CALCULATION OF THE ANGULAR DISTRIBUTION OF RESONANCE CHARGE EXCHANGE

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The angular distribution of resonance charge exchange for He$^+$ ions in helium is calculated in the quasiclassical approximation, which permits one to take into account the difference in the trajectories of the relative motion of the nuclei for symmetric and antisymmetric states of the quasimolecule. The conditions of applicability of the formulas employed are analyzed. The results are compared with the experiments and with rough calculations performed by Everhart[6]. Possible ways of refinement of the theory are discussed.

1. INTRODUCTION, INITIAL APPROXIMATION

THE dependence of the probability of resonance charge exchange w on the scattering angle θ_0 and on the energy of the incoming ion E_0 have recently been measured in detail[1] (see, for example, [1], where there are references to earlier papers). It was observed that w is an oscillating function of E_0 at fixed values of θ_0 and an oscillating function of θ_0 at fixed E_0. The adiabatic theory of resonance charge exchange, developed within the framework of the parametric method (classical approximation for nuclei)[2-4], also predicts the presence of such oscillations. In spite of this theory, however, the experimentally observed maxima and minima of the probability do not reach 1 and 0, respectively.

In addition, attempts to calculate the dependence of w on E_0 and θ_0[5,6] meet with difficulties of fundamental nature, for in the case of motion in fields we have

$$U_\pm(R) = Z^2 / R + \epsilon_\pm(R)$$

(Z is the nuclear charge, $\epsilon_\pm(R)$ the principal symmetrical and antisymmetrical electronic terms of the quasimolecule, and R the distance between nuclei). The same values of θ correspond to different impact parameters ρ, which we shall denote by $\rho_\pm(E, \theta)$. This difficulty arises in all collision-theory problems connected with the calculation of the angular distribution in the classical approximation. However, in the case of resonance charge exchange, which is in fact only the result of interference between molecular states and is not connected

with the electronic transitions between the states, this difficulty can be overcome (see below). This can be done also for inelastic processes characterized by a small transition region, for example, for transitions occurring during crossing (or pseudo-crossing) of molecular terms.

Recently Smith undertook[7-9] the study of charge exchange on the basis of the quantum-mechanical formulas of Massey and Smith[10]:

$$q(\theta) = \tfrac{1}{4}|f^+(\theta) - f^-(\theta)|^2, \tag{1}$$

$$q_1(\theta) = \tfrac{1}{4}|f^+(\theta) + f^-(\theta)|^2. \tag{2}$$

In (1) and (2), q_1 and q are respectively the differential cross sections for elastic scattering and resonance charge exchange, while $f^\pm(\theta)$ are the amplitudes of potential scattering in the fields[2] $U_\pm(R)$:

$$f^\pm(\theta) = \frac{1}{2Mv}\sum_{l=0}^{\infty}[\exp(2i\eta_l^\pm) - 1](2l+1)P_l(\cos\theta), \tag{3}$$

where M is the reduced mass, v the relative velocity at infinity, and $\eta_l^\pm$ the l-th scattering phase shifts.

Formula (1) gives the correct values of the total charge-exchange cross section σ for[3] $v \ll v_0$ (v_0 is the velocity of the "active" electron on the Bohr orbit). However, even if this condition is satisfied, the values obtained for $q(\theta)$ and $q_1(\theta)$ are incorrect when $\rho(E, \theta)$ is small, for it is necessary to take into account in this case all the states of the quasimolecule, which have the same energy when

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[1]Both quantities are taken in the laboratory frame; the same quantities in the c.m.s. will be denoted by E and θ.

[2]Unless otherwise stipulated, we are using atomic units throughout.

[3]We confine ourselves in this article to the indicated velocity interval.

R = 0 (see, for example [11], Ch. 18). This effect, called rotational coupling, is of no significance, and the electronic wave function varies adiabatically if

$$v/\rho \ll a\rho^2, \tag{4}$$

where the left side is equal in order of magnitude to the angular velocity of the quasimolecule, while the right side is equal to the splitting of the terms that are degenerate when R = 0, one of which is $\epsilon_-(R)$.

The cited articles by Smith present calculations for the charge exchange

$$H^+ + H \to H + H^+ \tag{5}$$
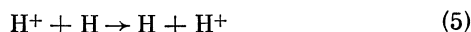
in a broad interval of energies and scattering angles. Smith also took into account the effect of rotation, using for this purpose the results of Bates and Williams [12], and obtained good agreement with experiment for $\theta_0 = 3°$ (there are no experimental data as yet for other angles).

In this article we consider the resonance process

$$He^+ + He \to He + He^+, \tag{6}$$

which is more interesting than (5) in many respects, and for which there are experimental data in a wide range of E and θ. Smith did not analyze with sufficient accuracy the conditions for further approximations, we shall discuss this in greater detail in the following section. This is all the more important because the results obtained can be developed in appreciable detail in different directions.

2. SEMICLASSICAL APPROXIMATION

In view of the large mass of the atoms, their motion in a sufficiently broad region is quasiclassical, that is, the phases satisfy the following formula (the Langer-Jeffries approximation):

$$\eta_l^\pm = Mv \left[\int_{R^\pm}^\infty \left(1 - \frac{\rho^2}{R^2} - \frac{U_\pm(R)}{E} \right)^{1/2} dR \right.$$

$$\left. - \int_\rho^\infty \left(1 - \frac{\rho^2}{R^2} \right)^{1/2} dR \right], \tag{7}$$

where

$$\rho = (l + 1/2)/Mv \tag{8}$$

is the classical impact parameter and $R^\pm$ are the largest roots of the corresponding integrands. Formula (7) is applicable at any rate if (see [13], Ch. 7)

$$\eta_l \gg 1. \tag{9}$$

In the quasiclassical region of E and θ, the series in (3) converge very poorly: it is necessary to include several hundred or even thousands of partial waves. We can therefore replace the summation in (3) by integration, use for $P_l(\cos \theta)$ the asymptotic expressions as $l \to \infty$, and calculate the resultant integrals by the stationary phase method. This procedure, described in detail in [14] (Sec. 126), leads to the expression

$$f^\pm(\theta) = A_l^\pm \exp\{i[2\eta_l^\pm - (l_\pm + 1/2)\theta]\}$$
$$= [q^\pm(\theta)]^{1/2} \exp(iS^\pm), \tag{10}$$

where

$$A_l^\pm = \frac{1}{Mv} \left[\frac{l_\pm}{\sin\theta} \left| \frac{d^2\eta_l^\pm}{dl_\pm^2} \right|^{-1} \right]^{1/2} = [q^\pm(\theta)]^{1/2} \tag{11}$$

is the square root of the classical differential cross section for scattering by the potential $U_\pm(R)$, and $l_\pm(E, \theta)$ is the root of the equation

$$2\frac{d\eta_l^\pm}{dl_\pm} = \theta. \tag{12}$$

It is useful to note that the quantity $S^+(S^-)$ contained in (10) has a simple physical meaning: it is equal to the difference between the classical action (calculated for the entire scattering process) for a particle deflected in the field $U_+(U_-)$ through an angle θ, and the action of a particle moving uniformly in a straight line. Expression (10) is valid for a monotonic dependence of $U_\pm$ on R, under the condition

$$l_\pm(E, \theta)\theta \gg 1. \tag{13}$$

On the other hand, if the potential has a minimum (like U_+ for the process (5) and U_- for (6)), then there exists an angle of extremal deflection θ_c = $2(d\eta_l/dl)l_c$, where $l_{c\pm}$ is the root of the equation $d^2\eta_l^\pm/dl_\pm^2 = 0$. When $\theta \lesssim \theta_c$ formula (10) cannot be employed, owing to the so called "rainbow effect" [15,9].

In helium, the rainbow effect can occur for scattering in the field $U_-(R)$. Therefore the conditions for the applicability of formula (10) turn out to be not (13) but

$$l_+\theta \gg 1, \quad l_-(\theta - \theta_c) \gg 1. \tag{14}$$

We now write the initial formula for the probability of resonance charge exchange

$$w(E, \theta) = q/(q + q_1), \tag{15}$$

or on the basis of (10)

$$w = 1/2[1 - G(E, \theta)\cos(S^- - S^+)], \tag{16}$$

$$G(E, \theta) = 2(q^+ q^-)^{1/2}/(q^+ + q^-) < 1. \tag{17}$$

Formulas (16) and (17) show that one of the causes of the smoothing of the interference extrema is the difference between the classical cross sections q^+ and q^-.

Assume further that

$$U_\pm(\rho_\pm) / E \ll 1. \tag{18}$$

Then formula (7) takes the simpler form:

$$\eta_l^\pm = -\frac{1}{v} \int_\rho^\infty \frac{U_\pm(R) R dR}{(R^2 - \rho^2)^{1/2}} = -\frac{1}{v} F_\pm(\rho) \tag{19}$$

(see, for example, [14], Sec. 123). Although we imposed no limitations on θ in the derivation of (19), this formula is actually applicable only when $\theta \ll 1$, as can be readily seen from (18) (see also Problem 1 of Sec. 20 in the book by Landau and Lifshitz [16]).

If conditions (4), (9), (14), and (18) are satisfied, then the determination of $w(E, \theta)$ reduces on the basis of (10)−(12) and (19) to the following operations: from the equation

$$\left| \frac{dF_\pm(\rho)}{d\rho} \right| = E\theta \tag{20}$$

we obtain $\rho_\pm(E\theta)$; we then calculate

$$G(E\theta) = 2 \frac{[\rho_+\rho_-(d^2F_+/d\rho_+^2)(d^2F_-/d\rho_-^2)]^{1/2}}{\rho_+ d^2F_-/d\rho_-^2 + \rho_- d^2F_+/d\rho_+^2}, \tag{21}$$

and

$$S^- - S^+ = \frac{2}{v} \beta(E\theta)$$
$$= \frac{2}{v} [F_+(\rho_+) - F_-(\rho_-) + E\theta(\rho_+ - \rho_-)]. \tag{22}$$

We substitute (21) and (22) in (16) and obtain the charge-exchange probability as a function of E and θ. We note that when $\theta \ll 1$ the quantities $\rho_\pm$, G, and β are functions of the product $E\theta$, and not of E or θ separately.

3. CHARGE EXCHANGE IN HELIUM

The numerical calculation procedure by means of the formulas of the preceding section is analogous in many essential features to that described by Everhart [6], who considered the process (6) within the framework of the parametric method. We shall discuss in what follows principally those aspects which are not treated in [6]. Like Everhart, we choose for $\epsilon_+(R)$ the "diabatic" curve calculated by Lichten [7]. This term, which in the molecular-orbital approximation corresponds to the configuration $(1\sigma_g)(1\sigma_u)^2$ of the He_2^+ molecule determines, according to Lichten, the process of
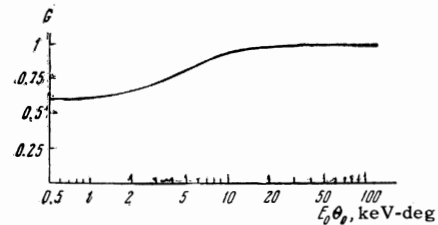

FIG. 1. 'Smoothing' function $G(E_0\theta_0)$.

resonance charge exchange of He^+ in He for not too small v. When R < 2, it crosses an infinite number of terms and then the boundary of the continuous spectrum of the He_2^+ quasimolecule.

However, the presence of the interference picture shows that the interaction with these terms and with the continuous spectrum is small. Therefore, for not too small v, we can calculate the process (6) by means of the formulas of Sec. 2, replacing, however, the adiabatic term Σ_g by the corresponding diabatic curve.

In view of the fact that the function $G(E\theta)$ varies very slowly (Fig. 1), the extrema of the $w(E, \theta)$ are determined, in accordance with (16), by the condition

$$S^- - S^+ = \pi k, \tag{23}$$

the even k correspond to the minima and the odd to the maxima.

Allowance for the coupling with the other states leads (see [9]) in lieu of (16) to the following formula for the total charge-exchange probability:

$$w = \tfrac{1}{2}[1 - G(E, \theta)A(E, \theta)\cos(S^- - S^+ + \Phi(E, \theta))], \tag{24}$$

$$A = [1 - P(1 + |f^+(\theta)|^2 / |f^-(\theta)|^2)]^{1/2}, \tag{25}$$

where P is the probability of all the inelastic processes except ionization, and Φ is the correction to the phase of $f^-(\theta)$.

In the case of process (5), which was considered by Smith [9], only the coupling between the states $2p\sigma$ and $2p\pi$ is essential (rotational coupling). Calculations show that $A(E, \theta)$ is close to unity, i.e., the rotational coupling changes the value of the extrema little. However, $\Phi(E, \theta)$ turned out to be close to π for $E\theta > 2$ keV/deg, when $\rho_+ \ll 1$, with $\Phi \sim 2$ at the lowest value $E\theta = 0.75$ keV-deg.

Since we do not calculate P and Φ in the present paper, we shall attempt to take account of the rotation effect indirectly. To this end we put A = 1 (P = 0) and $\Phi = \pi$. The first assumption is natural, while the second calls for some justification. We therefore turn to formula (4), putting in it, for rough estimates, a = 1 (an exact determination of a is a
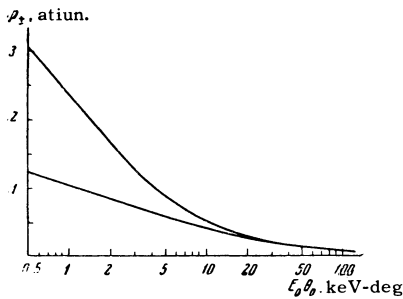
FIG. 2. Effective impact parameters $\rho_{\pm}$ $(E_0\theta_0)$.

laborious process). Using Fig. 2, we conclude that in the greater part of the experimentally investigated region of E and θ ($0.5° < \theta_0 < 5°$ and 0.4 keV $< E_0 < 25$ keV), a condition converse to (4) is satisfied. Since rotation of the quasimolecule is essentially a geometrical effect, it is natural to expect that the phase Φ is close to π for the indicated values of E and θ. In a smaller albeit significant part of the investigated range of E and θ, the inequality (4) is satisfied, i.e., the assumption that $\Phi = \pi$ is not justified. It can be thought, however, that at these values of E and θ the inequality $|\pi - \Phi| > \Phi$ is satisfied, as for process (5).

Putting[4] $\Phi = \pi$ in (24), we obtain in place of (23) the following extremum condition:

$$S^- - S^+ = \pi(k - 1). \tag{26}$$

For process (5) $S^- - S^+ < 0$, while for charge exchange (6) $S^- - S^+ > 0$. Therefore, in the former case the effect of rotation is to shift the extrema towards smaller values of E and θ, and in the latter towards larger ones.

Figure 3 shows the theoretical and experimental[18] curves that determine the maxima of the charge-exchange probability for k = 7, 9, 11, 13, and 15 ($S^- - S^+ = 6\pi$, 8π, 10π, 12π, and 14π, respectively). We see from the figure that the present results agree with experiment much better than the data of Everhart[6]. An exception is the region of very small $E\theta$, when Φ is probably much smaller than π.

It is also seen from these data that the agreement between Everhart's results[6] and experiment[18] is to some degree accidental. It is brought about by partial compensation of two effects which he did not take into account, the phase shift due to the rotation of the axis and the effect

of the non-coinciding trajectories for the fields U_+ and U_-. A similar compensation was observed by Smith[9] in the study of the process (5).

Lockwood et al.[18] give experimental curves of $w(\theta)$ for E = 25, 8.07, and 5.03 keV, and also $w(E)$ for $\theta = 1$, 3, and 5°. It is clear from Fig. 3 that the positions of the theoretical and experimental extrema are in the main close to one another. However, the corresponding w_{max} and w_{min} agree poorly, except for the $w(\theta)$ curve for E = 1 keV, where the agreement can be regarded as satisfactory.
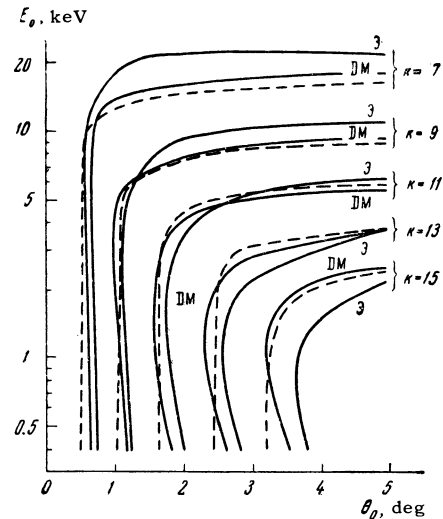

FIG. 3. Curves determining the position of the maxima of the charge-exchange probability; continuous curves — theory, dashed — experiment (DM — our results, E — Everhart's data).

To conclude this section, let us check the extent to which the remaining approximations used in the calculation are valid in the region of E and θ under consideration. It is easy to verify that conditions (9) and (18) are satisfied in our case. Going over to (14), we note that a limitation on the permissible values E and θ is imposed only by the second of these inequalities. Numerical estimates yield $l_c \approx 3Mv/2$ and $E\theta_c \approx 0.2$. Thus, this inequality can be rewritten in the form:

$$2v^{-1}\rho_-(E\theta - 0.2) \gg 1. \tag{27}$$

Using the $\rho_-(E\theta)$ curve of Fig. 2, we can verify that this condition is satisfied except for the smallest values of E and θ (the lower left corner of the region shown in Fig. 3). A typical value of the left side of (27) is several times ten. Thus, the transition to the classical limits described in Sec. 2 is justified and the rainbow effect is negligible here.

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[4]We tacitly imply in this case that the interaction between the state $(1\sigma_g)(1\sigma_u)^2$ and other states does not lead to additional phase multipliers in the amplitude $f^+(\theta)$, since a sudden flipping of the internuclear axis does not change in this case the sign of the wave function.

4. CONCLUSION

We did not take into account in this article the following causes for resonance smoothing, which were not investigated with sufficient consistency: 1) rotation of the quasimolecule; 2) transitions connected with the fact that the diabatic curve crosses an infinite number of terms of the same symmetry; 3) autoionization connected with the fact that the state $(1\sigma_g)(1\sigma_u)^2$ is quasistationary at at low values of R.

The first effect can be accounted for in a manner similar to that used by Bates and Williams[12] (see also formula (24)). The second effect can hardly be taken into account theoretically. It is more reasonable to solve the inverse problem, that of estimating the probabilities of the corresponding processes by comparing calculated w_{max} and w_{min} (with account of the rotational coupling) with the measured values and of estimating the addition to the phase $f^+(\theta)$ due to pseudocrossing by comparing their relative locations. A theoretical calculation of autoionization also entails great difficulties. It can be assumed however, that in our region of E and θ this process does not play a large role. In fact, from formulas (24) and (25), it is easy to see that the probability (averaged over some region of E and θ) (we denote it by $\widetilde{w}$) is close to $\frac{1}{2}$. On the other hand, the experimental values lie in the range $0.4 < \widetilde{w} < 0.6$. If autoionization were to play an important role, then a noticeable excess of w over $\frac{1}{2}$ would be observed[5]. The fact that $\widetilde{w} > \frac{1}{2}$ in many cases may be the consequence of either experimental errors or of the unjustified assumption used in the derivation of (24), that the transitions to the excited states do not lead to a change in the incident-particle scattering angle. If the velocities are sufficiently small, then the state of the quasimolecule changes in accordance with the adiabatic curve. A perfectly rigorous description of the transition to this state from the diabatic term is not a simple problem. It must be empha-

sized, incidentally, that such a transition can be realized only in large-angle experiments ($\theta \sim 1$ rad), when it is possible to attain not only an adiabatic change of the state of the quasimolecule, but also a sufficiently close approach of the ion to the atom.

It must be noted in conclusion that, owing to the presence of interference, measurements of the differential cross section of resonance charge exchange make it possible to obtain much more information on the behavior of the terms than from the usual measurements of the angular distribution in the absence of interference (elastic scattering of atoms, scattering of ions by atoms of a different gas, etc.).

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[5] As can be seen from (24) transitions to the excited bound states of He_2^+ do not change $\widetilde{w}$, since half of the particles that go through such a transition contribute to the charge-exchange cross section (with excitation), while the other half contribute to the excitation cross section.

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