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RESONANT CHARGE EXCHANGE IN SLOW COLLISIONS INVOLVING HALOGENS AND OXYGEN

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The coupling of electron momenta is considered for the resonant charge exchange process in slow collisions. Because the electron transfer in this process occurs at large distances between the colliding atomic particles, where ion–atom interactions are relatively weak, we can separate different types of interaction and find the character of coupling of the electron momenta in the quasi-molecule consisting of the colliding ion and its atom for real collision pairs. Since the real number of interaction types for colliding particles exceeds that used in the classical Hund coupling scheme, there are intermediate cases of momentum coupling outside the standard Hund scheme. This occurs for the resonant charge exchange involving halogens and oxygen where the quantum numbers of the quasi-molecule in the course of the electron transfer are the total momenta J and j of the colliding ion and atom and the projection M or M_J of the atom orbital or total momentum on the quasi-molecule axis. The ion–atom exchange interaction potential is independent of the ion fine state, and under these conditions, the resonant charge exchange process is not entangled with the rotation of electron momenta, as in the case «a» of the Hund coupling. The partial cross section of the resonant charge exchange process depends on quantum numbers of the colliding particles. The average cross sections weakly depend on the coupling scheme.

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1. INTRODUCTION

The process of resonant electron transfer in slow collisions of an ion and the parent atom results in transition of a valence electron from one core to another. In the simplest case of the transition of an s-electron, this process is determined by the interference of two electron terms of the quasi-molecule consisting of the colliding ion and the atom. Correspondingly, the probability of this transition is expressed through the energy difference for the even and odd quasi-molecule states and the cross section of this process [1]. In the case of the transition of a p-electron involving an ion and an atom with unfilled electron shells, the resonant charge exchange process becomes more complex, because the electron transfer can be entangled with the processes of rotation of electron momenta and transitions between fine states of the colliding particles. One can simplify the analysis of this process by constructing a hierarchy of interactions in the quasi-molecule and thus choosing a suitable case of the Hund coupling [2–4] that corresponds to certain quantum numbers of the quasi-molecule. Although this scheme is related to diatomic molecules when the distance between the nuclei is fixed, it can be extended to a quasi-molecule consisting of colliding particles [5–8]. According to the general method by Nikitin [5–8], the trajectory is then divided into several parts such that the Hund coupling of a certain type is realized in each part. If the transition range between different cases of the Hund coupling is narrow, one can construct the wave function of colliding particles and the S-matrix of the transition by sewing the wave functions on different sides of the transition range [7]. This allows one to separate different processes and to find the probabilities for the variation of quantum numbers of the colliding particles at a given collision trajectory.

This general scheme can be used in the analysis of the resonant charge exchange process involving an ion and an atom with unfilled electron shells when the electron momenta can be coupled via different schemes, and the resonant charge exchange process can therefore be entangled with other processes (rotation of the electron momenta and transitions between fine structure states) in different ways. Indeed, within the framework of the classical Mulliken scheme of the momentum sum-

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mation [2], three types of interactions are introduced for a quasi-molecule: the electrostatic interaction V_e is responsible for the energy splitting of different angular momentum projections onto the molecule axis; δ_f corresponds to the spin–orbit interaction and other relativistic interactions; and the rotational energy or the Coriolis interaction V_{rot} accounts for the interaction between the orbital and spin electron momenta with the rotation of the molecular axis. Depending on the ratio between these interaction energies, one can construct six cases of the Hund coupling [2–4]; each of these cases corresponds to a certain scheme of momentum summation and is characterized by certain quantum numbers of the diatomic molecule. These cases are used as model ones in the analysis of some transitions in atomic collisions [7–9].

For the resonant charge exchange in slow collisions, the electron transfer from one core to another proceeds at large distances between the colliding particles, where interactions of different types are weak. This simplifies the general analysis of the charge exchange process and gives an additional experience in understanding the momentum coupling. As a result of this analysis, we find a number of interactions to be actually greater than within the framework of the Hund scheme. Indeed, the electrostatic interaction V_e includes the exchange interaction V_{ex} inside the atom, which leads to certain orbital momenta L of the atom and l of the ion, and to certain spins S and s of these atomic particles, such that V_{ex} characterizes the energy splitting of states with different quantum numbers $LSls$. A long-range interaction $U(R)$ and the ion–atom exchange interaction $\Delta(R)$ are added to this. In addition, the fine splitting of levels refers to the atom (δ_a) and ion (δ_i) separately, and competition between all these interactions gives rise to many other cases of momentum coupling compared to the Hund coupling scheme. In analyzing this problem for real ion–atom systems, we deal with a restricted number of momentum couplings. Below, we consider this problem for the resonant charge exchange of halogens and oxygen in the case where the ions and atoms are found in the ground state and the collision energies vary from thermal ones up to tens of electronvolts (this energy range is of interest for a low-temperature plasma).

2. ASYMPTOTIC THEORY OF THE RESONANT ELECTRON TRANSFER

We first formulate a general method to analyze this problem. We use the asymptotic theory of the resonant charge exchange [9–12], where the electron transfer has the tunnel character and large impact parameters of collisions give the main contribution to the cross section of this process. A reciprocal value of a typical impact parameter of collisions is a small parameter in the asymptotic theory of the resonant charge exchange process. Expanding the cross section with respect to this small parameter and restricting ourselves by two expansion terms, we can express the ion–atom exchange interaction potential and the cross section through asymptotic parameters of the transferring electron in an isolated atom and quantum numbers of the ion and atom electron shells. In contrast to models, the asymptotic theory allows us to find the correct value of the cross section with an estimated accuracy. For p-electron transitions in the collision energy range under consideration, this accuracy is better than 10 % [13, 14].

Table 1. The cases of Hund coupling

Hund case	Relation	Quantum numbers
a	$V_e \gg \delta_f \gg V_{rot}$	Λ, S, S_n
b	$V_e \gg V_{rot} \gg \delta_f$	Λ, S, S_N
c	$\delta_f \gg V_e \gg V_{rot}$	Ω
d	$V_{rot} \gg V_e \gg \delta_f$	L, S, L_N, S_N
e	$V_{rot} \gg \delta_f \gg V_e$	J, J_N

The asymptotic theory allows us to determine the ion–atom exchange interaction potential. The cross section of the electron charge exchange is then expressed through the exchange interaction potential for given quantum numbers of the quasi-molecule consisting of the colliding ion and atom at a given distance R between them. In constructing this interaction potential, we start from the Hund coupling scheme [2–4] represented in Table 1 together with the quantum numbers of the quasi-molecule for each case of the Hund coupling. We introduce the following notation: $\mathbf{L}$ is the total electron angular momentum of the molecule, $\mathbf{S}$ is the total electron spin, $\mathbf{J}$ is the total electron momentum of the molecule, $\mathbf{n}$ is the unit vector along the molecular axis, $\mathbf{N}$ is the rotation momentum of nuclei, Λ is the projection of the angular momentum of electrons on the molecular axis, Ω is the projection of the total electron momentum $\mathbf{J}$ on the molecular axis, S_n is the projection of the electron spin on the molecular axis, and L_N, S_N, and J_N are projections of these momenta onto the direction of the rotation momentum $\mathbf{N}$

of the nuclei. We take this scheme as a basis in considering the momentum coupling of the quasi-molecule consisting of a colliding ion and the parent atom.

We note two momentum coupling schemes for the atom and ion [4, 15, 16]. In the LS scheme, which is realized under the condition $V_{ex} \gg \delta_a, \delta_i$, we have LM_LS as the quantum numbers of the atom and lms as the quantum numbers of the ion, where L and l are the angular momenta, M and m are their projections on the molecular axis, and S and s are the spins of the atom and the ion respectively. In the j–j coupling scheme for an individual atomic particle, which occurs at $V_{ex} \ll \delta_a, \delta_i$, we use JM_J as the atom quantum numbers and jm_j as the ion quantum numbers, where J and j are the total electron momenta and M_J and m_j are their projections on the molecular axis for the atom and the ion respectively. These quantum numbers are the basis for the limiting cases of electron terms of the quasi-molecule.

The possible interaction potentials in the quasi-molecule under consideration are

$$\begin{gathered} V_{ex}, \quad U_M = \frac{Q_{MM}}{R^3}, \\ U_m = \frac{Q_{MM} q_{mm}}{R^5}, \ \Delta(R), \ \delta_i, \ \delta_a, \quad \text{and} \quad V_{rot}. \end{gathered} \tag{1}$$

We divide the electrostatic interaction V_e in Table 1 into four parts: the exchange interaction V_{ex} inside the atom and ion responsible for electrostatic splitting of levels inside the isolated atom and ion with given electron shells; the long-range interaction U_M of the ion with the quadrupole atom moment; the long-range interaction U_m responsible for splitting of the ion level; and the ion–atom exchange interaction potential Δ that determines the resonant charge exchange cross section. The fine splitting δ_f of levels in Table 1 is written separately for the ion (δ_i) and the atom (δ_a). Here, M and m are the projections of the atom and ion angular momenta on the molecular axis, R is the ion–atom distance, Q_{ik} is the tensor of the atom quadrupole moment, and q_{ik} is the quadrupole moment tensor of the ion. As can be seen, a number of possible coupling cases increases significantly in this description in comparison with the classical case. Of course, a small part of these cases can be realized, and we verify this below for certain cases of the resonant charge exchange.

To find the suitable momentum coupling scheme, we evaluate the above ion–atom interaction potentials at distances that determine the resonant charge exchange cross section. Constructing the hierarchy of interactions, we find the quasi-molecule quantum numbers in this distance range and the partial cross sections corresponding to these quantum numbers. This allows us to ascertain the momentum summation scheme in slow ion–atom collisions with a resonant electron transfer. We note that the character of momentum coupling influences the value of the average cross sections. Below, we realize this operation for certain cases of ion–atom collisions where the colliding ion and atom are found in the ground electron states.

3. HIERARCHY OF ION–ATOM INTERACTIONS FOR HALOGENS

We start with the resonant electron transfer involving a halogen atom and an ion in the ground electron states,

$$\mathrm{X}^+ + \mathrm{X} \to \mathrm{X} + \mathrm{X}^+, \tag{2}$$

where X is the halogen atom. In this case, the atom and ion ground states are $(p^5)^2P$ and $(p^4)^3P$, respectively, and all the interactions in (1) are therefore realized in this case. In Table 2, we collect some parameters of the colliding atomic particles in this case. We note that the lower fine structure states include states with the total electron momenta $j = 2, 1, 0$. The parameter δ_i in Table 2 is the splitting of the ${}^2P_2$–${}^2P_0$ ion levels. Next, the value V_{ex} is the splitting between the ground ion level ${}^2P_2$ and the level ${}^1D_2$ in the notation of the LS momentum coupling scheme. Because the ratio δ_i/V_{ex} is small for all ions, the LS momentum coupling scheme is valid in the ion, and we take it as a basis.

As a characteristic of the resonant charge exchange process, we take the average cross section σ_{ex} of the resonant charge exchange in the case «a» of the Hund coupling [13, 14, 19], and the corresponding impact parameter R_0 of collision is determined from the relation

$$\sigma_{ex} = \frac{\pi}{2} R_0^2. \tag{3}$$

In Table 2, we give the values of R_0 (in units of the Bohr radius a_0) at the collision energies 0.1, 1 (in parentheses), and 10 eV (in square brackets). These energies pertain to the laboratory reference frame, where the atom is motionless and the parent ion has the energy indicated. Using these values of R_0, we evaluate various interaction potentials and refer them to a given collision energy. The value U_M in Table 2 is equal to

$$U_M = U_{00} - U_{11} = \frac{Q_{00} - Q_{11}}{R_0^3} = \frac{6e^2\overline{r^2}}{5R_0^3}, \tag{4}$$

where e is the electron charge and r is the distance of a valence atomic p-electron from the nucleus inside the atom. In Table 2, we list the corresponding values

Table 2. Parameters of halogen atoms and ions [17, 18]

	F	Cl	Br	I
δ_a, cm^{-1}	404	882	3685	7603
δ_i, cm^{-1} *	490	996	3840	7087
V_{ex}, cm^{-1}	20873	11654	11410	13727
δ_i/V_{ex}	0.023	0.085	0.34	0.52
R_0, a_0	11.7(10.6)[9.54]	15.1(13.8)[12.3]	16.5(15.1)[13.6]	19.1(17.2)[15.8]
$\overline{r^2}$, a_0^2	1.54	4.06	5.22	7.20
U_M, cm^{-1}	253(341)[467]	311(407)[575]	306(448)[546]	272(372)[481]
U_M/δ_a	0.63(0.84)[1.16]	0.35(0.46)[0.65]	0.08(0.12)[0.15]	0.036(0.049)[0.063]
V_{rot}, cm^{-1}	8.6(30)[106]	4.9(17)[60]	3.0(10)[36]	2.0(7.1)[25]
$\Delta(R_0)$, cm^{-1}	7.0(23)[78]	4.2(14)[46]	2.6(8.4)[29]	1.8(6.1)[21]

* Energy differences for levels of the states $^3P_2$ and $^3P_0$.

of U_M and the ratio U_M/δ_a that is usually less than unity, and therefore the fine structure of level splitting is important for processes involving halogens.

If the colliding particles move along straight trajectories, the quasi-molecule rotation energy is given by

$$V_{rot} = \frac{\hbar v}{R_0} \tag{5}$$

at closest approach and at the impact parameter R_0 of the ion–atom collision, where v is the relative ion–atom velocity. According to the data in Table 2, the rotation energy is smaller than the other interaction potentials (U_M, δ_i, δ_a). This determines the character of momentum coupling in this case.

Based on the above analysis, we can construct a hierarchy of interactions for the quasi-molecule consisting of a halogen ion and the parent atom at the distances between these particles that determine the cross section of resonant charge exchange (2). The following hierarchy of interactions is valid for more or less all halogens in the range of collision energies 0.1–10 eV:

$$V_{ex} \gg \delta_i, \delta_a \gg U_M \gg U_m, V_{rot}. \tag{6}$$

In terms of the data in Table 1, this is an intermediate case between cases «a» and «c» of the Hund coupling. In addition, we evaluate the exchange ion–atom interaction potential $\Delta(R)$ using the formula for the resonant charge exchange cross section σ_{ex} for the transition of an s-electron [1, 10, 11],

$$\sigma_{ex} = \frac{\pi R_0^2}{2}, \tag{7a}$$

where

$$\frac{1}{v}\sqrt{\frac{\pi R_0}{2\gamma}}\,\Delta(R_0) = 0.28 \tag{7b}$$

and γ is the asymptotic parameter of the wave function of the transferred valence electron ($\Delta(R) \propto \propto \exp(-\gamma R)$). From this, we can compare the exchange interaction potential $\Delta(R_0)$ at the distance R_0 (Eq. (3)) with rotation energy (5) of the quasi-molecule at the impact parameter R_0 of collision and the minimal distance between the colliding ion and atom,

$$\Delta(R_0) = 0.28v\sqrt{\frac{2\gamma}{\pi R_0}} = 0.22\sqrt{\gamma R_0}\,V_{rot}. \tag{8}$$

A small parameter of the asymptotic theory is

$$\frac{1}{\gamma R_0} \ll 1. \tag{9}$$

At collision energies of several electronvolts, we have $\gamma R_0 \approx$ 10–15, and the above values are therefore comparable, $\Delta(R_0) \sim V_{rot}$. The values of the exchange interaction potential given in Table 2 confirm this statement.

The hierarchy of interactions in (6) leads to the quantum numbers $LSJM_J lsjm_j$ of the quasi-molecule, where L and l are the atom and ion angular momenta, S and s are the atom and ion spins, J and j are the total electron momenta of the atom and ion, and M_J and m_j are their projections on the molecular axis. The wave function is then given by

$$\Psi_{LSJM_Jlsjm_j} = \sum_{MM_Smm_s} \begin{bmatrix} L & S & J \\ M & M_S & M_J \end{bmatrix} \times \\ \times \begin{bmatrix} l & s & j \\ m & m_s & m_j \end{bmatrix} \Phi_{LMSM_S}\psi_{lmsm_s}, \quad (10)$$

where Φ and ψ are the respective wave functions of the weakly interacting atom and ion; the atom $(LMSM_S)$ and ion $(lmsm_s)$ quantum numbers are given with the spin–orbit and other relativistic interactions neglected.

Guided by the hierarchy (6) of interactions, we now find positions of the energy levels for the quasi-molecule under consideration and apply this to the case of halogen atoms. In the first approach, $LSls$ are the quasi-molecule quantum numbers, and in the case of halogen atoms, we are restricted by the lowest electron terms ^2P for the atom and ^3P for the ion. The second approach gives the quantum numbers J and j of the quasi-molecule with the splitting between the fine-structure levels determined by the corresponding values δ_a and δ_i for the isolated atom and ion. The third approach leads to the quantum number M_J of the quasi-molecule, i.e., the projection of the total atom electron moment on the molecular axis; the splitting between the levels with different M_J is then determined by the interaction of the ion charge and the atom quadrupole moment,

$$\Delta U(M_J) = \\ = \left\langle \Psi_{LSJM_{J_j}lsjm_j} \left| \frac{Q_{MM}}{R^3} \right| \Psi_{LSJM_{J_j}lsjm_j} \right\rangle = \\ = \sum_{MM_S} \frac{Q_{MM}}{R^3} \begin{bmatrix} L & S & J \\ M & M_S & M_J \end{bmatrix}^2. \quad (11)$$

For the interaction of the halogen atom and the ion, $X(^2P) + X^+(^3P)$, where X is the halogen atom, this formula becomes

$$\Delta U(JM_Jjm_j) = V_0 \left(2 \begin{bmatrix} L & S & J \\ 0 & M_J & M_J \end{bmatrix}^2 - \\ - \begin{bmatrix} L & S & J \\ 1 & M_J - 1 & M_J \end{bmatrix}^2 \right), \quad U_0 = \frac{2\overline{r_a^2}}{5R^3}, \quad (12)$$

where Q_{MM} is the component of the quadrupole moment tensor of the atom, r_a is the distance of the valence electron from the nucleus, and the bar denotes the average over electron positions in the atom.

The fourth approach corresponds to the quantum number m_j of the quasi-molecule, with the interaction potential between the ion and atom quadrupole momenta given by

$$\Delta U(JM_Jjm_j) = \\ = \frac{1}{R^5} \sum_{MM_S} Q_{MM} \begin{bmatrix} L & S & J \\ M & M_S & M_J \end{bmatrix}^2 \times \\ \times \sum_{mm_s} q_{mm} \begin{bmatrix} l & s & j \\ m & m_s & m_j \end{bmatrix}^2, \quad (13)$$

where q_{mm} is the component of the ion quadrupole moment tensor. We note that the electron terms of the quasi-molecule under consideration are degenerate with respect to the sign of the total momentum projections. For the interaction of atomic particles, $X(^2P) + X^+(^3P)$, where X is a halogen atom, this formula can be rewritten as

$$\frac{\Delta U(JM_Jjm_j)}{V_0} = -\left(2 \begin{bmatrix} L & S & J \\ 0 & M_J & M_J \end{bmatrix}^2 - \\ - \begin{bmatrix} L & S & J \\ 1 & M_J - 1 & M_J \end{bmatrix}^2 \right) \left(2 \begin{bmatrix} l & s & j \\ 0 & m_j & m_j \end{bmatrix}^2 - \\ - \begin{bmatrix} l & s & j \\ 1 & m_j - 1 & m_j \end{bmatrix}^2 \right), \quad (14)$$

where

$$V_0 = \frac{4\overline{r_a^2}\,\overline{r_i^2}}{25R^5} \quad (15)$$

and the distances r_a and r_i pertain to the atom and ion correspondingly.

As an example, we construct the lowest-energy levels for the chlorine ion–atom system at $R = 14a_0$ that characterizes the resonant charge exchange cross section at the collision energy 1 eV (see Table 2). At this distance, the energy of charge–quadrupole interaction (Eq. (12)) and quadrupole–quadrupole interaction (Eq. (15)) are $U_0 = 130$ cm^{-1} and $V_0 = 0.6$ cm^{-1}. In this case, we therefore have the following hierarchy of interactions (6):

$$V_{ex} \gg \delta_i, \delta_a \gg U_M \gg \Delta, V_{rot} \gg U_m. \quad (16)$$

In Table 3, we list the level energies E for chlorine given by

$$E = \delta_i + \delta_a + U_M + \varepsilon_0, \quad (17)$$

where ε_0 is taken such that the lowest electron term has zero energy $E = 0$. The quasi-molecule energies

Table 3. The lowest energy levels E of the quasi-molecule Cl^+–Cl at the distance $R = 14a_0$ between the nuclei. The statistical weight g pertains only to even (or odd) quasi-molecule states

JM_Jj	g	$\Delta U(M_J)$, cm^{-1}	E, cm^{-1}
$\frac{3}{2}\frac{3}{2}2$	20	−130	0
$\frac{3}{2}\frac{1}{2}2$	20	130	260
$\frac{3}{2}\frac{3}{2}1$	12	−130	696
$\frac{3}{2}\frac{1}{2}1$	12	130	956
$\frac{3}{2}\frac{3}{2}0$	4	−130	996
$\frac{1}{2}\frac{1}{2}2$	20	0	102
$\frac{3}{2}\frac{1}{2}0$	4	130	1256
$\frac{1}{2}\frac{1}{2}1$	12	0	1708
$\frac{1}{2}\frac{1}{2}0$	4	0	2009

in Table 3 pertain to the ground electron state of the atom and ion, i.e., $L = 1$, $l = 1$, $S = 1$, $s = 1/2$ for this term. In this approximation, we include the quantum numbers JM_Jj for interacting particles; the states with other quantum numbers, i.e., $LSls$ are characterized by higher energies. Indeed, the excitation energy of the ion state $^1D_2$ is 11654 cm^{-1} and the excitation energy of the ion state $^1S_0$ is 27878 cm^{-1}; these ion states pertain to the same electron shell $3p^4$. A nonexcited electron shell of halogen atoms is characterized by one electron term $L = 1$, $s = 1/2$, which simplifies the analysis.

The data in Table 3 are obtained with the interaction potentials Δ, V_{rot}, and U_m neglected. These potentials give additional quantum numbers for the quasi-molecule, and therefore the accuracy of the data in Table 3 is determined by these values: $\Delta(R) \sim V_{rot} \sim \sim 10$ cm^{-1} and $U_m \sim 1$ cm^{-1}. In this approximation, the statistical weight of the quasi-molecule states is

$$g = 2 \cdot 2(2j+1), \qquad (18)$$

where the first factor accounts for the degeneration with respect to the sign of M_J, the second factor corresponds to the separation of quasi-molecule states into odd and even ones, and this degeneration is therefore removed by the exchange interaction $\Delta(R)$, and the third factor in (18) accounts for the degeneration with respect to m_j, which is removed by all the neglected interactions Δ, V_{rot}, and U_m.

Thus, the above analysis of the interaction of the halogen ion and atom at large separations shows that the character of electron momentum coupling differs from that of the Hund coupling scheme. Along with the quantum numbers of electron shells of an isolated atom and an ion in the framework of the LS coupling scheme, the quantum numbers of the quasi-molecule are JjM_J (the total electron momenta of the atom and the ion and the projection of the total atom momentum on the molecular axis). Other quantum numbers are mixed due to the rotation energy V_{rot}, exchange Δ, and quadrupole–quadrupole interaction potentials U_m between the atom and the ion.

4. ION–ATOM EXCHANGE INTERACTION FOR HALOGENS

We now determine the exchange ion–atom interaction potential that allows us to evaluate the resonant charge exchange cross section. For this, we represent the wave function of the atom having n valence electrons with the momentum l_e in the framework of the LS coupling scheme as [15, 16, 20]

$$\Phi_{LSM_LM_S}(1,2,\ldots,n) = \\ = \frac{1}{\sqrt{n}}\hat{P}\sum_{lmsm_s\mu\sigma} G^{LS}_{ls}(l_e,n)\begin{bmatrix} l_e & l & L \\ \mu & m & M_L \end{bmatrix}\times \\ \times\begin{bmatrix} \frac{1}{2} & s & S \\ \sigma & m_s & M_S \end{bmatrix}\varphi_{l_e\frac{1}{2}\mu\sigma}(1)\psi_{lsmm_s}(2,\ldots,n), \qquad (19)$$

where Φ, ψ, and φ are the respective wave functions of the atom, the ion, and the valence electron with the quantum numbers indicated; μ and σ are the projections of the angular momentum and spin of the valence electron; the argument of the wave function indicates the electrons contained by this atomic particle; the operator $\widehat{P}$ permutes the electrons; and the parentage coefficient $G^{LS}_{ls}(l_e,n)$ is responsible for addition of the valence electron to the ion for construction of an atom for given quantum numbers of these atomic particles.

The exchange interaction potential is given by [8, 12]

$$\Delta(R) = 2\left\langle \Psi_1 \left| \widehat{H} \right| \Psi_2 \right\rangle - \\ - 2\left\langle \Psi_1 \left| \widehat{H} \right| \Psi_1 \right\rangle \langle \Psi_1 | 1 | \Psi_2 \rangle, \qquad (20)$$

where Ψ_1 is the wave function of the quasi-molecule with the valence electron located near the first core (the atom is located near the first nucleus), Ψ_2 corresponds to the atom location near the second nucleus, and $\widehat{H}$ is the Hamiltonian of electrons. We note that an accurate evaluation of this interaction requires the accurate wave functions of the quasi-molecule that account for the interaction of the valence electron with both cores simultaneously; this is achieved in the framework of the asymptotic theory. Using the general method to evaluate the exchange interaction potential $\Delta(R)$ similarly to the case «a» of the Hund coupling in [12, 14, 19, 21], we obtain

$$\begin{gathered}
\Delta(R) = n\left(G_{ls}^{LS}\right)^2 \times \\
\times \sum_{\substack{\mu m m' M M' \\ \sigma\sigma' m_s m_s' m_s'' \\ M_S M_S' m''' m_s'''}} \begin{bmatrix} l_e & l & L \\ \mu & m' & M \end{bmatrix} \begin{bmatrix} \frac{1}{2} & s & S \\ \sigma & m_s^{'} & M_S \end{bmatrix} \times \\
\times \begin{bmatrix} L & S & J \\ M & M_S & M_J \end{bmatrix} \begin{bmatrix} l & s & j \\ m & m_s & m_j \end{bmatrix} \times \\
\times \begin{bmatrix} l_e & l & L \\ \mu & m'' & M' \end{bmatrix} \begin{bmatrix} \frac{1}{2} & s & S \\ \sigma' & m_s'' & M_S' \end{bmatrix} \times \\
\times \begin{bmatrix} L & S & J \\ M' & M_S' & M_J \end{bmatrix} \times \\
\times \begin{bmatrix} l & s & j \\ m''' & m_s''' & m_j \end{bmatrix} \Delta_{l_e\mu}. \qquad (21)
\end{gathered}$$

We here account for the character of coupling of the electron momenta in the quasi-molecule, such that the atomic core quantum numbers $lsm'm_s'$ and the valence electron atomic numbers $l_e\mu\frac{1}{2}\sigma$ are first summed in the atomic quantum numbers LSM_LM_S, and the atom quantum numbers are then summed over the quantum numbers $LSJM_J$, and the ion quantum numbers $lsmm_s$ are summed over the ion quantum numbers lsj. We sum or average over the other quasi-molecule quantum numbers and use the relations

$$\sum_{m m_s m''' m_s'''} \begin{bmatrix} l & s & j \\ m & m_s & m_j \end{bmatrix} \begin{bmatrix} l & s & j \\ m''' & m_s''' & m_j \end{bmatrix} = 1,$$

$$\sum_{\sigma\sigma' m_s' m_s''} \begin{bmatrix} \frac{1}{2} & s & S \\ \sigma & m_s^{'} & M_S \end{bmatrix} \begin{bmatrix} \frac{1}{2} & s & S \\ \sigma' & m_s'' & M_S' \end{bmatrix} = \\ = \delta_{M_S M_S'}$$

for the Clebsch–Gordan coefficients. In Eq. (21), $\Delta_{l_e\mu}$ is the one-electron exchange interaction potential that corresponds to the case where a valence electron with these quantum numbers is located in the field of two structureless cores and has the same asymptotic wave function as in real atoms. As a result, we obtain by analogy with [6, 12, 14, 19, 21] that

$$\begin{gathered}
\Delta(l_e\mu, ls, LS, JM_Jj, R) = n\left(G_{ls}^{LS}\right)^2 \times \\
\times \sum_{\mu m M M_S} \begin{bmatrix} l_e & l & L \\ \mu & m & M \end{bmatrix}^2 \times \\
\times \begin{bmatrix} L & S & J \\ M & M_S & M_J \end{bmatrix}^2 \Delta_{l_e\mu}(R), \qquad (22)
\end{gathered}$$

where the argument contains the quantum numbers of the quasi-molecule and the distance R between the interacting ion and atom. This formula reduces the problem of the exchange interaction between an atom and an ion with unfilled electron shells to the transition of one electron between structureless cores. It is important that the exchange interaction potential is independent of the ion moment j.

The one-electron exchange interaction potential $\Delta_{l_e\mu}$ is given by [6, 8, 12, 22]

$$\begin{gathered}
\Delta_{l_e\mu}(R) = A^2 R^{2/\gamma - 1 - |\mu|} \exp\left(-R\gamma - \frac{1}{\gamma}\right) \times \\
\times \frac{(2l+1)(l_e+|\mu|)!}{(l_e-|\mu|)!\,|\mu|!(\gamma)^{|\mu|}}. \qquad (23)
\end{gathered}$$

It decreases with the increase of μ as $R^{-|\mu|}$. Here, l_e and μ are quantum numbers of the valence electron and γ and A are the parameters of the asymptotic wave function of this electron. This formula contains the first term of the asymptotic expansion over the small parameter $1/\gamma R$ for the ion–atom exchange interaction potential at large distances between the nuclei.

From (22), we thus obtain the exchange interaction potential involving the halogen atom $\mathrm{X}(^2P)$ and its ion $\mathrm{X}^+(^3P)$,

$$\begin{gathered}
\Delta(l_e\mu, ls, LS, JM_Jj, R) = 3\Delta_{10}(R) \times \\
\times \sum_M \begin{bmatrix} 1 & 1 & 1 \\ 0 & M & M \end{bmatrix}^2 \begin{bmatrix} 1 & 1/2 & J \\ M & M_J - M & M_J \end{bmatrix}^2 = \\
= \frac{3\Delta_{10}(R)}{2} \begin{bmatrix} 1 & 1/2 & J \\ 1 & M_J - 1 & M_J \end{bmatrix}^2, \qquad (24)
\end{gathered}$$

where we extract the dominant term in the sum in (22) that is proportional to $\Delta_{10}(R)$ (see Eq. (23)). In Table 4, we give the values of the exchange interaction potential for the ground electron states of the halogen

Table 4. The exchange interaction potential $\Delta(R)$ for the halogen atom and ion in the ground electron states, Cl($^2P_{JM_J}$) + Cl$^+$(^3P_j), and different states of the fine structure JM_Jj for these particles. The total energy E of these states (Table 3) refers to the distance $R = 14a_0$ between the nuclei and is obtained in neglecting the quadrupole–quadrupole ion–atom interaction and the rotation energy of the quasi-molecule

JM_Jj	$\Delta(R)$	Δ, cm^{-1}	E, cm^{-1}	σ_{ex}, Å^2 $\varepsilon = 0.1$ eV	σ_{ex}, Å^2 $\varepsilon = 1$ eV	σ_{ex}, Å^2 $\varepsilon = 10$ eV
$\frac{3}{2}\frac{3}{2}2$	$\frac{3}{2}\Delta_0$	87	0	110	92	76
$\frac{3}{2}\frac{1}{2}2$	$\frac{1}{2}\Delta_0$	29	260	93	77	62
$\frac{3}{2}\frac{3}{2}1$	$\frac{3}{2}\Delta_0$	87	696	110	92	76
$\frac{3}{2}\frac{1}{2}1$	$\frac{1}{2}\Delta_0$	29	956	93	77	62
$\frac{3}{2}\frac{3}{2}0$	$\frac{3}{2}\Delta_0$	87	996	110	92	76
$\frac{1}{2}\frac{1}{2}2$	Δ_0	58	1012	104	86	71
$\frac{3}{2}\frac{1}{2}0$	$\frac{1}{2}\Delta_0$	29	1256	93	77	62
$\frac{1}{2}\frac{1}{2}1$	Δ_0	58	1708	104	86	71
$\frac{1}{2}\frac{1}{2}0$	Δ_0	58	2009	104	86	71

atom X(^2P_J) and ion X$^+$(^3P_j) with different fine-structure quantum numbers for these particles.

To demonstrate these results, we return to the above example of the interaction Cl(^2P) + Cl$^+$(^3P) at the distance $R = 14a_0$ between the nuclei. The energy splittings between even and odd quasi-molecule states are $\Delta_{10} = 14$ cm^{-1} and $\Delta_{11} = 2.0$ cm^{-1} if we consider the cores structureless. Table 4 contains the values of the exchange interaction potential under these conditions for given quantum numbers of the interacting particles. We ignore the quadrupole–quadrupole ion–atom interaction and the rotation energy; the energy of the even or odd state with given quantum numbers is $E \pm \Delta(R)/2$. The data in Table 4 confirm the above hierarchy of interactions between halogen atoms and their ions.

5. RESONANT CHARGE EXCHANGE FOR HALOGENS

The above results allow us to determine the resonant charge exchange cross section in slow collisions of halogen atoms and their ions in the ground electron states. To determine the partial cross section of the resonant charge exchange, we use the asymptotic formula [10, 11] (see Eq. (8))

$$\sigma_{res} = \frac{\pi R_0^2}{2}, \quad \text{where} \quad \frac{1}{v}\sqrt{\frac{\pi R_0}{2\gamma}}\Delta(R_0) = 0.28, \qquad (25)$$

where v is the collision speed and the asymptotic coefficient γ is expressed through the atom ionization potential I as $\gamma = \sqrt{2I}$ in atomic units (also see Eq. (23)). Equation (25) is valid for s-electron transitions or in the case where electron transfer transitions for states with given quantum numbers can be separated from other transitions. In particular, the partial cross sections of resonant charge exchange are given in Table 4 for chlorine.

We introduce the resonant charge exchange cross section averaged over fine states assuming the initial population of atom and ion fine states to be proportional to their statistical weights,

Table 5. The average cross sections (in 10^{-15} cm^2) for the halogen atom and ion in the ground electron states, X(^2P) + X$^+$(^3P) at the indicated collision energies ε in the laboratory reference frame for hierarchy (6) of interactions and in the case «a» of the Hund coupling [13, 19] (in parentheses)

	$\varepsilon = 0.1$ eV	$\varepsilon = 1$ eV	$\varepsilon = 10$ eV
F ($\gamma = 1.132$, $A = 1.6$)	6.2 (6.0)	5.1 (4.9)	4.1 (4.0)
Cl ($\gamma = 0.976$, $A = 1.8$)	10 (10)	8.7 (8.4)	7.1 (6.9)
Br ($\gamma = 0.932$, $A = 1.8$)	13 (12)	11 (10)	8.9 (8.2)
I ($\gamma = 0.876$, $A = 1.9$)	16 (16)	14 (13)	12 (11)

$$\overline{\sigma_{ex}} = \frac{1}{3}\sigma_{ex}\left(\frac{3}{2},\frac{3}{2}\right) + \\ + \frac{1}{3}\sigma_{ex}\left(\frac{3}{2},\frac{1}{2}\right) + \frac{1}{3}\sigma_{ex}\left(\frac{1}{2},\frac{1}{2}\right), \quad (26)$$

where the atom quantum numbers J and M_J are given in the partial cross section argument. If we expand the resonant charge exchange cross section with respect to the small parameter $1/R\gamma$, keep only two terms in the expansion, and take Eq. (24) into account, we can write Eq. (25) for average cross section (26) as

$$\sigma_{res} = \frac{\pi R_0^2}{2}, \quad (27a)$$

where

$$\frac{2.7}{v}\sqrt{\frac{\pi R_0}{2\gamma}}\,\Delta_{00}(R_0) = 0.28 \quad (27b)$$

and $\Delta_{00}(R)$ is the ion–atom exchange interaction potential for the transferred s-electron with the given asymptotic parameters γ and A of its wave function. This value is related to the one-electron exchange interaction potential $\Delta_{10}(R)$ for a transferred p-electron by $\Delta_{10}(R) = 3\Delta_{00}(R)$ in accordance with (23). Table 5 contains the average cross sections of resonant charge exchange for halogen atoms and their ions in the ground electron state, X(^2P) + X$^+$(^3P), for hierarchy (6) of interactions for the quasi-molecule constituted by the colliding atom and ion. These cross sections practically coincide with the average cross sections for the ground fine states of the colliding particles, i.e., for the process X($^2P_{3/2}$) + X$^+$($^3P_2$). Thus, averaging over fine states of the ground electron states and over momentum projections of the ground fine state of colliding particles leads to results that are close to each other. In addition, these data are compared with the cross sections in the case «a» of the Hund coupling taken from [13, 14]. As follows from the comparison, the real hierarchy of interactions in a quasi-molecule increases the resonant charge exchange cross section by several percent compared with the case «a» of the Hund coupling.

One more peculiarity of the resonant charge exchange for momentum coupling follows from hierarchy (6) of interactions. The exchange interaction potential that determines the cross section of this process is given by (24), where we restrict to only a transition of a p-electron with zero momentum projection on the molecular axis. As follows from this formula, such states are present in any fine-structure state, and we can therefore neglect the transition of the electron whose momentum projection on the molecular axis is unity. We note that in contrast to the case «a» of the Hund coupling, where rotation of the molecular axis leads to transitions between states with different momentum projections on the molecular axis, such transitions are absent in the case of halogen atoms and ions because of separation of fine-structure states by energy. Next, we evaluate the resonant charge exchange cross section with the momentum coupling of the transferred electron with momenta of atomic cores taken into account. We can estimate the error in the cross sections if we compare the cross sections with and without the momentum coupling taken into account. We take a p-electron that has the same asymptotic parameters as valence electrons of halogen atoms, but is located in the field of structureless cores. For the resonant charge exchange cross section in chlorine at the respective energies 0.1, 1, and 10 eV, we then obtain the values 87, 71, and 57 Å^2 instead of those in Table 5. We see that ignoring the coupling between the momenta of the transferred electron and atomic cores leads to a significant error.

Thus, it follows from the above analysis that in the course of collision and electron transfer, a quasi-molecule consisting of the colliding halogen ion and atom is characterized by the quantum numbers JM_Jj, and transitions between these states are absent during the

Table 6. The interaction potentials for the system $O^+(^4S_{3/2}) - O(^3P_{JM_J})$ at $R = 12a_0$ and the partial cross sections σ_{ex} of resonant charge exchange at the indicated collision energy ε

JM_J	$a(JM_J)$	$\Delta(JM_J)$	E, cm^{-1}	Δ, cm^{-1}	σ_{ex}, Å^2 $\varepsilon = 0.1$ eV	σ_{ex}, Å^2 $\varepsilon = 1$ eV	σ_{ex}, Å^2 $\varepsilon = 10$ eV
22	−1	$\frac{4}{3}\Delta_1$	0	6.8	63	51	40
21	1/2	$\frac{2}{3}\Delta_0$	153	20	77	62	49
20	7/6	$\frac{8}{9}\Delta_0$	220	27	81	66	52
11	1/2	$\frac{2}{3}\Delta_0$	311	20	77	62	49
10	−1/2	$\frac{2}{3}\Delta_1$	209	4.4	56	44	34
00	1/3	$\frac{4}{9}\Delta_0$	363	13	72	57	45

electron transfer. The partial cross sections of resonant charge exchange depend on quantum numbers, whereas the average cross sections for the correct scheme of momentum coupling and in the case «a» of the Hund coupling are close to each other.

6. RESONANT CHARGE EXCHANGE FOR OXYGEN

We now consider one more example of resonant charge exchange with a p-electron transition,

$$O^+(^4S) + O(^3P) \to O(^3P) + O^+(^4S), \qquad (28)$$

involving the oxygen atom and ion in the ground electron states. Constructing the hierarchy of interactions (1) in this case, we take the previous case in Eq. (6) as a basis, with the quantum numbers JM_J of the quasi-molecule consisting of the colliding particles. In accordance with (12), the interaction potential of the ion charge with the atom quadrupole moment is then given by

$$\Delta U(JM_J) = \frac{2\overline{r_a^2}}{5R^3} \times \\ \times \left(2 \begin{bmatrix} 1 & 1 & J \\ 0 & M_J & M_J \end{bmatrix}^2 - \begin{bmatrix} 1 & 1 & J \\ 1 & M_J{-}1 & M_J \end{bmatrix}^2 \right) = \\ = \frac{2\overline{r_a^2}}{5R^3} a(JM_J), \qquad (29)$$

where we use the same notation as in (12); the values $a(JM_J)$ are contained in Table 6. It follows from (22) that instead of Eq. (24) for the ion–atom exchange interaction potential for halogens, the exchange interaction potential for oxygen is given by

$$\Delta(JM_J, R) = \\ = \frac{4}{3} \sum_{\mu} \begin{bmatrix} 1 & 1 & J \\ \mu & M_J - \mu & M_J \end{bmatrix}^2 \Delta_{1\mu}(R), \qquad (30)$$

with the coefficients in this formula given in Table 6.

We note that the excitation energies of oxygen atom fine states from the ground fine state $^3P_2$ are 158 cm^{-1} for the state $^3P_1$ and 220 cm^{-1} for the state $^3P_0$. These values are comparable to the long-range ion–quadrupole interaction potential (29) at distances that give the main contribution to the resonant charge exchange cross section. Hence, we have an intermediate case of momentum coupling for oxygen, and in the above halogen example with $\delta_a \gg \Delta U$, the quantum numbers of the quasi-molecule are JM_Jj, while in the other limiting case where $\delta_a \ll \Delta U$, the quantum numbers of the quasi-molecule are JMj (where M and M_J are the projections of the orbital and total atom momentum on the quasi-molecule axis). We consider the first limiting case below; Table 6 contains the values of the quasi-molecule energies E calculated in accordance with Eq. (17), where we account for the fine-structure splitting of levels and charge-quadrupole ion–atom interaction in the limit $\delta_a \gg \Delta U$. The energies are taken at the ion–atom distance $R = 12a_0$ that corresponding to the impact parameters typical of the resonant charge exchange cross section at the collision energy

Table 7. The ion–atom exchange interaction potential for the quasi-molecule $O^+({}^4S_{3/2}) - O({}^3P_J)$ with its quantum numbers given by J and M (the total atom momentum and the projection of the atom orbital momentum on the molecular axis) and the partial resonant charge exchange cross sections σ_{ex} for the indicated quantum numbers and collision energies ε in the laboratory reference frame

JM	$\Delta(JM_J)$	σ_{ex}, Å^2 $\varepsilon = 0.1$ eV	σ_{ex}, Å^2 $\varepsilon = 1$ eV	σ_{ex}, Å^2 $\varepsilon = 10$ eV
20	$\frac{20}{9}\Delta_{11}$	69	56	40
21	$\frac{10}{9}\Delta_{10}$	84	68	55
10	$\frac{4}{3}\Delta_{11}$	63	51	45
11	$\frac{2}{3}\Delta_{10}$	77	62	49
00	$\frac{4}{9}\Delta_{11}$	51	41	31
01	$\frac{2}{9}\Delta_{10}$	63	50	38

about 1 eV. We note that rotation energy (5) is 29 cm^{-1} under these conditions and exceeds or is comparable to the exchange interaction potential.

The average resonant charge exchange cross section is

$$\overline{\sigma_{ex}} = \frac{2}{9}\sigma_{ex}(22) + \frac{2}{9}\sigma_{ex}(21) + \frac{1}{9}\sigma_{ex}(20) + \\ + \frac{2}{9}\sigma_{ex}(11) + \frac{1}{9}\sigma_{ex}(10) + \frac{1}{9}\sigma_{ex}(00), \quad (31)$$

where the quantum numbers of the fine-structure atom state are given in parentheses, and we assume the population of these states to be proportional to their statistical weights. If we neglect the electron transitions due to rotation of the molecular axis, we obtain by analogy with Eqs. (25) and (27) that the average cross section is given by

$$\sigma_{res} = \frac{\pi R_0^2}{2}, \quad (32a)$$

where

$$\frac{2.9}{v(R_0\gamma)^{1/3}}\sqrt{\frac{\pi R_0}{2\gamma}}\Delta_{00}(R_0) = 0.28 \quad (32b)$$

and we use the same notation as in (25) and (27). Averaging the cross sections in Table 6 in accordance with (31) and (32) gives the values 71, 57, and 45 Å^2 for the average cross section of resonant charge exchange at the respective collision energies 0.1, 1, and 10 eV. On the other hand, the respective cross sections in the case «a» of the Hund coupling are given by 73, 60, and 48 Å^2 [13, 14]. Similarly, for the partial cross sections for the ground fine-structure state, i.e., for the process $O^+({}^4S_{3/2}) + O({}^3P_2)$, we obtain the values 72, 55, and 46 Å^2 at the respective collision energies 0.1, 1, and 10 eV; the difference of these average cross sections from the above average cross sections does not exceed their accuracy. Next, if we ignore the coupling of the transferred electron with cores, i.e., if we consider the transition of a p-electron between structureless cores at the same asymptotic parameters of the electron wave function as in the above cases, we obtain the values 64, 51, and 40 Å^2 for the average cross sections of resonant charge exchange at the indicated collision energies.

The other limiting case of the interaction hierarchy, $\delta_a \ll U_0$, between the fine splitting of atom levels and the ion–atom quadrupole interaction potential leads to the molecular quantum numbers JMj, where M is the projection of the atom angular momentum on the quasi-molecule axis. In this limiting case, Eq. (22) for the ion–atom exchange interaction potential for process (28) becomes

$$\Delta(R) = \frac{4}{3}\sum_{\mu M_S}\begin{bmatrix} 1 & 1 & 1 \\ \mu & M-\mu & M \end{bmatrix}^2 \times \\ \times \begin{bmatrix} 1 & 1 & J \\ M & M_S & M_S+M \end{bmatrix}^2 \Delta_{1\mu}(R), \quad (33)$$

Averaging the cross sections in Table 7 over fine states of the ground electron state gives the values 70, 56, and 44 Å^2 for the average cross section of resonant charge exchange at the respective collision energies 0.1, 1, and 10 eV. For the ground fine state $J = 2$, these values are 79, 64, and 50 Å^2, respectively. In this case, the average cross section differs from that for the lowest fine state. Next, with the logarithmic dependence of the cross section on the collision velocity taken into account, we obtain that instead of (32), the cross section averaged over fine states is given by

$$\sigma_{res} = \frac{\pi R_0^2}{2}, \tag{34a}$$

where

$$\frac{2.6}{v(R_0\gamma)^{1/3}}\sqrt{\frac{\pi R_0}{2\gamma}}\Delta_{00}(R_0) = 0.28, \tag{34b}$$

which differs from Eq. (32) only slightly. We find that the average cross sections are close to those for the case «a» of the Hund coupling. But the partial cross sections can be different in these cases.

7. CONCLUSION

Studying the character of momentum coupling for the resonant charge exchange process in slow collisions, we have found that the number of real cases of momentum coupling is considerably larger than that following from the classical Hund scheme of momentum coupling. Constructing the hierarchy of interactions for the quasi-molecule consisting of the colliding ion and atom has allowed us to find a suitable scheme of momentum coupling. The strongest interaction for not heavy atoms is the exchange interaction of electrons inside these atomic particles, and therefore the quasi-molecule quantum numbers for the ion and atom electron shells are $LSls$ (the orbital momentum and spin of the atom and the same quantum numbers for the ion). For halogen and oxygen, the rotation energy V_{rot} of colliding particles is small compared to the fine splitting of the atom (δ_a) and ion (δ_i) levels; it is also small compared to the long-range charge–quadrupole interaction U between the ion and the atom. Hence, the resonant charge exchange proceeds at certain quantum numbers JM_Jj or JMj depending on the ratio δ_a/U (where J and j are the total atom and total ion momenta and M and M_J are the projections of the atom orbital and total atom momenta on the quasi-molecule axis). This character of momentum coupling does not correspond to cases in the Hund coupling scheme.

In contrast to the case «a» of the Hund coupling, where the electron transfer process is entangled with the rotation of the atom and ion orbital momenta, the resonant charge exchange process for halogen and oxygen proceeds separately for each fine-structure state. This increases the accuracy of evaluation of the electron transfer cross section. The resonant charge exchange cross section depends on the initial quantum numbers of the quasi-molecule; according to the analysis for halogens and oxygen, the cross sections in the case «a» of the Hund coupling scheme and for the real momentum coupling are close to each other.

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