PRODUCTION OF CHARGED AND EXCITED PARTICLES DURING COLLISIONS OF He^+, Ne^+, *AND* Ar^+ *IONS WITH CO MOLECULES*

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The total cross sections for the production of CO^+, C^+, and O^+ ions during collisions between He^+, Ne^+, and Ar^+ ions and CO molecules at ion energies between 3 and 50 keV have been measured mass-spectrometrically. Measurements have also been carried out of the cross sections for the formation of CO^+, C^+, and O^+ ions in excited states during the same collisions at energies between 0.12 and 30 keV. The results of measurements lead to the conclusion that many of the features of the $\sigma(v)$ curves (σ is the cross section for the collision process and v is the relative collision velocity) can be explained in terms of the adiabatic maximum rule. At low velocities the $\sigma(v)$ curves depart from the predictions of the Massey adiabatic hypothesis.

INTRODUCTION

THE production of charged and excited particles during collisions between slow heavy particles and N_2 and H_2 molecules was investigated in[1] and [2], by mass-spectrometric and spectroscopic methods, respectively.

We have carried out studies of this kind using the CO molecule. The incident particles were He^+, Ne^+, and Ar^+ ions with energies between 3 and 50 keV (mass-spectrometric method) and between 0.12 and 30 keV (spectroscopic method). The cross sections for the production of slow charged particles (CO^+, C^+, and O^+) and excited particles (CO^{+*}, C^{+*}, and O^{+*}) were measured in relative units. The cross sections for the excitation of certain spectral lines of helium, neon, and argon ions and atoms have also been determined. The processes in which the above charged and excited particles can appear are summarized in the table below. The cross sections for the production of the above particles were measured by the mass-spectrometric method described in[1], and by the spectroscopic method described in[3].

Carbon monoxide was produced by thermal decomposition of formic acid in the presence of sulfuric acid. Mass-spectrometric and spectroscopic analyses showed that the carbon monoxide produced in this way did not contain appreciable amounts of impurities.

No.	Process	Type of process
1	$A^+ + CO \rightarrow A + CO^+$	Charge exchange
2	$A^+ + CO \rightarrow A^+ + CO^+ + e$	Ionization
3a	$A^+ + CO \rightarrow A + C^+ + O$	Dissociative charge exchange
3b	$A^+ + CO \rightarrow A + C + O^+$	
4a	$A^+ + CO \rightarrow A^+ + C^+ + O + e$	Dissociative ionization
4b	$A^+ + CO \rightarrow A^+ + C + O^+ + e$	
5	$A^+ + CO \rightarrow A + CO^{+*}$	Charge-exchange with excitation of molecular ion
6	$A^+ + CO \rightarrow A^+ + CO^{+*} + e$	Ionization with the excitation of molecular ion
7a	$A^+ + CO \rightarrow A + C^{+*} + O$	Dissociative charge exchange with the excitation of fragment particle
7b	$A^+ + CO \rightarrow A + C + O^{+*}$	
8a	$A^+ + CO \rightarrow A^+ + C^{+*} + O + e$	Dissociative ionization with the excitation of fragment particle
8b	$A^+ + CO \rightarrow A^+ + C + O^{+*} + e$	
9	$A^+ + CO \rightarrow A^* + CO^+$	Charge-exchange with the excitation of the incident particle
10	$A^+ + CO \rightarrow A^{+*} + CO^+ + e$	Ionization with the excitation of the incident particle
11	$A^+ + CO \rightarrow A^{+*} + CO$	Excitation of incident particle

RESULTS

Figures 1–3 show the $\sigma(v)$ curves for the following pairs: He^+-CO, Ne^+-CO, and Ar^+-CO (σ is the cross section for the production of the particle under investigation and v is the relative collision velocity). The scales for the relative cross sections for the production of charged and excited particles are different.

Figure 1 shows both the present data and the $\sigma(v)$ curves for CO^+, O^+, and C^+ taken from the paper by Browning et al.[4] Figure 2 shows the corresponding curves taken from the paper by Ogurtsov and Flaks.[5] The data obtained as a result of our own measurements and those taken from[4,5] are normalized at the point corresponding to an incident energy of 20 keV.

In addition to the $\sigma(v)$ curves for the production of slow charged and excited particles, Figs. 1-3 give: (1) the $\sigma(v)$ curve for the excitation of the 3888 Å line of He I (Fig. 1), (2) the $\sigma(v)$ curves for the excitation of the 5852 and 4290 Å lines of Ne I and Ne II, respectively (Fig. 2), and (3) the $\sigma(v)$ curve for the 4764 Å line of Ar II (Fig. 3).

It is important to note that the ratio of the cross section for the production of the C^+ and O^+ ions to the cross section for the production of the molecular ions CO^+, as deduced from our own work, turns out to be much smaller than the value reported in[4,5]. This is probably due to the fact that the pumping system and the arrangement used to produce the slow ion beam in[4,5] has higher ion collection efficiency than the system used in the present work. It is well known that the C^+ and O^+ ions are produced with a higher initial energy than the molecular ions, so that if the ion collection efficiency is low, the above cross-section ratio turns out to be too low. However, the low fragment-ion collection efficiency of our equipment does not lead to appreciable distortion of the shape of the $\sigma(v)$ curves. This follows from a comparison of the $\sigma(v)$ curves obtained from our own measurements with the

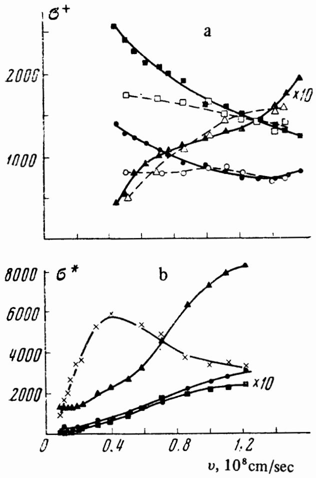

FIG. 1

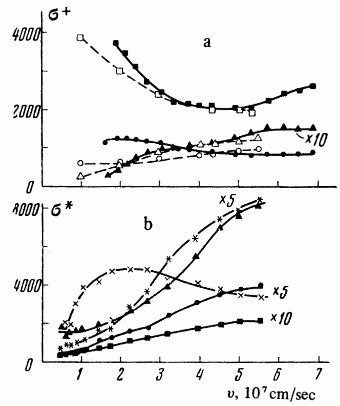

FIG. 2

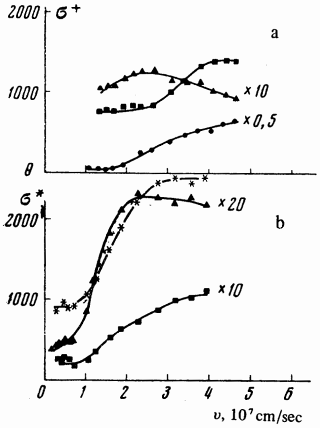

FIG. 3. σ(v) curves for Ar^+-CO (σ in relative units). a–Charged particles, b–excited particles: ●–O^+ ions, ■–C^+ ions, ▲–CO^+ ions, *–Ar^{+*} ions.

FIG. 1. σ(v) curves for He^+-CO (σ in relative units). a–Charged particles, broken curves taken from [4]; b–excited particles: ●, ○–O^+ ions, ■, □–C^+ ions, ▲, △–CO^+ ions, X–He* atom.

FIG. 2. σ(v) curves for Ne^+-CO (σ in relative units). a–Charged particles, broken curves taken from [5]; b–excited particles: ●, ○–O^+ ions, ■, □–C^+ ions, ▲, △–CO^+ ions, X–Ne* atom, *–Ne^{+*} ions.

data reported in[4,5] (see Figs. 1a and 2a).[1)] A simpler situation occurs in the case of the production of slow charged particles in nitrogen and hydrogen.[1,2]

DISCUSSION

Inspection of Figs. 1-3 will show that the shape of the $\sigma(v)$ curves is very different in this particular energy range. Whenever the experimental $\sigma(v)$ curve cannot be compared with a more or less precise theoretical analysis of the cross sections, all that can be done is to consider the general behavior of the $\sigma(v)$ curve in terms of the Massey adiabatic hypothesis. This involves two assumptions, namely, (1) in the velocity range where $a|\Delta E|/hv \gg 1$[2)] the cross section σ should fall monotonically and rapidly with increasing velocity of the colliding particles, and (2) the velocity v_{max} at which σ reaches a maximum is determined by the adiabatic maximum rule $v_{max} \approx a|\Delta E|/h$. The quantity a in the formula for v_{max} is not very dependent on the nature of the two colliding particles, but does depend on the type of process in which the particles participate.[6,7]

To explain the shape of the $\sigma(v)$ curves (Figs. 1-3) we must know the velocity v_{max} for all the processes (see the table) which contribute to the cross section for the production of the particular particles. For this purpose, we have therefore calculated the resonance defects for all these processes and then used the adiabatic maximum rule to calculate v_{max}. For all the nondissociative processes, the parameter a is assumed to be 7 Å,[6] whereas for the processes accompanied by differentiation of the CO molecule we assume that a = 3 Å.[8]

When the $\sigma(v)$ curves shown in Figs. 1-3 are analyzed it must be remembered that, in some cases, several processes contribute to the cross section for the production of the particular particles. For example, the cross sections σ_{C^+} and σ_{O^+} for the formation of the C^+ and O^+ ions include contributions due to processes 3, 4, 7, and 8, whereas the cross sections $\sigma_{C^{+*}}$ and $\sigma_{O^{+*}}$ for the formation of the excited ions C^{+*} and O^{+*} include contributions due to only processes 7 and 8. Processes 1, 2, 5, 6, 9, and 10 contribute to σ_{CO^+} whereas in the case of the production of the excited ion CO^{+*} only processes 5 and 6 provide a contribution. Moreover, when the relative contribution of the various processes to the cross section for the production of a particular particle are considered, it must be remembered that processes with the smaller resonance defect usually have a higher probability.

Let us now consider the shape of the individual $\sigma(v)$ curves for the various particle pairs.

He^+-CO

The maxima on the $\sigma(v)$ curves for processes 1, 2, 5, and 6 involving the production of CO^+ ions are, respectively, at velocities of 1.8×10^8, 2.4×10^8, 1.35×10^8, and 2.8×10^8 cm/sec. In the particular velocity range, only process 5 has a maximum. This process is responsible for the point of inflection near v = 1.3 $\times 10^8$ cm/sec on the $\sigma(v)$ curve for CO^+ ions. Process 5 has a still greater effect on the cross section curve for CO^+ ions reported in[4] (see Fig. 1a). This influence of process 5 on the shape of the $\sigma(v)$ curve for CO^+ ions is due to the fact that this process has the smallest resonance defect among processes 1, 2, 5, and 6.

The shape of the $\sigma(v)$ curve for the production of the excited ion CO^{+*} is determined only by processes 5 and 6 of which the former has a much lower resonance defect and, therefore, this process should affect the shape of the curve. The experimental $\sigma(v)$ curve for CO^{+*} ions which should have a maximum near

1)The only discrepancy is the presence of the maximum on the $\sigma(v)$ curve for the production of O^+ ions by Ne^+ ions near 2×10^7 cm/sec. This maximum is not present on the corresponding curve given in [5].

2)In these expressions ΔE is the resonance defect for the process, a is the range of the interaction between colliding particles, and h is Planck's constant.

$v = 1.2 \times 10^8 - 1.3 \times 10^8$ cm/sec does, in fact, confirm this prediction.

The shape of the $\sigma(v)$ curves for the production of the C^+ and O^+ ions is determined by processes 3, 4, 7, and 8. Of these, process 3 has much the smallest resonance defect and corresponds to small values of v_{max}. One would therefore expect that, in the corresponding velocity range, the cross sections for the production of these ions should increase with decreasing incident particle velocity. This is confirmed by experimental $\sigma(v)$ curves for these ions (Fig. 1a).

The cross section for the production of the excited ions C^{+*} and O^{+*} include contributions due to processes 7 and 8. Processes 7b and 8b, which determine the shape of the $\sigma(v)$ curve for O^{+*} ions have v_{max} respectively equal to 2.3×10^8 and 4.1×10^8 cm/sec. This means that, in this particular velocity range, the quantity $\sigma_{O^{+*}}$ should increase monotonically with increasing incident-particle velocity and this is, in fact, confirmed experimentally (Fig. 1b).

The shape of the $\sigma(v)$ curve for C^+ ions is determined by processes 7a and 8a. The former has a much smaller resonance defect. Process 7a corresponds to $v_{max} = 1.35 \times 10^8$ cm/sec and, consequently, the experimental $\sigma(v)$ curve for C^+ ions should have a maximum in the region of this velocity. This is also confirmed by experiment.

The particular feature of the $\sigma(v)$ curve for the excitation of the 3888 Å line of He I, determined by process 9, is the presence of the maximum at a velocity of 4×10^7 cm/sec. According to the adiabatic maximum rule, this curve should have a maximum at a velocity of 2.1×10^8 cm/sec. Therefore, the maximum at $v = 4 \times 10^7$ cm/sec lies in the adiabatic region of the $\sigma(v)$ curve for the production of the excited helium atoms.

Ne^+-CO

The shape of the $\sigma(v)$ curve for CO^+ ions (Fig. 2) shows that there should be a maximum at velocities that are not much greater than 5.5×10^7 cm/sec (this velocity corresponds to the maximum energy of the Ne^+ ions used in the experiment). It is quite probable that this maximum is connected with the production of the CO^{+*} ions in process 5, which has a low resonance defect ($v_{max} \sim 8 \times 10^7$ cm/sec). Process 6, which has a much greater resonance defect ($v_{max} = 2.8 \times 10^8$ cm/sec), appears to provide a small contribution to the cross section for the production of CO^{+*} ions in this particular velocity range.

The $\sigma(v)$ curve for CO^+ ions should have a maximum near 0.7×10^8 cm/sec. This maximum cannot be connected either with process 1 ($v_{max} = 1.8 \times 10^8$ cm/sec) or process 2 ($v_{max} = 2.4 \times 10^8$ cm/sec). The most likely situation is that this maximum is due to the predominant effect of process 5 on the cross section for the formation of CO^{+*} ions.

Some of the features of the cross section for the production of C^+ and O^+ ions as functions of the Ne^+ ion velocity (Fig. 2a) can be explained by the large contribution of processes 3a and 3b which have low resonance defects with $v_{max} = 0.6 \times 10^7$ and $v_{max} = 2 \times 10^7$ cm/sec, respectively. This can be used to explain the rising branch of the $\sigma(v)$ curve at low velocities for C^+ ions and the maximum near 2×10^7 cm/sec of the cross section curve for O^+ ions. The branch of the $\sigma(v)$ curve which rises with increasing velocity in the case of C^+ ions, and the shape of $\sigma(v)$ curve for O^+ ions at velocities in excess of 5×10^7 cm/sec depend on the following facts: (1) the increasing contribution of processes 7 and 8 for the production of C^{+*} and O^{+*} ions to the total cross section for the production of C^+ and O^+ ions, and (2) the presence of maxima at 1.6×10^8 and 1.8×10^8 cm/sec which are connected with processes 4a and 4b.

The shape of the $\sigma(v)$ curves for C^{+*}, O^{+*}, and Ne^{+*} shows that there is no rapid reduction in the cross section in the adiabatic collision region.

It is clear from Fig. 2b that the cross section curve for the excitation of Ne* atoms has a maximum at 2×10^7 cm/sec. The maximum for process 9, which determines the shape of the $\sigma(v)$ curve for Ne* atoms, should lie at about 1.9×10^8 cm/sec. Therefore, this maximum is located in the adiabatic region of the $\sigma(v)$ curve.

Ar^+-CO

Process 1 has a low resonance defect ($v_{max} = 3 \times 10^7$ cm/sec) and provides the main contribution to σ_{CO^+}, so that the maximum on the $\sigma_{CO^+}(v)$ curve near 2×10^7 cm/sec can be explained in terms of the adiabatic maximum rule. This is also the case for the maximum on the $\sigma_{CO^+}(v)$ curve since process 5 provides the main contribution to this cross section. The shape of the $\sigma(v)$ curves for C^+ and O^+ ions is in accordance with the fact that maxima connected with processes 3a and 3b, which provide the main contribution to the cross section for the production of these ions, lie at 5×10^7 and 6.5×10^7 cm/sec, respectively.

The cross sections for the production of C^{+*} and O^{+*} ions are very small because of the high values of the resonance defects for processes 7 and 8, which are responsible for the production of these ions. This is why one cannot detect the emission due to O^{+*} ions. For the C^{+*} ions the velocity range which we investigated is, in fact, the adiabatic range. However, we did not observe a rapid fall in the cross section $\sigma_{C^{+*}}$ with decreasing velocity. The maximum on the $\sigma(v)$ curve for the production of excited Ar^+ ions was found to lie in the adiabatic region of this curve.

The above results lead us to the conclusion that many of the features of the $\sigma(v)$ curves which we have investigated can be explained in terms of the adiabatic maximum rule. However, the shape of the $\sigma(v)$ curves at low velocities departs from the predictions of the adiabatic hypothesis. Instead of the monotonic rapid fall in the effective cross section with reducing velocity, we observe either additional maxima[3)] or a slower fall of the cross section with decreasing incident particle energy. Our analysis has shown that the deviation of the $\sigma(v)$ curves from the predictions of the adiabatic hypothesis cannot be explained by the presence of metastable ions in the incident particle beam. These

[3)]The phrase additional maximum refers to a maximum which cannot be explained in terms of the adiabatic maximum rule.

anomalies in the adiabatic region of the $\sigma(v)$ curves can, in principle, be explained by the intersection of the potential-energy surfaces for the initial and final states of the colliding particle system.

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