CHARGE EXCHANGE INVOLVING AN EXCITED STATE

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Submitted March 27, 1967

Zh. Eksp. Teor. Fiz. 54, 120–126 (January, 1968)

We consider the effect of transition through an intermediate level on the charge exchange cross section when the atom is produced in the excited state. It is shown that such a process may play an important role in the region of comparatively low velocities of relative motion. Calculations of the cross sections for the reactions $p + He \to H(2p) + He^+$ and $p + Ne \to H(2p) + Ne^+$ are in satisfactory agreement with the experiments. The possibility of using the results for a description of other atomic collisions, including transitions via an intermediate level, are considered.

THE authors together with Sobel'man[1] considered the problem of charge exchange (reaction of the type $A^+ + B \to A + B^+$, where A, B and A^+ and B^+ are atoms and singly-charged positive ions) in the two-level approximation. This approximation is justified for the calculation of charge exchange in the ground state of the atom, since the first excited level lies as a rule far from the ground level and interacts with the latter weakly. However, in the calculation of the cross section for charge exchange in the excited state it is necessary, in general, to take into account, besides the direct charge exchange (two-level approximation), also the competing transition via the intermediate level (we use the terminology of [2]). For charge exchange in the first excited state, this intermediate level is the ground state.

Recently a number of experimental papers[3-5] reported charge exchange in the 2p and 2s states of hydrogen in collisions between protons and inert-gas atoms. The experimental cross sections have a number of qualitative singularities which cannot be explained by any of the existing theories based on the Born or two-level approximation. In particular, in the region of not too large energies ($E \lesssim 20$ keV) the cross section can have two maxima and, in addition, at these energies the charge exchange in the 2p state is much more effective than in the 2s state.

In this paper we propose a method of calculating charge exchange in an excited state with allowance for a transition via an intermediate level, making it possible to explain qualitatively the indicated singularities of the experimental cross sections. The collision is regarded in the quasiclassical (parametric) approximation. We use a method previously developed for the excitation of atoms[6] and charge exchange,[1] as well as its generalization to include the case of several levels.[7] An atomic system of units is used.

1. To take into account the transition via an intermediate level (besides the direct transition) it is necessary to solve a system of three equations:

$$i\dot{a}_m = \sum_{n \neq m} U_{mn} \exp\left\{ i \int^t \alpha_{mn}(\tau)\, d\tau \right\} a_n, \qquad m, n = 0, 1, 2; \tag{1}$$

$$|a_n(-\infty)| = \delta_{0n}, \qquad \sum_n |a_n(t)|^2 = 1. \tag{2}$$

The indices 0, 1, and 2 denote respectively the initial, intermediate, and final states; $U_{mn}(t) = U^*_{nm}(t)$ are the off-diagonal matrix elements of the interaction, and their concrete form depends, in particular, on the basis functions, atomic or molecular, in terms of which the total wave function of the system is expanded.[1)] The phases $\alpha_{mn}(t)$ determine the difference between the energy levels of the system in the zeroth approximation.

Going over from the probability amplitudes $a_n(t)$ to the functions $R_n(t)$[6,7,9]:

$$R_n(t) \equiv \mu_n(t) \exp\left[-i\Omega_{n0}(t) - i\frac{\pi}{2} \right] = \frac{a_n(t)}{a_0(t)}, \qquad n = 1, 2, \quad R_0(t) \equiv 1, \tag{3}$$

where μ_n and Ω_{n0} are real functions, we obtain in place of (1) a system of two nonlinear equations

$$i\dot{R}_n = U_{n0} \exp\left\{ i \int^t \alpha_{n0}(\tau)\, d\tau \right\} - U_{0n} \exp\left\{ -i \int^t \alpha_{n0}(\tau)\, d\tau \right\} R_n^2 \tag{4}$$

$$+ U_{nm} \exp\left\{ i \int^t \alpha_{nm}(\tau)\, d\tau \right\} R_m - U_{0m} \exp\left\{ i \int^t \alpha_{0m}(\tau)\, d\tau \right\} R_n R_m,$$

$$n, m = 1, 2, \quad n \neq m,$$

and the normalization condition (2) is always satisfied:

$$|a_n(t)|^2 = \mu_n^2 / (1 + \mu_1^2 + \mu_2^2). \tag{5}$$

Separating in (4) the real and imaginary parts, we easily obtain a system of equations for $\mu_n(t)$:

$$\dot{\mu}_n = U_{n0} \cos \Phi_{n0} (1 + \mu_n^2) + \mu_m U_{nm} \cos \Phi_{nm} + \mu_n \mu_m U_{0m} \cos \Phi_{0m}, \tag{6}$$
$$m \neq n.$$

The phases Φ_{mn} are defined as follows:

$$\Phi_{mn}(t) = \int^t \alpha_{mn}(\tau)\, d\tau + \Omega_{mn}(t), \qquad \Omega_{mn} = \Omega_{m0} - \Omega_{n0}, \tag{7}$$

and in quasistationary conditions they determine the correction due to the interaction U_{mn}, to the distance between the terms m and n.[6,7] The corresponding equations for the phases Ω_{n0} are given in [7].

In our problem $\mu_2 \ll \mu_1 \ll 1$ (the probability of charge exchange in the ground state is much larger than in the excited state) so that we can neglect in the right side of (6) all the terms containing μ_2. In this case, μ_1 takes the form[7]

$$\mu_1(t) = \operatorname{tg} \int_{-\infty}^t U_{10}(t) \cos \Phi_{10}(t)\, dt \approx \int_{-\infty}^t U_{10}(t) \cos \Phi_{10}(t)\, dt. \tag{8}$$

[1)] The Hermitian formulation of the charge-exchange problem in the two-level approximation [1] can be readily generalized to a larger number of levels [8]. Without loss of generality, we can assume that $U_{mn} = U_{nm}$ and $U_{mn}(t)$ are even functions, that is, $U_{mn}(-t) = U_{mn}(t)$.

Substituting (8) in the equation for μ_2, we get

$$\mu_2(\infty) = \int_{-\infty}^{\infty} U_{20}(t)\cos\Phi_{20}(t)\,dt + \int_{-\infty}^{\infty} U_{21}(t)\cos\Phi_{21}(t)\,dt \int_{-\infty}^{t} U_{10}(t')\cos\Phi_{10}(t')\,dt'. \quad (9)$$

Inasmuch as under our conditions $\mu_2 \ll 1$, we can omit the normalization denominator for (5). The expression for the transition probability in state 2 takes the form

$$W_{02} = |a_2(\infty)|^2 = [\mu_2(\infty)]^2 \quad (10)$$
$$= \left[\int_{-\infty}^{\infty} U_{20}(t)\cos\Phi_{20}(t)\,dt + \int_{-\infty}^{\infty} U_{21}(t)\cos\Phi_{21}(t)\,dt \int_{-\infty}^{t} U_{10}\cos\Phi_{10}(t')\,dt'\right]^2.$$

The first term describes the direct transition $0 \to 2$, and the second term the transition via the intermediate level $0 \to 1 \to 2$.

2. The transition probability (10) was obtained from the nonlinear system (6) in the second order of perturbation theory. A successive application of the perturbation theory employed here calls for substitution in the right side of (10) of the phases $\Phi_{mn}(t)$ in the same form that they would possess under the assumption of paired interaction of the terms[2] m and n:

$$\Phi_{mn}(t) = \int_{t_{mn}^{(0)}}^{t} \alpha_{mn}(\tau)\,d\tau + \Omega_{mn}(t) = \int_{t_{mn}^{(0)}}^{t} \sqrt{\alpha_{mn}^2(\tau) + 4U_{mn}^2(\tau)}\,d\tau. \quad (11)$$

The question of the choice of the lower integration limit $t_{mn}^{(0)}$ was discussed earlier,[7,9] where it was shown that solution of the system (1) consisting of only two equations yields $t^{(0)} = 0$. As applied to the case considered here, this means that for direct transitions $t_{10}^{(0)} = t_{20}^{(0)} = 0$, but $t_{21}^{(0)} \neq 0$ and depends on the characteristic parameters of the problem. Its calculation in general form is hardly possible. We shall therefore estimate the role of $t_{21}^{(0)}$ qualitatively. When the right side of (10) is squared, a crossing (interference) term arises. The presence of $t_{21}^{(0)} \neq 0$ produces in this term a factor that oscillates rapidly with changing impact distance and with changing collision velocity. Its contribution to the total cross section is negligibly small, and in our approximation the interference term can be dropped. On the other hand, the concrete value of $t_{21}^{(0)}$ has little influence on the square of the second term in the right side of (10). Therefore in calculating this integral we put $t_{21}^{(0)} = 0$. Since the integrands are even, we get

$$\int_{-\infty}^{\infty} U_{21}(t)\cos\Phi_{21}(t)\,dt \int_{-\infty}^{t} U_{10}(t')\cos\Phi_{10}(t')\,dt' = \frac{1}{2}\int_{-\infty}^{\infty} U_{21}(t)\cos\Phi_{21}(t)\,dt \int_{-\infty}^{\infty} U_{10}(t)\cos\Phi_{10}(t)\,dt, \quad (12)$$

after which the expression for the transition probability takes the form

$$W_{02} = W_{0\to 2} + {}^1/_4 W_{0\to 1} W_{1\to 2}, \quad (13)$$

$$W_{m\to n} = \left(\int_{-\infty}^{\infty} U_{mn}(t)\cos\Phi_{mn}(t)\,dt\right)^2. \quad (14)$$

Thus, the total transition probability W_{02} is the sum of the probabilities of the direct transition $W_{0\to 2}$ and the $W_{0\to 1\to 2} = ({}^1/_4)W_{0\to 1}W_{1\to 2}$, which describes approximately the transition through the intermediate level. There is no interference between the indicated processes, and the probability of transition through the intermediate level is expressed in terms of the product of the probabilities of the transitions between the neighboring states. At larger collision velocities, the expressions $W_{m\to n}$ (14) go over into the first-order Born probabilities.[1,6] It is easy to verify that in the Born region the formula (13) coincides with the second Born approximation.

The effective cross section for charge exchange in the excited state is also a sum of two terms describing the indicated processes:

$$\sigma_{02} = \sigma_{0\to 2} + \sigma_{0\to 1\to 2}, \quad (15)$$

$$\sigma_{0\to 2} = 2\pi\int_0^{\infty} \rho\,d\rho\,W_{0\to 2}, \qquad \sigma_{0\to 1\to 2} = \frac{\pi}{2}\int_0^{\infty} \rho\,d\rho\,W_{0\to 1}W_{1\to 2}, \quad (16)$$

where ρ is the impact parameter.

The general formulas (13)–(16) can be used also to calculate the cross sections of other inelastic processes occurring in collisions of heavy particles. The simplest example is excitation from the ground state of atomic levels lying above the resonance level. In practice the problem reduces to a calculation of the matrix elements U_{mn} and of the separation between the terms Φ_{mn}.

3. We shall employ the obtained formulas to calculate the cross sections for the charge exchange of protons by atoms of inert gases with formation of hydrogen in the 2p state. We confine ourselves to the region of energies from 5 to 30 keV. At these energies it is advantageous to use the eigenfunctions of the unperturbed atoms as the basis functions for the expansion of the total wave function of the system.[3] The initial, intermediate, and final states are the ground state of the target atom B and the 1s and 2p states of the produced hydrogen atom. The ion B^+ is assumed here to be a structureless charged particle.

To calculate the cross section $\sigma_{0\to 2}$ of the "direct charge exchange" we can use the results of [1]. We shall therefore consider in detail only $\sigma_{0\to 1\to 2}$. The individual probabilities $W_{0\to 1}$ and $W_{0\to 2}$ are also known.[1,6,7,11] They are given by:

$$W_{0\to 1}(\rho, v) = \exp\left\{-2\left[\left(\frac{\pi\omega_{10}}{2\gamma v}\right)^2 + \left(\frac{\omega_{10}\rho}{v}\right)^2\right]^{1/2}\right\} \sin^2\frac{1}{v}\int_{-\infty}^{\infty} U_{01}(\rho, x)\,dx, \quad (17)$$

$$W_{1\to 2}(\rho, v) = \exp\left\{-2\left[\frac{\lambda\omega_{21}}{2v^2} + \left(\frac{\omega_{21}\rho}{v}\right)^2\right]^{1/2}\right\} \sin^2\frac{1}{v}\int_{-\infty}^{\infty} U_{12}(\rho, x)\,dx. \quad (18)$$

Here v—relative velocity, ω_{mn}—resonance defect between the levels m and n, and U_{01} and U_{12}—off-diagonal matrix elements connected with the charge exchange in the ground state and with the excitation from the ground state. At large distances $U_{10}(R) \exp(-\gamma R)$, where $\gamma = \sqrt{2I_{min}}$, and I_{min} is the smaller of the ionization energies of the states 0 and 1.[1,11] For an op-

[2] It is known that only two terms of a physical system can cross at one point of the complex plane; an exception is the harmonic oscillator [10].

[3] A lower energies on the order of thermal it is necessary to expand in terms of the eigenfunctions of the quasimolecule.

tically allowed transition $U_{12}(R)$ λ/R^2.[6,7] If the transition $1 \to 2$ is a transition of the $s \to s$ type, for example $1s \to 2s$ in the hydrogen atom, then the excitation probability $W_{1\to 2}$ takes the form (17) but $\gamma = \sqrt{2I_1} + \sqrt{2I_2}$.[7, 11]

An exact calculation of $\sigma_{0\to 1\to 2}$ using (17) and (18) calls for numerical integration. However, it is possible to obtain a simple analytic approximation for $\sigma_{0\to 1\to 2}$ by recognizing that the excitation probability $W_{1\to 2}(p)$ is a smoother function than the probability $W_{0\to 1}(\rho)$ of the charge exchange in the ground state. We take the function $W_{1\to 2}(\rho)$ at $\rho = \rho_0$ outside the integral sign in (16). The remaining integral gives the cross section for charge exchange in the ground state:

$$\sigma_{0\to 1\to 2} = \frac{\pi}{2}\int_0^\infty \rho d\rho W_{0\to 1}(\rho) W_{1\to 2}(\rho)$$

$$\approx \frac{W_{1\to 2}(\rho_0)}{4}\int_0^\infty 2\pi\rho d\rho W_{0\to 1}(\rho) = \frac{W_{1\to 2}(\rho_0)}{4}\sigma_{01}. \qquad (19)$$

Using the explicit form of $W_{0\to 1}(\rho)$[1] and retaining in the approximate calculation the correct dependence on the velocity at small and large velocities, we can get for $\rho_0 = \rho_0(v)$ the following value:

$$\rho_0(v) = \frac{\sqrt{1+v^2/4}}{2I_B - 1}, \qquad 2I_B - 1 > 0, \qquad (20)$$

where I_B is the ionization potential of the target atom. Within the framework of the rather rough approximation made here, it is meaningless to use the exact form of the pre-exponential factor in the probability $W_{1\to 2}(\rho_0)$ (18). Since at low velocities its order of magnitude is $\frac{1}{2}$ and it is proportional to $(\lambda/v\rho_0)^2$ at large velocities, we can obtain a qualitative description of this factor by replacing it with $[2 + (v\rho_0(v)/\lambda)^2]^{-1}$.

As a result we obtained for $\sigma_{0\to 1\to 2}$ the rather simple approximate expression

$$\sigma_{0\to 1\to 2} \approx \sigma_{01}\exp\left\{-2\left[\frac{\lambda\omega_{21}}{2v^2} + \left(\frac{\omega_{21}\rho_0}{v}\right)^2\right]^{1/2}\right\}\frac{1}{4}\left[2 + \left(\frac{v\rho_0}{\lambda}\right)^2\right]^{-1}. \qquad (21)$$

Here σ_{01} is the cross section for charge exchange in the ground state of the hydrogen atom. The constants in (19)–(21) for the charge exchange in the 2p-state of hydrogen are $\lambda = 0.7$ and $\omega_{21} = 0.375$.

The results of the calculation of the total charge exchange cross section in the 2p-state of hydrogen, for collision between protons and He and Ne atoms are shown in Figs. 1 and 2. We see that the developed method leads to satisfactory agreement with the experimental data.[3, 5] In the case of charge exchange of protons in He, the main contribution to the cross section in the 6–40 keV region is made by "charge exchange via the intermediate level" 1s of the H atom,

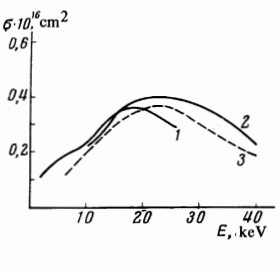

FIG. 1. Cross section of the process $p+He \to H(2p) + He^+$: 1 – experimental data [3], 2 – experimental data [5], 3 – result of present work.

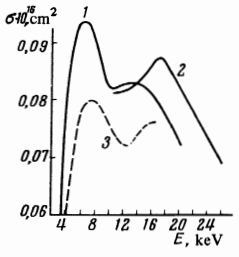

FIG. 2. Cross section of the process $p + Ne \to H(2p) + Ne^+$: 1 – experimental data [3], 2 – experimental data [5], 3 – result of present work.

and the "direct charge exchange" plays no role in the indicated region. In the case of the Ne atom, the maximum near 8 keV is due to "charge exchange via the intermediate state" 1s, and the next maximum at ~ 15 keV is due to "direct charge exchange." At energies lower than 10 keV the "direct charge exchange" plays practically no role, and at energies larger than 15 keV the contribution from the "charge exchange via the intermediate state" is in turn quite small.

It is impossible to write out a simple, approximate formula similar to (20) for the cross section $\sigma_{0\to 1\to 2}$ in the case of charge exchange in the 2s-state of hydrogen. We note only the main qualitative features of such a cross section. The cross section for "charge exchange via an intermediate level" is in this case also proportional to the cross section σ_{01} for charge exchange in the ground state, but the corresponding maximum of the cross section is reached at higher energies. At collision energies < 25 keV, we have $\sigma(2s) < \sigma(2p)$. The foregoing singularities result from the fact that the matrix element of the 1s–2s transition has an exponential, and not a dipole character as in the case of the 1s–2p transition.

Thus the competing transition via the intermediate level can lead, at medium energies, to the appearance of an additional maximum in the charge-exchange cross section in the excited state. At higher collision energies, on the order of 100 keV and more, the effects considered here, of the strong coupling between the initial, intermediate, and final states, cease to play any role. At high collision energies the charge exchange cross sections can be calculated by using the first Born approximation.

In conclusion we note that transition via the intermediate level can play an important role also in other problems of the theory of atomic collisions. The importance of this process for the excitation of levels lying above resonance by electrons in atoms of alkali elements has already been noted.[12]

We are sincerely grateful to I. I. Sobel'man for a useful discussion.

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Translated by J. G. Adashko
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