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DOUBLE CHARGE EXCHANGE BETWEEN SINGLY-CHARGED POSITIVE IONS AT LOW ENERGIES

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The effective cross sections for double charge exchange of H^+, Li^+, Na^+, and K^+ ions in several gases, in the energy range from 150 to 6000 eV, were measured using a method which made it possible to allow experimentally for the error associated with a difference in scattering of the primary and secondary particles.[11] The data of the present paper, together with those of earlier investigations[8, 12] of the same ion—molecule and ion—atom pairs made it possible to establish the form of the function $\sigma_{1-1}(v)$ over a fairly wide range of velocities, beginning from a velocity close to the threshold value and ending at a velocity close to v_{max} (v_{max} is the velocity at which the cross section reaches its maximum value). For the majority of the investigated ion—molecule and ion—atom pairs, a typical behavior of the $\sigma_{1-1}(v)$ function was observed, namely: near v_{max}, the cross section σ_{1-1} decreased exponentially when the ion velocity decreased, but in the pre-threshold region, the cross section σ_{1-1} decreased slowly when the velocity was decreased. It was found possible to describe the $\sigma_{1-1}(v)$ curve by a single empirical formula over the whole range of velocities considered here.

INTRODUCTION

HASTED[1] was among the first to investigate the function $\sigma(v)$ for inelastic processes in the range of slow (adiabatic) collisions. In his investigations, Hasted attempted to establish a qualitative agreement between his experimental data and the adiabatic theory([2], §87), which predicts that, in the range of velocities not too close to the threshold value and sufficiently far from the velocity corresponding to the maximum value of the cross section, the velocity dependence of the inelastic cross section is given by the formula

$$\sigma(v) \sim e^{-c/v}, \qquad (1)$$

where c is a constant.

By analyzing the experimental data on the cross section σ_{10} for the capture of a single electron by singly charged positive ions, Hasted established that, if the reaction energy ΔE was not too low, the curve $\sigma_{10}(v)$ could be approximated by the function (1). He also pointed out that such a dependence was observed only in a certain range of velocities, namely, in the range close to the velocity v_{max}. The formula (1) was not obeyed when the velocity was reduced and when the cross sections σ_{10} were low (10^{-18} – 10^{-19} cm^2); and value of the cross section σ_{10} decreased much more slowly than predicted by the law given above.

It is necessary to mention that the function $\sigma(v)$ has the same properties in the processes of excitation and ionization of atoms by ion impact, where again a region of rapid decrease is found, as well as a region of slow decrease at low velocities.[3, 4] Similar observations have been made also when an electron is captured by a neutral atom.[5] This suggests that such a behavior of the function $\sigma(v)$ is common to all inelastic processes. However, this conclusion must be justified more rigorously.

The main difficulty in the experimental proof of this conclusion was that the methods used by a number of investigators[1, 3-5] were based on the observation of a change in state of only one of the two interacting particles, and all the possible states of the particle not investigated were ignored altogether. The result of a quantitative analysis of such observations, from which the cross section of the investigated process is calculated, may represent the sum of contributions of all the possible ways in which the observed particle can be formed, which, naturally, gives a distorted representation of the actual form of the function $\sigma(v)$ corresponding to a particular variant of the process. Hence, there are some doubts as to whether the departure from the formula (1), reported in [1, 3-5], is due to the characteristic features of the inelastic interactions, because the same departure may be explained by the imperfection of the method used.

Moreover, the cited papers (except for [4]) report the data for processes with a relatively small resonance defect. In such a case, there may be a pseudo-intersection of the potential curves of the initial and final states of a system of colliding particles, which may also distort the form of the function $\sigma(v)$ in the investigated range of velocities.

It follows that an investigation of the function $\sigma(v)$ for inelastic atomic collisions in the adiabatic region requires, first of all, an improved method capable of isolating a given process.

However, we can point out another method of investigation based on the fact that, in some processes, either only a single variant is possible or the contribution of other possible channels is slight. To investigate the behavior of the function $\sigma(v)$ in the adiabatic region, we selected one of these processes: the double charge-exchange of singly charged positive ions. The distinguishing characteristic of this process is that the negative ion formed by such an exchange cannot be in an excited state,[6, 7] and, therefore, we can select ion–atom or ion–molecule pairs such that the process will be free of slow and fast excited particles. In other words, the process will take place in accordance with the only possible variant and there will be no distortion of the $\sigma_{1-1}(v)$ curve.

Another feature of this process is the relatively large resonance defect, reaching several tens of electron-volts, for some ion–atom pairs. Therefore the potential curves of the initial and final states of the system are unlikely to approach one another at those internuclear distances which are reached in the adiabatic collisions. Moreover, the high value of the resonance defect ensures the adiabaticity of collisions at high values of the colliding-particle velocities, which makes the investigation of the adiabatic region much easier.

In the case of a two-electron charge-exchange, the distortion of the $\sigma_{1-1}(v)$ curve in the adiabatic region due to the formation of slow excited ions may not appear in those cases when the additional maxima, associated with these processes, are far from the main maximum in the high-velocity region, and the investigated region of velocities lies in the low-velocity region, sufficiently far from the main maximum.

Hence, we may conclude that in the investigation of the dependence $\sigma_{1-1}(v)$ in the adiabatic region, the most reliable results can be obtained in the case of two-electron charge-exchange of protons or alkali-metal ions (obtained from a thermionic source) in hydrogen, helium, and other inert gases.

The first paper[8] on the function $\sigma(v)$ for the double charge-exchange of positive alkali-metal ions reported data which were in qualitative agreement with the results of other investigators.[1, 3-5] However, this paper gave only one $\sigma_{1-1}(v)$ curve (for the K^+–Ne pair) suitable for judging the behavior of the function $\sigma(v)$ in the adiabatic region, which, naturally, was quite insufficient. In view of this, attempts have been made to carry out measurements using a larger number of ion–atom and ion–molecule pairs at considerably lower velocities.[9] However, the method used[10] did not make it possible to measure the values of the σ_{1-1} cross sections less than 10^{-20} cm^2. Therefore, Kozlov et al.[9] were only able to measure the cross sections for the pairs H^+–H_2, H^+–Ar, H^+–Kr, which had σ_{1-1} values greater than 10^{-20} cm^2 in the energy range from 5000 to 500 eV.

Of the results obtained, only the data for the H^+–H_2 pair could be assigned to the adiabatic collision region. It was found that the $\sigma_{1-1}(v)$ curve for this pair was, in the investigated range of velocities, in complete agreement with Eq. (1), predicted by the adiabatic theory. Thus, it followed from [8, 9] that the function $\sigma_{1-1}(v)$ was different for the K^+–Ne and H^+–H_2 pairs. This difference could be only apparent, because the $\sigma_{1-1}(v)$ curve for the H^+–H_2 pair was not investigated experimentally in the range of velocities with the same values of the adiabatic parameter $a|\Delta E|/hv$,[1] as the $\sigma_{1-1}(v)$ curve for the K^+–Ne pair. It was also necessary to investigate more extensively the $J^+ \rightarrow J^-$ process in the adiabatic region (J^+ and J^- represent currents of positive and negative ions) in order to accumulate sufficient experimental data to establish reliably the form of the $\sigma_{1-1}(v)$ dependence in the adiabatic region.

The present investigation was intended to solve this problem

RESULTS OF MEASUREMENTS

The measurements of the σ_{1-1} cross sections were carried out for the following pairs:

H^+ — Ne, Li^+ — Ar, Na^+ — Ar,

H^+ — H_2, Li^+ — Kr, Na^+ — Kr, K^+ — Ne,

H^+ — He, Li^+ — H_2, Na^+ — H_2.

We used a method which allowed experimentally for the error associated with the difference in the scattering of the primary and secondary particles.[11] The energy range, in which the measurements of the σ_{1-1} cross sections were carried out, was different for each pair. The upper limit was 6000 eV and the lower limit was governed by the value of the random measurement error and was equal to the energy at which this error did not exceed 20%. We must mention that the value of the random error was governed mainly by the ratio J^-/J^-_r (J^- and J^-_r are the negative ion currents from charge exchange in the investigated and residual gases). At low energies (1000 eV or less), the value of J^-_r was practically constant and depended very weakly on the residual gas pressure in the charge-exchange chamber.

Before discussing the results, we must mention the degree of reliability of our data, which should be high in investigations of this type. Certain conclusions about this may be drawn by considering, as an example, the $\sigma_{1-1}(v)$ curve for the charge-exchange of protons in hydrogen (Fig. 1, upper curve). Figure 1 gives the results of earlier investigations[9, 12] and of the present study. In spite of the fact that in these three investigations, the value of σ_{1-1} was measured by different methods, the results were in good agreement. This indicated that the results of the present investigation were reliable and, consequently, they were suitable for drawing conclusions about the real behavior of the $\sigma_{1-1}(v)$ function in the investigated range of velocities.

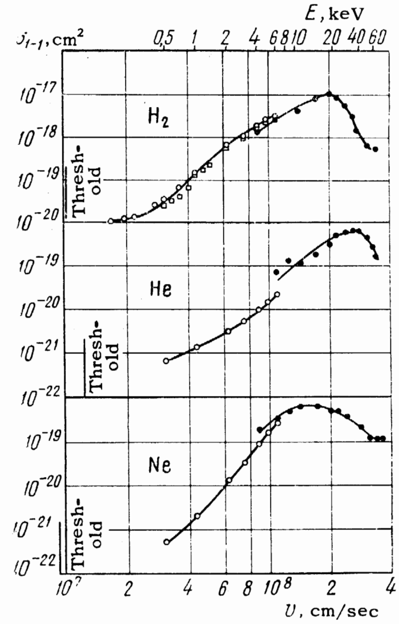

FIG. 1. $H^+ \rightarrow H^-$ process in hydrogen, helium, and neon: ○ – data of the present investigation; □ – [9]; ● – [12].

An examination of the data shows that the $\sigma_{1-1}(v)$ curves for the Li^+–H_2 and Na^+–H_2 pairs differ considerably from all the other curves (Fig. 2). The difference lies in the presence of a maximum in the $\sigma_{1-1}(v)$ function for these two pairs at low velocities. The presence of this maximum cannot be explained by the participation of excited particles in the charge-exchange process. We cannot assume either that this maximum is associated with the intersection of the potential curves of the initial and final states of the system, because the investigated process is known to have a relatively large resonance defect. Evidently, the observed behavior of the $\sigma_{1-1}(v)$ function of the Li^+–H_2 and Na^+–H_2 pairs should be associated

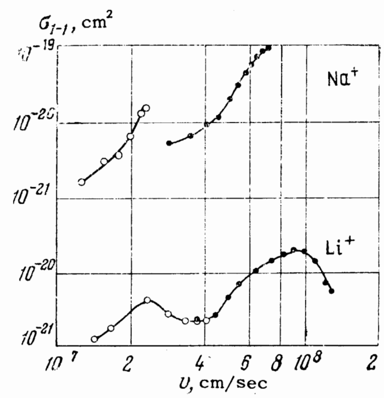

FIG. 2. $Li^+ \rightarrow Li^-$ and $Na^+ \rightarrow Na^-$ processes in hydrogen: ○ – data of the present investigation; ● – [8].

[1]Here, a is the effective radius of interaction, ΔE is the resonance defect, h is Planck's constant, and v is the relative velocity of the colliding particles.

with the specific features of the interaction mechanism, which are characteristic of these pairs only.

It should be mentioned that similar behavior was observed for the $\sigma(v)$ function in the formation of metastable hydrogen atoms in collisions of protons with a rarefied gas.[13] Jaecks, Van Zyl, and Geballe[13] suggested that, in the investigated range of velocities, there were two different mechanisms of the formation of metastable hydrogen atoms. The appearance of an additional maximum was ascribed to a multistage mechanism of the formation of metastable hydrogen atoms, with the colliding particles forming a short-lived molecule, which dissociated into various fragments among which were the metastable hydrogen atoms.

The behavior of the $\sigma_{1-1}(v)$ curves for the processes $Li^+ \rightarrow Li^-$ and $Na^+ \rightarrow Na^-$ in hydrogen can be explained in the same way. For this purpose, it is necessary to assume that the colliding particle pairs form quasimolecules which dissociate into fragments, including negative Li^- and Na^- ions, and that the probability of such a multistage mechanism of the Li^- and Na^- ion formation is maximum at velocities corresponding to the positions of the additional maxima in the $\sigma_{1-1}(v)$ curves.

The explanation of the additional maxima in the $\sigma(v)$ curves at low velocities, given in [13], is only a hypothesis, requiring rigorous quantitative proof.

We shall now consider the results of measurements of the effective cross sections σ_{1-1} for other pairs, given in Figs. 1 and 3–5.

First of all, we must stress the qualitative agreement between the the $\sigma_{1-1}(v)$ curves, obtained in the present investigation, and the curves for other inelastic processes described in [1, 3–5]. In fact, the region of rapid decrease in the $\sigma_{1-1}(v)$ curves obtained in the present investigation may be approximated by a function of the Eq. (1) type, in the same way as the region of rapid decrease in the $\sigma(v)$ curves reported in [1, 5]. This can be seen clearly in Figs. 6–9, which give the dependences of $\log \sigma_{1-1}$ on the value of $1/v$ for various ion–molecule pairs. It is evident that the points in the region of rapid decrease in the $\sigma_{1-1}(v)$ curves well fit a straight line, indicating that the behavior of the σ_{1-1} cross section in this range of velocities is described by Eq. (1).

Furthermore, in the region of pre-threshold values of the velocity, there is a systematic departure from this dependence in the direction of larger cross sections, which is similar to the behavior observed for the σ_{1-1} curves reported in [1, 5].

The similarity of the behavior of the $\sigma(v)$ function for such very different inelastic processes allows us to conclude that this behavior is a general

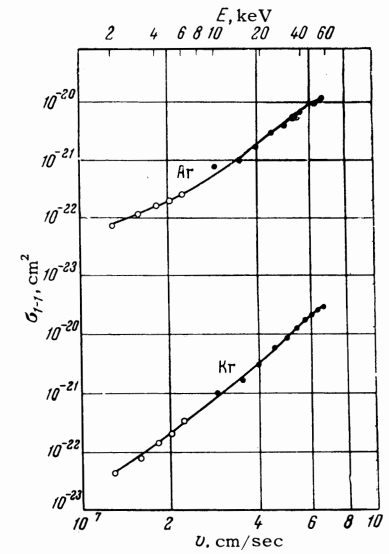

FIG. 4. $Na^+ \rightarrow Na^-$ process in argon and krypton: ○ – data of the present investigation; ● – [8].

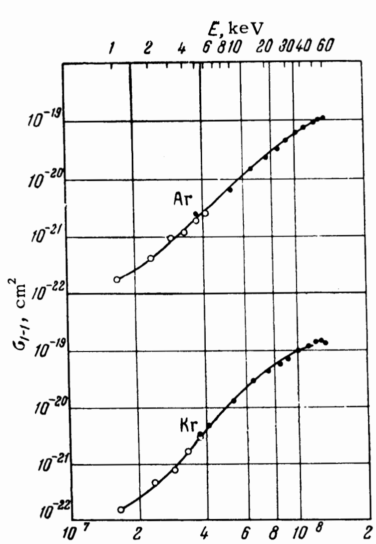

FIG. 3. $Li^+ \rightarrow Li^-$ process in argon and krypton: ○ – data of the present investigation; ● – [8].

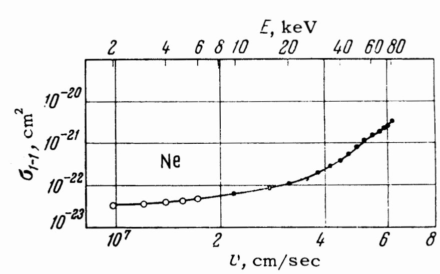

FIG. 5. $K^+ \rightarrow K^-$ process in neon: ○ – data of the present investigation; ● – [8].

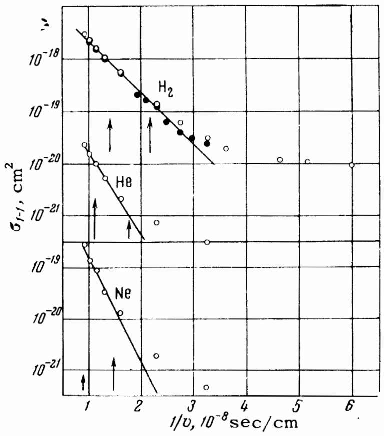

FIG. 6. Dependence of σ_{1-1} on 1/v for the $H^+ - H_2$, $H^+ - He$, $H^+ - Ne$ pairs: o – data of the present investigation; ● – [8].

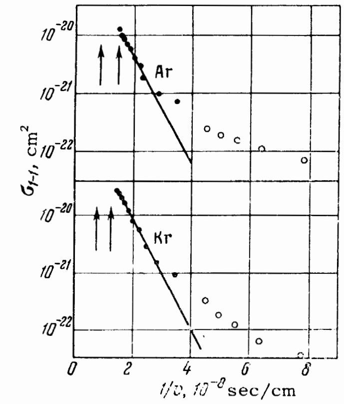

FIG. 8. Dependence of σ_{1-1} on 1/v for the $Na^+ - Ar$ and $Na^+ - Kr$ pairs: o – data of the present investigation; ● – [8].

characteristic of all possible elementary inelastic interactions. In view of this, it becomes interesting to find to what extent the behavior of the $\sigma(v)$ function agrees with the theory describing inelastic collisions in the adiabatic region.

From theoretical considerations of the mechanism of inelastic processes in slow (adiabatic) collisions, it follows that:

1) The effective cross section for a collision between two particles with an initial relative velocity v, in which the internal energy changes by ΔE, is small compared with the gas-kinetic cross section if

$$a|\Delta E| / hv \gg 1. \tag{2}$$

2) At constant values of ΔE and a, the effective cross section increases rapidly as the relative velocity increases until $a|\Delta E|/hv$ reaches a value of the order of unity.[14]

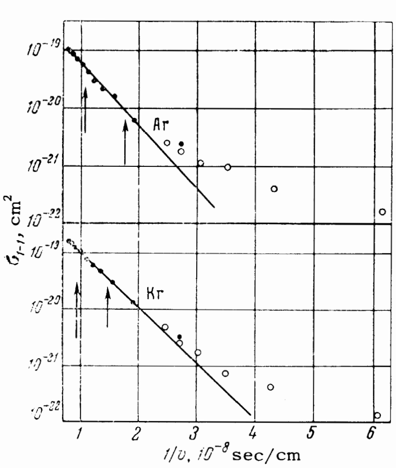

FIG. 7. Dependence of σ_{1-1} on 1/v for the $Li^+ - Ar$ and $Li^+ - Kr$ pairs: o – data of the present investigation; ● – [8].

To these conclusions, we must add that the calculations carried out within the framework of the so-called adiabatic perturbation theory[2] give a dependence of the transition probability on the relative velocity in the form

$$f(v)\, e^{-k/v},$$

where f(v) is a slowly varying function, and k is a constant.

The criterion of the applicability of this theory is the condition (2). Therefore, it would seem that, in principle, the theory may be applied to any inelastic process with not too low a value of the resonance defect, since for such processes we can always find a range of velocities at which the condition (2) would be satisfied. However, Bates et al.[15, 16] showed that the condition (2) was not satisfied for short distances of approach between interacting particles and, therefore, the head-on and near-head-on collisions were not adiabatic.

Smirnov[17] calculated the dependence of the cross section of an inelastic process on the relative velocity of the colliding particles, allowing for a departure from the condition (2) at short distances between interacting atoms. The results of his calculations show that the function $\sigma(v)$ con-

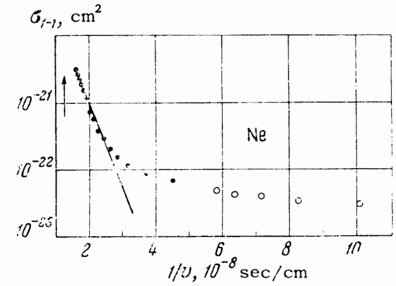

FIG. 9. Dependence of σ_{1-1} on 1/v for the $K^+ - Ne$ pair: o – data of the present investigation; ● – [8].

sists of two parts. The first part, in the velocity range[2)]

$$v > (\Delta E)^{1/2}(Z_1Z_2/\mu)^{1/4} \equiv v_0,$$

decreases with velocity as v^8 (for the s–s transitions), while the second part is a monotonic function of the velocity and is constant for the s–s transitions. Therefore, the $\sigma(v)$ curve decreases rapidly and, at $v \approx v_0$, becomes either a monotonic function of the velocity or tends to a constant value. Smirnov[17] has shown that the range of velocities where the dependence $\sigma(v)$ should become weaker lies between the limits v_1 and v_2, where

$$v_1 = \left(\frac{\Delta E Z_1 Z_2}{\mu}\right)^{1/3}, \quad v_2 = (\Delta E)^{1/2}\left(\frac{Z_1Z_2}{\mu}\right)^{1/4}.$$

In this range of velocities, the cross section for the s–s transitions is proportional to $1/\mu^2$.

It follows from what has been said above that the experimental data of the present measurements confirm qualitatively the main conclusions of the theoretical calculations of Smirnov.[17] In fact, the experimental curves of $\sigma_{1-1}(v)$ exhibit, in agreement with the theoretical prediction, a region of rapid decrease which, when v decreases, changes smoothly into a region of slow decrease (cf. Figs. 6–9). Hence, it follows that in the double charge-exchange of singly charged positive ions, the nuclei approach each other to distances at which the condition for adiabatic collisions is not obeyed. This departure from the adiabatic condition, predicted by the theory[17] and confirmed experimentally, causes a departure of the function $\sigma(v)$ from Eq. (1), which was obtained from the adiabatic perturbation theory.

We must, however, mention that the behavior of the $\sigma_{1-1}(v)$ curve in the region of rapid decrease does not agree with the v^8 law, which was predicted theoretically.[17] In fact, in this range of velocities, the $\sigma_{1-1}(v)$ curve is well represented by the function (1) (cf. Figs. 6–9). Moreover, the transition of the $\sigma_{1-1}(v)$ function from the region of rapid decrease to that of slow decrease occurs at lower velocities than those predicted by Smirnov.[17] This can be seen clearly in Figs. 6–9, where the arrows indicate the range of velocities within which the slow variation of σ_{1-1} with velocity should be observed. An examination of Figs. 6–9 shows that the velocity at which the slow decrease of σ_{1-1} begins differs from the theoretically predicted value and the larger the masses of the colliding particles, the greater is this difference.

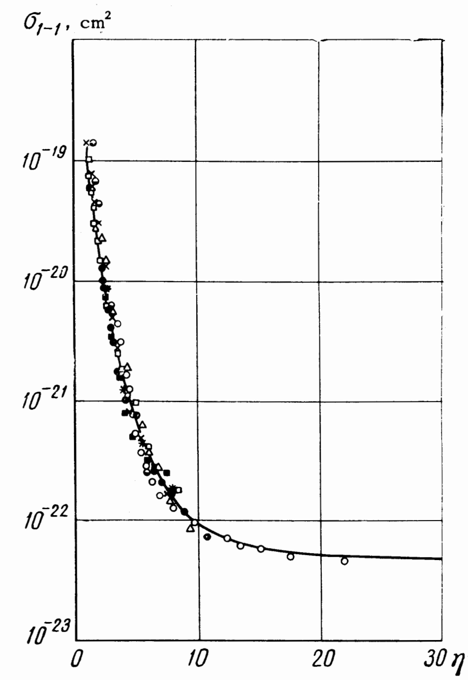

FIG. 10. Dependence of $\sigma_{1-1}(\eta)$ for the pairs: ■ – $H^+ - H_2$; * – $H^+ - He$; ◒ – $H^+ - Ne$; □ – $Li^+ - Ar$; × – $Li^+ - Kr$; ● – $Na^+ - Ar$; △ – $Na^+ - Kr$; ○ – $K^+ - Ne$.

Hasted[1] stressed that the rate of rise of the $\sigma_{10}(\eta)$ curve ($\eta = a|\Delta E|/hv$ for the processes $J^+ \rightarrow J^0$, characterized by not too small a value of the resonance defect, was independent of the nature of the partners in a pair of colliding particles. A similar conclusion may be drawn by considering the data on the $J^0 \rightarrow J^-$ processes which were reported by Fogel'.[5] In view of this, it is interesting to find to what extent the same conclusion is justified in the case of the $J^+ \rightarrow J^-$ process. A special analysis of the experimental data made it possible to select a function by means of which the $\sigma_{1-1}(v)$ curves could be approximated for any investigated pair (except the Li^+-H_2 and Na^+-H_2 pairs) over the whole investigated range of velocities. If the adiabatic parameter η is used as the variable, this function will have the form

$$\sigma_{1-1} = \sigma_0 \exp\left[Ae^{-c\eta}\right], \tag{3}$$

where σ_0, A and c are constants.

The continuous curve in Fig. 10 shows the form of the function $\sigma_{1-1}(\eta)$, calculated using Eq. (3). The values of the coefficients σ_0, A and c were obtained by analyzing the experimental data for the Na^+-Ar pair. These coefficients were found to be 5×10^{-23} cm², 10, and 0.282, respectively. Figure 10 includes also the experimental data for all the investigated pairs. The $\{\sigma_{1-1}(\eta)\}_m$ curve for a given pair m was made to coincide at some point

[2)]The atomic system of units is used; Z_1 and Z_2 are nuclear charges, and μ is the reduced mass.

η with the calculated curve $\{\sigma_{1-1}(\eta)\}_{Na^+-Ar}$, the conversion coefficient

$$k_m = \{\sigma_{1-1}(\eta)\}_m / \{\sigma_{1-1}(\eta)\}_{Na^+-Ar}$$

was found and then the values of the quantity $(\sigma_{1-1})_m$, multiplied by the corresponding value of the coefficient k_m, were plotted in the figure. It is evident from Fig. 10 that the experimental points lay quite near the calculated curve over the whole range of the parameter η, which varied from 1 to 22. The good agreement between the experimental data and the calculated curve indicated that the law of the decrease of the $\sigma(\eta)$ curve when η increased was the same for all the pairs considered.

Thus, the processes

$$J^+ \to J^-, \quad J^+ \to J^0, \quad J^0 \to J^-$$

are represented by a single relationship which states that in these processes the rate of rise of the $\sigma(\eta)$ curve as the value of η falls is independent of the nature of the partners in a pair of colliding particles. In the case of the $J^+ \to J^-$ process, this means that the coefficients A and c, which occur in Eq. (2), are the same for all the investigated pairs and are, on the average, equal to 10 and 0.282, respectively. This makes Eq. (3) very convenient for the extrapolation of given effective cross sections σ_{1-1}, measured near a maximum, to a region of considerably lower velocities, where experimental measurements are exceptionally difficult.

It should be mentioned that Eq. (3) does not reflect the behavior of the function $\sigma_{1-1}(v)$ near the threshold of the process because, even when $v = 0$, it gives a non-zero result. Nevertheless, as shown in Fig. 10, the theoretical curve describes the actual $\sigma_{1-1}(v)$ curve over quite a wide range of values of η. An examination of the curve in Fig. 10 shows that the function $\sigma_{1-1}(\eta)$ changes from a region of rapid decrease to one of slow decay at $\eta \approx 4$ for all the investigated beams.

It is interesting to find how far the data of the present investigation are in agreement with the theoretical conclusion,[17] according to which the following relationship should apply in the region of slow variation in $\sigma(v)$:

$$(\sigma_{1-1})_i / (\sigma_{1-1})_k = (\mu_k / \mu_i)^2. \quad (4)$$

(Here, $(\sigma_{1-1})_i$ and $(\sigma_{1-1})_k$ are the values of the effective cross sections for the i-th and k-th pairs of colliding particles, respectively; μ_k and μ_i are the reduced masses of these particles.) Since the nature of the dependence $\sigma_{1-1}(\eta)$ is the same for all the investigated pairs, the condition (4) is equivalent to the condition

$$(\sigma_0)_i / (\sigma_0)_k = (\mu_k / \mu_i)^2. \quad (5)$$

An analysis of the experimental data showed that the value of the coefficient σ_0 changed markedly from one pair to another. Both the incident ion and the target atom governed the value of this coefficient. However, the law of variation of the quantity σ_0 from one pair to another was not in agreement with the condition (5). It was found that the value of σ_0 decreased when the binding energy of an electron to a negative ion decreased. For a given ion, i.e., for a constant binding energy of electrons to a negative ion, the value of the coefficient σ_0 decreased when there was an increase in the binding energy of electrons in the particles which lost electrons. Unfortunately, the small number of the investigated pairs prevented us from establishing the law of the dependence of the quantity σ_0 on the binding energy of electrons in the negative ion and on the binding energy of electrons in the particle which lost electrons. Therefore, it would be desirable to carry out such measurements for a larger number of colliding particle pairs.

In conclusion, the authors express their gratitude to Ya. M. Fogel' for his valuable advice and help in this investigation.

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