TABLE II. The ratio of twice the frequency shift  $2\delta\nu_0$  to the electron linewidth  $\Delta\nu$  at different temperatures.

The polarization of the Rb atoms was estimated from the absorption of pumping light.

Temperature (°K)	$2\delta  u_0/\Delta  u$	Polarization
190	$0.19 \pm 0.05$	$0.2 \pm 0.1$
300	$0.15 \pm 0.01$	$0.2 \pm 0.1$
600	$0.055 \pm 0.015$	$0.1 \pm 0.05$

tained in Ref. 2. Using a particular vapor-pressure curve<sup>9</sup> to determine the Rb density N at this temperature one finds that

\*Work supported by National Science Foundation.

<sup>1</sup>H. G. Dehmelt, Phys. Rev. <u>109</u>, 381 (1958).

<sup>2</sup>L. C. Balling, R. J. Hanson, and F. M. Pipkin, Phys. Rev. <u>133</u>, A607 (1964).

<sup>3</sup>L. C. Balling and F. M. Pipkin, Phys. Rev. <u>136</u>, A46 (1964).

<sup>4</sup>L. C. Balling, Phys. Rev. 151, A1 (1966).

<sup>5</sup>L. C. Balling, R. H. Lambert, J. J. Wright, and

PHYSICAL REVIEW A

# $\sigma_{\rm SF} = \frac{8.8 \times 10^{-7}}{v} \ \rm cm^2$ .

From Table II one sees that the ratio of the frequency shift to linewidth decreases with increasing temperature, but that this change may be entirely due to a decrease in the Rb polarization.

A smaller frequency shift was observed in the experiment described in Ref. 2 and the polarization of Rb atoms were estimated to be close to unity. We believe this estimate to be wrong and that a better estimate would have made the result quoted in Ref. 2 consistent with ours.

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R. E. Weiss, Phys. Rev. Letters 22, 161 (1969).

<sup>6</sup>J. J. Wright, L. C. Balling, and R. H. Lambert, Phys. Rev. <u>1</u>, 1018 (1970).

<sup>7</sup>H. M. Gibbs, Phys. Rev. <u>139</u>, A1374 (1965).

<sup>8</sup>F. Grossetete, J. Phys. Radium <u>25</u>, 383 (1964).

 ${}^9\mathrm{R}$ . E. Honig and D. A. Kramer, RCA Review, 1969, p. 285 (unpublished).

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## Charge-Changing Cross Sections of 5-25-MeV Iodine Ions in Hydrogen and Oxygen<sup>†</sup>

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Iodine ions, accelerated to energies between 5 and 25 MeV, have been passed through thin targets of hydrogen and oxygen, and nonequilibrium charge-state distributions have been measured in order to obtain charge-changing cross sections. Results of cross sections for capture of one and two electrons, and for loss of one to eight electrons are presented for ions with initial charge states ranging from  $2^{*}$  to  $18^{*}$ . Multiple-electron loss of iodine ions has been found to be relatively large in oxygen, but small in hydrogen. This systematic difference reveals the influence of different excitation mechanisms in energetic ion-atom collisions. The dependence of the cross sections on initial charge and velocity of the ions is discussed and, whenever possible, compared with theoretical expectations.

#### I. INTRODUCTION

Electron loss and capture by fast heavy ions in collisions with target atoms have been the subject of studies ever since fission fragments have been available. Bohr,<sup>1</sup> Bell,<sup>2</sup> and Bohr and Lindhard<sup>3</sup> presented theoretical formulations prior to 1955, i.e., at a time when essentially no experimental information on charge-exchange cross sections was available. No further comprehensive theoretical approaches have been reported. Experimental results on cross sections for heavy ions with nuclear charges Z > 18 have been measured in a few cases for krypton<sup>4</sup> and iodine ions<sup>5</sup>; more extensive investigations have been performed using iodine ions in N<sub>2</sub> by Angert *et al.*<sup>6</sup> on capture cross sections, and by Möller *et al.*<sup>7</sup> on loss cross sections. Datz *et al.*<sup>8</sup> reported mainly single-electron-caplogg grogg ge

ture and -loss cross sections for bromine ions at 13.9 and 25 MeV in  $H_2$ , He, and Ar, and Betz *et al.*<sup>9</sup> measured mult<sup>2</sup>ple-electron-capture and -loss cross sections for bromine and iodine ions between 6 and 15 MeV in  $H_2$  and He. Recently, a comprehensive review article on charge states and charge-changing cross sections of fast heavy ions penetrating through gaseous and solid media has been prepared.<sup>10</sup>

In this paper, we report extensive sets of cross sections  $\sigma(q, q')$  for initial projectile charge states q ranging from  $2^+$  to  $18^+$  and for transfer of n = q'-q electrons, where *n* ranges from -2 to +8. This is the first experiment in which multiple-electron loss has been measured very accurately for many adjacent initial charge states of a heavy ion in a relatively heavy target. Single-electron-capture cross sections are presented for a particularly wide range of initial charge states (in one case, 15 charge states have been used), enabling systematic studies of the charge-state dependence  $\sigma_c(q)$ . Double-capture cross sections have been studied in detail, as well as single- and multipleelectron-loss cross sections. The dependence of the cross sections on the ion velocity and on the target species is discussed. We demonstrate that our results cannot be adequately described by means of existing theories. It appears necessary to develop substantially revised and more comprehensive models for charge exchange of fast heavy ions. Furthermore, the need for more extensive experimental results for other ion species and at higher projecticle velocities is pointed out, especially because existing data are too limited to serve as a basis for satisfactory quantitative interpolations and extrapolations with regard to the basic parameters Z, q, ion velocity v, and target species  $Z_{t}$ .

#### **II. EXPERIMENTAL PROCEDURE AND DATA ANALYSIS**

The apparatus has been described previously,<sup>9,11</sup> as well as the procedure and technique of analysis.<sup>8,9</sup> It is sufficient to say that nonequilibrium charge-state fractions Y(q, x) have been measured for varying target thicknesses and for varying incident charge states, and the cross sections have been determined by a least-squares fit using the exact solution of the well-known system of differential equations,

$$\frac{dY(q, x)}{dx} = \sum_{q'} \left[ \sigma(q, q') Y(q) - \sigma(q', q) Y(q') \right], \quad (1)$$

where x denotes the target thickness in molecules/  $cm^2$  and  $\sigma$  is given in units of  $cm^2/molecule$ . Residual-ion excitation has been taken into account and the reported cross sections refer to the state of excitation in which the ions enter the target cell. It is believed that this is the ground state in only a few cases. Thus, our capture cross sections, though determined with an accuracy usually better than ~10%, may be too small by a factor of ~2 as compared to the ground-state values. The effects of residual-ion excitation on electron-capture and -loss cross sections have been discussed previoulsv.<sup>12,13</sup>

The capture cross sections associated with very high initial charge states have been obtained by means of the attenuation method as described in Ref. 14 and are, therefore, total-capture cross sections. Since they have been fitted to the individually obtained capture cross sections in the region of overlap, errors may, in these cases, be large because added contributions, especially of the double-capture cross sections, can only be estimated.

#### **III. RESULTS**

Table I summarizes the experimental parameters, and Tables II and III list all of the investigated cross sections for hydrogen and oxygen targets, respectively, arranged in the order of increasing values of E, n, and q. Errors are generally below 10% for relatively large cross sections for single capture or loss (except for the values obtained from the attenuation method where  $\delta\sigma_c/\sigma_c$  may reach a factor of perhaps 2), are somewhat larger for multiple loss, and are largest for double capture (~ 50%).

Figures 1-6 illustrate some typical trends which are discussed in the following paragraph. Figures 1 and 2 show the dependence of  $\sigma$  on q, Fig. 3 shows the particular case of 15-MeV iodine ions in oxygen for which multiple-loss cross sections have been measured in detail. The velocity dependence  $\sigma(v)$  is illustrated in Fig. 4. Some relative cross sections

TABLE I. List of experimental parameters;  $q_0$  denotes the charge states of the incident ions for which individual cross sections  $\sigma(q_0, q_0 \pm n)$  have been measured;  $n_{\max}^l = \max(n)$  for multiple loss. Attenuation cross sections  $\sigma_t(q_0^*)$  have been measured for values of  $q_0^*$  within the indicated range.

E (MeV)	Target	$q_0$	$q_0^*$	$n_{\max}^l$
5	$H_2$	2-7	6-15	3
10	-	3-9	9-18	3
15		5-11	11 - 17	3
20		6-10	12 - 16	2
25		10	14-15	1
5	. O <sub>2</sub>	2-7	6-14	4
10		3-10	9-16	3
15		5 - 11	11-18	8
20		6-10	12 - 16	3
25		10	14-15	1

 $k_n = \sigma(q, q+n) / \sigma(q, q \pm 1)$ 

are shown as a function of q and n in Figs. 5 and 6, respectively.

The interpretation of the capture cross sections is often difficult because it is generally not known whether the measured values refer to the ground state or to the excited states of the ions; it has been noted above that  $\sigma_c$  may decrease significantly when the excitation of the capturing ions is increased.<sup>13</sup> Whenever our capture cross sections have been measured for different residual-ion excitation, only the largest values are listed in Tables II and III, corresponding to the ground state or to the states of lowest investigated excitation.

A comparison of characteristic trends in the

data with theory is necessarily hindered because most theoretical results are valid only in restricted ranges of the basic parameters, and these limitations are, in general, not clearly spelled out, or because no theory exists which appears to be applicable to the experimentally investigated cases. For example, explicit theoretical formulas give simple power functions for the dependence of single-capture and -loss cross sections as a function of v, but most of the measurements have been performed in velocity ranges where the cross sections are near a maximum. Furthermore, the important processes of multiple-electron loss and multiple-electron capture have not yet received sufficient attention from theorists.

TABLE II. Charge-changing cross sections  $\sigma(q, q')$  of iodine ions passing through hydrogen at energies between 5 and 25 MeV, in units of  $10^{-16}$  cm<sup>2</sup>/molecule. Columns list initial charge state q, final charge state q', and the corresponding value of  $\sigma$ .

$\overline{q}$	q'	σ	q	q'	σ	q	q'	σ	q	q'	σ	q	q'	σ
5 MeV			10 MeV			15 MeV			20 MeV			25 MeV		
2	1	3.540	3	2	3.210	5	4	2.810	6	5	1.490	10	9	3.510
3	2	9,390	4	3	5.990	6	5	3.080	7	6	1.270	14	13	13.000
4	- 3	14,600	5	4	9.080	7	6	5.250	8	7	2.790	15	<b>14</b>	16.000
5	4	18.000	6	5	13,700	:8	<b>7</b>	7.720	9	8	6.330	10	8	0.010
6	5	20.800	7	6	18.100	9	8	11.500	10	9	9.000	10	, ,	
7	6	18.500	8	7	18.300	10	9	16.600	12	11	14.400	10	11	0.205
8	7	22.000	9	8	24.000	11	10	20.900	13	12	18.000			
9	8	31.000	10	9	27.000	12	11	26,000	14	13	25,000			
10	9	36,000	11	10	31.000	13	<b>12</b>	29.000	15	14	29.000			
11	10	41.000	12	11	36.000	14	13	39.000	16	15	35.000			
12	11	47.000	13	12	41.000	15	<b>14</b>	44.000	. 0	17	0 150			
13	12	53.000	14	13	53.000	16	15	51.000	9	· •	0.150			
14	13	67.000	15	<b>14</b>	62.000	17	16	60.000	6	7	0.780			
15	14	85.000	16	15	72.000	6	4	0 134	7	8	0.560			
0	0	0 025	17	16	88.000	7	5	0.101	8	9	9.464			
2	1	0.025	18	17	99.000	8	6	0.287	9	10	0.267			
3 ⊿	2	0.200	9	1	0 144	9	7	0.095	10	11	0.217			
4 5	3 3	1 990	3 1	2	0.144	10	8	0.298	7	9	0.052			
6	J 4	1.650		2	0.300	10	Ũ	0.200	9	11	0.045			
7	т 5	1 190	6	4	0.735	5	6	0.778	0		0.010			
		1.100	7	5	1 06	6	7	0.703						
2	3	2.740	، د	6	0 272	7	8	0.391						
3	4	1.650	0	Ũ	0.212	8	9	0.221						
4	5	0.065	3	4	1.890	9	10	0.140						
5	6	0.345	4	5	1.320	10	11	0.106						
6	7	0.197	5	6	0.807	5	7	0.201						
7	8	0.044	6	7	0.724	6	8	0.172						
2	4	0.700	7	8	0.669	7	9	0.099						
3	5	0.266	8	9	0.372	8	10	0.064						
4	6	0.104	3	5	0.686									
5	7	0.023	4	6	0.405	5	8	0.041						
6	8	0.019	5	7	0.332									
2	5	0.136	6	8	0.183									
4	7	0.019	7	9	0.319									
			3	6	0.369									
			4	7	0.230									
			5	8	0.152									
			6	9	0.348									

TABLE III. Charge-changing cross sections  $\sigma(q, q')$  of iodine ions passing through oxygen at energies between 5 and 25 MeV, in units of  $10^{-16}$  cm<sup>2</sup>/molecule. Columns list initial charge state q, final charge state q' and the corresponding value of  $\sigma$ .

$\overline{q}$	q <b>'</b>	σ	q	q'	σ	q	<i>q</i> ′	σ	q	q'	σ	q	q'	σ
	5 1	MeV		10	MeV		15 1	MeV		20	MeV		25	MeV
2	1	2.860	3	2	3.240	5	4	5.800	6	5	4.580	10	9	7.690
3	2	8.330	4	3	6.980	6	5	7.710	7	6	5.510	14	13	21.000
4	3	14.100	5	4	9.950	7	6	10.700	8	7	7.520	15	14	25.000
5	4	17.100	6	5	14.600	8	7	13.400	9	8	13,500	10	8	1.080
6	5	20.300	7	6	15.100	9	8	16.100	10	9	16.000	10	Ũ	
7	6	16.700	8	7	20.500	10	9	21.500	12	11	20.000	10	11	0.533
8	7	30,000	9	8	24.100	11	10	24.400	13	12	23.500			
9	8	34.000	10	9	30.200	12	11	28.000	14	13	33.000			
10	9	40.000	11	10	34.000	13	12	32.000	15	14	38.000			
11	10	44.000	12	11	40.000	14	13	40.000	16	15	43.500			
12	11	51.000	13	12	45.000	15	14	45.000	7	5	0.100			
13	12	62.000	14	13	59.000	16	15	55.000	8	6	0.602			
14	13	81.000	15	14	70.000	17	16	63.000	9	7	0.830			
3	1	0.219	16	15	77.000	18	17	72.000						
4	2	1,150	3	1	0.062	5	3	0.222	6	7	2.200			
5	3	2,800	4	2	0.312	6	4	0.332	7	8	1.050			
6	4	2.000	5	3	0.738	7	5	1,230	8	9	0.860			
7	5	5.950 6 140	6	4	1 320	8	6	0.685	9	10	0.659			
•	. 0	0.110	7	5	3 160	9	7	1,600	10	11	0.538			
2	3	3.620	8	6	3 450	10	8	3 310	6	8	1,120			
3	4	2.730	9	7	4 050	11	9	5 220	7	9	0.525			
4	5	1.860	10	8	6.090	11	0	0.220	. 8	10	0.316			
5	6	1.010	10	0	0.000	5	6	1.730	9	11	0.295			
6	7	0.764	3	4	2.690	6	7	1.430	10	12	0.260			
7	8	0.511	4	5	2.110	7	8	0.889		~				
		1 000	5	6	1.400	8	9	0.774	7	10	0.345			
2	4	1.620	6	7	1.070	9	10	0.583	8	11	0.255			
3	5	1.460	7	8	0.700	10	11	0.444	9	12	0.244			
6	6	1.060	8	9	0.600	11	12	0.415						
5	7	0.545	9	10	0.414	5	7	1.020						
6	8	0.321				6	8	0.662						
7	9	0.189	3	5	1.370	7	9	9.487						
2	5	1.02	4	6	0.992	. 8	10	0.442						
3	6	1.010	5	7	0.705	9	11	0.316						
4	7	0.776	6	8	0.366	10	12	0.268						
5	8	0.184	7	9	0.390	11	13	0.245						
6	9	0.124	8	10	0.220		20	•••===						
			9	11	0.247	5	8	0.579						
3	7	0.247	9	6	1 010	6	9	0.413						
			ن ۸	7	0.600	7	10	0.361						
			- <del>4</del>	6	0.090	8	11	0.349						
			0 6	0	0.397	9	12	0.270						
			0	9	0.250	10	13	0.199						
						5	9	0.399						
						6	10	0.356						
						7	11	0.328						
						8	12	0.236						
						9	13	0.151						
						-	10	0 000						
						5	10	0.362						
						6	11	0.215						
						7	12	0.227						
						8	13	0.120						
						5	11	0.303						
						6	12	0.173						
						7	13	0.147						
						5	12	0.182						
						6	13	0,074						
						v	10							
-						5	13	0.094	·					



FIG. 1. Charge-changing cross sections  $\sigma(q, q+n)$  in units of  $10^{-16}$  cm<sup>2</sup>/molecule for iodine ions passing through hydrogen at energies of 5, 10, 15, and 20 MeV, as a function of the initial ionic charge state q. The closed symbols refer to electron capture, n = -1 (•), and n = -2 (•), and the open symbols refer to electron loss, n = 1 ( $\bigcirc$ ), n = 2 ( $\square$ ), and n = 3 ( $\triangle$ ).

#### IV. DISCUSSION

#### A. Single-Electron Capture

As is to be expected from simple theoretical considerations,  $\sigma_c(q)$  generally increases with the initial charge state q, though shell and excitation effects may in some cases disturb that trend (Figs. 1 and 2). The latter effects, obvious especially at  $q = 7^*$  of the iodine ions, are consistent with earlier observations<sup>8,9</sup> and have been attributed mainly to the  $O \rightarrow N$  shell transition. Although this qualitative explanation which is discussed in Refs. 8 and 9 appears quite plausible, the effect is far from being satisfactorily understood.

Single-electron-capture cross sections are often approximated by the simple formula

$$\sigma_c(q,v) \propto q^{ac}/v^m , \qquad (2)$$

where the parameters  $a_c$  and m can be determined from both experimental and theoretical work. As regards  $a_c$ , we find values of 1.7, 2.0, 3.1, and 3.7 in hydrogen at 5, 10, 15, and 20 MeV, respectively, but no systematic increase is found in oxygen targets where  $a_c$  lies between 2.0 and 2.5. Theoretically, Bohr and Lindhard<sup>3</sup> estimated  $a_c$ = 2 in heavy targets and  $a_c \approx 3$  in very light targets. Deviations from  $a_c = 2$  in targets with  $Z_T \approx 7$  towards higher values have been observed,<sup>6</sup> and it may be argued<sup>4, 6, 10</sup> that this is an indication for a basic insufficiency of the underlying theoretical model.

It is evident from Fig. 4 that in order to describe our data the exponent m in Eq. (2) cannot have a constant value because the investigated ion velocities are low and  $\sigma_c(v)$  lies close to a maximum near  $v \simeq v_0$ . We find that m still decreases at the highest investigated velocities,  $v \simeq 6 \times 10^8$  cm/sec, and that this decrease is somewhat stronger in hydrogen than in oxygen. Bohr and Lindhard<sup>3</sup> estimate m = 3 for typical fission fragments ( $v \simeq 4v_0$ ), but corresponding experiments which have been performed at these projectile velocities lead to at least m = 4 and possibly to larger values.<sup>6</sup>

In the present velocity range, corresponding values of  $\sigma_c$  differ little in hydrogen and oxygen. This is perhaps somewhat surprising since all theoretical estimates give considerably smaller cross sections in hydrogen; these estimates are based on the assumption that those electrons which have orbital velocities near v are preferentially captured. For  $v \gg v_0$ , such electrons are not present in hydrogen and  $\sigma_c$  will thus be reduced. In the present case, however, the ion velocities are small and are comparable with orbital electron



FIG. 2. Charge-changing cross sections  $\sigma(q, q+n)$  in units of  $10^{-16}$  cm<sup>2</sup>/molecule for iodine ions passing through oxygen at energies of 5, 10, 15, and 20 MeV, as a function of the initial ionic charge state q. The closed symbols refer to electron capture, n=-1 (•), and n=-2 (•), and the open symbols refer to electron loss, n=1 (O), n=2 ( $\Box$ ), n=3 ( $\Delta$ ), and n=4 (×). The location of the values of the multiple-loss cross sections for  $n \le 7$  at 15 MeV is indicated by solid lines. (The details of this particular case are shown separately in Fig. 3.)

velocities in both  $H_2$  and  $O_2$ . One should expect, therefore, that  $\sigma_c$  will differ little in  $H_2$  and  $O_2$ and that this difference increases with increasing ion velocity, as is indeed observed experimentally (Fig. 4).

#### **B. Multiple-Electron Capture**

Figures 1-3 demonstrate that  $\sigma(q, q-2)$  generally increases with q. In most cases, there is a characteristic discontinuity at  $q = 8^*$  which has also been found previously and has been attributed to the  $O \rightarrow N$  shell transition.<sup>9</sup> The ratio

$$k_{-2} = \sigma(q, q-2) / \sigma(q, q-1)$$

generally increases with q in the investigated ranges of q. This is seen in Figs. 1 and 2, and especially in Fig. 5, where  $k_{-2}$  increases from ~4 to 21% for 15-MeV iodine ions in the range of charge states between 5<sup>+</sup> and 11<sup>+</sup>. The largest values have been found at the lowest ion velocity. For example, 5-MeV iodine in O<sub>2</sub> gives  $k_{-2} = 36\%$ for  $q = 7^+$ . In hydrogen,  $k_{-2}$  always remains below ~8% and is typically in the order of 1% and below. In fact, in many cases  $\sigma(q, q - 2)$  was too small to be measured with reasonable accuracy.

We believe that the smallness of effective double-



FIG. 3. Charge-changing cross sections  $\sigma(q, q+n)$  in units of  $10^{-16}$  cm<sup>2</sup>/molecule for iodine ions passing through oxygen at 15 MeV, as a function of the initial ionic charge state q. The values of n are indicated near each curve.



FIG. 4. Velocity dependence of cross sections for capture (closed symbols) and loss (open symbols) of a single electron by iodine ions passing through hydrogen and oxygen. The initial ionic charge state of the projectiles is indicated near each curve. Also shown are a few data points from Refs. 15 and 16.

capture cross sections relative to double-electron loss is partly a result of the fact that electrons are generally captured into excited states.<sup>13</sup> The total excitation  $I_t^*$  of an ion of initial charge q after effective capture of n electrons must be smaller than the ground-state ionization energy for the final charge state  $I_{q-n}$  and, thus, capture must proceed largely into ground states especially when n > 2. Otherwise,  $I_t^* > I_{q-n}$  will lead to rearrangement processes which result with high probability in the ejection of one or more electrons. Since electron capture by not fully ionized fast heavy ions is likely to occur preferentially into excited states at least for charge states  $q \simeq \overline{q}$ , one may argue that  $I_t^*$  exceeds  $I_{q-n}$  after most collisions where  $n \approx 2$ , i.e., it is not possible that all of the initially captured electrons remain bound. This implies that the probabilities for *initial* capture of more than one electron could be much higher than the actually observed small values. Furthermore, any residual excitations of the ions prior to the capture process must be expected to reduce multiple-electron-capture probabilities by a substantial amount.

#### C. Single-Electron Loss

The probability  $\sigma(q, q+1)$  decreases when the ions are in higher charge states (Figs. 1 and 2). It appears that  $\sigma(q, q+1)$  decreases more steeply in hydrogen than in oxygen, especially when  $q \gg \overline{q}$ . The rate of decrease varies with q, and a simple power law,  $\sigma_l \propto q^{-\alpha_l}$ , is a good approximation only



FIG. 5. Ratios of cross sections,  $k_n = \sigma(q, q+n)/\sigma(q, q \pm 1)$ , in percent, between multiple- and single-electron transfer for 15-MeV iodine ions passing through oxygen, as a function of the initial ionic charge state q. The values of n are indicated near each curve.

within limited ranges of charge states. When q is close to  $\overline{q}$ , i.e., close to the charge where the single-capture and -loss cross-section curves intersect,  $a_i$  is mostly of comparable but somewhat smaller magnitude than the corresponding exponents  $a_c$  for single capture [Eq. (2)]. Our values of  $a_i$  in oxygen are smaller than Bohr and Lindhard's estimate of  $a_i = 3$ .

Figure 4 illustrates the velocity dependence of single-electron-loss cross sections. In the investigated velocity range and for the most abundant charge states,  $\sigma(q, q+1, v)$  shows a broad maximum whenever the velocity of the most weakly bound electron in the ion,  $u_a = (2I_a/m_e)^{1/2}$ , approaches the ion velocity. This agrees with the finding discussed in Ref. 9. Based on Massey's adiabatic criterion as discussed by Nikolaev,<sup>4</sup> one should expect the maximum to occur at  $v = u_a$ . Deviations from this rule, found quite generally in the literature<sup>4,9</sup> as well as in our present data, are most likely a consequence of the approximate character of the criterion. In particular, effects of differing target atoms and of contributions of more tightly bound inner electrons in the ion to the single-electron-loss probability per ion have been disregarded. A more serious question arises due to the discrepancy of Massey's criterion and experimental results with Bohr and Lindhard's prediction  $\sigma_1 \propto v^2$ . On one hand,  $\sigma(q, q+1, v)$  is expected to show a maximum near  $v = u_a$ ; on the other hand, theory predicts just in that velocity range a dependence  $\sigma_l \propto v^2$ , i.e., in the range where our

and similar experiments have been carried out, namely, where q is close to  $\overline{q}$  and  $\sigma_c(q) \simeq \sigma_l(q)$ .<sup>3</sup> In view of the data, it must be concluded that neither theoretical approach is strictly applicable, although the Massey adiabatic criterion is at least qualitatively of considerable usefulness. Furthermore, we point out that in heavy ions with many bound electrons the velocities  $u_q$ , which are calculated from binding energies, may dramatically differ from those velocities which are determined from the kinetic energies,  $T_a$ , of the electrons in question.<sup>10</sup> For example, for ground-state iodine ions,  $I_q$  and  $T_q$  amount to 18 and 76 eV for charge 1<sup>+</sup>, and to 39 and 105 eV for charge 3<sup>+</sup>. This leads to considerable uncertainty as to what kind of electron velocity should be used in the framework of theoretical descriptions.

The dependence of  $\sigma_i$  on  $Z_T$  is not easily assessed from our results. In appears that  $\sigma_i$  of iodine ions stripped in hydrogen and oxygen differ more for higher charge states, but are very close to each other for low charge states.

#### **D.** Multiple-Electron Loss

The probabilities for loss of several electrons as a result of a single collision of a heavy ion with a target atom or molecule are complex quantities which have received only passing attention in the past, but which are of interest not only for those investigators who are primarily concerned with phenomena of charge exchange. Bohr<sup>1</sup> and Bohr and Lindhard<sup>3</sup> assumed that the maximum energy



FIG. 6. Relative cross sections,  $k_n = \sigma(q, q+n)/\sigma(q, q \pm 1)$ , in percent, for multiple-electron capture and loss of 15-MeV iodine ions with initial charge  $q = 5^+$  passing through hydrogen, helium, and oxygen, and with initial charge  $q = 6^+$  passing through oxygen. Also shown is the dependence  $k_n = k_0^{n-1}$  with  $k_0 = 0.6$ .

transfer in an ion-atom collision is given by  $2m_ev^2$ , corresponding to a classical impact of a heavy target core on an electron in an ion with relative velocity v. Thus, in their calculation of single-electron-loss cross sections, only those ion electrons which have orbital velocities  $u \le 2v$  are assumed to contribute to the total loss. Consequently, multiple-electron loss is severely hindered. Nevertheless, Bohr and Lindhard state that there is a considerable probability that several electrons are lost or captured by the ion, though they do not give further estimates—and neither do other authors.

Our experimental results show that simultaneous loss of *n* electrons occurs with extremely high probability even when *n* is large. This agrees with previous experimental findings.<sup>5,7,8</sup> Our results are unique insofar as we have, for the first time, measured multiple loss with good accuracy for large values of *n* and for many adjacent charge states. The most illustrative results are shown in Fig. 6. The ratio  $k_2$  between double and single loss can be as large as ~ 60% in oxygen and ~ 25% in hydrogen. For comparison,  $k_2 \lesssim 50\%$  has been found for iodine in helium.<sup>9</sup> The dependence on *q* is weak and no clear-cut trend is observed (Fig. 5).

As regards  $k_n$  especially for n > 2, there is a dramatic difference between light and heavy targets. In hydrogen and helium,  $k_n$  decreases rapidly with increasing n, but in heavier gases such as nitrogen, oxygen, or argon,  $k_n$  decreases slowly with n. Figure 6 illustrates, for example, that the probability  $\sigma$  (5, 13) for simultaneous loss of eight electrons by 15-MeV iodine ions of initial charge  $5^{+}$  in oxygen amounts to almost 6% of the single-loss cross section  $\sigma$  (5, 6). Likewise, Moak *et al.*<sup>5</sup> found  $k_8 \simeq 4\%$  for 110-MeV iodine ions with initial charge 12<sup>+</sup> in argon. Though multipleloss cross sections have not been measured directly for n > 8, it is known that the maximum number of lost electrons can be much higher; for example, 12-MeV iodine ions with initial charge 5<sup>+</sup> may lose as many as 27 electrons in a single encounter with a xenon atom.<sup>17</sup> The large probabilities for multiple-electron loss result (in the absence of equivalent multiple-capture cross sections) in pronounced asymmetries of equilibrium charge-state distributions; this can be explained on simple mathematical grounds<sup>10</sup> and agrees well with experimental evidence.

There is little doubt about a qualitative explanation of the observed effects. Following the discussion by Dmitriev *et al.*,<sup>18</sup> we distinguish two basically different processes, (i) direct ionization and (ii) quasimolecular collisions. As regards (i), individual electrons in an ion are lost via a direct interaction with atoms of the medium. In particular, the loss of an individual electron occurs quite independent of the presence of other electrons in the ion. Obviously, this mechanism applies primarily to collisions in which one or a few electrons are lost and is the only relevant mechanism of electron loss by light ions or, more generally, by ions which contain very few electrons. However, this mechanism of direct excitation is not likely to give rise to exceedingly large multipleelectron-loss cross sections and, in fact, light ions show relatively modest values of  $k_n$  for  $n > 1.^{18}$ Collisions of type (ii) may be described as follows. When two heavy ions with many bound electrons collide. electrons in interpenetrating shells are promoted to higher bound levels or even to continuum states, provided that the relative nuclear velocity is not too large with respect to the orbital velocities of the electrons involved. (For the present purpose, this is no serious restriction because ions with initial charge states close to the average equilibrium charge are already stripped off most outer electrons with low velocities  $u \stackrel{<}{\scriptstyle\sim} v$ .) When the ions separate, many of the excited electrons remain in excited states or become ionized. This promotion- and level-crossing mechanism has been discussed, for example, in Refs. 19 and 20, and has also been referred to as Pauli excitation.<sup>21</sup> A detailed prediction about the actual charge states which result from these collisions has not yet been possible. There is evidence that two cases have to be considered. First, innershell vacancies can be created which may decay via Auger effects and, thus, lead to additional ionization. Second, it has been argued on the basis of experimentally observed energetic shifts of heavy-ion x rays that extremely high charge states are already present before the radiative decay of an inner-shell vacancy occurs.<sup>22</sup> Despite the lack of complete understanding of these complex phenomena it must be assumed that processes of the kind in (ii) are responsible for the large multipleelectron-loss cross sections with  $n \gg 1$ . In this light, it is understandable that  $k_n$  remains small in light targets which cannot produce sufficient shell overlap in collisions with heavy ions. Since processes (i) and (ii) are dominant for low and high values of n, respectively, one may speculate that the region of overlap of the two processes is just that range of n in which  $k_n$  would show a decrease with n which is significantly weaker than for both lower and higher values on n. This is perhaps the effect which is visible near n = 5 in Fig. 6.

Finally, we note that charge exchange in relatively hard collisions which involve significant penetration of shells cannot be described adequately by existing theories which treat either collision partner as a point charge. It is also interesting to point out that multiple-electron loss and capture are based on essentially different processes. In particular, high excitation of ions in collisions re-

1492

duces the probability of multiple capture but enhances multiple loss. Little is known about the times which are necessary to complete the rearrangement processes of ions initially highly excited. When these times are longer than the times between two successive collisions, multiple-loss cross sections would to some extent change into

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<sup>1</sup>N. Bohr, Kgl. Danske Videnskab. Selskab, Mat.-Fys. Medd. <u>18</u>, No. 8 (1948).

<sup>2</sup>G. I. Bell, Phys. Rev. <u>90</u>, 548 (1953).

<sup>3</sup>N. Bohr and J. Lindhard, Kgl. Danske Vindenskab. Selskab, Mat.-Fys. Medd. <u>28</u>, No. 7 (1954).

<sup>4</sup>See review by V. S. Nikolaev, Usp. Fiz. Nauk <u>85</u>, 679 (1965) [Sov. Phys. Usp. <u>8</u>, 269 (1965)].

<sup>5</sup>C. D. Moak, H. O. Lutz, L. B. Bridwell, L. C. Northcliffe, and S. Datz, Phys. Rev. 170, 427 (1968).

<sup>6</sup>N. Angert, B. Franzke, A. Möller, and Ch.

Schmelzer, Phys. Letters <u>27A</u>, 28 (1968).

<sup>7</sup>A. Möller, N. Angert, B. Franzke, and Ch. Schmelzer, Phys. Letters <u>27A</u>, 621 (1968).

<sup>8</sup>S. Datz, H. O. Lutz, L. B. Bridwell, C. D. Moak, H. D. Betz, and L. D. Ellsworth, Phys. Rev. A <u>2</u>, 430 (1970).

<sup>9</sup>H. D. Betz, G. Ryding, and A. B. Wittkower, Phys. Rev. A <u>3</u>, 197 (1971).

<sup>10</sup>H. D. Betz, Rev. Mod. Phys. <u>44</u>, 465 (1972).

<sup>11</sup>G. Ryding, A. B. Wittkower, and P. H. Rose, Phys.

PHYSICAL REVIEW A

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## Theory of Low-Energy Scattering by a Long-Range $r^{-8}$ Potential

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The low-energy scattering by a potential consisting of a long- and a short-range part is discussed. A general expression for the phase shift  $\delta_L$  is derived starting from the radial Schrödinger equation. Effective-range expansions are presented for partial waves of all orders for the case of a long-range  $\gamma^{-8}$  potential. The quantity  $\tan \delta_L$  is calculated up to and including the term  $k^{2L+7} \ln k$ . It is found that in the low-energy limit,  $\tan \delta_L$  is proportional to  $k^{2L+1}$  for s, p, and d waves, and  $\tan \delta_L$  is proportional to  $k^6$  for all the higher partial waves.

#### I. INTRODUCTION

The purpose of this paper is to present a derivation of an effective-range theory for  $r^{-8}$  potential scattering. The significance of the  $r^{-8}$  potential lies in the fact that it appears as a correction of the van der Waals potential in the description of the long-range interaction of two atoms, and represents dipole-quadrupole effects.<sup>1,2</sup> It also appears as a correction to the polarization potential in the description of long-range electron-atom interaction.<sup>3-5</sup> It is generally a repulsive potential, while the van der Waals and polarization potentials are attractive.

In Sec. II we derive a general formula for the phase shift due to scattering by a potential consisting of a short- and a long-range part. In Sec. III we present effective-range expansions for  $r^{-8}$  potential scattering for all partial waves, based on the formula derived in Sec. II.

<u>6</u>

Ya. A. Teplova, Zh. Eksperim. i Teor. Fiz. <u>43</u>, 361 (1962) [Sov. Phys. JETP <u>16</u>, 259 (1963)].

Rev. 185, 129 (1969).

Rev. A 3, 1658 (1971).

Rev. 184, 93 (1969).

(1971).

Rev. Letters 24, 123 (1970).

<sup>19</sup>U. Fano and W. Lichten, Phys. Rev. Letters <u>14</u>, 627 (1965).

excitation cross sections; this would particularly

influence the balance of electron capture and loss

by heavy ions penetrating through large molecules

may hope that qualitatively satisfactory theories

can be worked out in the future.

or solids. Given the major qualitative understanding of multiple-electron loss of heavy ions, one

<sup>12</sup>G. Ryding, H. D. Betz, and A. B. Wittkower, Phys.

<sup>15</sup>G. Ryding, A. B. Wittkower, and P. H. Rose, Phys.

<sup>16</sup>A. B. Wittkower and H. D. Betz, J. Phys. B <u>4</u>, 1173

<sup>18</sup>I. S. Dmitriev, V. S. Nikolaev, L. N. Fateeva, and

<sup>13</sup>H. D. Betz, Phys. Rev. Letters <u>25</u>, 903 (1970).
 <sup>14</sup>G. Ryding, A. Wittkower, and P. H. Rose, Phys.

<sup>20</sup>W. Lichten, Phys. Rev. <u>164</u>, 131 (1967).

<sup>17</sup>Q. C. Kessel, Phys. Rev. A <u>2</u>, 1881 (1970).

 $^{21}W.$  Brandt and R. Laubert, Phys. Rev. Letters  $\underline{24},$  1037 (1970).

<sup>22</sup>H. D. Betz, J. P. Delvaille, K. Kalata, H. W. Schnopper, A. R. Sohval, K. W. Jones, and H. E. Wegner, in *Proceedings of the International Conference* on Inner Shell Ionization Phenomena, Atlanta, 1972 (North-Holland, Amsterdam, to be published).