CHARGE EXCHANGE OF MULTI-CHARGED IONS

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Results previously obtained by the author in the theory of pseudo-crossing of potential energy curves are used to derive for the charge exchange cross sections of multiply charged ions [reaction (1)] formulas in which the time dependence of the interaction matrix element and the non-separation of the terms at infinity are taken into account. The agreement between the results and the experimental data is satisfactory within a broad range of variation of the parameters of the problem.

THERE are various inelastic processes that occur in collisions of heavy particles and whose large cross sections are attributed to the crossing of electron levels. These include, for example, charge exchange and recombination of ions, detachment of electrons from negatively charged ions, predissociation, dissociative recombination, etc. The crossing of levels is encountered in solid state physics (transition of electrons from the valence into the conduction band), in the study of the behavior of various quantum systems in slowly varying fields, etc.

Recently Bates[1] and Hasted and Chong[2] made some critical remarks concerning the application of the Landau-Zener theory to collisions of heavy particles. These remarks were essentially aimed again the Zener model. This model was refined subsequently by various workers[3,4]; some used excessively arbitrary approximations for the matrix element of the interaction and the frequency[3], thereby greatly modifying the structure of the formulas and the character of the dependence of the corresponding cross sections on the parameters of the problem.

In the present paper we present a more complete derivation of the theory, and consider its application to the specific case of simple charge exchange of multiply charged ions.

1. The charge exchange of multiply charged ions, a process of interest for many practical problems (see, for example,[2]), was investigated experimentally by many authors[1,5-8]. It is known that a reaction of this type can be quantitatively described by means of the concept of pseudo-crossing of the potential-energy curves[9,10]. Indeed, for reactions of the type

$$A^{n+} + B \to A^{(n-1)+} + B^{+} + \Delta E \qquad (1)$$

the difference between the unperturbed electron levels at large distances can be expressed with good approximation by

$$\alpha = \Delta E - (n-1)/R. \qquad (2)$$

The interaction matrix element V has then the following behavior:

$$V = P_s(R)\, e^{-\beta R}, \qquad (3)$$

where $P_S(R)$ is a power function of the order of R^S, β is the smallest of the ionization potentials of the atom B and of the ion $A^{(n-1)+}$, and ΔE is the resonance defect. When $R = (n-1)/\Delta E$, crossing of the unperturbed levels takes place, and the static perturbation V separates these levels at the crossing point and causes non-adiabatic transitions. The approximate formula previously obtained[11] for the probability of this transition (after averaging the oscillating factor) is:

$$W = \frac{4\pi F(k)}{|\Gamma(1/2 + i\nu)|^2}\left|\exp\left\{-\frac{3\pi\nu}{2} + i\nu\ln\nu - i\nu\right\}\right|^2, \qquad (4)$$

$$\nu = \frac{1}{2\pi i}\oint \frac{\Omega}{v_R}\,dR, \quad F(k) = \left(1 + \frac{k^2}{4}\right)^{-1}, \quad k = \frac{2\beta v}{\Delta E}, \qquad (5)$$

$$\Omega = \left[\frac{\omega^2}{4} - \frac{iv_R V}{2}\frac{d}{dR}\frac{\alpha}{V} - v_R^2 S\left(\frac{V}{v_R}, R\right)\right]^{1/2},$$

$$\omega^2 = \alpha^2 + 4V^2, \qquad (6)$$

where v_R—radial component of the velocity of the colliding particles, S—Schwartz derivative, and the integral in (5) is taken along a contour circuiting the roots of the function Ω in the complex R plane in a counterclockwise direction. Using (2) and (3) we can obtain the following approximate expression for Ω, discarding in this case the terms of higher order of smallness:

$$\Omega = (A_0 + A_1/R + A_2/R^2)^{1/2};$$

$$A_0 = (\Delta E^2 + 4V^2)/4 - iv_R{}^0\Delta E\beta/2 - v_R{}^{02}\beta^2/4, \qquad (7)$$

$$A_1 = -\Delta E(n-1)/2 + iv_R^0[s\Delta E + \beta(n-1)]/2 + v_R^{02}\beta s/2,$$
$$A_2 = (n-1)^2/4 - iv_R^0(n-1)(l+1)/2 - v_R^{02}(s^2+2s)/2. \quad (8)$$

Here v_R^0—radial component of the relative velocity, assumed for simplicity constant in the vicinity of point of pseudo-crossing. We note that in expressions (7) and (8) account is taken of the non-separation of the terms at infinity and of the dependence of the matrix element V on R in accordance with (3).

As to the first term in the expression for A_0, if $V \ll \Delta E$ we can assume approximately that near the point of pseudo-crossing $V = V(R_X)$; we then have

$$\nu = \frac{1}{2\pi i v_R^0}\oint\left[A_0 + \frac{A_1}{R} + \frac{A_2}{R^2}\right]^{1/2} dR = \frac{1}{v_R^0}(A_1/2\sqrt{A_0} - \sqrt{A_2}). \quad (9)$$

Confining ourselves in (8) to terms linear in the velocity, we then obtain, in accordance with (9)

$$\nu = -\frac{\Delta E(n-1)}{2(\Delta E^2 + 4V_0^2)^{1/2}} + \frac{n-1}{2} - \frac{i}{2}. \quad (10)$$

When $V_0 \ll \Delta E$ we have

$$\nu = (n-1)\Delta U^2(R_x)/4\Delta E^2 v_R^0 - i/2, \quad (11)$$

where $\Delta U(R_X)$—separation of the levels at the points of pseudo-crossing. In the approximation considered here, we then have for the transition probability

$$W = 2F(k)e^{-\tau}(1-e^{-\tau}), \quad (12)$$
$$\tau = \pi(n-1)\Delta U^2(R_x)/2\Delta E^2 v_R^0. \quad (13)$$

Recognizing that $v_R^0 = v(1-\rho^2/R_X^2)^{1/2}$, we obtain for the single-charge exchange cross section

$$\sigma = 4\pi R_x^2 F(k)\int_1^\infty e^{-\eta x}(1-e^{-\eta x})x^{-3}dx; \quad (14)$$
$$\eta = 247(n-1)m^{1/2}[\Delta U(R_x)]^2/\Delta E^2 E^{1/2},$$
$$k = 0.34\beta E^{1/2}/m^{1/2}\Delta E. \quad (15)$$

Here ΔE, $\Delta U(R_X)$, and E are in electron volts, and m—mass of the ion A^{n+} in units of O^{16}.

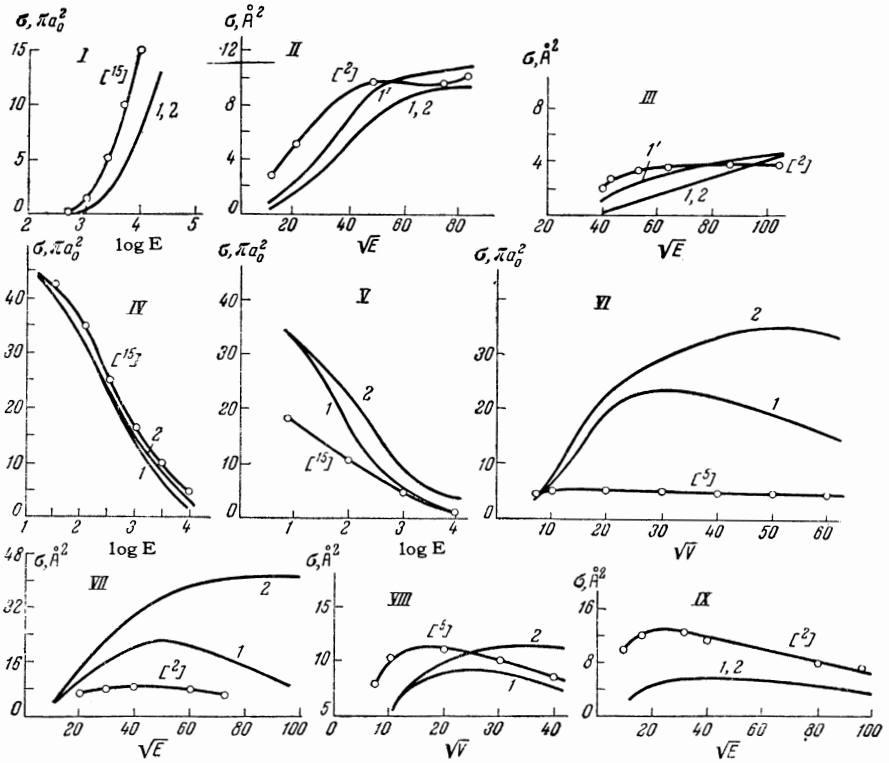
We thus obtain in the adiabatic approximation a formula (12) which coincides, apart from the factor F(k), with the Landau-Zener formula, although we have taken into account from the very beginning the dependence of V on R and the non-separation of the terms at infinity. The transition probability, as a function of the velocity v, has here one maximum in the adiabatic region. It can also be shown that allowance for the term proportional to the velocity in expression (11) for ν does not change the situation. In light of the foregoing, we regard as unfounded the attempts made by Bates[1] to connect the adiabatic theory with ordinary perturbation theory and to conclude therefrom the character of the dependence W(v). This circumstance was already pointed out by Dykhne and Chaplik[12]. The situation changes when there are several points of crossing or pseudo-crossing. In this case W(v) can have a more complicated structure (see, for example,[13]).

We now proceed to concrete calculations of the charge-exchange cross sections, based on the simplified theory given by formulas (14) and (15). We shall use here for $\Delta U(R_X)$ the numerical data obtained with the aid of the Slater wave functions in [2,9,10], and calculate β with the aid of the rule for the screening constants. The figure shows the results of the numerical calculations for different reactions, in order to verify the agreement with theory and experiment over as large an interval of variation of the characteristic parameters of the problem as possible. (The values of the employed parameters are indicated in the figure caption). The obtained calculated curves (1) are compared with experiment (curves with points) and with the Landau-Zener theory (curves 3).

For process I the present calculation gives, like the Landau-Zener theory, good agreement with experiment.

For processes II and III, the agreement is worse: the theoretical curves lie below the corresponding experimental curves. We made a numerical calculation for these reactions, in accordance with a more exact formula (4) (curves 1′); the agreement with experiment is in this case better. Nevertheless, for reactions of this type, where $\eta > 1$ over the entire interval of investigated energies and we are working in the region where the cross section increases, the calculations undervalue the cross section systematically, in spite of the fact that the theory should agree here better with experiment. The reason for this lies apparently in the values of the parameter $\Delta U(R_X)$, on which the calculations depend very strongly in the case of low velocities, for calculations of this parameter with the aid of the Slater wave functions cannot be regarded as sufficiently accurate. It must also be noted that the experiments are less reliable at low velocities.

For processes IV and V we operate in the region where the cross section decreases ($\eta < 1$). Here, too, the theory yields the correct character of the behavior of $\sigma(v)$. For the process V, the present



Charge exchange cross sections of multiply charged ions for the following processes: I – $Al^{2+} + H \rightarrow Al^{+} + H^{+}$ ($\Delta E = 5.29$ eV, $\Delta U = 1.54$ eV, $\beta = 1$); II – $Kr^{3+} + He \rightarrow Kr^{2+} + He^{+}$ ($\Delta E = 11.1$ eV, $\Delta U = 1.17$ eV, $\beta = 1.30$); III – $Kr^{4+} + He \rightarrow Kr^{3+} + He^{+}$ ($\Delta E = 19.0$ eV, $\Delta U = 2.52$ eV, $\beta = 1.30$); IV – $Si^{2+} + H \rightarrow Si^{+} + H^{+}$ ($\Delta E = 2.75$ eV, $\Delta U = 0.1$ eV, $\beta = 1$); V – $Li^{2+} + H \rightarrow Li^{+}(^3S) + H^{+}$ ($\Delta E = 3.02$ eV, $\Delta U = 0.124$ eV, $\beta = 1$); VI – $Ar^{2+} + H \rightarrow Ar^{+} + H^{+}$ ($\Delta E = 3.02$ eV, $\Delta U = 0.42$ eV, $\beta = 1.30$); VII – $Kr^{2+} + Ne \rightarrow Kr^{+} + Ne^{+}$ ($\Delta E = 2.7$ eV, $\Delta U = 0.35$ eV, $\beta = 2.32$); VIII – $N^{2+} + He \rightarrow N^{+} + He^{+}$ ($\Delta E = 5.02$ eV, $\Delta U = 0.77$ eV, $\beta = 1.30$); IX – $Kr^{4+} + Ne \rightarrow Kr^{3+} + Ne^{+}$ ($\Delta E = 22$ eV, $\Delta U = 1.02$ eV, $\beta = 2.51$). The experimental curves are identified by the literature references. (The energy E of the incoming particles is given on the figures in electron volts, and the accelerating potential difference V is in volts; a_0 – Bohr radius).

calculation gives a somewhat better result than calculation by the Landau-Zener formulas.

In the case of reactions VI—IX in the energy interval in question, up to 1 keV, the value of η changes from several units to several tenths, and here we can verify the agreement between calculation and experiment in the region of the maximum cross section. It is seen from the figure that in these cases the theory yields correctly the position of the maximum of the cross section, while the use of the Landau-Zener formulas is known to give worse results in several cases (VI—VIII).

We see from the foregoing examples that in general the simplified theory based on formulas (14) and (15) describes correctly the character of the dependence $\sigma(v)$ in the kilovolt region of energies and over a sufficiently wide range of variation of the parameters of the problem:

$$2.75 \text{ eV} \leqslant \Delta E \leqslant 22 \text{ eV}, \quad 3.75 \text{ at. un.} \leqslant R_x \leqslant 10 \text{ at. un.},$$
$$0.1 \text{ eV} \leqslant \Delta U(R_x) \leqslant 2.5 \text{ eV},$$

in many cases the results are noticeably better than those of the Landau-Zener theory. The position of the maximum of the cross section, determined by the condition $\eta \sim 1$, is confirmed experimentally. This condition can serve as an approximate criterion in a qualitative analysis of several reactions, replacing the known Massey criterion for adiabatic reactions.

Refinement of the theory can proceed in the following directions:

a) the use of the more general formula (4) and of more exact values of the parameters [especially $\Delta U(R_X)$] for pseudo-crossing of two electron levels;

b) account of electron capture in excited states of the ion $A^{(n-1)+}$.

In the latter case it is necessary to consider a problem with several pseudo-crossing points; the theory also can be generalized to this case [14].

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