Detachment of weakly bound electrons in slow collisions between atomic systems

F. I. Dalidchik

Institute of Chemical Physics, USSR Academy of Sciences
(Submitted July 10, 1973)
Zh. Eksp. Teor. Fiz. **66**, 849–856 (March 1974)

The processes leading to the detachment of weakly bound electrons in slow collisions between atomic systems are discussed. The role played in these processes by the excited states of the core electrons is elucidated. It is shown that in a number of cases the auto-ionization of the quasimolecule is largely determined by the direct core-core interaction (by the diagonal and off-diagonal parts). The spectrum of the liberated electrons both in the presence and absence of pseudocrossing of the terms of the core electrons is found. The effects which are connected with the role of the "inelastic" ionization channels, and which can manifest themselves both in an anomalous dependence of the initial part of the spectrum on the colliding-particle energy and in the appearance of characteristic near-threshold properties, are discussed. The results of the theory are used to interpret the available experimental data on electron detachment in collisions of negative ions and neutral alkali-metal atoms with inert-gas atoms.

1. The theory of the detachment of a weakly bound electron from a negative ion A^- or a neutral alkali-metal atom A in slow collisions with unexcited atoms B has been developed in a number of papers[1-4]. It is assumed in these papers that as the atoms approach each other the one-electron term of the ground state of the system touches or (in the zeroth approximation in the interaction) intersects the boundary of the continuous spectrum, acquiring an auto-ionization width that determines the ionization probability and the spectrum of the liberated electrons. These processes have also been systematically investigated experimentally[5-11].

In the experiments, besides instances confirming the main theoretical predictions, numerous cases were discovered which significantly disagreed with the theory. Thus, for example, in accordance with the Smirnov-Firsov theory[2], the cross section for detachment of an electron from a negative halogen ion in a collision with an inert-gas atom in the region of relative-motion energies W significantly exceeding the repulsive potential U_0 at the point of tangency ($W \gg U_0$) is practically a constant[5]. For the ions of the alkali metals, however, experiment yields cross sections that increase significantly with the energy[6]. The measured electron spectra[7-11] in the initial part (up to 2–4 eV) of the spectrum agree with fairly simple and universal theoretical dependences[1,3,4], but at high energies the spectra in many cases reveal a structure containing pronounced "prolongations" and resonance peaks[7-11] whose locations in many cases correspond to the well-known auto-ionization states of the isolated A atoms, whereas the threshold for their appearance does depend on the choice of the collision partner B.

The fairly large width of the initial part of the spectrum, a part which corresponds to the auto-ionization of the quasimolecule, the substantial population probability for the atomic auto-ionization states (up to 50%[9]), the pronounced energy dependences of the total cross sections[6], and the excitation functions of the resonance peaks[7]—all this apparently indicates a major role for the inner-shell electrons in the processes leading to the detachment of the weakly-bound outer electron. However, the role of the inner-shell electrons in these processes has thus far not been considered, although the presently available experimental data on the inelastic scattering of the neutral group-I atoms and their positive ions by inert-gas atoms[12-15] directly indicate a strong nonadiabatic coupling between the ground and excited states, a coupling which is localized over the same internuclear distances at which the electron detachment is accomplished in A^--B and A—B collisions.

The role of the excited states of the inner-shell electrons in the outer-electron detachment processes that occur in slow collisions of negative ions and neutral alkali-metal atoms with inert-gas atoms is considered in the present paper. Since the cross section for excitation of the atomic cores depends to a decisive degree on the presence or absence of pseudocrossing in the system composed of their terms $E_n^{AB}(R)$, these two cases will be analyzed separately below. The possibility of the simultaneous convergence of more than two terms is not excluded, but less likely; therefore, we shall, in the main, restrict ourselves to only a two-channel description of the motion of the weakly bound electron.

2. If there is no pseudocrossing in the system of terms $E_n^{AB}(R)$, then electron detachment in a slow collision between the unexcited atomic systems is possible only if the outer electron ceases at some internuclear distance to be in a bound state in the field of the unexcited cores[1,2] (i.e., if $\epsilon_1(R) = 0$ for $R \leq R_0$, where ϵ_1 is the binding energy), or if there exists a region where the Rydberg states change abruptly[3]. Channel coupling, however, is important in these cases only for electron energies E satisfying the conditions

$$|E-\omega_{12}+\varepsilon_2| \sim |V_{12}| \quad \text{or} \quad E>\omega_{12}, \tag{1}$$

where $\omega_{12} = E_2^{AB} - E_1^{AB}$; ϵ_2 is the binding energy of the electron in the field of the excited (n = 2) core, and therefore we shall consider it to be time independent.

The system of equations describing the situation u under consideration has the form

$$\begin{aligned} i\frac{\partial \Psi_1(r,t)}{\partial t} &= \left[-\frac{\Delta_r}{2}+V_{11}(r,t)\right]\Psi_1(r,t)+V_{12}(r)\,\Psi_2(r,t), \\ i\frac{\partial \Psi_2(r,t)}{\partial t} &= \left[-\frac{\Delta_r}{2}+V_{22}(r)\right]\Psi_2(r,t)+V_{21}(r)\,\Psi_1(r,t). \end{aligned} \tag{2}$$

Here

$$V_{11}(r,t) = [\hat{S}_{AB}^{-1}(t)\,\hat{V}_{e-AB}(r,t)\,\hat{S}_{AB}(t)]_{11} \tag{3}$$

which depends on the internuclear distance R(t) and the core-core interaction $U_{AB}(R)$, is the interaction between

the weakly bound electron and the quasi-molecular core in the ground state (adiabatic basis), $\hat{V}_{e\text{-}AB}(r, t)$ is the internuclear-distance-dependent interaction matrix describing the interaction between the electron and the noninteracting atomic cores, and $\hat{S}_{AB}(t)$ is the matrix diagonalizing the Hamiltonian of the interacting cores. Similarly,

$$V_{22}(r)=[\hat{S}_{AB}^{-1}(t_0)\hat{V}_{e-AB}(r, t_0)\hat{S}_{AB}(t_0)]_{22}+\omega_{12}(t_0) \tag{4}$$

is the interaction between the electron and an excited core; the moment of time t_0 is determined by the condition

$$R(t_0)=R_0. \tag{5}$$

The channel-channel interaction

$$V_{12}(r)=\left\langle \Phi_2(\xi_{AB}, t)\left| V_{e-AB}(r, \xi_{AB}, t)-iv_R\frac{\partial}{\partial R}\right| \Phi_1(\xi_{AB}, t)\right\rangle_{t=t_0},$$

where the $\Phi_n(\xi_{AB}, t)$ are the adiabatic wave functions of the cores and v_R is the radial velocity of the nuclei.

The solution of the system of equations (2) is, in the general case, not possible; therefore, we shall use the fact that the most important part of the electron-core interaction is the time-dependent short-range part, which can be taken into account through a nonstationary boundary condition at some finite r[1]:

$$\frac{\partial \ln \Psi_1(r, t)}{\partial r}\Big|_{r=a}=f(t); \tag{6}$$

here a is the effective radius of the short-range part of the interaction, which, for simplicity, is assumed to be spherically symmetric.

For the linear approximation

$$f(t)=\beta(t-t_0), \tag{7}$$

which corresponds to a slow passage through the point R_0, the system of equations (2) is exactly soluble by the Demkov-Osherov method[16], and the spectrum of the liberated electrons has the form

$$n_{1l}(E)=2\beta^{-1}(2E)^{1/2}P_{1l}(E, a)w_l(E). \tag{8}$$

Here $P_{1l}(E, a)$ is the penetration factor for an electron of energy E in the field of the unexcited core of the quasimolecule, the factor being determined by the relation[17]

$$(2E)^{1/2}P_{1l}(E, a)=\mathrm{Im}\frac{d}{dr}\chi_l^+(E, r)\Big|_{r=a}, \tag{9}$$

where $\chi_l^+(E, a)$ is that solution of the Schrödinger equation with the long-range part V_{11} of the potential and with angular momentum l that is irregular at the origin and has a diverging asymptotic form at infinity. The quantity

$$w_l(E)=w_{1l}(E)w_{2l}(E) \tag{10}$$

is the "survival" factor determining the fraction of electrons that are liberated with energy higher than E.

For $E<\omega_{12}$,

$$w_{1l}(E)=\exp\left\{-\frac{2}{\beta}\int_0^E(2E)^{1/2}P_{1l}(E, a)\,dE\right\}, \tag{11}$$

$$w_{2l}(E)=\exp\left\{-\frac{2}{\beta}\sum_m|\langle\chi_l^+|V_{21}|\chi_2^m\rangle|^2\eta(E-E_m)\right\}, \tag{12}$$

$$E_m=\omega_{12}-\varepsilon_m, \tag{13}$$

where χ_2^m and ϵ_m are respectively the wave function and binding energy of the m-th bound state of the electron in the field of the excited core; $\eta(x) = 0$ for $x < 0$ and $\eta(x) = 1$ for $x > 0$. Notice that for $r \to \infty$ $V_{21} \neq 0$ and is determined by the nonadiabatic channel-channel coupling.

A similar spectrum was found earlier[18] in the framework of the phenomenological approach with a channel-channel coupling that is localized at r = a. The factor $w_{2l}(E)$, which depends on the excitation energy of the inner-shell electrons ($\omega_{12} = E_2^{AB} - E_1^{AB}$) and the binding energy ϵ_m of the outer electron in the field of the excited core, determines the energy dependence of the cross section $\sigma(W)$ and the excitation function I(W) of the resonance electrons. Allowing for a double crossing of the transition region ($R \sim R_0$), we have for the negative-ion decay process, for example, the expressions:

$$\sigma(W)\approx\pi R_0^2[1-w_1^2(E_1)(1-w_2(E_1))^2], \tag{14}$$

$$I(W)\sim w_1(E_1)(1-w_2(E_2))w_2(E_1). \tag{15}$$

For $w_1(E_1) \ll 1$, the cross section weakly depends on the energy of the colliding particles, and the contribution of the resonance electrons to the integrated spectrum is small. This case corresponds, for example, to collisions between negative halogen ions and inert-gas atoms ($E_1 \sim 7$ eV, $I \sim 5\%$[7]). An alternative possibility is apparently realized for negative alkali-metal ions, which corresponds to the presently available indications that the terms of the ground and excited states of such quasimolecules as LiHe, NaAr, etc., closely approach each other and, possibly, intersect[12,13]. In these cases the energy $E_1(R_0)$ (see (13)) can be very small (and even negative), and then $w_1(E_1) \sim 1$, which guarantees at low velocities a pronounced growth of the electron-detachment cross section with increasing energy[6]. Analysis of the low-energy part of the differential spectrum of the electrons would allow in this case the reestablishment of the disposition of the terms of the quasimolecule at small distances ($R \sim 10^{-8}$ cm), since for $E_1 > 0$, according to (8), (11), and (12), there should be a discontinuity at $E = E_1$ in the spectrum of the emitted electrons, whereas for $E_1 < 0$ the spectrum will be smooth.

When $E > \omega_{12}$, a new ionization channel opens which is accompanied by the simultaneous excitation of the core electrons and whose spectrum has the form

$$n_{2l}(E)=2\beta^{-1}|\langle\chi_{l,E+\omega_{12}}^{-}|V_{12}|\chi_{E,2}^{+}\rangle|^2\rho_2(E)w_l(E+\omega_{12}). \tag{16}$$

Here $\chi_{E,2}^{+}$ is the normalized—with respect to momentum—wave function of an electron of energy E in the field of the excited (in the adiabatic representation) core of the quasimolecule and ρ_2 is the corresponding density of states.

3. In the presence of quasicrossing in the core-term system $E_{1,2}^{AB}(R)$, decisive importance is assumed by the possibility of the emergence of the discrete single-electron terms of the first channel (of the zeroth approximation in the coupling) in the continuous-spectrum zone of the second channel. We shall, for the analysis of this case, use the Landau-Zener model for the pseudocrossing, and describe the system by the following equations:

$$\begin{aligned} i\frac{\partial\Psi_1(r, t)}{\partial t}&=\left[-\frac{\Delta_r}{2}-V_{11}(r)+\beta t\right]\Psi_1(r, t)+V_{12}(r)\Psi_2(r, t),\\ i\frac{\partial\Psi_2(r, t)}{\partial t}&=\left[-\frac{\Delta_r}{2}+V_{22}(r)\right]\Psi_2(r, t)+V_{21}(r)\Psi_1(r, t).\end{aligned} \tag{17}$$

Here $V_{nn}(r) = \langle\Phi_n^0(\xi_{AB})|V_{e\text{-}AB}|\Phi_n^0(\xi_{AB})\rangle$ is the interaction between the electron and the noninteracting atomic cores in the state $\Phi_n^0(\xi_{AB})$; βt is the time-dependent

splitting of the intersecting core terms:

$$\omega_{12}(t)=\beta t; \tag{18}$$

finally

$$V_{12}(r)=\langle\Phi_1^0(\xi_{AB})\,|\,V_{e-AB}+U_{AB}(\xi_{AB}R_0)\,|\,\Phi_2^0(\xi_{AB})\rangle \tag{19}$$

is the complete adiabatic channel-channel coupling ($R_0 = R(0)$).

The solution of the system (17) by the Laplace method amounts to finding the solution to the integro-differential equation

$$F_1(E,r)=i\beta\frac{d}{dE}G_1F_1+V_{12}G_2V_{21}G_1F_1, \tag{20}$$

in terms of which the electron wave functions can be expressed:

$$\begin{aligned}\Psi_1(r,t)&=\int G_1F_1e^{-iEt}\,dE,\\ \Psi_2(r,t)&=\int G_2V_{21}G_1F_1e^{-iEt}\,dE;\end{aligned} \tag{21}$$

here $G_n = (E + \Delta_r/2 - V_{nn})^{-1}$ is the Green operator. For a weak channel-channel coupling $|V_{12}| \ll |\epsilon_{1n_0} - \epsilon_{1n}|$, when the virtual transitions of the electron in the field of the quasi-molecular core of the initial configuration can be neglected, G_1 can be approximated by a single meromorphic term:

$$G_1\approx\frac{|f_{1n_0}\rangle\langle f_{1n_0}|}{E-\varepsilon_{1n_0}}, \tag{22}$$

and in this case the spectrum is of the form

$$\begin{aligned}n_2(E)=&\frac{2\pi}{\beta}|\langle f_{1n_0}|V_{12}|f_{2E}^{+}\rangle|^2\rho_2(E)\\ &\times\exp\Big[-\frac{2}{\beta}\operatorname{Im}\int_{-\infty}^{E}\langle f_{1n_0}|V_{12}G_2V_{21}|f_{1n_0}\rangle dE\Big].\end{aligned} \tag{23}$$

The width of the spectrum (23) is, generally speaking, greater than the width in the absence of pseudocrossing, and is directly determined by the total channel-channel coupling, which is due both to the inelastic electron-core interaction (the term $\sim V_{e\text{-}AB}$) and the possibility of a nonadiabatic transition in the core-electron system (the term proportional to the direct core-core interaction U_{AB}). In this case the total probability of transition of the outer electron to all the possible states in the case $V_{e\text{-}AB} \ll U_{AB}$

$$w_2=\int_{-\infty}^{\infty}n_2(E)\,dE \tag{24}$$

coincides, as expected, with the probability of a nonadiabatic transition in the core system:

$$w_2=w_{AB}=\exp\,[-2\pi\beta^{-1}|\langle\Phi_1^0|U_{AB}|\Phi_2^0\rangle|^2]. \tag{25}$$

In the opposite case, when $V_{e\text{-}AB} \gg U_{AB}$, the spectrum (23) corresponds to electron emission as a result of the decay of the initial state, which, owing to the shift of the boundary of the continuous spectrum when $t > |\epsilon_{1n_0}|/\beta$, becomes of the resonance Feshbach type with an auto-ionization width

$$\Gamma_2=2\pi|\langle f_{1n_0}|V_{e-AB}|f_{2E}^{+}\rangle|^2\rho_2(E). \tag{26}$$

Electron detachment in collisions between atomic systems is possible both when the nuclei approach each other—as a result of the autoionization of the quasimolecule—and when they separate—as a result of the auto-ionization of the atoms. For the disintegration of a negative ion, for example, to the first possibility corresponds the cross section

$$\sigma_{ion}=\sigma_{11}+\sigma_{12}. \tag{27}$$

Here

$$\sigma_{11}\approx\pi R_0^2w_{12}^{e}(1-w_{AB}) \tag{28}$$

is the cross section for electron detachment that is not accompanied by the excitation of the atoms A and B, where

$$w_{12}^{e}=\exp[-2\pi\beta^{-1}|\langle f_{1n_0}|V_{12}|f_{2n}\rangle|^2], \tag{29}$$

f_{2n} is the wave function of the bound electron in the field of the core in the configuration corresponding, upon the separation of the nuclei, to the excited state of one of the atoms, and w_{AB} is the probability of a nonadiabatic transition in the A—B atomic system. Further,

$$\sigma_{12}\approx\pi R_0^2w_{12}^{e}w_{AB} \tag{30}$$

is the cross section for the process of electron detachment with excitation of the atoms:

$$A^-+B\to A^*+e+B. \tag{31}$$

The cross section for electron detachment upon separation of the atoms is given by

$$\sigma_{ion}\approx\pi R_0^2w_{12}^{e}(1-w_{12}^{e}), \tag{32}$$

which, like the cross section (21), can give a pronounced growth at low velocities. If at infinity the excited state of the quasimolecule correlates with the excited term of the particle B, and the collision time is sufficiently long, i.e., if

$$\Gamma R_0/v_R>1, \tag{33}$$

then the cross section (32) corresponds to a peculiar Penning effect in collisions between unexcited atoms. A similar process for the case of neutral-particle ionization has, in fact, recently been experimentally observed (for the Rb-Ar pair at T = 200 eV[11], when the higher-lying terms of the Rydberg crowding are, apparently, still not accessible). The presence of the Rydberg crowding for the case of ionization significantly complicates the determination of the spectrum of the electrons emitted upon separation of the nuclei, but it should be expected that for

$$\Gamma R_0/v_R<1$$

the high-energy part of the spectrum ($E \gtrsim 10$—20 eV) contains, to a comparable degree, a contribution from many of the autoionization states of the atoms. The threshold for their appearance and the excitation threshold for the core electrons in collisions between the corresponding ions

$$A^++B\to(A^+)^*+B$$

should then roughly coincide, which is well corroborated by the available experimental data. For the K-He, Rb-He, Cs-He, and Cs-Ne pairs the thresholds for the appearance of bunches of resonance electrons[9] and the excitation thresholds for the corresponding K^+-He, Rb^+-He, Cs^+-He, and Cs^+-Ne ions[14], for example, are respectively equal: 84 and 75 eV, 67 and 81 eV, 116 and 116 eV, and 340 and 390 eV.

The distinctive features of the spectrum of the electrons emitted as the nuclei approach each other and the quasimolecule-autoionization probability are determined by the magnitudes and energy dependence of the matrix elements entering into (23). The role of the centrifugal barrier and the Rydberg crowding has already been discussed[1,3,4]; therefore, we only note that the presence in the field of the unexcited quasimolecular core of fine levels (real or virtual) and quasi-stationary states leads to the appearance in the low-energy electron spectrum of resonance peaks:

$$n_2(E)=\frac{aE^{1/2}}{\varepsilon_0+E}\exp\Big[-\int_0^E\frac{aE^{1/2}\,dE}{\varepsilon_0+E}\Big],$$

$$n_2(E)=\frac{b}{(\varepsilon_0-E)^2+\Gamma^2/4}\exp\Big[-\int_0^E\frac{b\,dE}{(\varepsilon_0-E)^2+\Gamma^2/4}\Big] \qquad (34)$$

(a and b are constants) whose experimental detection would allow us to obtain unique information about the spectrum of the excited states of the corresponding quasimolecules.

4. Let us now discuss the question of the role of the "inelastic" ionization channel, i.e., of ionization which is accompanied by the simultaneous excitation of the inner-shell electrons (excitation energy $\omega_{23} = E_3^{AB} - E_2^{AB}$) and which becomes possible at outer-electron energies $E > \omega_{23}$. In the presence of pseudocrossing of the ground-state term with two excited-state ones, the integral spectrum of the emitted electrons evidently has the form

$$n(E)=n_2(E)+n_3(E)=\frac{\Gamma_2(E)}{\beta}\Big[\exp\Big(-\int_0^E\Gamma_2(E)\frac{dE}{\beta}\Big)+q(E)\exp\Big(-\int_{\omega_{23}}^{\omega_{23}+E}\Gamma_2(E)\frac{dE}{\beta}-\int_0^E\Gamma_3(E)\frac{dE}{\beta}\Big)\Big], \qquad (35)$$

$$q(E)=\frac{\Gamma_3(E)}{\Gamma_2(E)}\exp\Big(-\int_0^{\omega_{23}}\Gamma_2(E)\frac{dE}{\beta}\Big),$$

i.e., for a sufficiently low threshold for $q \gtrsim 7$ the number of electrons emitted with low energies can grow with the energy of the colliding particles. This interesting fact was recently observed experimentally for the Rb-Kr, Na^+-Kr, and some other pairs [11].

5. It is also of interest to consider the question of the possibility of the appearance of near-threshold anomalies in the electron spectrum of the elastic channel. For this purpose it is sufficient to consider a model three-channel system with a parabolic term[2)]:

$$E_1^{AB}(t)=-\alpha t^2+\beta,\quad \alpha>0,\ \beta>0;$$
$$E_2^{AB}(t)=0,\quad E_3^{AB}(t)=\omega_{23}<\beta.$$

The electron spectrum of the elastic channel in this case has, as can be shown, the form

$$n_2(E)=2\pi(\alpha\beta)^{-1/2}|\langle f_{2E}|V_{12}|f_{1n_0}\rangle|^2|F(E)|^2\rho_2(E). \qquad (36)$$

Here F(E) is a damped—as $E \to +\infty$—solution of the equation

$$d^2F/dE^2+\Omega^2(E)F=0,$$
$$\alpha\Omega^2(E)=\beta-E+\sum_{s=2,3}\langle f_{1n_0}|V_{1s}G_sV_{s1}|f_{1n_0}\rangle. \qquad (37)$$

Below we shall be interested only in the near-threshold energy region ($E \approx \omega_{23}$), and therefore let us represent $\Omega^2(E)$ in the form

$$\Omega^2(E)=A+B[2(E-\omega_{23})]^{1/2}, \qquad (38)$$

where A and B are electron-energy independent quantities. Then using the standard-equation method (see, for example, [19]) for finding the asymptotic form of F(E), we obtain the form of the spectrum near the threshold of the "inelastic" ionization channel:

$$n_2(E)=n_2(\omega_{23})\Big\{1-\frac{(2|E-\omega_{23}|)^{1/2}}{(\beta+\mathrm{Re}\,A)^2+(\mathrm{Im}\,A)^2}[D_1\eta(\omega_{23}-E)+D_2\eta(E-\omega_{23})]\Big\};$$
$$D_1=\mathrm{Im}\,A\cdot\mathrm{Re}\,B-\mathrm{Im}\,B\cdot\mathrm{Re}(A+\beta),$$
$$D_2=\mathrm{Im}\,A\cdot\mathrm{Im}\,B+\mathrm{Re}\,B\cdot\mathrm{Re}(A+\beta).$$

The threshold anomaly is described by the second term in the square brackets and vanishes when we go over to the conditions of the linear approximation.

The author is deeply grateful to Yu. F. Bydin and V. I. Ogurtsov for making the results of their experiments available to us, as well as for a discussion of the interpretation offered in the present paper, to O. B. Firsov for a discussion of part of the expounded results, and to A. S. Kompaneets and the participants of the seminar supervised by him for a discussion of the paper as a whole.

1) A number of authors [1, 3, 16] formulate the boundary conditions at the origin (i.e., at a = 0).

2) We note that the single-channel decay in the small-radius-potential model with a quadratic approximation for the boundary condition was recently considered by Devdariani [19].

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Translated by A. K. Agyei
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