

# ANALYSIS AND MODELLING OF THE DATA ON CHARGE EXCHANGE CROSS SECTIONS



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ANALYSIS AND MODELLING OF THE DATA ON CHARGE EXCHANGE CROSS SECTIONS • • •

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#### SUMMARY

Section 1 presents a brief survey of the available experimental data on charge exchange cross sections, including the most recent work on resonant processes.

In Section-2 the dependence of the electron capture cross section on the relative velocity, the degree of ionization and isoelectronic sequence of the projectile, and the atomic number of the target atom is discussed in phenomenological terms.

In the high energy region one may fit  $\sigma$  - v curves with a power law or a B.K.-type function. Examples of both kinds of fit are given, as well as the results of an attempt to scale empirical curves with respect to  $\sigma$  and v. Curve-fitting in the adiabatic region is also considered.

A correlation between the capture cross section and the electron affinity I of the ion is observed to hold at fixed velocity. The dependence  $I^{3/2}$  seems to be valid at high velocities, with capture into the L shell being more probable than capture into the K shell. Further periodic effects at lower velocities are indicated.

The influence of the resonance defect on the cross section is discussed for endothermic and exothermic reactions. The concept of effective

number of capturable target electrons is considered in view of the experimental evidence.  $\overline{\Box}$ 

Section 3 is devoted to empirical evaluations of the existing models for computing charge exchange cross sections,  $\nu \tau \nu \tau = 0 \tau \nu \tau = \eta \tau v$ .

The substantial data on the differential capture cross section for several cases of resonance is interpreted in terms of the capture probability and compared with the results of the usual adiabatic model. Deviations from the expected oscillatory behavior, attributed to excitation and ionization processes, are found in the capture probability, but may not be too important in the total charge exchange cross section. Several models of non-resonant reactions are reviewed and compared with empirical results.

The high energy classical models of Bohr and Lindhard, Bell and Gluckstern, and Thomas are described. Only Thomas' model gives a rapid enough decrease with velocity, indicating that the classical picture of the capture process given by the first two sets of authors is inadequate.

In Section 4 the conclusions of this study are presented, with sug-

#### SECTION 1

#### THE EXPERIMENTAL DATA

The data pertaining to charge transfer reactions has been surveyed most recently by Allison and Garcia-Munoz.<sup>(1)</sup> Further bibliographic material is given in the article of Hasted.<sup>(2)</sup> A brief review of the considerable data on electron capture available at present will form an introduction to the empirical and theoretical treatment of this data.

#### 1.1 SYMMETRICAL RESONANCE

Bibliographies for the earlier work on resonant reactions as well as cross section curves versus velocity are given by Hasted<sup>(3)</sup> and Rapp and Francis.<sup>(4)</sup> Table 1 is a summary of data, including recent work not mentioned in the above articles, along with relevant velocity ranges.

Since the He-He resonant cross section is known from velocities of  $2 \times 10^6$  cm sec<sup>-1</sup> to  $8.5 \times 10^8$  cm sec<sup>-1</sup>, it is an ideal case for comparing theory with experiment. Both the adiabatic and high energy regions are covered by the data, and there is reasonable agreement between independent measurements.

DATA SUMMARY

Resonance Reaction	Velocity Range (units 10 <sup>7</sup> cm sec <sup>-1</sup> )	References
H	2.8-28	Fite <u>et al.</u> <sup>(5)</sup>
	0.6-6.0	Fite, Smith and Stebbings <sup>(6)</sup>
He	22-46	Allison, Cuevas and Murphy <sup>(7)</sup>
	6-31	Barnett and Stier <sup>(8)</sup>
	0.7-4.2	Gustafsson and Lindholm <sup>(9)</sup>
	40-80	Nikolaev et al. <sup>(10)</sup>
	31-85	Pivovar, Tubaev and Novikov <sup>(11)</sup>
	22-44	Gilbody <u>et al</u> . <sup>(12)</sup>
0	0.22-3.5	Stebbings, Smith and Ehrhardt <sup>(13)</sup>
Ne	2.4-5.4	Fedorenko, Flaks and Filippenko <sup>(14)</sup>
	0.16-0,94	Gustafsson and Lindholm <sup>(9)</sup>
	7.6-20	Gilbody <u>et al.</u> (12)
Na	0.35-1.3	Bukhteev and Bydin <sup>(15)</sup>
A	0.05-0.68	Kushnir, Palyukh and Sena <sup>(16)</sup>
	1.5-3.4	Sluyters, De Haas and Kistemaker (17)
	0.11-0.65	Gustafsson and Lindholm <sup>(19)</sup>
	2.6-4.1	Nikolaev <u>et al.</u> (10)
	5.4-14	Gilbody et al. <sup>(12)</sup>
	0.44-1.2	Neff <sup>(18)</sup>
ĸ	0.05-0.68	Kushnir, Palyukh and Sena <sup>(16)</sup>
	0.45-1.4	Bydin and Bukhteev <sup>(19)</sup>
	1.5-3.8	Chkuaseli et al. <sup>(20)</sup>

TABLE 1 (continued)

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Kr	0.04-0.48	Kushnir, Palyukh and Sena <sup>(16)</sup>
	0.075-0.45	Gustafsson and Lindholm <sup>(9)</sup>
	4.8-7.3	Gilbody <u>et_al.</u> (12)
ወኤ	1 0-2 6	Chkuranali at al (21)
RD	1.0-2.0	$\frac{et at}{(15)}$
	0,15-0,55	Bukhteev and Bydin <sup>(13)</sup>
Xe	0,03-0,38	Kushnir, Palyukh and Sena <sup>(16)</sup>
Cs	0,03-0,38	Kushnir, Palyukh and Sena <sup>(16)</sup>
	0.2-0.7	Bukhteev and Bydin <sup>(15)</sup>
	0.12-0.54	Kushnir and Buchma <sup>(22)</sup>
	0.8-1.9	Chkuaseli <u>et al</u> . <sup>(21)</sup>
	0.07-0.8	Marino, Smith and Caplinger <sup>(23)</sup>
Hg	0.025-0.32	Kushnir, Palyukh and Sena <sup>(16)</sup>

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#### 1.2 NON-RESONANCE

Not only are simple electron exchange reactions to be included under the category of non-resonant processes, but also single and multiple electron capture by ions in high charge states. A majority of the experimental work on non-resonant reactions has involved hydrogen and rare gas ions incident on gaseous targets. There have been some recent exceptions to the use of permanent gas targets; sodium, calcium, potassium, rubidium, and cesium vapors have been employed by several investigators.

One can discuss separately the data in the near-adiabatic region, i.e., for velocities close to the velocity of maximum cross section  $v_m$ and the high energy region  $v >> v_m$ .

1.2.1 <u>Near-Adiabatic Region</u>. The value of  $v_m$  is important for tests of the Massey criterion; hence, much data has been taken in the near-adiabatic region. Hydrogen and helium ion data are very good in this region; e.g., Stedeford and Hasted,<sup>(24)</sup> Gilbody and Hasted,<sup>(25)</sup> and Allison, Guevas and Garcia-Munoz<sup>(26)</sup> show typical behavior of cross sections for  $v \simeq v_m$ .

There is evidence from the data in diatomic target gases that anomalous behavior implies a failure of the Massey criterion. Stebbings, Smith and Ehrhardt<sup>(27)</sup> present evidence of near-resonant dissociative reactions in the (He<sup>+</sup>, O<sub>2</sub>) and (He<sup>+</sup>, N<sub>2</sub>) charge exchange cross sections. If the total cross section contains a separate dissociative component, then inferences about v<sub>m</sub> can only be made on the individual component reactions.

1.2.2 <u>High Energy Region</u>. When the relative velocity becomes equal to or greater than orbital velocities, we will speak of the high energy region. A convenient unit is the Bohr velocity

$$v_o = \frac{e^2}{\hbar}$$
 (= 2.2 x 10<sup>8</sup> cm sec<sup>-1</sup>).

The data on H and He is very good. For hydrogen the work of Barnett and Reynolds <sup>(28)</sup> involves velocities up to  $6v_0$ . For helium ions Barnett and Stier<sup>(8)</sup> give  $\sigma_{10}$  up to  $1.4v_0$  while Pivovar, Tubaev and Novikov<sup>(11)</sup> bring the data up to  $4v_0$ . The latter authors (Pivovar, Novikov and Tubaev<sup>(29)</sup>) also give the single and double electron capture cross sections for He<sup>2+</sup> up to  $3.5v_0$ .

An important contribution to the high energy data is the work of Nikolaev <u>et al.</u>, <sup>(10,30)</sup> In the velocity range  $1.2v_o < v < 5.5v_o$  they studied the impact of various ions on helium, nitrogen, argon, and krypton targets. The  $\sigma$  - v curves for He, Li, B and N ions in different charge states were obtained, and data on Be, C, O, Ne, Na, Mg, Al, P, A, and Kr ions at selected velocities were also taken. Because of this mass of data, certain regularities can be detected which will be developed below. These are also the only data with ions in high charge states (up to septuply ionized).

#### SECTION 2

#### ANALYSIS OF THE DATA

#### 2.1 GENERAL DESCRIPTION OF VARIATIONS

The one electron capture cross section can be regarded as a function of four independent parameters:

(1) the relative velocity v;

(2) the ionic charge i;

(3) the number of electrons  $N_e$  belonging to the ion (or some identifying parameter for a given ioselectronic sequence);

(4) the atomic number  $Z_a$  of the target atom.

Molecular targets will clearly involve other parameters. We will discuss briefly the general nature of the functional dependence on these parameters as shown by the existing data.

The velocity dependence for resonance reactions is simple: the charge exchange cross section increases monotonically with decreasing velocity. However, in non-resonant reactions the cross section increas.  $\sim$ with decreasing v until a maximum is reached at velocity v<sub>m</sub>. As v



continues to decrease, o also decreases. When some diatomic gases are used as targets, two maxima are observed.

For most reactions the logarithmic derivative  $-\frac{d \ln \sigma}{d \ln v}$  is a monotonically increasing function of v. However, there is evidence from the work of Barnett and Reynolds<sup>(28)</sup> and Nikolaev <u>et al</u>.<sup>(10)</sup> that the logarithmic derivative achieves a maximum for  $v \gtrsim 10^9$  cm sec<sup>-1</sup> in heavy target gases such as argon and krypton. Since this effect seems to be related to the structure of the target atom, it will be discussed further below.

For an incident ion in charge state i the cross section  $\sigma_{i,i-1}$  describes single electron capture into state i-1. At high energies the capture cross section always increases for more highly charged ions. However, in the near-adiabatic region this rule may fail, as one sees in the cases Li<sup>2+</sup> in He (Allison, Cuevas and Garcia-Munoz)<sup>(26)</sup> and Xe<sup>4+</sup> in Ne (Ogurtson and Flaks).<sup>(31)</sup> The dependence on i will thus depend on the velocity.

One might expect that the charge exchange cross section would depend on the electronic structure of the incident ion in a way consistent with its position in the periodic table. Such periodicity seems to be borne out by the data of Nikolaev <u>et al.</u>;<sup>(10)</sup> plots of  $\sigma$  versus atomic number of the ion at fixed velocity show definite structure effects. The same periodicity effects appear in data of Ormrod and Duckworth,<sup>(32)</sup> who measured electronic stopping cross sections in carbon films. The

anomalously low cross sections of Li, Na, and Mg ions at  $v = 9 \times 10^7$  cm sec<sup>-1</sup> may be related to the low value of the charge exchange cross sections of those elements in carbon.

To what extent these periodicities are correlated to basic atomic parameters will be discussed below. There is evidence that periodic effects are associated with the most weakly bound electron and disappear for highly charged ions.

As a general rule the charge transfer cross section per atom increases with an increase in  $Z_a$ . This rule is particularly true at high energies as shown in data of Pivovar, Novikov and Tubaev. <sup>(29)</sup> The work of Nikolaev <u>et al.</u>, <sup>(10,30)</sup> also reveals this effect. However, there are exceptions, particularly in the adiabatic and near-adiabatic regions. It is reasonable to suppose that the increase of the cross section is related to the increased number of electrons in the target which can be captured.

#### 2.2 SEMI-EMPIRICAL METHODS

2.2.1 <u>Velocity Function Fits: High Energy Region</u>. In the high energy region it has often been assumed that the data could be fitted by a power law; i.e.,  $\sigma \propto v^{-n}$ . Barnett and Reynolds<sup>(28)</sup> estimated for H<sup>+</sup> the exponents to be n = 6.5 in nitrogen and n = 3.7 in argon. For He<sup>2+</sup> Rutherford<sup>(33)</sup> estimated n = 6 in nitrogen and Pivovar, Novikov and Tubaev<sup>(29)</sup> concluded that n = 6.5 for nitrogen, n = 6.3 for argon and n = 4.8 for krypton targets.

Exponents have been determined from the available data for various ions in helium. Table 2 lists 5 isoelectronic sequences with the parameters n and  $Q_0$  in the semiempirical formula

$$\sigma = Q_o (v/v_o)^{-n} \qquad (1)$$

Although fits to a power law can be made, one is using data at the upper end of the velocity range. If one wishes to extend the fit down to lower velocities, an additional parameter must be utilized. There are several functional types which exhibit the correct behavior in the high energy region. For example, the exponential fit  $\sigma \propto \exp \left[-(v/v')\right]$  was suggested by Stier and Barnett.<sup>(34)</sup> However, the exponential functional function will not join to a power law fit at higher velocities.

Therefore, an attempt was made to construct a semiempirical function based on the BK functions (Bates and McCarroll).<sup>(35)</sup> The simple analytic form

$$\sigma = Q_1 [1 + (v/v_1)^2]^{-n}$$
 (2)

was chosen. It was found that the (H<sup>+</sup>, He) cross section calculated by Mapleton<sup>(36)</sup> could be well approximated in the range  $1.4v_o \le v \le 6v_o$  by

$$\overline{Q}_{c} = \pi a_{o}^{2} \frac{10.3}{\left[1 + \left(\frac{v}{2v_{o}}\right)^{2}\right]^{5}}$$

where  $\pi a_{0}^{2} = 0.88 A^{02}$ .

Ion	Ne	Q <sub>o</sub>	n.
		(units 10 <sup>-14</sup> cm <sup>2</sup> /atom)	
н+	0	2.23	8.4
He <sup>2+</sup>	0	1.26	5.8
Li <sup>3+</sup>	0	5.18	6.0
в <sup>5+</sup>	0	55	6.95
He <sup>+</sup>	1	0.081	6.8
Li <sup>2+</sup>	1	148	8.75
в <sup>4+</sup>	1	23	6.75
.N <sup>6+</sup>	1	55	6.5
в <sup>3+</sup>	2	1.53	5.5
N2+	2	41	6.7
в <sup>2+</sup>	3	0.65	5.1
N <sup>4+</sup>	3	44	7.05
N <sup>3+</sup>	4	5.27	6.1

POWER LAW FITS IN HELIUM

TABLE 2

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In Table 3 are listed the parameters  $Q_1$ ,  $v_1$ , and n for selected capture cross sections in the high energy region. These values were obtained by fitting curves on log-log paper by eye. Consequently, there is a certain amount of subjective judgment in the fits, and the values are meant to be suggestive only.

The original Brinkman-Kramers result would predict n = 6, but the value of n in formula (2) is not a sensitive parameter in fitting. The fact that different values of n gave equally good fits for the same empirical curves led to the decision to use integral values only.

The smooth variation of cross section with target gas suggested that a scaling procedure might be developed to predict the cross sections of ions in one target gas by data taken in another gas. In order to investigate this possibility, all data for a given isoelectronic sequence were plotted together for a given target gas. Then data from different targets were compared.

The results can be expressed in the ratios  $\alpha_{ZZ'}$ ,  $\gamma_{ZZ'}$  such that

$$\sigma_{Z} (\alpha_{ZZ}, v) = \gamma_{ZZ}, \sigma_{Z}, (v) , \qquad (3)$$

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where Z and Z' are the atomic numbers of the compared targets. Table 4 gives selected ratios obtained by matching high energy data in various target gases. Omissions indicate bad comparisons or lack of data. Most comparisons involved simply an adjustment of a single scale, rather than both at once, since there seemed to be less subjective error in one-parameter scaling.

TABLE	3
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BK FUNCTION FITS

Ion	Target Gas	Q <sub>1</sub>	v	n
		(units A <sup>2</sup> /atom)	(units 10 <sup>8</sup> cm sec <sup>-1</sup> )	
H+	н <sub>2</sub>	11,2	4.6	7
	He	7.2	5 <b>.2</b>	6
	N <sub>2</sub>	7.6	5.9	6
	A	24	5.05	6
Het	He	5.2	4.15	4
	N <sub>2</sub>	5.0	5.6	5
He <sup>2+</sup>	<sup>H</sup> 2	35	5.0	7
	He	26	4.4	4
	<sup>N</sup> 2	13.7	5.5	4
	A.	40	5.5	5

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	Z	Ζ/	Ne	α <sub>zz</sub> ′	γ <sub>ZZ</sub> ,	
4- <u>1-4</u> -1	1(H <sub>2</sub> )	2(He)	0	0.825	1.0	
	. °		0 1 1	0.78 0.83 1.0	1.85 1.0 0.455	
	2	7 (N <sub>2</sub> )	0 0 1 1 2 2	0.885 1.0 0.90 1.0 0.83 1.0	1.0 0.555 1.0 0.61 1.0 0.59	
	7	18	0 0 1 1	0.93 1.0 1.15 0.96 1.0	1.0 0.785 0.30 1.0 0.72 <sup>+</sup>	
	2	18	0 0 1 1	0.86 1.0 0.895 1.0	1.0 0.53 1.0 0.475	
	18	32	0 0 1 1	0.93 1.0 0.92 1.0	1.0 0.75 1.0 0.695	

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TABLE 4

SCALING RATIOS

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\* σ taken per atom

poor match

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Adiabatic Region. It is again necessary to separate resonance and nonresonance reactions. For symmetrical resonance the formula

$$\sigma^{1/2} = A - B \ln v \tag{4}$$

has ample theoretical justification (Firsov, (37) Dalgarno(38)). This formula has been used by Popescu and Ionescu(39,40) to fit data from different sources. Since A and B also have theoretical expressions, such fitting allows a check on the theoretical basis of the formula.

In nonresonant reactions Hasted (41) attempted to fit some of his data to a curve of the form

$$\sigma \propto \exp\left[-\frac{K}{v}\right] . \tag{5}$$

In Table 5 the values of K given by Hasted are expressed in velocity units.

2.2.2 <u>Constant Velocity Fits.</u> In an effort to correlate periodic effects in the charge exchange cross section with other periodic atomic parameters, the data of Nikolaev <u>et al.</u>, <sup>(10)</sup> was supplemented with others, so that  $\sigma_{i,i-1}$  at v = 8 x 10<sup>8</sup> cm sec<sup>-1</sup> could be correlated with the electron affinity I of the incident ion. Here I is the I.P. of the ion of charge (i-1).

A striking correlation was found, particularly in helium gas. Figure 1 shows the results which are discussed here. Each isoelectronic

Reaction	K(units 10 <sup>7</sup> cm sec <sup>-1</sup> )
H <sup>+</sup> , He	5.7
H <sup>+</sup> , Ne	3.5
H <sup>+</sup> , Kr	1.22
H <sup>+</sup> , Xe	1.07
$H_2^+$ , Ne	3.5
$H_{2}^{+}$ , Kr	0.88
H <sup>+</sup> <sub>2</sub> , Xe	0.96
He <sup>+</sup> , Ne	4.7

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HASTED EXPONENTIAL FITS

TABLE 5

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The electron capture cross section of ions of charge i and velocity 8 x  $10^8$  cm/sec in helium gas plotted versus the electron affinity of the ion. Second Second

sequence seems to fall on a curve. The N<sub>e</sub> = 0 sequence seems to have somewhat high values for He<sup>2+</sup>, Li<sup>3+</sup> and B<sup>5+</sup>. The He II (N<sub>e</sub> =1) sequence fits very well the curve

$$\sigma_{\rm K} = 8.5 \times 10^{-21} \, {\rm I}^{3/2} \, {\rm cm}^2 / {\rm atom}$$

if I is given in eV. The value of  $\sigma_{10}$  for H<sup>+</sup> also falls close to this curve.

The isoelectronic sequences for  $N_e = 2-6$  are fitted by a function

$$\sigma_{\rm T} = 6.5 \times 10^{-20} \, {\rm I}^{3/2} \, {\rm cm}^2 / {\rm atom}$$

We may interpret this result as saying that the cross section for capture into the L shell is 7.6 times greater than that for capture into the K shell. The dependence on  $I^{3/2}$  must be considered as an empirical law, and as yet has no theoretical justification.

The data in nitrogen gas shows less correlation than in helium as may be seen from Figure 2. Yet the function

$$\sigma_{\rm c} = 1.75 \times 10^{-20} \, {\rm I}^{3/2} \, {\rm cm}^2/{\rm atom}$$

adequately describes K shell capture. Capture into the L shell has a higher cross section than K capture and seems to be correlated to I by a three-halves power law. Similar conclusions follow for argon and krypton targets, but there is less straight-line behaviour on the log-log plots in Figures 3 and 4.



Figure 2. The electron capture cross section of ions of charge i and velocity 8 x  $10^8$  cm/sec in nitrogen gas plotted versus the electron affinity of the ion.











It is interesting to record here that the K shell capture cross sections in He,  $N_2$ , A and Kr are in the ratio 1:2.06:2.24:3.9 at the same electron affinity.

Plots of cross section versus I for lower velocities do not reveal the same degree of correlation as those at 8 x  $10^{9}$  cm sec<sup>-1</sup>. If any relation of the form I<sup>m</sup> is satisfied, m must be chosen for many isoelectronic sequences together and is probably less than 3/2 in any case.

One suspects that the data on He at high velocities reveals the underlying correlation with I, which becomes less prominent in heavier targets because there are many nonequivalent electrons to be captured.

#### 2.3 CORRELATIONS WITH ATOMIC PARAMETERS

2.3.1 <u>Electron Affinities.</u> It has been stated above that, at velocities of order  $4v_0$  in light element targets, the electron capture cross section correlates strongly with the electron affinity I. This correlation has also been noted by Allison and Garcia-Munoz<sup>(1)</sup> for capture by the members of the HI isoelectronic sequence in hydrogen and nitrogen targets. For resonance reactions at  $v = 10^7$  cm sec<sup>-1</sup>, Hasted<sup>(3)</sup> demonstrates another correlation of  $\sigma$  with I, but in this latter case  $\sigma \propto I^{-1.2}$  seems to be the approximate behavior.

There are other suggestions of the effect of electron affinity on cross sections. Nikolaev <u>et al.</u><sup>(30)</sup> show a correlation between I and the ratio  $\sigma_{i,i-2}$  of single and double electron capture cross sections.

The  $I^{3/2}$  rule seems to apply here. Dmitriev <u>et al.</u> <sup>(42)</sup> report a dependence of the single electron loss cross section on the reciprocal of I.

The significance of these correlations becomes more understandable when one realizes that the electron affinity is related to the scale of the radial wavefunction for the captured electron in its final orbit.

2.3.2 <u>Resonance Defect.</u> At very high energies it seems that the resonance defect  $\triangle E$  is not a determining factor in the charge transfer cross section. However, in the adiabatic region the resonance defect plays a dominant role in the behavior of the cross section.

The Massey near-adiabatic criterion states that the velocity of maximum cross section  $v_m$  is proportional to the energy defect:

$$\mathbf{v}_{\mathrm{m}} = \frac{\mathbf{a}}{\mathbf{h}} \left| \Delta \mathbf{E} \right| \,, \tag{6}$$

The proportionality constant a with dimensions of length is the so-called Massey parameter. A discussion of the near-adiabatic criterion is found in the article by Hasted.<sup>(2)</sup>

One can conclude from this work that there is a definite correlation between  $v_m$  and  $\triangle E$ , but that it may not necessarily be linear (Gilbody and Hasted<sup>(25)</sup>). Furthermore, the values of the Massey parameter show considerable variation among comparable reactions and between different types of capture processes. Modification of the value  $\triangle E$  by including initial and final state interactions seems to give somewhat better correlations (Hasted and Lee<sup>(43)</sup>) and reduce the variation in the Massey paramet

Dependence of the magnitude of the cross section on  $\triangle E$  has also been investigated. Fedorenko and Belyaev<sup>(44)</sup> have shown that the maximum cross section  $\sigma_m$  for two sets of endothermic reactions was a decreasing function of  $|\triangle E|$ , while for the exothermic He<sup>+</sup> reactions the cross section increased with  $|\triangle E|$ . However, in the latter case the increase of cross section with heavier target atom could easily account for this difference. The work of Chkuaseli <u>et al</u>.<sup>(45)</sup> on alkali and alkali-earth reactions at  $v = 1.67 \times 10^7$  cm sec<sup>-1</sup> shows the normal decrease of  $\sigma$  with increasing  $|\triangle E|$ . Their data also indicate that the exothermic reactions have a higher cross section than the endothermic. Plots of  $\sigma$  versus  $\triangle E$  at a fixed velocity were first introduced by Wolf.

2.3.3 <u>Target Atom Parameters.</u> In order to explain the monotonic increase of cross section with  $Z_a$  that is observed at high velocities, one can introduce a phenomenological parameter F(v), the average number of electrons per atom capturable at velocity v. Bates and McCarroll<sup>(35)</sup> have presented an argument to show that in resonant collisions F(v) depends on the distribution of orbital velocity of atomic electrons projected along the trajectory of the ion. The idea that capture occurs chiefly for atomic electrons moving with the same velocity as the ion in the atomic rest frame has often been used in classical models of charge exchange (see below).

One would expect, therefore, that at velocities less than  $v_0$  only electrons from the outermost atomic shell will participate in charge exchange reactions; but for  $v \lesssim Z_a v_0$  substantial capture of inner electrons will occur. Nikolaev <u>et al.</u><sup>(10)</sup> suggest that the capture cross sections

in different targets for  $v \sim (2-3)v_0$  reveal a proportionality to the number of electrons in the outer shell; while at  $v \sim 6v_0$ , F equals the number of electrons in the next outer shell. As noted in Section 2.2 above, similar data at  $v = 8 \times 10^8$  cm sec<sup>-1</sup> does not show as good agreement with the shell hypothesis. A possible explanation for such a discrepancy is that capture from different shells has a different velocity dependence (the BK approximation reveals such differences). The capture cross section in heavy target gases at  $v \sim 5v_0$  shows (Nikolaev <u>et al.</u>)<sup>(10)</sup> a decreasing logarithmic velocity derivative, an indication perhaps that capture from an inner shell is occurring.

Such considerations suggest that F(v) reflects the number of electrons in a given shell when v approximately equals the mean orbital velocity of the shell, but also that there may be a different velocity dependence for different shells.

#### SECTION 3

#### EVALUATION OF EXISTING MODELS

#### 3.1 LOW ENERGY MODELS

In the adiabatic region the resonance cross sections have been found theoretically using the impact parameter method - Bates and McCarrol<sup>(35)</sup> has a good treatment of this method. Several assumptions introduced introduced by Firsov<sup>(37)</sup> are used to evaluate the formula

$$\sigma = 2\pi \int_{0}^{\infty} \sin^{2} \left[ \int_{-\infty}^{\infty} \frac{(E_{a} - E_{s})}{2\hbar v} dx \right] b db .$$
 (7)

One assumption involves a model for the difference of antisymmetric and symmetric energies:

$$E_a - E_s = 2 I R \exp[-I^{1/2} R]$$
, (8)

where R is the internuclear distance and I is the electron affinity; atomic units are used. The resulting integral over impact parameter is approximated by putting

 $\sigma \stackrel{\simeq}{=} \frac{\pi}{2} b_1^2$ 

where  $b_1$  is determined by letting

$$\int_{-\infty}^{\infty} \frac{(E_a - E_s)}{2\hbar v} dx = 2\pi\beta$$

The value of  $\beta$  is chosen differently by different authors (Rapp and Ortenburger, <sup>(46)</sup> Rapp and Francis<sup>(4)</sup>). Using the asymptotic value of the integral over x for a large impact parameter, one obtains the result (in atomic units).

$$b_{1} \stackrel{\simeq}{=} \frac{1}{I^{1/2}} \ln \left[ \frac{I^{3/4}}{\beta(2\pi)^{1/2}} \frac{\overline{b}_{1}^{3/2} (1 + \overline{b}_{1}^{-1} I^{-1/2})}{v} \right], \quad (9)$$

where  $\overline{b_1}$  is an average value of  $b_1$  over a range of velocity.

The expression (9) shows the dependence of cross section on the electron affinity I and the velocity v. For v > I the approximation in (9) with an average  $\overline{b_1}$  cannot be used, but also the validity of assumption (8) is questionable.

Comparisons of formula (9) with experimental results have been given by Rapp and Francis<sup>(4)</sup> [cf. Hasted<sup>(3)</sup>]. Their curves agree fairly well with the data, although discrepancies in the data make confirmation difficult. If anything, Rapp and Francis calculate cross sections which are smaller than the experimental ones. This is further borne out by the work of Fite, Smith and Stebbings<sup>(6)</sup> on hydrogen symmetrical resonance and the (0<sup>+</sup>, H) accidental resonance. They found a fit of the form (4)

to both sets of data. Also Marino, Caplinger and Smith<sup>(23)</sup> investigated the cesium resonance reaction, confirming the form (4), but found values higher than the theoretical ones.

There is independent experimental data on the validity of equation (7). Measurements of the resonance capture probability versus energy at a fixed scattering angle were carried out by Ziemba and Everhart<sup>(47)</sup> and Lockwood, Helbig and Everhart<sup>(48)</sup> on helium, Lockwood and Everhart<sup>(49)</sup> on hydrogen, and Ziemba <u>et al.<sup>(50)</sup></u> with several reactions. This work and that of Jones, Costigan and Van Dyk<sup>(51)</sup> on neon shows that the capture probability takes the empirical form

$$P(b,v) = A + B \sin^2 \left[ \frac{1}{2\hbar v} \int_{-\infty}^{\infty} (E_a - E_s) dx - \beta \right], \quad (10)$$

where A and B are slowly varying functions of velocity and impact parameter.

The experimental values of  $\int_{-\infty}^{\infty} (E_a - E_s) dx$  for different collision paths can be obtained from the scattering data. With a reasonable assumption about the ion-atom interaction one can derive  $(E_a - E_s)$  as a function of internuclear distance R. Jones, Costigan and Van Dyk found

$$E_a - E_c = 28.6 \exp(-2.4R)$$
,  $0.4 < R < 1.5$  (11)

in atomic units. It appears that the result in Equation (11) disagrees considerably with Equation (8), which would be in this case

$$E_a - E_s = 3.2 \exp(-1.26R)$$
 (12)

However, a fit to experimental data of the form in Equation (11) may not disagree with Equation (12) as much as appears, since the integral  $\int_{-\infty}^{\infty} (E_a - E_s) dx$  may not be sensitive to the precise functional form of  $(E_a - E_s)$ . Thus, it is not evident how the difference between Equations (11) and (12) would affect the cross section  $\sigma_{10}$ . A thorough comparison of experiment with theory in the case of helium is made by Everhart.<sup>(52)</sup>

The presence of parameters A and B in (10) can at present only be predicted on empirical grounds, although the phase  $\beta$  has some theoretical justification (Bates and McCarroll<sup>(35)</sup>).

Models have also been developed for nonresonance reactions. Most of these stem from the previous adiabatic model of symmetrical resonance using the impact parameter method. Gurnee and Magee<sup>(53)</sup> developed their own approximate solution of the two-state differential equations, which has been questioned by Skinner,<sup>(54)</sup> who compared exact numerical solutions with the conjectured solution. Rapp and Francis<sup>(4)</sup> and Demkov<sup>(55)</sup> have employed the Rosen-Zener model to solve the two-state differential equations. Rydnik and Yavorskii<sup>(56)</sup> have employed a semi-classical approach to obtain the nonresonance cross section.

All nonresonance theories predict the Massey criterion, and all consider the resonance cross section as basic. The solutions of Rapp and Francis and Rydnik and Yavorskii give a capture probability of the form

$$P = P_{res} w , \qquad (13)$$

where w can be interpreted as a transition probability and  $P_{res}$  is the capture probability for the resonance process. In contrast, Gurnee and Magee proposed

$$P = \sin^2 \left[ \frac{1}{2\hbar v} \int_{-\infty}^{\infty} (E_a - E_s) \cos \left( \frac{\Delta E}{\hbar v} x \right) dx \right] . \quad (14)$$

The data of Ziemba <u>et al.</u><sup>(50)</sup> give some indication that both types of correction (to the amplitude and phase of the sine function) will occur in general. For the case of protons on different targets, changes of amplitude and phase from those of the hydrogen resonanct process can be seen, but there are surprising qualitative similarities between hydrogen and helium targets, between nitrogen and oxygen targets, and among Ne, A and Kr targets.

The capture probability of helium ions in nonresonant processes does not seem as closely related to  $P_{res}$  as in the hydrogen data.

The specific form of the transition probability w for the Rydnik-Yavorskii model for small resonance defect is

w = exp 
$$\left[ -\frac{1}{2} \left( 1 + \frac{M_i}{M_a} \right)^{1/2} \frac{1}{v} \left( 1 - \frac{I_i^{1/2}}{I_a^{1/2}} \right) \right]$$
 (a.u.)(15)

where M<sub>i</sub> and M<sub>a</sub> are the ionic and atomic masses, I<sub>i</sub> is the electron affinity of the ion, and I<sub>a</sub> the I.P. of the atom. For the Rapp-Francis model

w(b) = f sech<sup>2</sup> 
$$\left[ \frac{\Delta E}{v} \left( \frac{\pi b}{2} \right)^{1/2} (I_{i,a})^{-1/4} \right]$$
 (a.u.)(16)

where f is a statistical weight and  $I_{i,a}$  is an effective mean I.P. for ion and atom. If  $b \simeq b_1 \propto I^{-1/2}$ , then the Massey parameter in each model is

$$a_{\rm RF} \quad I_{i,a}^{-1/2} ; a_{\rm RU} \propto \left(1 + \frac{M_i}{M_a}\right)^{1/2} I_{i,a}^{-1}$$

Rydnik and Yavorskii present tables to compare their predictions with experimental results; the correspondence is qualitatively good at least. Similarly, the Rapp-Francis model predicts well the shape of the nonresonant cross section curves but not the magnitudes of  $\sigma$ .

#### 3.2 HIGH ENERGY MODELS

Simple classical models for charge transfer collisions were developed by Bohr and Lindhard (Bohr, <sup>(57)</sup> Bohr and Lindhard <sup>(58)</sup>). We will discuss their theory of electron capture by fast fission fragments.

An essential concept of the Bohr-Lindhard model is the factorization of the capture cross section:

$$\sigma_{i,i-1} = Q_{ex} f_{i,i-1} N_{eff}$$
 (17)

Here  $Q_{ex}$  is an effective cross section for excitation of the atomic electrons by the incident ion, f is a probability of capture for a.single electron, and  $N_{eff}$  is the effective number of electrons which can be captured. Equation (17) can be regarded as an approximation to the general formula in the impact parameter method

$$\sigma = \pi \int_{o}^{\infty} P(b) d(b^2) N_{eff} , \qquad (18)$$

where P(b) is the probability of capture at impact parameter b. The Bohr-Lindhard expression (17) follows from (18) by letting  $f = P(b_1)$ ,  $Q_{ex} = \pi b_1^2$ , as if the entire integral were concentrated at  $b_1$ . In order to compute  $b_1$ , Bohr and Lindhard assume that an electron with orbital velocity v is released from the atom by interaction with the passing ion of charge i. Their result is

$$b_1 \simeq \frac{2ie^2}{mv^2} , \qquad (19)$$

which may be derived as follows: An ion of charge i and velocity v passing at distance b from the electron will impart to it an impulsive momentum change

$$\Delta p \sim \frac{ie^2}{vb}$$

In order to excite the electron, its energy change v  $\Delta p$  must be greater than 1/2 mv<sup>2</sup>; hence, b must be less than b<sub>1</sub> as given in Equation (19).

Bohr<sup>(57)</sup> assumes further that the probability of capture is the fractional volume of phase space available to the electron in the final state relative to the volume of phase space in the initial state. If  $u_{i-1}$  is the orbital velocity of the captured electron in the final state, then we have

$$f_{i,i-1} = \left(\frac{u_{i-1}}{v}\right)^3, v > u_{i-1}$$

and  $f_{i,i-1} = 1$  for  $v \le u_{i-1}$ . For bare nuclei  $u_{i-1} = iv_0$ , but more generally one can let

 $u_{i-1} = \frac{2I_{i-1}}{m}$  1/2

where  $I_{i-1}$  is the I.P. of the ion of charge i-1.

Finally Bohr and Lindhard take  $N_{eff}$  to be approximately  $Z_a^{1/3} \frac{v}{v_o}$ for  $1 \le \frac{v}{v_o} \le Z_a^{2/3}$ . Putting these results together, one has the Bohr-Lindhard formula for bare nuclei (in atomic units)

$$\sigma_{i,i-1} = 4 \quad i^{5} z_{a}^{1/3} v^{-6} \quad . \tag{20}$$

This model has been modified by Nikolaev<sup>(50)</sup> to correct for shielding by ionic electrons in inner shells. Although Equation (17) may be a valid form in which to write the cross section, the particular Bohr-Lindhard interpretation of  $b_1$  is inadequate. Typically excitation and ionization cross sections go as ( $v^{-2}$  ln v) for high velocities (Seaton<sup>(60)</sup>). This would imply  $b_1 \propto v^{-1}$  instead of  $v^{-2}$ . Now the capture probability in the BK approximation goes as  $b^3v^{-7} \exp(-bv)$  for large b and v (Bates and McCarrol1<sup>(35)</sup>). In this case one would take  $b_1 \simeq \frac{1}{v}$  in agreement with the excitation cross section. Furthermore, the BK capture probability depends on  $v^{-7}$  against the Bohr-Lindhard  $v^{-3}$ .

It seems, therefore, that a factorization of the type in Equation (I is not too meaningful in charge exchange processes. It is better to thin in terms of the impact parameter formula (18). The classical theory of Bohr and Lindhard would predict a constant value of P(b) equal to f for  $b \leq b_1$ , and zero otherwise. The quantum theory of capture always leads to  $P \propto \exp(-\lambda b)$  for large b, a result unobtainable in any classical theory.

Another model derived from the Bohr-Lindhard approach was due to Bell<sup>(61)</sup> and Gluckstern,<sup>(62)</sup> In Bell's treatment the capture probability for impact parameter b would be given by an integral of the Thomas-Fermi distribution function over the region of phase space defined by the two inequalities

$$\frac{\partial \phi(\mathbf{r})}{\partial \mathbf{r}} \leq \frac{\mathrm{i}e^2}{b^2} \quad ; \quad \frac{1}{2} \text{ m } (\vec{v} - \vec{w}) \leq \frac{\mathrm{i}e^2}{b}$$

where r and  $\vec{w}$  are respectively the radius and velocity of an atomic electron and  $\phi(r)$  is its potential energy.

Gluckstern replaced the integration over phase space by a sum over the target electrons, assuming them to be located at radii determined from the Thomas-Fermi charge distribution. Calculations were performed for  $v/v_0 = 3,5,7$ , i = 3-6, and  $Z_a = 1,7,18,80$ . Capture in hydrogen was calculated from the Bohr model of the H atom.

The results of Gluckstern can be summarized in an approximate way as follows: The velocity dependence is roughly as  $(v/v_0)^{-3.5}$ ; the dependence on Z<sub>a</sub> ranges from Z<sub>a</sub><sup>0.16</sup> to Z<sub>a</sub><sup>0.7</sup>. The dependence on charge i is close to i<sup>2</sup>: at v = 8 x 10<sup>8</sup> cm sec<sup>-1</sup> a plot of  $\sigma$  versus I<sub>i</sub> gave powerlaw fits I<sup>n</sup>, where n = 0.98 for H, N = 0.90 for N and n = 0.87 for A.

In order to obtain better agreement with experimental results, Gluckstern arbitrarily reduced his calculated cross sections by 40%. This unjustified reduction should be discounted in evaluating the Gluckstern calculation. Certainly the Bell-Gluckstern results are too high at high velocities, but this is probably due to the exponent n of  $(v/v_0)^{-n}$  being too small. Since, even for hydrogen, this defect gives overly large cross sections, it must be traced to the classical picture of the capture probability. The capture probability in a quantum treatment will decrease more strongly with v than in a classical picture. However, it must be noted that Thomas, <sup>(63)</sup> by considering the capture process to be divided into two scattering processes, predicted a dependence on velocity with n = 11. Perhaps there is an appropriate classical picture probabilities for high impact velocities.

#### SECTION 4

#### CONCLUSIONS

#### 4.1 DISCUSSION

Any theory or model of charge exchange processes will provide an expression for the probability of electron capture P(b,v) as a function of impact parameter b and relative velocity v. For resonant processes the oscillatory nature of P as a function of either variable has been amply demonstrated both theoretically and experimentally. Similar evidence of oscillations in the nonresonant capture probability has been found empirically (Ziemba et al.<sup>(50)</sup>) in the adiabatic velocity range.

On the basis of the empirical studies one might attempt to write the capture probability in the general form

$$P = A + B \sin^2 \zeta$$
 (21)

For resonance one would expect A = 0. B = 1; however, the data suggest (Jones, Costigan and Van Dyk<sup>(51)</sup>) that  $A \neq 0$  and B < 1 because of the effect of nonresonant processes, particularly excitation and ionization of the target atom. These processes, in heavy atoms such as neon,

involve interpenetration of the K and L shells. Abrupt changes in the coefficients A and B as inner shells are disturbed may be related to corresponding abrupt rises in the mean energy loss per collision (Morgan and Everhart<sup>(64)</sup>).

However, the data have been taken only for collisions at angles such that inner electron shells are penetrated. There is no evidence to disprove the assumption A = 0, B = 1 for large impact parameter, where the predominant contribution to the capture cross section occurs. At high velocities the capture probability at small b is more important, and one might expect to find effects of excitation on the capture cross section.

At present the phase  $\zeta$  can be predicted by several theories of resonance and the model of Gurnee and Magee<sup>(53)</sup> for nonresonance. The amplitude B for nonresonant processes is also given by several models. However, there is no predictive theory for A. The lack of a simple model for charge exchange with excitation prevents one from stating definite conclusions about the possibility of understanding the empirical results before the present theories (Bates and McCarroll<sup>(35)</sup>) are applied to this problem. The attempts to develop models for ionization processes (Russek)<sup>(65)</sup> are certainly not applicable to the charge exchange process.

As v increases the phase  $\zeta$  decreases, until the capture probability is approximately  $\zeta^2$ . Of course, in the high velocity region,  $\zeta$  is no longer proportional to  $v^{-1}$ , but decreases more rapidly with v. Bates and McCarroll<sup>(35)</sup> have argued that the behavior of P for high velocities

is governed by the probability of finding an electron moving parallel to the ion with velocity v at distance b from the nucleus of the target atom. This assumption does reproduce the behavior of P for large b and v, in particular, the exponential decay with impact parameter characteristic of the BK approximation.

If the Bates-McCarroll conjecture were applied to a Thomas-Fermi model of the atom, one would predict a cross section that went as  $v^{-8}$  for large velocity. This faster dropoff with v than in the other classical models may show the usefulness of this picture. However, the experimental data seems less sensitive to the target atom than the properties of the projectile ion. How one would construct a good model with this conjecture is not too clear.

#### 4.2 CONCLUSIONS

Since most theories of charge exchange are confined to either the adiabatic or the high energy region, the possibility of developing empirical or semiempirical models to bridge the gap in velocities becomes attractive. For this reason it is important to establish firmly any empirical correlations of charge transfer cross sections with atomic parameters. It has already been noted above that at high velocities the electron capture cross section seems to have an  $I^{3/2}$  dependence on the electron affinity of the ion. The significance and generalization of this  $I^{3/2}$  law are certainly worth further study. Similarly the resonance defect appears to control the magnitude of the cross section in the adiabatic region; quantification of this relationship would be desirable.

An empirical verification should be made of the rule stating that a target of greater atomic number gives a higher cross section. It would be important to obtain evidence bearing on the relation of this increase to the increased number of outer shell electrons.

The case of resonance charge exchange in helium should be studied more closely to check the applicability of the various models for both adiabatic and high energy domains, inasmuch as one has data on the capture probability at different values of b and v.

The different models for nonresonant processes which are discussed above should be tested on their predictive values. Especially important would be to verify the asymptotic approach of the nonresonant to the resonant cross section at high velocities.

Since the classical high-energy models of charge exchange predict slower variation with velocity than is observed, they cannot be satisfactory in their present form. However, a better classical model should not only provide a more rapid velocity dependence, but also give some insight into the observed dependence on the charge i. Perhaps, the picture of classical orbits would provide understanding of the capture process in different velocity domains.

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