking to the future, though, three stage tandem accelerators (Van de Graaff, 1960), which can accelerate negative ions up to a few MeV, have recently become available, so that it is now possible to study the charge changing processes of the negative ion in such high energy regions.

In many experimental analyses of charge change, a diatomic molecular gas such as hydrogen, nitrogen, or oxygen is assumed to be equal to two free atoms in order to make comparisons with theoretical calculations. Some experiments have been done directly on atomic hydrogen, although these are restricted to a limited energy range and for only a few processes. Beyond these, no experiments using atomic nitrogen or oxygen as target atom have been made, except for a recent measurement of σ_{-10} in atomic oxygen by Snow *et al.* (1969) in a limited energy range (not reported here). Works on atomic hydrogen are concerned mainly with single electron capture by a proton or single electron loss by a hydrogen atom below an energy range of a few hundred keV. As shown by Wittkower *et al.* (1966) the ratio of the cross section for atomic hydrogen to that for molecular hydrogen is constant [0.42 ± 0.03, compared with the theoretical estimate of 0.41 (Tuan and Gerjuoy, 1960)] for energies over 100 keV, showing that molecular hydrogen may not simply be assumed to be two free hydrogen atoms.

From a practical point of view alkali vapors as target gases will be useful in the production of intense beams of neutral atoms or of negative ions at a desired energy. Alkali atoms have one electron in their outermost shell which easily interacts with energetic projectiles. Therefore, the cross section for electron capture is expected to have a large value. Actually, as shown in Sec. 6.8, these alkali atoms have large cross sections down to relatively low energies and, as a consequence, are often used as target atoms in the production of negative ions. It is reported that a Cs vapor target can produce He negative ions with an intensity about two orders of magnitude higher than that which can be obtained from inert gases (Massey, 1949). This behavior is also found in the processes exhibited by a hydrogen beam, as mentioned in Sec. 6.8.

However, in some applications, such as the injection of a negative ion into tandem accelerators, an appropriate energy of the negative ion is required so as to match the ion optics of the accelerator. Thus, even though a target atom may have a large cross section, it can often not be used properly for such purposes, if the large cross section obtains at a beam energy

which does not match with the energy required for beam transmission through the ion optics. The loading effect on the accelerator becomes severe in such cases.

Fortunately, various alkali-metal vapors have different characteristics; that is, the maximum cross sections for charge change occur at different energies, depending on the alkali vapor. Therefore, a proper alkali-metal vapor can be chosen in accordance with experimental requirements.

Since the maximum cross sections for production of negative He ions occur at 2 and 17 keV for Cs and K, respectively, it is expected that the cross section might occur at even higher energy for Li vapor (the ionization potentials of these alkali vapors are 3.87, 4.32, and 5.36 eV, respectively). Lithium also has the following advantages, compared with other alkalis:

(1) it has lower Z and, therefore, multiple scattering effects are very small,
(2) it is less active and is therefore easy to handle.

As Middleton and Adams (1968) pointed out, in the production of negative He ions, the use of Li vapor as charge changer improved the transmission of the beam through the tandem accelerator and the loading effect of the accelerator became very small. The same thing is expected to be valid in the case of a hydrogen beam.

Oparin *et al.* (1967) investigated the yield of highly excited atoms in Mg, Ca, Zn, and Cd vapors and concluded that Mg is the most effective target. Wells *et al.* (1969) recently utilized alkaline-earth metals (Mg, Ca, Ba, and Sr,) as charge changing vapors and reported them to be superior to ordinary gases in negative He ion production. As mentioned before, the charge changing processes are requisite in some thermonuclear research facilities. For example, PHOENIX (Kuo *et al.*, 1964) a research apparatus at Culham Laboratory, and ALICE (Post, 1958) at Lawrence Radiation Laboratory require neutral beams for injection into the magnetic bottle to heat the confined plasma. In these cases, neutral beams are produced either by charge capture or by dissociation of positive molecular ions. In such a thermonuclear apparatus, the leakage of the neutral target gas into the plasma chamber must be restricted to be as small as possible. For this reason ordinary gases such as hydrogen or He are not adequate. As an example, hydrogen gas was used in the early stages of PHOENIX. It was abandoned, however, and replaced by H_2O, C_8H_{16}, or CO_2 vapor (Geller and Prevot, 1954; Borovik, 1967), which are easily condensed and trapped by liquid nitrogen and, therefore, can be kept away from the main plasma chamber. Recently, it has been reported that a magnesium vapor or jet (Butusov, 1967, 1968) is even more favorable than the gases mentioned above as charge neutralizer. Unfortunately, there are few measurements of the cross section for charge capture in magnesium vapor (Sellin and Granoff, 1969; Futch and Moses).

On the other hand, in DCX (Luce, 1958) at Oak Ridge National Laboratory, research is progressing toward finding effective dissociators of a molecular hydrogen beam with simultaneous small charge changing (electron capture) cross section. At Oak Ridge National Laboratory, a carbon arc was first used as dissociator. However, the loss of the neutral beam component due to charge capture by the carbon arc was very severe. Therefore, many other arcs, for example, H_2, D_2, Li, or Ar are being developed with small charge changing cross section. A theoretical estimate (Obedkov and Parlov, 1967) which can be compared with experiments, is reported on the charge changing cross section of a proton in collision with a lithium arc. However, basic data on the cross sections for charge change with arcs have not yet been reported except in one work by Bogdanov *et al.* (1965). Again in accelerator application, the use of water vapor as charge changer has been reported (Roos, 1965). Nearly 100% transmission of the incident beam was obtained by using a large aperture in the charge change canal in the tandem high voltage terminal. This was possible because the water vapor can be easily condensed on liquid nitrogen traps. However, the cross section itself for water vapor has not been measured except by Toburen *et al.* (1968a) who measured the cross section for single and double electron capture by a proton in the energy ranges 100–2500 keV and 75–250 keV, respectively.

A mercury vapor or jet is often used as the charge changer in heavy-ion linear accelerators (Beringer and Rall, 1957) and negative ion beam injectors (Fogel *et al.*, 1960, Dawton, 1961). Mercury vapor can be condensed on a liquid nitrogen trap and it also has pumping action, thus giving it some superiority to the usual gases. But it has a high atomic number. Therefore, scattering in the charge changing process becomes significant, especially at low energies. In such cases, mercury has to be ruled out.

Organic gases such as C_3H_8 are interesting possibilities as target gases although they are not reported on here. Data on organic gases are very limited, except for those by Farrokhi (1966) and Toburen *et al.* (1968a, b). Farrokhi measured the cross section for double electron capture by a proton in such gases as CH_4, C_2H_6, C_3H_8 and C_4H_{10} in the energy range of 10–50 keV and found some structure of the cross section which could be explained by Massey's adiabatic theory. Toburen *et al.* also determined the cross sections for single and double electron catpure by a proton in CH_4, C_2H_2, C_2H_6, and C_4H_{10} in the energy range of 50–2500 keV. Dimov and Dudnikov (1967) measured the cross sections for the charge change of hydrogen beams (σ_{01}, σ_{-10} and σ_{-11}) for C_3H_8 at energies of around 1 MeV and found that the cross sections for C_3H_8 were about

one order of magnitude larger than those for usual gases such as hydrogen or helium.

Other inorganic gases such as SF_4, SF_6, or CCl_2F_2 are expected to have large electron loss cross section because they are electronegative. According to the measurement by Dimov and Dudnikov, SF_4 gas has cross sections over one order of magnitude larger than do the inert gases. However, no other measurements for such gases are reported. It is hoped that more measurements in more target gases and in metal vapors will be done in the future. From these, the dependence of the cross sections on the target elements will become empirically clear. Then, the calculation or estimation of the cross sections for complex molecular gases should become possible.

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