Quasimolecular State Interference in Collisions between Rubidium Ions and Argon

S. V. Bobashev, V. I. Ogurtsov, and L. A. Razumovskiĭ

A. F. Ioffe Physico-technical Institute, USSR Academy of Sciences

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The cross sections for excitation of argon resonance lines emitted in collisions between rubidium ions and argon are measured. The oscillatory structure of the cross sections which is observed is regarded as a new experimental confirmation of the effect of interference between quasimolecular states in the ion-atom system.

In the study of the processes of excitation of different spectral lines in ion-atom collisions, oscillations of the total cross sections of the excitation as functions of the energy were observed[1-5]. Rosenthal[6], and somewhat later Bobashev[3], advanced the hypothesis that the cause of the oscillations of the total excitation cross sections is interference of two vacant excited states of the quasimolecule.

Rosenthal and Bobashev proposed that when the colliding atomic particles come very close together the ground-state term of the system (term 0 in Fig. 1) intersects in succession two vacant excited terms of the quasimolecule in the vicinity of the internuclear distance $R = R_1$. The terms are populated in this case in coherent fashion. When the particles move apart, an interaction occurs between the molecule terms E_1' and E_2' either because of the intersection of the terms[6] or because of their coming together[3]. An important assumption in this case is that the additional interaction between the terms occurs at a sufficiently large internuclear distance $R = R_2$, i.e., that $R_1 \ll R_2$. As a result of the interaction, the probability of each of the two inelastic channels of the reaction (1 and 2) is a harmonic function of the reciprocal velocity of relative motion of the particles v^{-1}. The oscillation frequency is proportional to the area bounded by the two paths on the plot of the system energy against the internuclear distance (the area of the "loop" $ABE_2'CE_1'A$ in Fig. 1). The oscillation frequency depends little on the impact parameter, since the impact parameters, which are important for the excitation process, are much smaller than those distances at which interaction of the terms occurs when the particles move apart. It is precisely for this reason that the oscillations remain also in the total cross sections.

Ankudinov, Bobashev, and Perel'[7] have considered the simplest model of the collision of two atomic particles, leading to oscillations of the total cross sections of the processes with large resonance defect. We recall certain conclusions that follow from the theoretical calculation in[7] and have a direct bearing on the purpose of the present paper. It has been noted in[7] that the modulation depths of the total cross sections can be appreciable, and consequently the oscillations of the total cross sections can be observed experimentally only in the case when the terms interact sufficiently effectively in the vicinity of $R = R_2$. It is also obvious that in the case of effective interaction in the region R_2, the probability of an inelastic atomic collision leading to a transition from the initial state 2 (or 1) into the state 1 (or 2) should be large, or, more accurately, much larger than the probability of the transition $0 \to 2$ or $0 \to 1$, since the cross sections of the process $2 \to 1$ (or $1 \to 2$) and the processes $0 \to 1$ and $0 \to 2$ are proportional to R_2^2 and R_1^2, respectively. In addition, in accordance with[7], the dependence of the total cross sections of the processes $2 \rightleftarrows 1$ on the energy should not have a noticeable structure, unlike the cross sections of the processes $0 \to 2$ and $0 \to 1$.

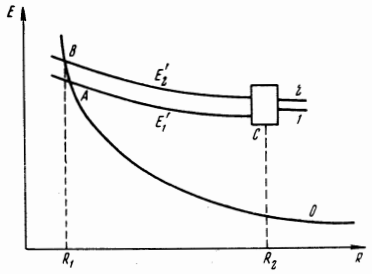

FIG. 1. Schematic representation of the behavior of the terms of an ion colliding with an atom as a function of the internuclear distance. The rectangle bounds the region of additional nonadiabatic interaction.

On the other hand, the large value of the cross section of the transitions $2 \rightleftarrows 1$ indicates that it is possible to observe the structure of the total cross sections by experimentally investigating atomic collisions that lead to transitions $0 \to 2$ and $0 \to 1$[1)].

Figure 2 shows the cross section for the charge exchange of argon ions with rubidium atoms, taken from the paper of Peterson and Lorents[8], who assumed that the measured curve corresponds to the charge exchange of Ar^+ ions in excited and metastable states of the argon atom, i.e., to the process

$$\begin{aligned} &Ar^+ + Rb \to Ar^* + Rb^+, \\ &\Delta E_1 = 0.03 - 0.23\,\text{eV}. \end{aligned} \qquad (1)$$

The quantity ΔE_1 is the energy difference between the levels of the argon atom, populated in the collision process, and the ground level of the Rb^+ ion, i.e., the value of the resonance defect.

The large value of the cross section of the process (1) has induced us to measure the excitation cross sections of two resonance lines of the argon atom, excited in the reaction

[1)]It was also emphasized in [7] that oscillations can be expected in the total cross sections not only when the interaction of the terms occurs when the particles move apart, but also when such an interaction occurs when the particles come together, i.e., in transitions $2 \to 0$ and $1 \to 0$.

$$Rb^+ + Ar \rightarrow Rb^+ + Ar^* \begin{cases} \nearrow \lambda\ 1067\ Å \\ \searrow \lambda\ 1048\ Å. \end{cases} \quad (2)$$

The resonance defects (ΔE_2) for this reaction are equal to 11.83 and 11.62 eV, respectively. We started here from the assumption that since the excitation process (2) is accompanied by the charge exchange of the rubidium ions with the argon atoms

$$Rb^+ + Ar \rightarrow Rb^0 + Ar^+, \quad \Delta E_3 = 11{,}57\ эв, \quad (3)$$

it can be assumed on the basis of the considerations advanced above that the two inelastic channels (2) and (3) will interfere in the region $R = R_2$. The region R_2 is responsible for the large cross section of the process (1).

If we do not distinguish, for simplicity, between the two excited states of the argon atom, regarding them as one term, then, by using the scheme of Fig. 1, we can easily see that the three collision processes under consideration, (1), (2), and (3), correspond to the transitions $2 \rightarrow 1$, $0 \rightarrow 1$, and $0 \rightarrow 2$. Thus, when starting the measurements, we hoped that the experimentally observed structures of the cross sections for the excitation of the resonant states could be regarded as an experimental confirmation of the qualitative hypothesis explaining the nature of the oscillations of the total cross sections[3,6].

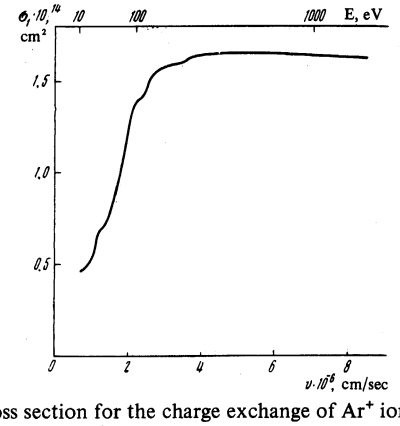

FIG. 2. Cross section for the charge exchange of Ar^+ ions on rubidium atoms, taken from [8].

The measurements were performed with the setup described in[9]. Figure 3 shows the absolute cross sections for the excitation of two resonant states of the argon atom, $3p^5 4s(^3P_1)$ (λ 1067 Å) and $3p^5 4s(^1P_1)$ (λ 1048 Å), as functions of the reciprocal velocity v^{-1} of the relative motion of the rubidium ions and argon atoms in the reaction (2). The absolute values of the excitation cross sections were estimated on the basis of data on the transmission coefficients of the vacuum monochromator for the wavelengths λ = 1067 Å and λ = 1048 Å, obtained from a comparison of the previously measured[10] spectrum of the radiation produced when K^+ ions collide with argon, and the data given in[11] concerning the inelastic energy loss spectra for the K^+ + Ar pair. The estimated error of the presented absolute cross sections is above 100%, whereas the angle of the relative measurements does not exceed 10%.

The measurement results (Fig. 3) show that the

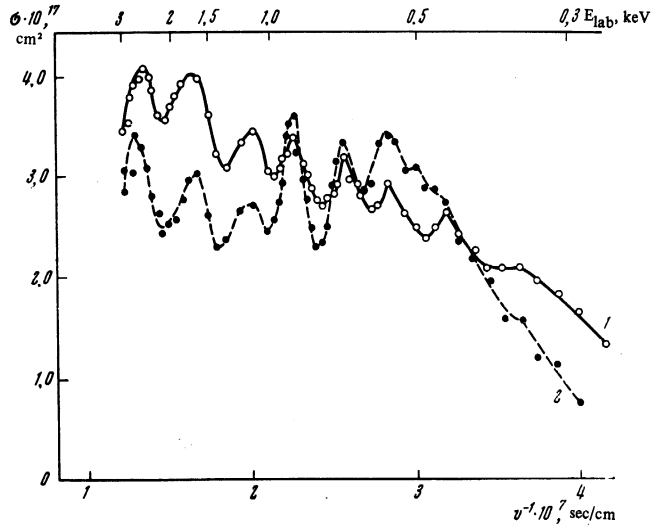

FIG. 3. Cross sections for the excitation of the resonant lines ArI λ 1067Å (curve 1) and ArI λ 1048Å (curve 2) as functions of the reciprocal rubidium ion velocity, in process (2) (E_{lab} is the energy of the Rb^+ ions in the laboratory frame).

cross sections for the excitation of the resonant lines actually oscillate in terms of the reciprocal velocity, with a period $\Delta(v^{-1}) = (3.0 \pm 0.3) \times 10^{-8}$ sec/cm. The total absolute value of the excitation cross sections of the two resonant lines of argon (2) is smaller by two orders of magnitude than the cross sections for the charge exchange of the argon ions (1). These two experimental facts, predicted on the basis of the theory of[7], are convincing proof of the interference between the quasimolecular states of close energy in the ion-atomic collision process $Rb^+ + Ar$. Knowing from experiment the oscillation period and the cross sections σ_1 and σ_2 of (1) and (2), we can estimate the splitting between terms $E_2' - E_1' = \Delta E$ in the region $R < R_2$ and the distance $R_2 - R_1 = \Delta R$ for the quasimolecular system ($RbAr^+$). The connection between ΔE and ΔR has been established in[7], namely $\Delta E \Delta R = 2\pi\hbar/\Delta(v^{-1})$eV-cm. If the term interaction in the regions R_1 and R_2 is sufficiently effective, then $\sigma_1 = (1/2)\pi R_2^2$ and $\sigma_2 = (1/2)\pi R_1^2$, which yields the estimates $\Delta E = 1.65$ eV, $\Delta R = 10$ Å, and $R_2 \approx 10$ Å.

In conclusion we note two circumstances. First, it is of interest to investigate experimentally the cross section of the charge exchange (3), which should definitely have an oscillatory structure. Second, the role played in the collision process by metastable states that are energywise close to the investigated resonant levels of argon is still unclear. An experimental investigation of the metastable states is in our opinion of considerable interest.

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