Transfer Ionization in $H^+ + H^-$ Collisions

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(Received 20 July 1987)

Absolute cross sections for transfer ionization in $H^+ + H^-$ collisions have been measured in the center-of-mass energy range 50 eV to 40 keV, with the use of merged- and crossed-beam techniques, together with coincident detection of the products. A mechanism is proposed for the process in the low-energy region. The cross section, calculated accordingly, is found to be in satisfactory agreement with the experimental data.

PACS numbers: 34.70.+e

Transfer ionization (TI) is a two-electron process in which one of the target electrons is captured into the projectile and the other one is ejected. In atom–multiply-charged-ion collisions, this process has been extensively studied in the last years (see, e.g., the review by Salzborn and Müller[1]). It is strongly exothermic, and its mechanisms are rather well understood, at least at low energy, in terms of the different Auger relaxation channels of a superexcited quasimolecular state.[2]

In this Letter, we report on a fundamental investigation of transfer ionization in simple, endothermic conditions, namely, in collisions between H^+ and H^- ions. This is, to our knowledge, the first work on TI in ion-ion collisions. With only two electrons, the H^+-H^- system is simple enough hopefully to be treated theoretically without major uncertainties regarding the electronic structure, so that attention can be focused on the dynamical aspects. On the other hand, it is complex enough to offer intricate rearrangement possibilities. Whereas mutual neutralization[3–6] and electron detachment[7–10] in this system have been successfully studied in the past, there are, to our knowledge, no data available yet for the transfer-ionization process.

In the present work, the cross section of the transfer-ionization reaction

$$H_a^+ + H_b^- \rightarrow H_a + H_b^+ + e$$

has been measured over a wide energy range extending from 50 eV to 40 keV. Furthermore, a two-step mechanism has been considered and a model calculation carried out to estimate the cross section.

The absolute cross section at center-of-mass energies from 880 eV to 40 keV has been measured in Giessen by means of the crossed-beam technique and a coincidence method in the detection of the reaction products H^0 and H^+. Apart from some minor changes for the present measurements, the experimental arrangement used has been previously[11] described in full detail. In short, two well-collimated and momentum-analyzed ion beams of adjustable energies are arranged to intersect at an angle $\theta = 45°$ in ultrahigh vacuum of $\simeq 10^{-11}$ mbar. The collision products formed in both beams are charge analyzed after the interaction region by electrostatic deflection. Both H^0 atoms and H^+ ions are counted individually by identical single-particle detectors. In order to reduce the background, both ion beams are cleaned, shortly before intersection, by electrostatic deflection of particles in other charge states which result from charge-changing collisions in the residual gas. In spite of this precaution and the prevailing ultrahigh vacuum in the interaction region, a coincidence technique had to be employed for signal recovery.

The cross section at energies below 700 eV has been measured in Louvain-la-Neuve with a merged-beam set-up. The apparatus and the experimental method have been described in an earlier paper on the mutual neutralization of H^+ and H^- (Ref. 3). Only a brief description is needed here. Two well-collimated equivelocity beams of H^+ and H^- are merged over a distance of 10 cm in high vacuum (10^{-10} mbar). The actual interaction length (7 cm) is a part of the merging region on which a polarization voltage is applied. This voltage, by its discriminating action on the velocity of ions and atoms, at the same time controls the energy of the colliding particles and differentiates the reaction products formed over the reaction length from those created elsewhere. The beams are electrostatically demerged and the neutral and positive components are further analyzed magnetically. This energy analysis completely separates the primary positive ions, which are measured as a current, from the protons created in the stripping of H^-. These are detected individually as are the neutral products.

A twofold time-correlation analysis is performed of the signals delivered by the detectors: one part dealing with pairs of neutrals and the other with neutral-ion pairs. The former yields the contribution of mutual neu-

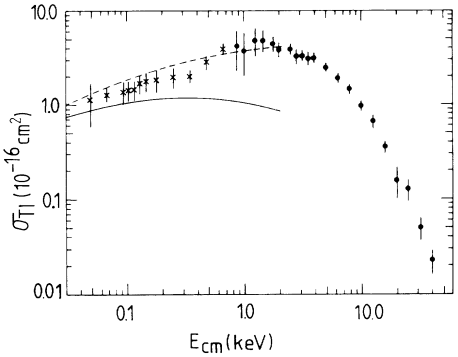

FIG. 1. Total cross section for the transfer ionization: $H^+ + H^- \rightarrow H + H^+ + e$. Experiment: Giessen results, circles; Louvain-la-Neuve results, crosses. Error bars correspond to 90% confidence limits. Theory: present model, full curve; present model with all f_K set equal to $\frac{1}{2}$ (see text), dashed curve.

tralization,

$$H^+ + H^- \rightarrow H + H,$$

while the latter simultaneously provides the contribution of transfer ionization. The ratio of the two cross sections so obtained is not affected by the systematic error that easily arises in the determination of the geometrical overlap of the beams. The cross section for transfer ionization is then derived by use of the results of Szücs *et al.* for mutual neutralization. The experimental results over the whole energy range are presented in Fig. 1.

A full theoretical treatment of TI is an extremely difficult task since many discrete states and a whole continuum are strongly coupled. However, in the low-energy range, up to a few kiloelectronvolts, the transfer ionization in H^+-H^- collisions can be regarded as the result of the autoionization of antibonding molecular states eventually created after mutual neutralization in the colliding system. Such an approach has been used by Urbain *et al.*[12] in the theoretical study of associative ionization. An estimate of the cross section will be given on the basis of a rather simple model.

In that model, we assume that the relevant autoionizing states are those correlated with $H(1s)+H(n=2,3)$ states, since these are the only ones likely to be populated from the initial ionic channel. There are three such states, as shown in Figs. 2(a) and 2(b): one *gerade* (state I) and two *ungerade* (states II and III). In contrast to state III, the states I and II can be reached in different ways, corresponding to the different possibilities of transfering the outer, weakly bound electron of H^- and promoting the inner electron into the $(2p\sigma_u)$ molecular orbital. Each of these states becomes autoionizing when its energy curve enters the H_2^+ continuum, that is, after crossing the series of $(1s\sigma_g nl\sigma_{g,u})$ Rydberg bonding levels. This implies a loss of flux that must be taken into account in calculation of the probability of effective autoionization.

The first step in the calculation deals with the mutual neutralization into $H(1s)+H(n=2)$ or $H(1s)+H(n=3)$, at large internuclear distances. We define p_2 and p_3 as the respective probabilities of nonneutralization. At low energy (below 20 eV), the use of the Landau-Zener (LZ) approximation is well justified to calculate p_2 and p_3 at the crossing points, $R_3=36$ a.u. and $R_2=11$ a.u.[13] At higher energy, however, it has been established[5] that an additional dynamical radial coupling is active between the ionic and the $n=3$ covalent states

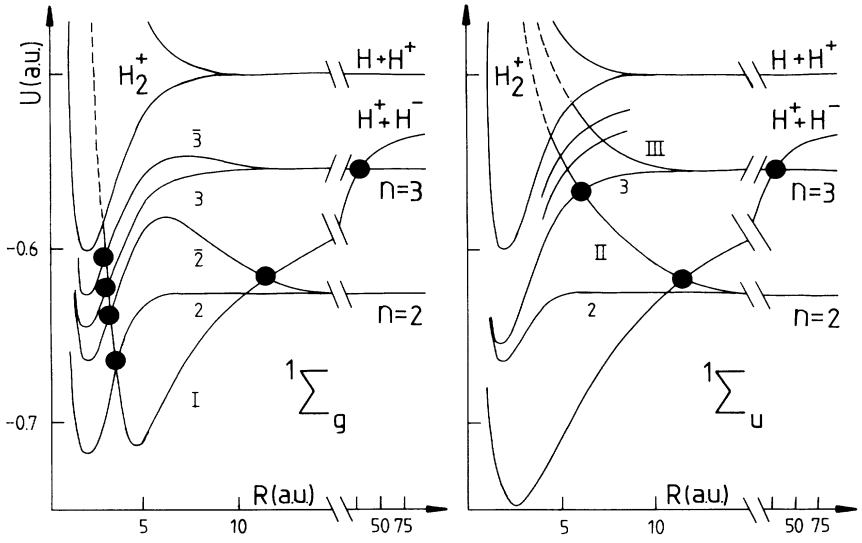

FIG. 2. Relevant potential energy curves with associated crossings (circles). (a) $^1\Sigma_g$ symmetry. (b) $^1\Sigma_u$ symmetry. [$n=2,3$ stand for $H(1s)+H(n=2,3)$ limits, respectively.]

at values of R around 15 a.u. We therefore express p_2 and p_3 as

$$p_2 = p_2^{\mathrm{LZ}}, \quad p_3 = g(b,v_R)p_3^{\mathrm{LZ}}.$$

The function $g(b,v_R)$ depends upon the impact parameter b and the radial velocity v_R at $R=15$ a.u. and is chosen in such a way as to reproduce the actual neutralization cross section for collision energies below 2 keV.

At shorter distances, the probability flux is equally shared between *ungerade* and *gerade* molecular states. In the $^1\Sigma_g$ symmetry [Fig. 2(a)], the ionic state diabatically goes over to the well known $(2p\sigma_u)^2$ antibonding state. It should be noticed that the latter state can also be populated around $R=3$ a.u. from the bonding states correlated with $H(1s)+H(n=2,3)$. The related probabilities are expressed as p_{nK}, where $K=$I, II, or III. In the $^1\Sigma_u$ symmetry [Fig. 2(b)], the neutralization populates the states II and III. State III consists of two quasidegenerate autoionizing states $(2p\sigma_u 4d\sigma_g)$ and $(2p\sigma_u 5g\sigma_g)$. State II is associated with the $(2p\sigma_u 3d\sigma_g)$ configuration.

The next point is to calculate, for each of the states I, II, and III, the probability that it actually autoionizes in the collision. This probability is the product of two terms: one, s_K, featuring the survival probability of state K across the Rydberg series below the continuum, and the other, f_K, giving the probability of autoionization over the time spent in the continuum (that probability is halved to take into account the fact that the dissociation leads with equal probabilities to detachment and TI). The latter is expressed in the form

$$f_K = \tfrac{1}{2}\left[1-\exp\left(-2\int_{r_K}^{R_K}\Gamma_K(R)\,dR/v_R\right)\right],$$

where Γ_K, the autoionization rate to the H_2^+ $(1s\sigma_g)$ continuum, is taken from Tennyson and Noble,[14] and r_K and R_K are the nuclear separations at the turning point and at the crossing with the continuum edge, respectively. The former term is given by the LZ formula for multiple crossing:

$$S_K = \exp(-\textstyle\sum_n \gamma_{nK}),$$

where γ_{nK} characterizes the interaction between state K and the Rydberg state with principal quantum number n. The value of γ_{nK} is linked to Γ_K through n^{-3} scaling. The probabilities p_{nK} are evaluated in the same way:

$$p_{nK} = \exp(-\gamma_{nK}).$$

Finally, we obtain the transfer-ionization probability P_{TI} as a sum:

$$P_{\mathrm{TI}}(b) = \textstyle\sum_{i,K} P_{iK},$$

where P_{iK} relates to the way (i) of reaching the continuum via the autoionizing state K. We get

$$P_{1\mathrm{I}} = \tfrac{1}{2}p_3p_2p_{2\mathrm{I}}p_{\bar{2}\mathrm{I}}p_{3\mathrm{I}}p_{\bar{3}\mathrm{I}}s_{\mathrm{I}}f_{\mathrm{I}},$$

$$P_{2\mathrm{I}} = \tfrac{1}{2}p_3(1-p_2)(1-p_{\bar{2}\mathrm{I}})p_{3\mathrm{I}}p_{\bar{3}\mathrm{I}}s_{\mathrm{I}}f_{\mathrm{I}},$$

$$P_{3\mathrm{I}} = \tfrac{1}{2}(1-p_3)[\tfrac{1}{3}(1-p_{3\mathrm{I}})p_{\bar{3}\mathrm{I}} + \tfrac{2}{3}(1-p_{\bar{3}\mathrm{I}})]s_{\mathrm{I}}f_{\mathrm{I}},$$

$$P_{1\mathrm{II}} = \tfrac{1}{2}p_3(1-p_2)p_{3\mathrm{II}}s_{\mathrm{II}}f_{\mathrm{II}},$$

$$P_{2\mathrm{II}} = \tfrac{1}{2}(1-p_3)\tfrac{1}{3}(1-p_{3\mathrm{II}})s_{\mathrm{II}}f_{\mathrm{II}},$$

$$P_{1\mathrm{III}} = \tfrac{1}{2}(1-p_3)\tfrac{2}{3}s_{\mathrm{III}}f_{\mathrm{III}}.$$

The total cross section is then obtained as

$$\sigma_{\mathrm{TI}} = 2\pi\int_0^\infty P_{\mathrm{TI}}(b)\,b\,db.$$

The results of our calculations are shown in Fig. 1 (full curve). At low energy, they are in satisfactory agreement with the experimental data although they systematically underestimate them. This might be an indication that some autoionization is produced by a coupling to other doubly excited configurations. An upper limit to the effect of such couplings is readily obtained by setting all f_K equal to $\frac{1}{2}$. The result of this is shown in Fig. 1 (dashed curve). At high energies (above 2 keV), the present model is not expected to be adequate: It neglects electron momentum translational effects which affect the probabilities p_2 and p_3. It also ignores the direct detachment of H^- which, at high energy, can supersede the mutual neutralization as the primary mechanism.

In summary, the main result of the present work is the report of reliable experimental values for the cross section of transfer ionization in $H^+ + H^-$ collisions, over a wide energy range. Their reliability is attested by the consistency of the results obtained by the use of two different experimental methods. The wide energy range that is covered reveals the entire energy dependence of the cross section, which, at high energy, is rather similar to the one observed in multiply-charged-ion–atom collisions, but which, at low energy, decreases in a specific way. The low-energy behavior is well rendered when a two-step mechanism is considered, where mutual neutralization is followed by autoionization.

This work was supported by the Bundesministerium für Forschung und Technologie, Germany, and the Fonds National de la Recherche Scientifique, Belgium.

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