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High-energy behavior of the vibrational excitation cross section σ_{01} for H^+-H_2

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We present calculations of the cross section σ_{01} for the vibrational excitation of H_2 by proton impact with several interaction potentials. The calculation with a long-range anisotropic potential shows features very similar to the calculation with the H_3^+ potential surface, but both calculations fail to account for the observed persistence of the cross section at high velocities (proton energy range from 200 to 1000 eV). A trajectory-dependent potential is introduced semiempirically, and it is found that the main features of the observed cross section can be explained by its short-range parts ($r \lesssim 2$ Å). The significance of this potential is discussed. The approximation scheme involves first-order time-dependent perturbation theory and considers only rectilinear trajectories. Therefore, for the case reported here, the calculation is valid for energies higher than about 200 eV, where the trajectories are essentially rectilinear. The semiempirical deduction of the trajectory-dependent potential was greatly simplified by representing the time-dependent potential by a Gaussian pulse. The results are compared with a previous calculation.

I. INTRODUCTION

Recent experimental measurements of the cross sections for vibrational excitation of the diatomic molecule H_2 by proton impact,[1] and of the molecular ion H_2^+ by impact on neutral targets[2] have revealed some features which do not appear in the theoretically calculated cross sections.[3]

The most outstanding feature of the cross sections for a single quantum transition, besides their large magnitude, is their persistence in the high-velocity region ($v > 2\times 10^7$ cm/sec) as evidenced by a broad maximum in the energy region of 300–1000 eV. The cross sections involving multiquantum transitions are smaller, and are generally monotonically decreasing with energy in the high-velocity region.

The broad maximum in the cross section associated with single quantum transitions has been interpreted qualitatively on the basis of Massey's adiabatic criterion.[4] However, the collision distances obtained with this simple idea are somewhat large. For example, for the H^+-H_2 system, the collision distances obtained for the single quantum excitation of H_2 is about 8×10^{-8} cm. This distance is longer than the range of the potential, which is less than 4×10^{-8} cm.

So far, the calculations of vibrational excitation cross sections reported by Ritchie[5] for the system H^+-H_2 have used a long-range isotropic potential of interaction with a suitably chosen short-range cutoff. The calculation done by Ritchie and those reported here with a long-range anisotropic potential do not account for the broad maximum observed in the cross section for the single quantum ($\nu' : 0 \rightarrow 1$) excitation collision in the H^+-H_2 system. Also, as we report in the present work, calculations employing the potential surface of H_3^+ yield a cross-section energy curve similar in character to that obtained with the long-range potentials.

None of the above calculations account for the very broad maximum observed in the experimental cross section. Thus, it seems that the observed behavior of the cross section cannot be explained in terms of the static character of a simple adiabatic potential surface or in terms of the expected anisotropies of the long-range potential.

At least two reasons can be given for the lack of agreement using the potentials mentioned above. First, the H^+-H_2 potential curve crosses the potential curve for the charge-exchanged system H-H_2^+ at approximately 1.2 Å. Transitions occurring in this region should influence the outcome of the collision.[6,7] In the energy range 200–1000 eV the probability of excitation of the electronic cloud of H_2 is expected to be large. In fact, the two adiabatic electronic systems ($H^+ + H_2$) and ($H + H_2^+$) are both energetically possible at short distances. The presence of the ($H + H_2^+$) channels, communicated by electron exchange with the original ($H^+ + H_2$), increases the probability of excitation of the vibrational modes of H_2, making the "effective" potential between H^+ and H_2 softer at short distances. The collision starts at large distances with an ($H_2 + H^+$) configuration and as the H^+ gets closer to H_2 (in the region of 1.2 Å) the (H_2^+

$+H$) configuration becomes very competitive. In fact, in this region the nonadiabatic coupling between the two surfaces reaches its maximum. Thus it is plausible that the system for these close distances may have one or another configuration, and may undergo jumps from one to another by "virtual" electron transitions. The coupling between both configurations is due to the effect of the relative motion of projectile versus target on the electronic wave function. The Q value of the reaction $H_2 + H^+ \rightarrow H_2^+ + H$ is 1.835 eV; then, for large separation, with H_2 in vibrational states having $\nu' \leq 4$ before the interaction, the configuration remains $(H_2 + H^+)$ and so only one potential surface will be possible. The crossing of the two-potential surfaces, which occurs at about 1.2 Å, makes the effective two-body interaction between H^+ and H_2 quite nonlocal in character.

The second reason for expecting a nonlocal potential is that the time the proton spends in the vicinity of the H_2 molecule is of the order of the vibrational period of the molecule. This means, of course, that the nuclear motion of H_2 is no longer independent of the motion of the incident proton, and it becomes extremely difficult to obtain the wave function for the system. Thus, the molecular nuclei may respond differently to two identical instantaneous distances of the incident proton (which correspond to the same proton-molecule distance) if those two instantaneous distances belong to different trajectories (different impact parameters). As we shall see below, this characteristic justifies the potential that is finally suggested by this work.

In the light of these arguments it is not surprising that calculations with ordinary potentials have failed. However, precisely because of the reasons given above, it is very difficult at the moment to obtain from first principles a potential which will describe the process of vibrational excitation of H_2 by proton impact adequately.

We have found that we can account for the broad maximum of the cross section mentioned above by properly adjusting the potential at short distances in such a way that it conforms qualitatively to the arguments given above regarding the dependence of the motion of the three nuclei and the possibility of electron exchange. In order to deduce the proper form of the potential it was necessary to allow the potential to have a simultaneous dependence on the proton-molecule distance r as well as the impact parameter of the collision b. This nonseparability of the projectile from the motion of the internal coordinates of the target may be associated with a dynamical description of the collision in terms of a nonlocal potential.

In the following section on theory (Sec. II), the general method of calculation of transition probabilities of cross section is described. In Sec. III we discuss in some detail the following potentials: (i) the anisotropic long-range interactions with cutoff at short distances, (ii) two possible potentials deduced from the H_3^+ potential surface of Conroy, and (iii) the potential proposed in this work which fits the experimental data. In Sec. IV, we present the results and conclusions.

II. THEORY

Consider the collision between a proton and a hydrogen molecule. We designate the proton-molecule distance by the vector $\vec{r}$. The symbol y represents the distance between the nuclei of H_2, and θ_0 is the reference orientation angle between the internuclear axis of the molecule and the asymptote of the proton trajectory (see Fig. 1). The instantaneous angle between the internuclear axis and the vector $\vec{r}$ is designated by θ, and b is the impact parameter of the collision. Since we are interested in the vibrational excitation of the molecule, we write the potential as a function of $\vec{r}$ and y:

$$V(\vec{r}, y) = V(\vec{r}_0, y_0) + (\nabla_r V)_{r_0, y_0} \nabla \vec{r} + (\nabla_y V)_{r_0, y_0} \nabla y + \cdots, \tag{1}$$

where y_0 is the equilibrium displacement of the molecule. If we define the dimensionless quantity

$$\xi = \frac{y - y_0}{y_{\max} - y_0} = \frac{y - y_0}{(2\pi/m\omega)^{1/2}} \tag{2}$$

and express the effect of the angle θ explicitly, we can write

$$V(r, y, \theta) = V_0(r, \theta) + V_1(r, \theta)\xi + \tfrac{1}{2} V_2(r, \theta)\xi^2 + \cdots. \tag{3}$$

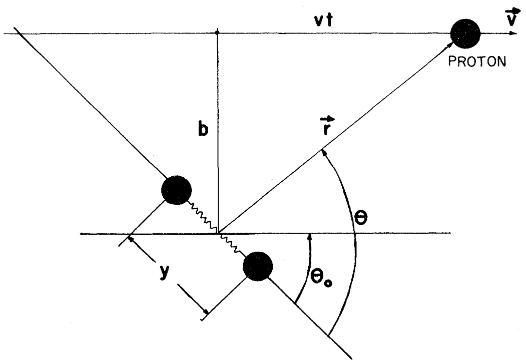

FIG. 1. Coordinates for the H^+-H_2 collision.

Here,

$$V_0(r,\theta)=V(r,\theta,\xi=0)\,,$$

$$V_1(r,\theta)=\left.\frac{\partial V}{\partial \xi}\right|_{\xi=0},$$

$$V_2(r,\theta)=\left.\frac{\partial^2 V}{\partial \xi^2}\right|_{\xi=0},\quad \text{etc.}$$

After averaging in angle, we may write the previous expressions as

$$\overline{V}_0(r,y)=\overline{V}_0(r)+\overline{V}_1(r)\xi+\tfrac{1}{2}\overline{V}_2(r)\xi^2+\cdots\,. \tag{4}$$

The proton kinetic energies to be considered here will always be larger than 200 eV. At these energies the de Broglie wavelength of the proton is much smaller than the molecular target size, and the total scattering cross section is strongly forward peaked.[1] Therefore, the proton trajectory can be represented by a classical straight-line trajectory of the form

$$r^2=v^2t^2+b^2\,, \tag{5}$$

where v is the proton velocity, t is the time measure from the classical turning point, and b the impact parameter. Substitution of Eq. (5) into Eq. (3) allows us to write the coefficients $V_i(r,\theta)$ as explicit functions of time.

We are interested in determining the cross section for the excitation of one vibrational quantum ($\nu'=0\rightarrow\nu'=1$) of H_2 as a result of the proton collision. We denote the cross section with the symbol σ_{01} using the initial and final vibrational quantum numbers $\nu'=0$ and $\nu'=1$ as subscripts.

In our case of classical trajectories, the number of projectile angular momenta contributing to the cross section will be very large, and we can substitute the angular momentum quantum number l by the relation $k_0b=l+\frac{1}{2}$ where k_0 is the projectile wave vector. In these cases the cross section σ_{0n} may be obtained from the transition probability P_{0n} at a given energy from

$$\sigma_{0n}=2\pi\int P_{0n}(b)b\,db\,. \tag{6}$$

The expression for $P_{0n}(b)$ will be obtained from time-dependent perturbation theory. In the Schrödinger picture, the state vector $|\psi(t)\rangle$ at a time t evolves from the state vector $|\psi(t_0)\rangle$ at a prior time t_0 by the time evolution operator $U(t,t_0)$:

$$|\psi(t)\rangle=U(t,t_0)|\psi(t_0)\rangle\,. \tag{7}$$

In our case $|\psi(t_0)\rangle$ is the state before the interaction, and it is given by $|k_0\rangle|H_0\rangle$ where $|k_0\rangle$ is the plane wave corresponding to the projectile and $|H_0\rangle$ is the ground state of the H_2 molecule. At any other time after t_0 the state of the system $|\psi(t)\rangle$ will be a linear combination of $|k_n\rangle|H_n\rangle=|\phi_n\rangle$, where $|H_n\rangle$ is an excited state of H_2 which may include electronic, vibrational, and rotational excitation. Since we are considering pure vibrational excitation, only $|H_n\rangle$ represents the nth excited state of the harmonic oscillator.

The transition probability is then given by

$$P_{0n}(t)=|\langle\psi(t)|\phi_n\rangle|^2=|\langle\phi_n|U(t,t_0)|\phi_0\rangle|^2\,. \tag{8}$$

This probability can be expressed also in terms of the evolution operator in the interaction picture $U_I(t,t_0)$, which is given by

$$U_I(t,t_0)=1-\frac{i}{\hbar}\int_{t_0}^{t}V_I(\tau)U_I(\tau,t_0)\,d\tau\,, \tag{9}$$

where $V_I(\tau)$ is the potential $V(\tau)$ expressed in the interaction picture; $V(\tau)$ is the potential given by Eq. (3) [or Eq. (4) in the case that we average in angle] but as a function of time. The passage from configuration space to time dependence may be done, for high energy, by Eq. (5). If this is done the interaction will be a function of time, and will be parametric on the impact parameter b and on the velocity v.

In the interaction picture then

$$P_{0n}(t)=|\langle\phi_n|U_I(t,t_0)|\phi_0\rangle|^2\,. \tag{10}$$

To approximation in first order, Eq. (9) gives

$$P_{0n}(t)=\frac{1}{\hbar^2}\left|\int_{t_0}^{t}V_I^{0n}(\tau)\,d\tau\right|^2, \tag{11}$$

where

$$\begin{aligned}V_I^{0n}(\tau)&=\langle\phi_n|e^{iH_0\tau/\hbar}V(\tau)e^{-iH_0\tau/\hbar}|\phi_0\rangle\\&=e^{i(E_n-E_0)\tau/\hbar}V_{0n}(\tau)\,,\end{aligned} \tag{12}$$

where H_0 is the Hamiltonian without the interaction, and

$$V_{0n}(\tau)=\langle\phi_n|V(\tau)|\phi_0\rangle\,. \tag{13}$$

Substituting Eq. (4) into Eq. (13), we obtain the expression

$$\begin{aligned}V_{0n}(\tau)=&\langle\phi_n|\overline{V}_0\{r(\tau)\}|\phi_0\rangle\\&+\langle\phi_n|\overline{V}_1\{r(\tau)\}\xi|\phi_0\rangle+\cdots\,.\end{aligned} \tag{14}$$

In this series only the nth-order term is nonzero. That is,

$$V_{0n}(\tau)=\langle\phi_n|\overline{V}_n\{r(\tau)\}(\xi^n/n!)|\phi_0\rangle\,. \tag{14a}$$

Hence, Eq. (11) becomes

$$\begin{aligned}P_{0n}(t)=\frac{1}{\hbar^2}\Bigg|\int_{t_0}^{t}&e^{i(E_n-E_0)\tau/\hbar}\\&\times\langle\phi_n|\overline{V}_n\{r(\tau)\}(\xi^n/n!)|\phi_0\rangle d\tau\Bigg|^2\end{aligned} \tag{15}$$

As we have said previously below Eq. (9), V_n will be parametric on b and also on v. We have found it particularly convenient for the development presented here to approximate V_n by Gaussian pulses in time, of the form

$$V_n = A_{0n}(b)\, e^{-\beta t^2/T^2}\,, \tag{16}$$

where

$$T = b/v\,.$$

For the case of single quantum ($\nu' = 0 \rightarrow \nu' = 1$) excitation of H_2 by proton impact at energies higher than 200 eV the main contribution to the scattering comes from forward scattering.[1] So the direction of $\vec{k}_0$, the proton momentum after the collision, is virtually the same as the direction of $\vec{k}_1$, the momentum after the collision. Furthermore, since the energy lost per excitation collision is only 0.5 eV, we set $\vec{k}_1 \approx \vec{k}_0$. Therefore, in this approximation, the matrix element P_{0n} [Eq. (15)] is independent of the scattering angle but dependent on the impact parameter. Thus, the angular momentum dependence of the wave functions is explicitly shown in the amplitude A_{01} and in the characteristic time $T/\beta^{1/2}$ of Eq. (16).

Finally, the first-order transition probability for the $\nu' = 0 \rightarrow \nu' = 1$ excitation of the ground state H_2 by proton impact for energies higher than 200 eV is given by

$$P_{01}(b,t) = \frac{1}{\hbar^2} \left| \int_{t_0}^{t} e^{i\omega_{10}\tau} B_{01} A_{01}(b)\, e^{-\beta\tau^2/T^2(b)}\, d\tau \right|^2, \tag{17}$$

where

$$B_{01} = \langle H_1 | \, \xi \, | H_0 \rangle$$

and (18)

$$\omega_{01} = (E_1 - E_0)/\hbar\,.$$

Now, letting $t_0 \rightarrow -\infty$ and $t \rightarrow \infty$ we obtain

$$P_{01}(b) = \frac{\pi}{\hbar^2} \frac{T^2}{\beta(b)} e^{-(T\omega_{10})^2/2\beta(b)} |B_{01} A_{01}(b)|^2\,. \tag{19}$$

The inelastic cross section can be found using Eq. (6).

In Sec. III we carry out the above analysis for the various cases of interest here, and we also show the utility of the Gaussian pulse approximation in obtaining the final potential.

III. POTENTIALS

A. Long-range potential with cutoff at short distances

We have calculated σ_{01} using a long-range potential similar to one used by Ritchie[5] for $H^+ + H_2$ and also Takayanagi and Geltman[8] for $e^- + H_2$. This potential was taken as

$$V(r,\theta) = -\frac{1}{2}\frac{\alpha e^2}{r^4} - \frac{1}{2}\frac{e^2}{r^4}\alpha' P_2(\cos\theta) + \frac{1}{2}\frac{e^2 Q}{r^3} P_2(\cos\theta)\,, \tag{20}$$

where α and α' are related to the polarizability of the molecule along its axis and perpendicular to it. P_2 is the Legendre polynomial of second order, and Q the electric quadrupole moment of the molecule. The molecule H_2 has no permanent dipole moment; for this reason there is no term proportional to the Legendre polynomial of first degree. After the linear approximation, from which we may write

$$r^2 = v^2 t^2 + b^2 = b^2(1 + \tau^2) \tag{21}$$

and (see Fig. 1)

$$\cos\theta = (\tau b/r)\cos\theta_0 \pm (b/r)\sin\theta_0\,, \tag{22}$$

where

$$\tau = vt/b\,, \tag{23}$$

the interaction potential given by Eq. (20) becomes, for $\theta = 0$, a simple function $W(\tau, b)$, that is

$$W(\tau,b) = \frac{A_1}{r^3} + \frac{A_2}{r^4} + \frac{A_3 b^2 \tau^2}{r^5} + \frac{A_4 b^2 \tau^2}{r^6}\,. \tag{24}$$

The coefficients A_i are

$$\begin{aligned} A_1 &= -\tfrac{1}{4} e^2 Q = -0.19\ \text{Å}^3\,\text{eV}\,,\\ A_2 &= -\tfrac{1}{2} e^2(\alpha + \tfrac{1}{2}\alpha') = -1.04\ \text{Å}^4\,\text{eV}\,,\\ A_3 &= \tfrac{3}{4} e^2 Q = 0.58\ \text{Å}^3\,\text{eV}\,,\\ A_4 &= -\tfrac{3}{4}\alpha' e^2 = -0.64\ \text{Å}^4\,\text{eV}\,. \end{aligned} \tag{25}$$

The calculation was done for $\theta_0 = 0$, $\theta_0 = \frac{1}{4}\pi$, and $\theta_0 = \frac{1}{2}\pi$. For the last two angles Eq. (24) takes a slightly different form. After the probabilities were calculated using Eq. (10), the average cross section was obtained using the same criterion as Takayanagi and Geltman.[8] That is, assuming that

$$\sigma(\theta_0) = \sigma_0 + \sigma_2 P_2(\cos\theta_0) + \sigma_4 P_4(\cos\theta_0)\,, \tag{26}$$

the average cross section becomes

$$\bar{\sigma} = \tfrac{1}{15} |\sigma(0) + 8\sigma(\tfrac{1}{4}\pi) + 6\sigma(\tfrac{1}{2}\pi)|\,. \tag{27}$$

We have predicted the cross section for several cutoffs ranging from 0.95 to 2.0 Å. Although the shape of the cross-section curves is similar for all of them, their magnitudes are different. In fact, (see Fig. 2) in all cases the cross sections show a sharp peak at around 250 eV and positive curvature for energies greater than 400 eV.

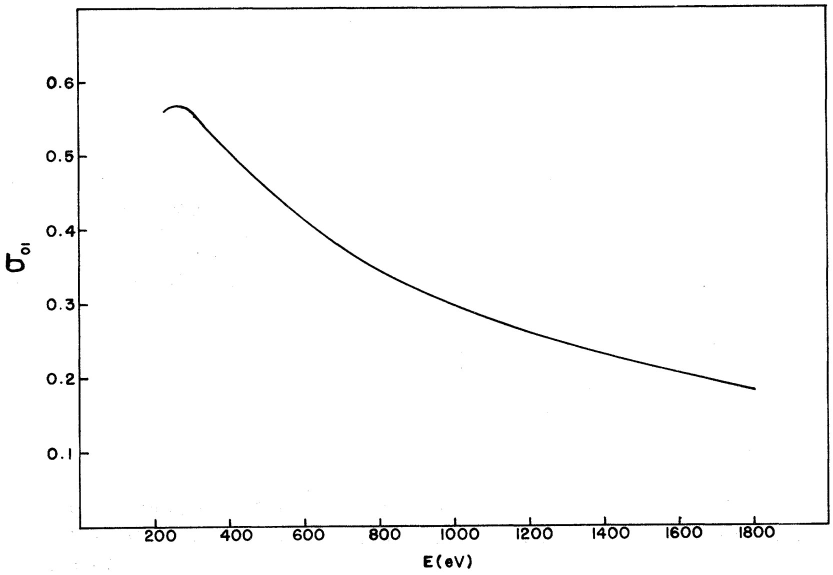

FIG. 2. Vibrational excitation cross section σ_{01} in Å^2 for the transition $0 \rightarrow 1$ as function of the laboratory energy of H$^+$, corresponding to the anisotropic long-range potential given by Eq. (20).

B. Potential deduced from the H_3^+ configuration

Assuming that we are dealing with an atomic configuration, where the process may be assumed to develop adiabatically, we may deduce the $H^+ \rightarrow H_2$ potential from the energy of the H_3^+. The potential energy will be equal to the energy of the H_3^+ system minus the energy of the $H_2 + H^+$ when both elements are at infinite distance. This was done by several authors[6,7] under certain limitations of the geometric configuration of the three atoms. We have also deduced such a potential using the work of Conroy[9] for the isosceles-triangle configuration of H_3^+.

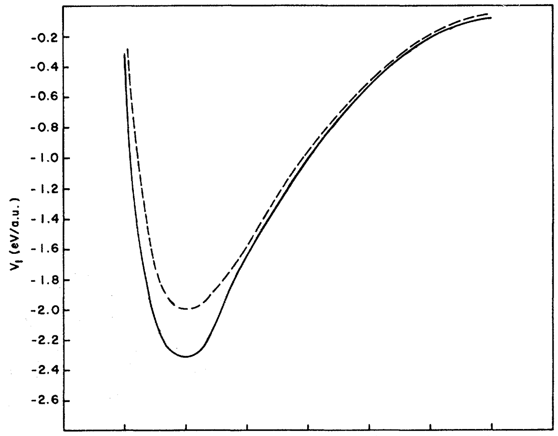

FIG. 3. V_{01} potential deduced from the H_3^+ surface energy calculated by Conroy (Ref. 9). The solid curve corresponds to values of $y = y_{\min}$ such that the energy of the H_3^+ system presents a minimum, for the given r. The dashed curve corresponds to $y = y_0 = 1.4$ a.u.

The potential $V(r, y, \theta)$ (see Fig. 3) may be written as

$$V(r, y, \theta) = E(r, y, \theta) - E(\infty, y, \theta), \qquad (28)$$

where E is the energy of the H_3^+ system. It was obtained by the polynomial expansion given by Conroy as the best fit to the H_3^+ system.

Since we are interested in the transition $0 \rightarrow 1$, we must be concerned with the first derivative of $V(r, y, \theta)$ with respect to y, that is $V_1(r, \theta)$ in Eq. (3).

This derivative should be taken at the value of y which makes E minimum, if we assume that for each position of the projectile the atoms rearrange themselves to get a configuration corresponding to a minimum energy of H_3^+. However this may not be the case, and the equilibrium position of the molecule may be different due to the short time used by the projectile in crossing the interaction area. This will produce a variation in the effective spring constant of the molecule. For this reason we have also considered the extreme possibility where the H_2 nuclei do not change their equilibrium position, as the projectile gets closer. Thus we have built two potentials by taking the derivative of $V(r, y, \theta)$ first at $y = y_{\min}$, where E is minimum, and then at $y = y_0 = 1.4$ a.u. In the last case, the spring constant is assumed to be unaffected by the interaction.

Figure 3 gives the potential matrix element V_{01} [see Eq. (14a)] deduced from the work of Conroy for $y = y_{\min}$ corresponding to the distance between the nuclei of H_2 where the energy of the H_3^+ sys-

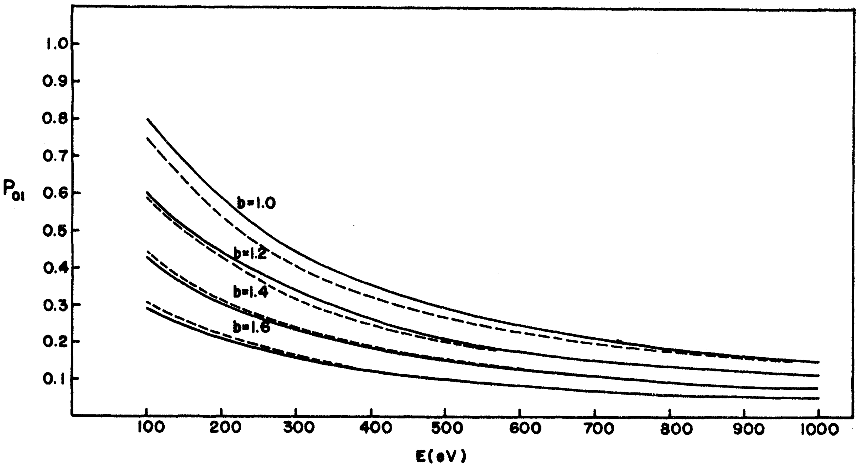

FIG. 4. Probabilities P_{01} corresponding to the potentials given in Fig. 3. The dashed curves correspond to $y=y_0$ and the solid curves to $y=y_{\min}$.

tem has a minimum, and the potential corresponding to $y_0=1.4$ a.u., which is the equilibrium vibrational distance for the free H_2 molecule. The corresponding predictions are shown in Figs. 4 and 5.

C. The new potential

In the potentials used above, the time pulse $V_1(t)$, is obtained by direct substitution of Eq. (5) into the potential form wherever the distance r appears. This pulse depends parametrically on b, and the shape of potential pulse is strictly related to the dependence on r. That is, in the calculations above the static character of the potential was preserved by the transformation from r to the time t.

We now proceed along the following lines. We assume that the potential should retain its static character for long distances r because of the weak coupling between the potential surfaces at long distances, and we further assume that at long distances the projectile motion is separable from the motion of the molecular nuclei. Because a complete quantum-mechanical solution to the dynamic problem of the H^+-H_2 system does not exist, it is impossible to determine *a priori* the region where short-range effects begin to play an important role in the collision. The short-range effects in this problem may be many in number; however, those primarly responsible for an alteration of the static character of the potential are the two effects already mentioned above.

In order to include the short-range effects in the potential, we must find the proper way to alter the static potential at short distances. Indeed, we have found that by adjusting the parameters

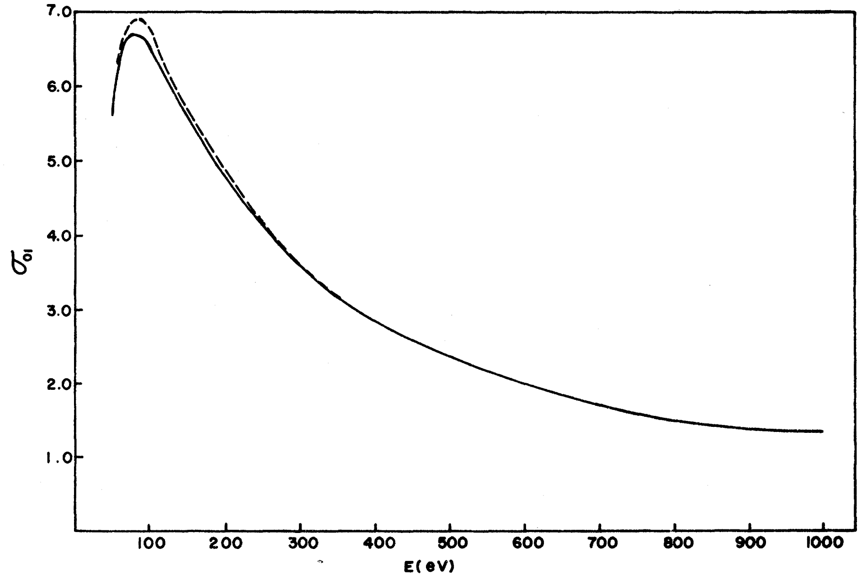

FIG. 5. Vibrational cross section σ_{01} in $Å^2$ corresponding to the potentials given in Fig. 3. The dashed curve is for $y=y_0$ and the solid curve is for $y=y_{\min}$.

TABLE I. Parameters in table are defined in Eq. (16). β is dimensionless, the impact parameter b is given in Å, and the amplitude A_{01} in eV/a.u.

b	β	A_{01}
0.6	0.02	0.40
1.0	0.06	0.78
1.4	0.13	0.50
1.8	0.13	0.36
2.0	0.20	0.38
2.4	0.20	0.29
2.8	0.20	0.25

$\beta(b)$ and $A_{01}(b)$ in the Gaussian pulse, in such a way that r and b behave as variables which are completely independent from each other, we can explain the observed cross section without the necessity of an unduly large collision distance.

The Gaussian-pulse representation of the potential is particularly convenient for carrying out the procedure of adjusting the potential at short distances. Several attempts soon show that the values given in Table I for β and A_{01} are the "best" values with regard to reproducing the broad high-energy maximum observed in the energy dependence of the cross section. The potential surface of Conroy was taken as the potential at large distances and as the starting point for the adjustment procedure at short distances.

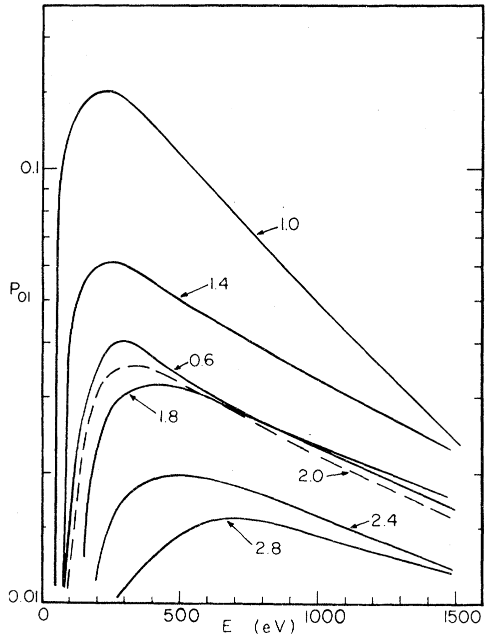

FIG. 6. Probability for the transition $\nu' = 0 \rightarrow \nu' = 1$ as a function of the energy for the potential proposed in this work. The number next to each probability curve gives the value of the impact parameter in Å.

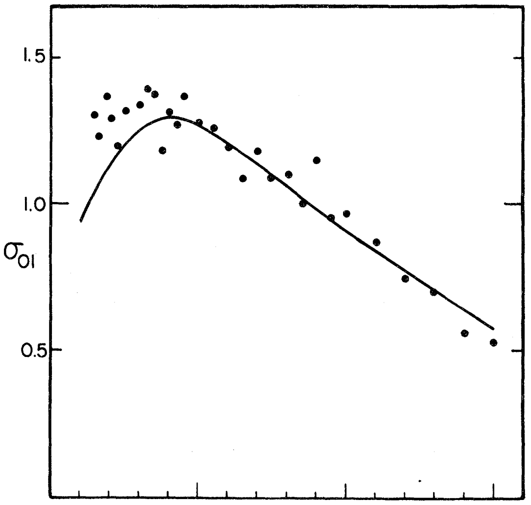

FIG. 7. Vibrational excitation cross section σ_{01} in Å^2 for the transition $0 \rightarrow 1$ as a function of the laboratory energy of H$^+$. Closed circles, experimental results of Ref. 1; smooth curve, predicted cross section corresponding to the potential given in Table I.

The values obtained were found to be somewhat critical in the sense that a small deviation from these led to an appreciable change in the cross-section shape.

IV. RESULTS AND CONCLUSIONS

From Figs. 2 and 5–7 we see that the only potential which accounts for the broad maximum of the cross section is the one given in Table I. The others will not even account for the negative curvature of the experimental points. This is an indication of the importance of the short-range part of the potential around 1 Å, where the contribution to the probability P_{01} is also maximum. At these distances the sensibility of the cross section to the potential has been found critical.

The set of parameters given in Table I were found quite unique for fitting the experimental data. This sort of uniqueness gives a sense of reliability for our approach.

As it is shown in Table I the parameter $\beta(b)$ is fairly constant for large impact parameters while the amplitude $A_{01}(b)$ varies significantly throughout the entire range of b. This suggests that we are not working on the same potential surface. In fact, the potential $V_1(r)$ equivalent to these pulses is not unique in this region. Thus, the possibility of electron exchange and correlation of different potential surfaces is supported by this work. For larger impact parameters however, A_{01}, inferred from the Gaussian pulse, goes to zero following a unique envelope which agrees pretty well with $V_1(r)$, deduced from the work of Conroy. As it was expected, in this region there is only one potential surface corresponding to the system ($H_2 + H^+$).

This work suggests the possibility of studying the coexistence of different potential surfaces by the type of experiments analyzed here, which reveal effective contributions from the short-distance part of the interaction. In addition, the fact that deviations from the static potential become important already at an impact parameter of 2 Å, indicates that the coupling of the motions of the three nuclei is already significant at these distances.

An indication of the importance of the short-range region is given by Fig. 6, where the transition probability reaches a maximum for impact parameter $b \simeq 1$ Å and an energy E of the order 200 eV.

A further fitting of the 0-2, 0-3, etc. vibrational excitation cross sections may give us some more information on the potential. However, since in these cases we would be concerned with the second derivative of the potential with respect to y we would also find similar uncertainties at short distances. The conclusions may not fold in with the others, and we will just have two uncoupled sets of information. The uncoupling could be removed by removing the uncertainties of $V(r, y, \theta)$ at short distances, and this is, at the moment, quite a difficult task.

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