Ionization and capture of electrons in collisions of Na⁺ ions with Ne atoms

Z. Z. Latypov and A. A. Shaporenko

A. F. Ioffe Physico-technical Institute, USSR Academy of Sciences
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Experimental results are presented of an investigation of the charge exchange $Na^+ + Ne \rightarrow Na + Ne^+$ and ionization $Na^+ + Ne \rightarrow Na^+ + Ne^+ + e$ in the collision energy range from thresholds to 450 eV in the c.m.s. The experimentally determined energy threshold of the investigated processes, the structures of the effective cross sections, and the mechanisms of the inelastic processes are discussed in light of the molecular-orbital model and of a diabatic correlation diagram, constructed by the authors, of the terms of the $(NaNe)^+$ quasimolecule.

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1. Inelastic collisions of Na⁺ ions with Ne atoms have attracted attention because of many singularities of this system. Of great interest among them are effects of quantum-mechanical phase interference of charge-exchange and excitation processes, and also polarization of the optical radiation produced in the Na⁺ Ne collisions. In [1-4] these effects are explained on the basis of the molecular-orbital model, and the relations between them are explained. Nevertheless, many details of these phenomena are not yet clear and further experimental investigations of the inelastic processes in the (naNe)⁺ system are needed for an unambiguous quantitative description.

In the analysis of the experimental data on inelastic collisions of heavy nuclei, frequent use is made of the diabatic correlation phase diagram of the intermediate quasimolecule; this diagram is constructed on the basis of the molecular-orbital model. [5] To estimate the quality of these diagrams, great interest attaches to a comparison of the predictions of the diagrams with the experimental data. From this point of view it is of interest to investigate in detail the process of ionization in the (NaNe)⁺ system, in which the total cross section of these processes at low energies, including the thresholds, has not yet been investigated.

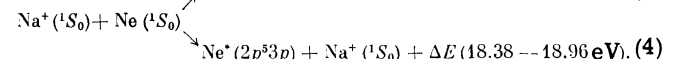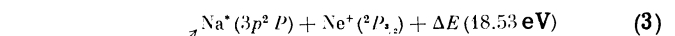We have studied experimentally the charge-exchange and ionization processes

$$Na^+ + Ne \nearrow^{Na + Ne^+}_{\searrow Na^+ + Ne^+ + e,} \quad (1)(2)$$
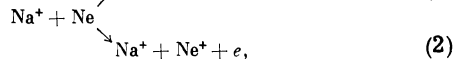
constructed the correlation diagram of certain levels of the (naNe)⁺ system, and discussed the obtained data.

2. The effective cross section of process (1) is the sum of the cross sections of the charge-exchange processes in the ground and excited states of the Na atoms. This cross section was determined in the energy interval from the thresholds of the processes to 450 eV (the energy is in the c.m.s.) as the difference between the combined cross section of processes (1) and (2), measured by the method proposed in [6], and the cross section of process (2), measured by the method described in [7]. Curve A of Fig. 1 is the combined cross section of processes (1) and (2), while curve B is the cross section of process (2). The characteristic mean-squared measurement errors are indicated on the curves in the form of vertical bars. The absolute values of the cross sections were determined accurate to a factor of two.

It follows from Fig. 1 that the cross section of the charge exchange (1) oscillates. The maxima of the oscillations are equidistant in the scale of the reciprocal collision velocities v^{-1}. The average distance between the maxima is $\Delta(v^{-1}) \approx 3.0 \times 10^{-8}$ sec/cm. In the energy interval considered here, the fundamental oscillation frequency of the process (1) is close in magnitude to the oscillation frequency of the cross section for light emission from the Na⁺(3p) atoms (and from certain levels of the Ne*(3p) atom), excited in the Na⁺-Na collisions, [3,4] and is in phase with the oscillation of the Na*(3p) emission cross section. It can therefore be concluded that the oscillation of the combined cross section (1) with the indicated frequency is due to oscillation of the cross section for the charge exchange of the Na⁺ ion into the excited state: Na*(3p). It is shown in [3,4] that oscillation of the total cross sections of emission of light by the Na*(3p) atoms and certain states of the Ne*(3p) atoms can be explained within the framework of the Rosenthal-Foley method [8] and is due mainly to interference between the (NaNe)⁺-quasimolecule states corresponding to the charge-exchange and excitation processes:

$$Na^+(^1S_0) + Ne\,(^1S_0) \nearrow^{Na^*(3p^2\,P)\,+\,Ne^+(^2P_{,})\,+\,\Delta E\,(18.53\,\text{eV})}_{\searrow Ne^*(2p^53p)\,+\,Na^+\,(^1S_0)\,+\,\Delta E\,(18.38\,-\,18.96\,\text{eV}).} \quad (3)(4)$$

To discuss the mechanisms of the inelastic processes in the (NaNe)⁺ system, and also the causes of the structures that appear on the cross sections of these processes, we consider the diabatic correlation diagram of some terms of the quasimolecule (NaNe)⁺. Figure 2 shows this diagram, which was constructed by us on the basis of the (NaNe)⁺ molecular-orbital diagram given in [3]. The zero energy level on the diagram (Fig. 2) is taken to be the energy of the diabatic term of the Σ state of (NaNa⁺), which dissociates into Na⁺ + Ne and corresponds to the initial state of the colliding particles. For the sake of simplicity, the diagram does not show the quasimolecule Σ terms that dissociate into Na⁺(4s) + Ne⁺ and Na⁺ + Ne*(4s), which cross the ground-state Σ term at $R \approx 1.2$ at. un. and $R \approx 0.9$ at. un. respectively (the Π terms of these quasimolecule states approach the ground-state term at much lower R). For the same purpose, the electron configurations of the limit of the unified atom are not indicated for all the presented terms. To describe the course of the terms corresponding to certain auto-ionization states of the Ne atom, plots of the Σ and Π states are given for (NaNe)⁺ with electron configuration $1s^22s^22p^43s3p$ and $1s^22s2p^63s$ of the Ne atom. It should be noted that the (NaNe)⁺ energy
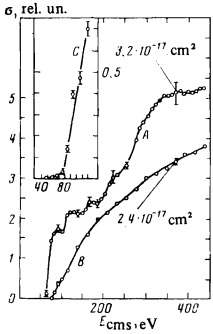
FIG. 1   FIG. 2

FIG. 1. Curve A is the combined cross section of processes (1) and (2), curve B the cross section of process (2), and curve C the threshold part of cross section (2).

FIG. 2. Diabatic correlation diagram of the $(NaNe)^+$ levels. The solid lines show the levels of the Σ states, and the dashed ones those of the Π states. Na^+ and Ne correspond to the ion and atom in the ground states: A_1–$Na^+ + Ne$, B_1–$Na + Ne^+$, C_1–$Na^+ + Ne^*$ (3s), D_1–Na^* (3p) + Ne^+, E_1–$Na^+ + Ne^*$ (3p), F_1–Na^* (4s) + Ne^+, H_1–$Na^+ + Ne^+$, K_1–$Na^+ + Ne^{**}(1s^2 2s2p^6 3s)$, L_1–$Na^+ + Ne^{**}(1s^2 2s^2 2p^4 3s3p)$, A_2–Sc^+ $(1s^2 2s^2 2p^6 3p^2 3d^6 4f^2)$, B_2–$Sc^+(1s^2 2s^2 2p^6 3s3p 3d^5 4f^2)$, C_2–Sc^+ $(1s^2 2s^2 2p^6 3p^2 3d^6 4p4f)$, D_2–$Sc^+(1s^2 2s^2 2p^6 3p^2 3d^6 4d4f)$, D_3–Sc^+ $(1s^2 2s^2 2p^6 3p^2 3d^6 4f)$, H_2–$Sc^{+*}(1s^2 2s^2 2p^6 3p^2 3d^6 4f)$, H_3–Sc^{++} $(1s^2 2s^2 2p^6 3p^2 3d^5 4f^2)$, K_2–$Sc^+(1s^2 2s^2 2p^6 3p3d 3d^6 4p4f^2)$, L_2–Sc^+ $(1s^2 2s^2 2p^6 3p^2 3d^6 4p4d)$, L_3–$Sc^+(1s^2 2s^2 2p^6 3p^2 3d^6 4p5f)$.

corresponding to the configuration $1s^2 2s^2 2p^4 3s3p$ of the Ne atom for large R, when determined from the molecular orbitals, turned out to be too low. Therefore the corresponding Σ and Π terms at large R are represented by dash-dot lines with allowance for the value of the energy of the auto-ionization state $2p^4(^3P)3s(^2P)3p(^1P)$ of the Ne atom, which is given in [9].

Estimates of the closest-approach distance in "head-on" collisions, realized in our case, were obtained by using Firsov's potential, [10] and we obtained for relative-motion energies E equal to 70 and 450 eV the respective values $R_{min} \approx 1.5$ at. un. and $R_{min} \approx 0.8$ at. un.

It follows from the correlation diagram that in our energy interval there exist at least two possibilities for the appearance of oscillations on the total cross sections:

a) The Π states of $(NaNe)^+$, which dissociate into $Na(3s) + Ne^+$ and $Na^+ + Ne^*(3p)$, are populated at internuclear distances $0.6 \lesssim R \lesssim 0.8$ at. un. from the initial Σ state, owing to rotation of the internuclear axis, and then interact with each other diabatically at $R \sim 3$ at. un., owing to the radial motion. The interference of these states (Π states) leads to oscillations of the corresponding cross sections with a frequency proportional to the area $\langle \Delta R \Delta E \rangle \approx 0.6$ (at. un.)^2 determined from the diagram. However, the indicated states differ from each other in two orbitals, and therefore the connection between them at $R \sim 3$ at. un. due to the radial motion, will
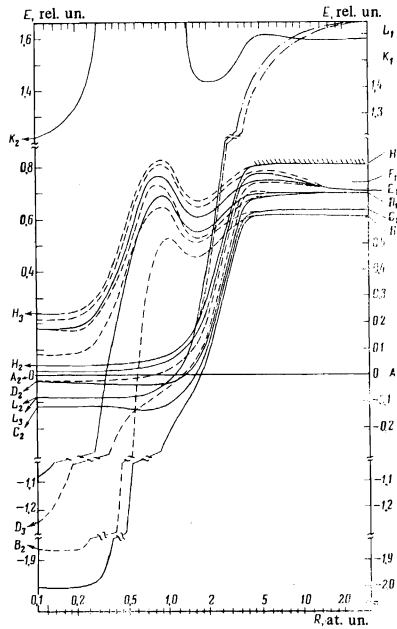
be weak, and consequently the amplitude of the oscillations will also be small. In addition, this interference can arise only in an energy region that starts in the upper limit of our interval, where approach distances $R \lesssim 0.8$ at. un. are reached. It appears that superposition of this oscillation on the main curve explains the more complicated variation of the cross section (1) at high energies (Fig. 1) and the deviation of the oscillation frequency of the charge-exchange cross section at higher energies [2] from the fundamental frequency in this energy interval.

b) Π states of $(NaNe)^+$ that dissociate into $Na^+ + Ne^*(3p)$ and $Na^*(3p) + Ne^+$ are populated at $0.8 \lesssim R \lesssim 1.2$ at. un. from the initial Σ state, owing to rotation of the internuclear axis, and subsequently interact diabatically as $R \to \infty$. Owing to the interference of these Π states, the cross sections of the processes (3) and (4) will oscillate with a frequency proportional to the area between the Π terms. The value $\langle \Delta R \Delta E \rangle \approx 0.6$ (at. un.)^2, determined from the diagram, agree satisfactorily with the value $\langle \Delta R \Delta E \rangle \approx 0.88$ (at. un.)^2 obtained from the oscillation frequency of the experimental cross section, if account is taken of the limited accuracy of the diagram of Fig. 2.

The apparent threshold of the combined charge-exchange cross section, observed at our experimental sensitivity, lies near 62 eV (Fig. 1) and is lower than the thresholds ~93 and ~102 eV determined for the processes (3) and (4) in [3, 4]. The threshold energy ~62 eV, according to our data, corresponds to a closest-approach distance $R_{min} \sim 1.5$ at. un. When Na^+ and Ne come so close together then, as seen from Fig. 2, the states of $(NaNe)^+$ corresponding to the processes of charge exchange into $Na(3s)$ and $Na^*(3p)$ become populated. The first maximum of the oscillation on the charge-exchange cross section (Fig. 1) is located at E = 89 eV, corresponding to our estimate $R_{min} \lesssim 1.5$ at. un. Starting with these disatnces, Σ–Π transitions corresponding to processes (3) and (4) set in with increasing E, in accordance with the diagram (Fig. 2), followed by interference of two Π states. This explains the appreciable excess of the threshold of the oscillating cross sections over the values of ΔE in the processes (1), (3), and (4).

3. The effective ionization cross section measured by us is, within the limits of our measurements, a smooth curve. Particular interest attaches to the fact that the cross section has a threshold (curve C on Fig. 1). Extrapolation of the cross section to zero value yields the threshold value of the energy, ~45 eV, although a second threshold can be discerned at ~70 eV, where the energy increases abruptly. The minimum value of ionization cross section, measured by us, is $\sim1.5 \times 10^{-20}$ cm^2 (at $E \approx 48$ eV). We see thus that the process of simple ionization of the Ne atom, with a threshold ~21.5 eV, makes no noticeable contribution to the cross section of the ionization (2), and the processes mostly realized have higher thresholds, namely excitations of auto-ionization states of the Ne atom, and possibly ionization of the Ne atom with excitation.

An examination of the correlation diagram (Fig. 2) confirms this conclusion. Indeed both the Σ and Π terms of $(NaNe)^+$, which correspond as $R \to \infty$ to the state of the continuous spectrum of $Na^+ + Ne^+ + e$, do not cross the Σ term of the initial state of $(naNe)^+$ in the entire interval of R, and the transition from the initial state to a continuous one has, according to diagram, low probability.

The main contribution to the ionization process (2) is made by the excitation of the group of auto-ionization states of the atoms Ne $2p^4(^3P)3s(^2P)np(^1P)$, $2p^4(^3P)3p(^2S)np(^{1,3}P)$, $2p^4(^1S)3s(^2S)nd(D)$, $2p^4(^1S)3s(^2S)np(^1P)$ with excitation energies from ~ 45 to ~ 55 eV. It is seen from Fig. 2 that the Σ and Π terms corresponding to excitation of the Ne atom with electron configuration $1s^2 2s^2 2p^4 3s3p$ cross the Σ term of the initial state of $(NaNe)^+$ at R ~ 1.1 at. un. The terms corresponding to the other indicated auto-ionization states of Ne also take place in this region of R. This agrees, within the limits of the accuracy of the Firsov potential, with our estimates of the distances R_{min} obtained from the experimental threshold energies. In the energy interval investigated by us, there is low probability of production of auto-ionization states of Ne with excitation of the internal shell 2s, namely $2s2p^6(^2S)np(^{1,3}P)$ and $2s2p^6(^2S)ns(^{1,3}S)$ and with thresholds from ~ 43 to ~ 48 eV. This follows from two circumstances, first, in our energy interval the corresponding Σ terms (as seen from the diagram for the configurations $1s^2 2s2p^6 3s$) are isolated from the ground-state Σ term at small R. Second the interaction of the Σ term corresponding to excitation of the internal 2s shell of Ne and the Σ term corresponding to the double excitation of 2p electrons of Ne at large R is ineffective, since these two types of Σ terms differ by two orbitals. It should be noted, finally, that in our energy interval the probability of the ionization of the Na^+ ion is low, as follows from the molecular-orbital diagram [3] (in order not to complicate the diagram, the corresponding term is not shown in Fig. 2).

The dominant role of excitation of the auto-ionization states of the Ne atoms in the processes of ionization of NaNe)$^+$ in our energy interval is indirectly confirmed by results of an investigation [11] of the differential inelastic energy losses in (NaNe)$^+$ in the region of higher energy.

Our concluding deduction is the following: The experimentally observed thresholds of the charge-exchange processes (1) and of the ionization processes (2), the structures of the cross sections, and the mechanisms of the mentioned inelastic processes can be satisfactorily explained within the framework of the correlation diabatic diagram of terms of the (NaNe)$^+$ quasimolecule.

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