## IONIZATION OF GASES BY FAST HELIUM ATOMS AND SINGLY-CHARGED HELIUM IONS

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The ionization of helium, argon, hydrogen, and nitrogen gases by fast 15-180-keV He atoms and He<sup>+</sup> ions is studied. The ionization cross sections and the cross sections for the production of slow ions with different e/m ratios are measured. The ratios of the ionization cross sections and the positions of peaks on the cross-section vs. velocity curves are discussed.

## INTRODUCTION

ELECTRON capture and loss by fast He atoms and ions traversing gases have been studied very thoroughly. The cross sections obtained up to 1958 are given in Allison's review article;<sup>[1]</sup> several additional investigations have been reported since that time.<sup>[2-5]</sup> The ionization of gases by fast He<sup>+</sup></sup>ions has been investigated in [6-10], of which only <sup>[6]</sup> and <sup>[8]</sup> contain information regarding the mass-spectrometric composition of the slow ions formed in the gas. Ionization by fast He atoms has not been studied. A comparison of the cross sections for gas ionization by fast ions and atoms of identical velocities is of interest for the purpose of evaluating the contributions of different processes and arriving at an understanding of the atomic collision mechanism. In earlier work  $\lfloor 11 \rfloor$ we investigated fast H atoms and protons.

In the present work we performed parallel measurements of cross sections for gas ionization by fast He atoms and fast He<sup>+</sup> ions along with a mass-spectrometric analysis of the slow ions. These measurements were obtained in two atomic gases (helium and argon) and two molecular gases (helium and argon) and two molecular gases (hydrogen and nitrogen) differing greatly in molecular weight. The investigated velocity range includes the Bohr orbital velocity  $v_0$ . ( $v_0 = 2.2 \times 10^8$  cm/sec is the electron velocity in the Bohr hydrogen atom; for helium ions and atoms this velocity corresponds to an energy of 100 keV.)

## 1. PROCEDURE

In measuring cross sections we used the condenser method, combined with an analysis of e/m for slow ions, which we employed in our previous work.<sup>[8,11,12]</sup> The beams of fast helium atoms and ions were rendered homogeneous with regard to both composition and energy. Fast-atom beam intensities were measured by means of thermistors. <sup>[12]</sup> The fast-ion currents were in the range  $7 \times 10^{-9} - 1 \times 10^{-6}$  A. The fast-atom fluxes were were equivalent to currents in the range  $5 \times 10^{-9}$   $-2 \times 10^{-7}$  A. Currents to the plates of the measuring condenser were in the range  $2 \times 10^{-11}$ - $5 \times 10^{-8}$  A.

The total cross sections measured by the condenser method,  $\sigma_+$  for the formation of slow positive ions and  $\sigma_-$  for the formation of free electrons, are related to the cross section  $\sigma_i$  for gas ionization, the detachment cross section  $\sigma_l$ , and the cross section  $\sigma_c$  for electron capture by fast particles as follows:

$$\sigma_{+} = \sigma_{i} + \sigma_{c}, \qquad (1)$$

$$\sigma_{-} = \sigma_{i} + \sigma_{l}. \tag{2}$$

For fast atoms  $\sigma_c$  is determined only by the transition He  $\rightarrow$  He<sup>-</sup> and is small compared with  $\sigma_i$ ;<sup>[5]</sup> therefore,

$$\sigma_i \approx \sigma_+, \quad \sigma_l \approx \sigma_- - \sigma_+.$$
 (3)

For fast He<sup>+</sup> ions we have, according to Allison, <sup>[1]</sup>  $\sigma_l \ll \sigma_i$ ; therefore

$$\sigma_i \approx \sigma_-, \quad \sigma_c \approx \sigma_+ - \sigma_-.$$
 (4)

The data regarding the total cross section  $\sigma_+$ and line ratios in the mass spectrum of slow ions produced in a gas enabled us to determine the cross sections for the production of the corresponding slow ions.

The beams of fast helium atoms produced by charge exchange in the gases contained no appreciable admixture of atoms in metastable states. This is indicated by the equality  $\sigma_i = \sigma_i$  of the measured cross sections for the symmetric interaction pair He-He and by other control experiments.

The low gas pressure  $(1-4) \times 10^{-4}$  mm Hg in

342

the collision chamber ensured single collisions. Differential pumping enabled the maintenance of pressures under  $5 \times 10^{-6}$  mm in the other vacuum compartments. Pressures were measured with a Knudsen gauge and were monitored with a McLeod gauge. All cross sections are given in cm<sup>2</sup> per gas molecule; the errors did not exceed 15%.

## 2. RESULTS AND DISCUSSION

A. Cross sections  $\sigma_i$  (He) for gas ionization by fast helium atoms and  $\sigma_i$  (He<sup>+</sup>) by fast singlycharged helium atoms. Figure 1 shows our present measurements of the cross sections for helium, argon, hydrogen, and nitrogen ionization by fast He atoms; Fig. 2 shows the corresponding cross sections in the case of fast He<sup>+</sup> ions. Figure 2 also shows cross sections for the ionization of helium and molecular hydrogen at lower<sup>[3]</sup> and at higher <sup>[10]</sup> energies. These curves agree with our present work within experimental error limits. The cross sections for argon ionization measured in <sup>[9]</sup> are 30-40% larger than in the present work. The results obtained for these cross sections in <sup>[8]</sup> are in better agreement with our results.

The curves of the cross section for ionization by fast He atoms (Fig. 1) show that the maximum



FIG. 1. Cross section for gas ionization by fast He atoms.

of this cross section can be expected for  $v > 3 \times 10^8$  cm/sec. For  $\sigma_i$  (He<sup>+</sup>) in helium the maximum is observed at  $v \approx 4 \times 10^8$  cm/sec (Fig. 2); for molecular hydrogen the maximum is located at  $v \approx 3 \times 10^8$  cm/sec.<sup>[10]</sup>

The experimental data can be compared with specific theoretical calculations in only one case, hydrogen ionization by fast He<sup>+</sup> ions. In <sup>[13]</sup> a Born approximation was used to calculate the cross section for the process



FIG. 2. Cross section for gas ionization by fast  $He^+$  ions. Solid lines – present work; dashed lines – at 10-42 keV from [<sup>9</sup>], and at 150-1000 keV from [<sup>10</sup>].

$$He^+ + H \rightarrow \overline{He^+} + H^+ + e_{\bullet}$$
 (5)

(The bar denotes fast particles.) Figure 3 shows the corresponding theoretical curve, derived using the simplest hypothesis that the H<sub>2</sub> molecule is equivalent to two H atoms. This figure also includes experimental curves for hydrogen ionization by fast helium atoms and singly-charged ions obtained in the present work, and for fast He<sup>+</sup> ions in <sup>[10]</sup>. It is easily seen that the theory agrees with experiment for velocities  $v > 2v_0$ .

A comparison of the experimental cross sections for ionization by fast helium atoms and ions shows that for  $v > v_0$  we have  $\sigma_i(He^+) > \sigma_i(He)$ , while for  $v < v_0$  we have  $\sigma_i(He^+) < \sigma_i(He)$ . We observed completely analogous relations for the ionization cross sections in our earlier work with



FIG. 3. Cross section for the ionization of molecular hydrogen by fast He and H atoms and fast He<sup>+</sup> and H<sup>-</sup> ions. Heavy solid curves – present work; light solid curve – for H from  $[^{11}]$ ; dashed curves – for He<sup>+</sup> from  $[^{10}]$  and for H<sup>-</sup> from  $[^{14}]$ ; dot-dash curve – theoretical calculation from  $[^{13}]$ .

fast H atoms and protons.<sup>[11]</sup> This indicates the unsuitability of the Born approximation for velocities  $v < v_0$  and is of independent theoretical interest.

Figure 3 shows for comparison, the curve for fast H atoms which we obtained previously, <sup>[11]</sup> and the curve for fast  $H^-$  ions given in <sup>[14]</sup>. The H atom and the He<sup>+</sup> ion are one-electron systems, while H<sup>-</sup> and He are two-electron systems with different nuclei. The figure shows that for  $v > v_0$ the cross section for ionization by fast H atoms is smaller than that for ionization by fast He<sup>+</sup> ions. The cross section for ionization by fast H<sup>-</sup> ions is considerably smaller than in the case of fast He atoms but is close to the value for  $\sigma_i(H)$  in the entire compared velocity range. It appears from these comparisons that for the simplest atomic systems the ionization cross sections are strongly dependent on nuclear charge. Similar properties have been observed in other gases.

It is also interesting to note that the ionization cross section reaches its maximum sooner with fast H atoms than with fast He atoms and ions.

B. Cross sections for the production of slow ions with different values of e/m. The cross sections for the production of slow ions with different e/m ratios were measured when helium, argon, molecular hydrogen, and molecular nitrogen were ionized by fast atoms and singly-charged helium ions. It is seen from the results, shown in Fig. 4, that the cross sections for the production of slow ions having identical charges are always larger in the case of fast ions. This occurs because interactions of fast ions with gas molecules are possible both as "pure" ionization and as charge exchange, while in the case of fast atoms only pure ionization is possible. This is also indicated by the character of the cross section vs velocity curves. For example, in the case of helium Fig. 4 shows that for the He<sup>+</sup>-He pair the cross section  $\sigma_{01}$  decreases continuously as the velocity increases. This is associated with the relatively large contribution of resonance charge exchange compared with the cross section for single-electron ionization.

The results obtained in our earlier work and in the present investigation permit certain general comments concerning the pure ionization processes observed in atom-atom collisions. Figure 5 shows the curves for the cross sections of single (n = 1), double (n = 2), and triple (n = 3) pure ionization of argon by different fast atoms. The data for fast He atoms were obtained in the present work, for fast H atoms in <sup>[11]</sup>, and for fast Ne and Ar atoms in <sup>[12]</sup>. It is easily seen that for the same



FIG. 4. Cross sections for the production of slow ions with different e/m ratios in different gases: helium, argon, molecular hydrogen, and molecular nitrogen. Solid curves – for fast He atoms; dashed curves – for fast He<sup>+</sup> ions.

ionizing atom the cross section for the removal of the next electron from the argon atomic shell is usually 5 to 10 times smaller. The relative contribution of deeper pure ionization processes increases with the atomic number of the ionizing atom. This occurs particularly when we pass from fast H to fast He atoms. For example, for fast H atoms the cross section ratios  $\sigma_{02}/\sigma_{01}$  and  $\sigma_{03}/\sigma_{02}$ are ~ 0.2 and ~ 0.07, respectively, while in the case of fast He atoms the same ratios become considerably larger, the values being ~ 0.5 and ~ 0.18.

The cross section  $\sigma_{01}$  for single ionization of argon by fast He atoms remains practically constant in the entire investigated velocity range (Fig. 5); the maximum is evidently smeared out in this region. The maxima of the  $\sigma_{02}(v)$  and  $\sigma_{03}(v)$  curves obviously lie in the region  $v > v_0$ . On the  $\sigma_{02}(v)$  and  $\sigma_{03}(v)$  curves for fast hydrogen

344



FIG. 5. Cross sections for "pure" ionization of argon by different fast atoms. Solid curves – single ionization ( $\sigma_{01}$ ); dashed curves – double ionization ( $\sigma_{02}$ ); dot-dash curves – triple ionization ( $\sigma_{03}$ ).

atoms the maximum is observed at  $v \gtrsim v_0$ , and on the  $\sigma_{01}(v)$  curve it obviously is found at  $v < v_0$ ,<sup>[11]</sup> the determination of its exact location being of decided interest.

We shall now consider a little more thoroughly the cross sections for the production of slow ions in the molecular gases hydrogen and nitrogen as a result of collisions with fast He atoms and ions (Fig. 4). We observed slow  $H_2^+$  and  $H^+$  ions in hydrogen, and  $N_2^+$ ,  $N^+$ , and  $N^{2+}$  ions in nitrogen. The maxima for the production of the molecular ions  $H_2^+$  and  $N_2^+$  in the ionization of the molecular gases by fast He<sup>+</sup> ions lie in the region  $v \leq v_0$ , which is the region of the maximum for charge exchange in <sup>[1]</sup>. On the analogous curves for ionization by fast He atoms the maxima appear to lie at higher velocities ( $v \geq v_0$ ), since the molecular ions are formed only as a result of pure single ionization.

The curves for the production of slow atomic ion fragments  $[\sigma(H^+) \text{ and } \sigma(N^+)]$  are more complex; some of these possess a maximum at higher velocities  $[(2-2.5) \times 10^8 \text{ cm/sec}]$ , while with decreasing velocity a minimum is observed, followed by growth of the cross section at still lower velocities (for example, see Fig. 4 for hydrogen). Atomic ion fragments should result from dissociative ionization:

$$\overline{\mathrm{He}}^{+} + \mathrm{H}_{2} \to \overline{\mathrm{He}} + \mathrm{H} + \mathrm{H}^{+}.$$
 (6)

$$\overline{\mathrm{He}} + \mathrm{H}_2 \to \overline{\mathrm{He}} + \mathrm{H} + \mathrm{H}^+ + e. \tag{7}$$

These processes can occur both as a result of electronic transitions of molecules to unstable states, and as a result of electronic transitions accompanied by changes of vibrational energy. It can be expected from general considerations<sup>[15]</sup> that the contribution of electronic excitation is greater at high velocities, while the vibrational factor is greater at low velocities. This fact

probably accounts for the complex shapes of the curves representing the production of slow atomic ion fragments in the investigated velocity range.

Using our present experimental results regarding the total cross sections for the production of positive and negative particles ( $\sigma_+$  and  $\sigma_-$ ), we also determined [see Eqs. (3) and (4)] the loss cross section  $\sigma_l$  of fast helium atoms and the capture cross section  $\sigma_{C}$  for fast  $\mathrm{He}^{*}$  ions in helium, argon, hydrogen, and nitrogen. In the entire velocity range  $\sigma_{C}$  agrees well with the results of Stier and Barnett, <sup>[16]</sup> but our present result for  $\sigma_l$  is 20-40% greater than the measurements of other investigators. [2,3,17] It is possible that the method of determining  $\sigma_l$  through the registration of fast ions resulting from electron loss in the mentioned investigations was not free of systematic errors resulting from incomplete collection of these ions because of scattering.

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91