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Influence of symmetry on the exchange of one and two electrons in atomic collisions

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A theoretical investigation is made of one-and two-electron charge exchange in collisions between an atom and an ion of the same element, the latter with two missing electrons. It is shown that the probability of one-electron exchange in the case when the initial term crosses the final ground-state term is half the usual probability for reasons of symmetry: the atom and ion are identical. The occurrence of term crossing in this system alters the physical nature of resonant two-electron exchange. There is a new channel for two-stage exchange of two electrons. The first electron is released on the first pseudocrossing of terms and the second on the second pseudocrossing. In the case of crossing with terms of the excited state, this exchange occurs if the excitation is transferred during the time between the two pseudocrossings. The experimental cross section for the exchange of two electrons in a collision of a negative hydrogen atom with a proton can be ascribed completely to this new channel.

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1. We shall consider the exchange of one and two electrons in slow ($v < 2\times10^8$ cm/sec) collisions between an atom and an ion of the same element but with two missing electrons:

$$A + A^{++} \nearrow A^+ + A^+ \quad (1a)$$

$$A + A^{++} \searrow A^{++} + A. \quad (1b)$$

It is found that the symmetry resulting from the fact that the particles A and A^{++} represent the same element has an important influence on the physical nature of these processes and their effective cross sections.

The exchange of one electron, process (1a), is nonresonant and due to the crossing of the energy terms of the initial and final states. The exchange of two electrons is a resonant process and its probability in the absence of one-electron term crossing has already been studied.[1-2] However, the occurrence of such crossing greatly alters this process.

The absolute magnitude of the term repulsion responsible for the two-electron exchange[2,3] is considerably less than the splitting in the one-electron exchange. Therefore, when an atom approaches an ion, the one-electron exchange may occur earlier. This would seem to block the two-electron exchange channel (1b) to an extent increasing with the probability of the one-electron exchange during the collision time, i.e., the time interval between two passages of an atom through the same pseudocrossing. In fact, the situation is different. When the probability of the one-electron exchange is high, new ways of exchanging two electrons become possible:

$$A + A^{++} \nearrow A^+ + A^+ \to A^{++} + A, \quad (2a)$$

$$A + A^{++} \searrow A^+ + (A^+)^* \to (A^+)^* + A^+ \to A^{++} + A, \quad (2b)$$

which will be investigated below; it should be noted that the chain of events (2) occurs during the same collision.

Let us assume that, in the process (2b), the probability of a nonadiabatic transition on pseudocrossing of one-electron terms is small. Then, after the first passage of the atom through this pseudocrossing, an electron is very likely (probability ~1) to be captured by the ion. If there is no event up to the second passage through the pseudocrossing, the probability of the electron returning to the atom as a result of the second pseudocrossing is equally high and the probability of one-electron charge exchange is low. If excitation is exchanged between the

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two passages through the same quasicrossing, the first electron can no longer return to the atom since it is now in the ground state. The probability of finding the second electron at the same adiabatic term as the first electron is equally high, i.e., the second electron is transferred to a new atom. This gives rise to the two-electron exchange.

This simple discussion is sufficient to show that, at low collision rates, the effective two-electron exchange cross section of the channel (2b) may be larger than the one-electron charge-exchange cross section or the cross section of the channel (1). This is explained by the relatively large splitting of the terms responsible for the transfer of excitation. For example, when the dipole transition $(A^+)^* - A^+$, is allowed for, the splitting decreases proportionally to R^{-3} (R is the internuclear distance), whereas the splitting responsible for the two-electron exchange decreases exponentially.[2,3]

2. The probability of two-electron exchange in accordance with the direct channel (1b), like the probability of the one-electron exchange, is due to dephasing of the even and odd quasimolecular states, governed by the difference between their energies (we have in mind here the parity in respect of inversion of the coordinates of all the electrons at the center of the molecule AA^{++}). Komarov and Yanev[2] report the asymptotic result that, for large internuclear distances $R \to \infty$, this difference between the energies decreases proportionally to $\exp(-2\alpha R)$, where $\alpha = (2I_1)^{1/2}$ and I_1 is the first ionization potential of the atom A. It should be stressed that the argument of this exponential function does not include the second ionization potential, although we are dealing with the two-electron exchange. This result is due to crossing transitions of electrons in which the energy of each of them changes[3] (but the total energy remains constant). Such a transition occurs only as a result of the electron–electron interaction, which reduces the preexponential term by the factor R^{-3} (Ref. 3).

The cross section of the two-electron exchange is[1]

$$\sigma_1 = \frac{\pi R_0^2}{2}, \quad \left(\frac{\pi R_0}{4\alpha}\right)^{1/2} \Delta E_1(R_0) = 0.28v, \tag{3}$$

where ΔE_1 is the difference between the energies of the even and odd states, associated with the simultaneous exchange of two electrons between atoms. It was calculated earlier.[2,3]

A term of the $A+A^{++}$ system may intersect (generally speaking, excited) terms of the system $A^+ + (A^+)^*$. If the two-electron exchange radius R_0 in Eq. (3) exceeds the distances at which the crossing takes place, the influence of the one-electron exchange on the two-electron process is small and, in this case, Eq. (3) applies. In the opposite case, the two exchange processes should be considered simultaneously, as below.

3. We shall first consider the case (2a) of the crossing of the terms of the system $A+A^{++}$ with the ground-state term of the $A^+ + A^+$ system, when both A^+ ions are in the ground state. We have to consider here the interaction between three terms: two terms of the $A+A^{++}$ system—the terms even $E_+(R)$ and odd $E_-(R)$ relative to the inversion of the coordinates of the two electrons at

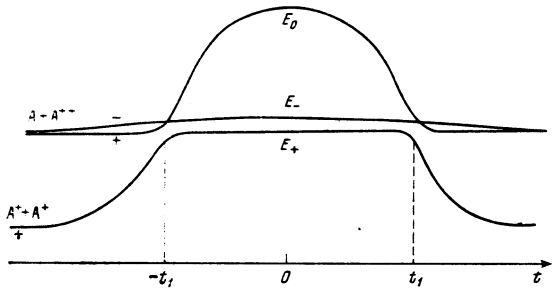

FIG. 1. Crossing of the initial state of the $A+A^{++}$ system with the final ground state of the $A^+ + A^+$ system.

the center of the quasimolecule, i.e., at the point which halves the internuclear axis—and one term $E_0(R)$ of the $A^+ + A^+$ system, which has a definite parity, either + or − (it is assumed that the ground state of the A^+ ion is not degenerate). These terms are represented schematically in Fig. 1 as a function of the collision time. We have to consider only that state of $A^+ + A^+$ whose spin is equal to the spin of the atom A because the total electron spin is conserved in the nonrelativistic approximation. For example, for particles A with two excess electrons (in addition to those in the filled shells), the spin of two electrons is either zero or unity. Consequently, in the $A^+ + A^+$ system, we have to consider either the singlet or triplet states, and the singlet state is even relative to inversion at the center of the quasimolecule, whereas the triplet term is odd. Figure 1 shows the case of the even state of $A^+ + A^+$. The odd state is described by all the formulas given below if + is exchanged with −; all the conclusions still apply to this case.

In general, the triplet state of the $A^+ + A^+$ system, which is odd relative to inversion, also interacts with the odd state of the $A + A^{++}$ system. This interaction is of the spin–orbit type; it is weak and we shall ignore it for large interatomic distances.

We shall divide the time axis into five intervals, as shown in Fig. 2. The one-electron charge exchange occurs in the intervals II and IV. In the interval I for $t \to -\infty$, the initial conditions correspond to an $A + A^{++}$ collision:

$$\Psi(r_1, r_2, t) \xrightarrow[t\to-\infty]{} \frac{1}{\sqrt{2}}\left[\Psi_+(r_1, r_2)\exp\left\{-i\int_{-\infty}^{t} E_+\,dt'\right\} + \Psi_-(r_1, r_2)\exp\left\{-i\int_{-\infty}^{t} E_-\,dt'\right\}\right]. \tag{4}$$

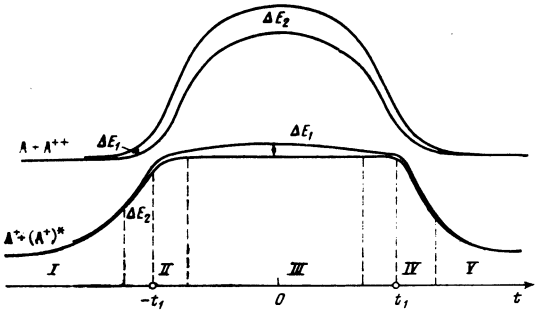

FIG. 2. Crossing of terms with an excited state of the $A^+ + (A^+)^*$ system.

The quasimolecular functions $\Psi_\pm$ reduce, in the limit $R\to\infty$, to combinations of the atomic functions $\Psi_{I,II}$ corresponding to the case when both electrons are near the nucleus I or II:

$$\Psi_\pm \underset{R\to\infty}{\longrightarrow} \frac{1}{\sqrt{2}}(\Psi_I \pm \Psi_{II}). \tag{5}$$

For identical atoms, the center of the quasimolecule coincides with the center of gravity. Therefore, the parity in respect of inversion is retained for the whole collision. This means that only the states of the same parity interact with one another. States of different parity are not mixed. However, this conclusion is not self-evident and we shall demonstrate it. In the adiabatic basis of wave functions, a transition is governed by the matrix elements of the derivative $\partial/\partial t$, i.e., by the quantities

$$\int \Psi_-(\mathbf{q},t)\frac{\partial}{\partial t}\Psi_+(\mathbf{q},t)\,d\mathbf{q} = \langle - | \partial/\partial t | + \rangle.$$

Here, $\mathbf{q}$ is the set of coordinates of all the electrons.

The derivative $\partial\Psi_+/\partial t$ has the same parity as the function being differentiated because the parity of the functions is conserved at any moment t. Hence, it follows that the matrix elements vanish. $\langle + | \partial/\partial t | - \rangle = 0$, so that the + and − functions are not mixed.

Let ω be the probability of a nonadiabatic transition for a single passage through a pseudocrossing. In the region III, subject to the initial condition (4), the wave function is then

$$\Psi|_{-t_1<t<+t_1} = \left(\frac{\omega}{2}\right)^{1/2}\Psi_+ \exp\left\{-i\int_{-\infty}^{t} E_+\,dt'\right\}$$
$$+\left(\frac{1-\omega}{2}\right)^{1/2}\Psi_0 \exp\left\{-i\int_{-\infty}^{-t_1} E_+\,dt' - i\int_{-t_1}^{t} E_0\,dt'\right\} + \frac{1}{\sqrt{2}}\Psi_- \exp\left\{-i\int_{-\infty}^{t} E_-\,dt'\right\}; \tag{6}$$

Here, $\pm t_1$ are the moments of passage through a pseudocrossing.

Similarly, if we consider the system passing through a pseudocrossing at a time $+t_1$ when the particles fly apart, we obtain the probability of the two-electron exchange during the whole collision ($A+A^{++}\to A^{++}+A$):

$$P_2 = \sin^2\left(\frac{\chi_1-\chi_2}{2}\right)$$
$$+\omega\left[\sin\frac{\chi_1+\chi_2}{2}\sin\frac{\chi_1-\chi_2}{2} - \sin^2\frac{\chi_1-\chi_2}{2}\right] + \omega^2\sin^2\frac{\chi_1}{2}, \tag{7}$$

where the phases $\chi_{1,2}$ are

$$\chi_1 = \int_{-t_1}^{+t_1}(E_0 - E_+)\,dt', \tag{8}$$

$$\chi_2 = \int_{-\infty}^{+\infty}(E_- - E_+)\,dt'. \tag{9}$$

The phase χ_2 determines the contribution of a simultaneous jump of two electrons. If this phase is small, Eq. (7) reduces to

$$P_2 \approx (1+\omega^2)\sin^2\frac{\chi_1}{2}, \qquad \chi_2 \ll \chi_1. \tag{10}$$

The interference phase χ_1 is large if it is goverened by the Coulomb displacement of the term E_0: $(E_0 - E_+) \propto R^{-1}$ if the crossing occurs in the asymptotic range of large interatomic distances. Therefore, averaging Eq. (10) over a small interval of the impact parameters $\Delta\rho$, we find that

$$\bar{P}_2 \approx (1+\omega^2)/2. \tag{11}$$

When the probability of a nonadiabatic transition is $\omega \to 0$, the formulas (10) and (11) reduce to

$$P_2 \approx \sin^2(\chi_1/2), \tag{12}$$

$$\bar{P}_2 \approx 1/2. \tag{13}$$

In the limit $\omega \to 0$, the even component of the $A+A^{++}$ system most probably follows the adiabatic term $E_+ \to E_0 \to E_+$. In this case, the dephasing of the quasimolecular + and − states and the exchange of two electrons are goverened only by the phase difference $E_0 - E_+$, which is reflected in Eq. (12).

The probability P_1 of the one-electron charge exchange throughout the whole collision is

$$P_1 = \omega(1-\omega)[1+\cos\chi_1]. \tag{14}$$

Averaging this expression over the small interval $\Delta\rho$, we obtain

$$\bar{P}_1 = \omega(1-\omega). \tag{15}$$

This quantity is half the usual value because, in the symmetric $A+A^{++}$ case, half the colliding atoms are in the noninteracting state Ψ_-. The result (15) is only valid for a collision of an atomic particle A with a corresponding ion A^{++}, which has two electrons less, when the $A^+ + A^+$ state is practically the only one. For example, in an $A+A^{+++}$ collision, even after one-electron charge exchange to the ground state, $A+A^{+++}\to A^+ + A^{++}$, the resultant particles are different so that the $A^+ + A^{++}$ quasimolecule has two (+ and −) degenerate states in the limit $R\to\infty$ for each value of the total spin and charge exchange is possible to either of these states.

If $\omega \to 1$, the system jumps a pseudocrossing without being affected by it and then the probability of the two-electron charge exchange (7) tends to the earlier value[1-3]:

$$P_2 \underset{\omega\to 1}{\longrightarrow} \sin^2(\chi_2/2). \tag{16}$$

It follows from Eqs. (12) and (13) that, in the case of adiabatically low collision rates so that $\omega \to 0$, the effective cross section for the two-electron exchange is $\sigma_2 \to \pi R_1^2/2$ (R_1 is the pseudocrossing radius) and it is greater than even the maximum one-electron charge exchange cross section to the ground state, irrespective of the term splitting $\Delta E_1 = E_- - E_+$ responsible for the two-electron exchange.

We can thus see that the symmetry of the molecules formed as a result of a collision has a considerable influence on the probabilities of exchange of one and two electrons.

The above results are derived on the assumption that the laws governing the changes in the phases of the adiabatic states, i.e., the functions $E(t)$, change pre-

cisely at the pseudocrossing moments $\pm t_1$. This is justified if the size of the region where the exact adiabatic terms differ greatly from the unperturbed terms is small compared with R_1. This condition is satisfied if the repulsion of terms in a pseudocrossing is small compared with $E_0 - E_\pm$ for $\rho \lesssim R_1$, which is clearly valid because $(E_0 - E_\pm) \propto R^{-1}$ and the repulsion of the terms is exponentially small for large values of R_1.

4. We shall now consider the case (2b) when the terms of the initial system cross the terms of the $A^+ + (A^+)^*$ excited states. In this case, the atomic particles are in different states and, therefore, quasimolecular states are always degenerate relative to inversion, i.e., there are always even and odd states with the same energy in the limit $R \to \infty$ [even if the atomic excited state $(A^+)^*$ is not itself degenerate].

Let us assume that the atomic excited state $(A^+)^*$ is not degenerate. In this case, the terms behave as shown in Fig. 2. Applying the above procedure, we obtain the probability of the two-electron exchange:

$$P_2^- = \omega^2 \sin^2\left(\frac{1}{2}\int_{-\infty}^{+\infty} \Delta E_1\, dt'\right) + (1-\omega)^2 \sin^2\left[\int_{-\infty}^{-t_1} \Delta E_1\, dt' + \frac{1}{2}\int_{-t_1}^{t_1} \Delta E_2\, dt'\right], \tag{17}$$

where ΔE_2 is the splitting of the terms responsible for the excitation transfer $A^+ + (A^+)^* \to (A^+)^* + A^+$, which occurs during the motion of the particles between two pseudocrossing moments $\pm t_1$. It is assumed that the splittings $E_- - E_+$ are much smaller than the matrix element of the one-electron exchange, so that the repulsion of the terms and the probability of the one-electron transition ω are the same for the + and − states. Averaging the interference terms has already been carried out in Eq. (17).

The probabilities of the one-electron charge exchange under conditions such that the excitation remains on the particle which is initially a doubly charged ion, P_{1I}, and in the case when it remains at the other particle, P_{1II}, are

$$P_{1I} = \omega(1-\omega)[1+\cos\chi_3 \cos\chi_4], \tag{18}$$

$$P_{1II} = \omega(1-\omega)[1-\cos\chi_3 \cos\chi_4], \tag{19}$$

where the phases $\chi_{3,4}$ are

$$\chi_3 = \frac{1}{2}\int_{-\infty}^{+\infty} (\Delta E_1 + \Delta E_2)\, dt, \tag{20}$$

$$\chi_4 = \frac{1}{2}\int_{-t_1}^{t_1} (\Delta E_2 - \Delta E_1)\, dt. \tag{21}$$

The total probability of the one-electron charge exchange is $P_1 = P_{1I} + P_{1II} = 2\omega(1-\omega)$, which differs from the preceding case because the final quasimolecular state is now, like the initial state, doubly degenerate (+ and −).

In the general case of crossing with N terms of the $A + A^{++}$ system, the formula (17) becomes

$$P_2^N = \omega_1^2\omega_2^2 \dots \omega_N^2 \sin^2\left(\frac{1}{2}\int_{-\infty}^{+\infty} \Delta E_1\, dt\right) + \sum_{k=1}^{N} \omega_1^2\omega_2^2 \dots \omega_{k-1}^2 (1-\omega_k)^2 \sin^2\left[\int_{t_k}^{\infty} \Delta E_1\, dt + \frac{1}{2}\int_{-t_k}^{t_k} \Delta E_2^{(k)}\, dt\right]. \tag{22}$$

We shall now consider the above expressions for the charge-exchange probabilities. The first term in Eq. (17) is the probability of direct charge exchange in accordance with Eq. (1). It is ω^2 times smaller than the usual probability because of the presence of the one-electron charge-exchange channel. In the range of adiabatically low rates $\omega \to 0$, even the direct channel is closed. However, a second channel then opens up: the second term in Eq. (17) becomes more important. This second channel is also important for $\omega \sim 1 - \omega$ because the splitting of the terms resulting in excitation transfer is usually greater than the two-electron exchange splitting $|\Delta E_2| > |\Delta E_1|$.

It is worth noting some analogy as well as the difference between the simultaneous jump of two electrons from one particle to another, represented by the channels (1) and (2). In both channels, the transfer of two electrons occurs apparently through the outer orbits of the atoms and, therefore, we have $\Delta E_1 \propto \exp(-2\alpha R)$, except that, in the channel (2), the two electrons are not transferred simultaneously. After the first charge exchange in the region II, the atoms move in the region III and excitation transfer takes place: the electron remaining at the first atom becomes excited and the electron transferred to an excited orbit of the ion drops to the ground state. In the region IV, a second electron is then transferred again to the outer orbit.

5. We shall now consider the case of a collision of a negative hydrogen atom with a proton, $H^- + H^+$, for which the two-electron charge-exchange cross section was determined experimentally[4] in the relative energy range 50–190 eV or at relative velocities $v \approx 1.5$–2.5×10^7 cm/sec (≈ 0.07–0.12 a.u.). For this case, the term splitting of the direct channel is[3]

$$\Delta E_1 \approx 7.9 \cdot 10^{-7} R^{3.49} e^{-0.47R}. \tag{23}$$

This expression has a maximum at $R \approx 7.4$ and its value is then $\Delta E_1(7.4) = 2.7 \times 10^{-5}$ a.u. Such small splitting means that, even if the direct charge exchange does occur, this will happen outside the asymptotic range of the interatomic distances.

We shall now estimate the probability product $\omega_1^2 \cdots \omega_\nu^2$, which occurs in the first term of Eq. (22) and which represents the probability of the evolution of the system in accordance with the channel (1). The ionic term $H^- + H^+$ crosses three terms of the $H(1s) + H^*(n)$ system with $n = 4$, 3, and 2. The crossing with the $n = 4$ level can be ignored as too distant. We shall estimate the probabilities ω_3 and ω_2 using the Landau–Zener model:

$$\omega = \exp\left[-\frac{\pi}{2}\frac{R^2 \delta E^2}{v}\right]. \tag{24}$$

According to the δ-potential model of a negative ion, the charge exchange occurs only to one of the n^2 degenerate states. Using the parameters $R_2 = 11.1$, $\delta E_2 = 1.00$ eV, $R_3 = 35.6$, and $\delta E_3 = 0.0165$ eV, obtained earlier,[5] we have

$$v = 0.1 \text{ a.u.}, \quad \omega_3 = 0.993; \quad \omega_2 = 0.074 \tag{25}$$

for the case when the radial velocity is $v_{rad} \approx v$ ($\rho < R_{2,3}$).

If the phase difference required for the direct charge-exchange process is accumulated at $\rho \sim 1$ a.u. (or $\sim a_0$, which is the Bohr radius), then, subject to allowance for weakening by a factor $\omega_2^2\omega_3^2 = 5.5\times10^{-3}$, we can estimate the contribution of the direct channel to the total two-electron charge-exchange cross section, which gives the value $\sigma_1(v=0.1) \leq 5\times10^{-19}$ cm^2 two orders of magnitude less than the experimental cross section: $\sigma_{exp}(v=0.1) \approx (4\pm2)\times10^{-17}$ cm^2. Consequently, in this case, the whole cross section is goverened by the second channel.

We shall now estimate the cross section for the two-electron exchange $H^- + H^+ \to H^+ + H^-$, in accordance with the second channel for the case when $v=0.1$ and we shall do this using the second term of Eq. (17). The contribution of the term $H^*(n=3)$ can be ignored and allowance need only be made for the crossing with the term $H^*(n=2)$, which is characterized by $(1-\omega)^2 \approx 1$. We can then see that the probability of the two-electron exchange is simply equal to the probability of the exchange of excitation in a time between $-t_1$ and $+t_1$:

$$P_2^* \approx \sin^2\left(\frac{1}{2}\int_{-t_1}^{t_1}\Delta E_2\,dt\right)|_{H^-+H^+;\,v=0.1}. \tag{26}$$

The application of Eq. (17) to the case under consideration requires further discussion because the excited states $H^*(n)$ are degenerate. This application is possible if there is no mixing of the degenerate states and this will be assumed here. The probability (26) becomes ~1 for the impact parameters $\rho < R_2 = 11.1a_0$ (the trajectory is assumed to be rectilinear), so that (according to Ref. 6, $\Delta E_2 = 2d_{12}/R^3$)

$$\frac{1}{2}\int_{-t_1}^{t_1}\Delta E_2\,dt \leqslant \frac{|d_{12}|^2}{v\rho^2}\int_{-\infty}^{+\infty}\frac{d\xi}{(1+\xi^2)^{3/2}} = \frac{2|d_{12}|^2}{v\rho^2}, \tag{27}$$

where $|d_{12}|^2$ is the square of the matrix element of the dipole moment: $|d_{12}|^2 = |\langle\psi_{1s}|r\cos\theta|\psi_{2p0}\rangle|^2 = 0.551$ for the $2p_0 \to 1s$ transition in the hydrogen atom. Estimating the cross section from $\sigma \sim \pi a_0^2\rho_0^2/2$, where ρ_0 is the impact parameter for which the phase (26) becomes equal to $\pi/2$, we obtain $\sigma(v=0.1) \approx 3\times10^{-16}$ cm^2 ($\rho_0 \approx 2.6$). The degenerate states are most likely to be mixed in the $\rho \sim \rho_0$ case, so that the probability of charge exchange decreases by a factor of n^2. We then obtain $\rho \sim 7.5\times10^{-17}$ cm^2, which is close to the experimental value $(4\pm2)\times10^{-17}$ cm^2. These estimates indicate that the two-electron exchange $H^- + H^+ \to H^+ + H^-$ does indeed occur in accordance with the channel (2b). The exact theoretical value of the cross section can be obtained by solving the many-level problem.

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