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Impact-Parameter Calculation of Hydrogen-Hydrogen Double-Excitation Collisions*

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Cross sections for the double-excitation collisions $H(1s)+H(1s) \rightarrow H(2lm)+H(2l'm')$ are calculated over the range of incident energy 6.25 keV to 3.0 MeV by means of the impact-parameter method. The first Born, distorted Born, and two-state approximations are employed. At low-incident velocities, the inclusion of distortion terms decreases all cross sections below the first Born values. At high-incident velocities, the distorted Born cross sections for excitation of the projectile atom to the $2p_0$ state exceed the Born values, while all other distorted Born cross sections approach the Born values from below. Back coupling from the final state to the initial state has negligible effect on the double-excitation cross sections. The polarization of the radiation emitted from the excited $H(2p)$ projectiles is calculated for the double-excitation collisions and found to dominate the polarization for incident energies exceeding 25 keV.

I. INTRODUCTION

Direct inelastic atom-atom collisions are of great importance to the study of excitation and ionization in meteor trails, auroras, and other atmospheric phenomena.

Over a wide range of incident energies, the scattering solution is well represented by a product of atomic wave functions, with no electron exchange between them, combined with a suitable description of the heavy-particle motion. In the slow-collision or adiabatic region, the representation of the scattering function and the determination of the electronic coupling terms are much more difficult.[1-3]

At high energies, the inelastic cross section can be calculated by using either the first Born wave approximation or the first Born impact-parameter approximation assuming a straight-line trajectory. At high energies, the cross sections are equivalent though not identical.[4]

To improve the calculation for intermediate velocities, one can progress to higher-order Born wave approximations[5] or include distortion, coupling to the ground state, and coupling to other excited states in the impact-parameter treatment. The use of a classical rather than a straight-line trajectory is a significant improvement in atom-atom collisions only at very low velocities.[6]

Recently, Flannery and Levy[7] derived analytic forms for the matrix elements needed in an impact-parameter treatment of H-H collisions and initiated a series of cross-section calculations. Flannery[8,9] has since completed impact-parameter calculations of single-excitation cross sections that consider distortion, back coupling to the initial state, and couplings between degenerate excited states.

This paper will consider the effects of distortion and back coupling on a number of double-excitation (excitation of projectile and target) cross sections that make important contributions to the total projectile excitation and to the polarization of the radiation emitted by the excited projectile. Electron and nuclear exchanges, both of which are necessary for antisymmetrization of the electronic wave function, are not included in this treatment.

II. THEORY

In the application of the impact-parameter method to the collision of two atoms, one assumes that the projectile atom travels with a constant velocity v in a straight line at a constant distance ρ, the impact parameter, from the Z axis of a fixed cylindrical coordinate system with the target atom at the origin. The electronic part of the scattering solution is described by a time-dependent wave function

$$\Psi_n(\vec{r}_1, \vec{r}_2, t) = \sum_m a_{n,m}(t)\psi_m(\vec{r}_1, \vec{r}_2)\, e^{iE_m t/\hbar} , \tag{1}$$

where ψ_m is a product of the hydrogenic wave function $\Phi_{nlm}(\vec{r}_1)$ with energy ϵ_n and the hydrogenic wave function $\Phi_{n'l'm'}(\vec{r}_2)$ with energy $\epsilon_{n'}$, and $E_m = \epsilon_n + \epsilon_{n'}$.

With the use of the electrostatic interaction between the two hydrogen atoms,

$$V[\vec{r}_1, \vec{r}_2, \vec{R}(t)] = [R(t)]^{-1} + [|\vec{R}(t) + \vec{r}_1 - \vec{r}_2|]^{-1} - [|\vec{R}(t) - \vec{r}_2|]^{-1} - [|\vec{R}(t) + \vec{r}_1|]^{-1} \tag{2}$$

as the time-dependent perturbation, an infinite set of coupled differential equations is generated. Truncating the summation to include only the initial and final states (two-state approximation) gives the equations

$$i\hbar \frac{\partial}{\partial t} a_n(t) = \sum_{m=1}^{2} a_m(t) V_{n,m}[\vec{R}(t)] \exp\left[\frac{it(E_n - E_m)}{\hbar}\right], \quad n = 1, 2, \tag{3}$$

$$\text{with} \quad V_{n,m}[\vec{R}(t)] = \int d\vec{r}_1 \int d\vec{r}_2 \{\Phi^*_{n_1}(\vec{r}_1)\Phi^*_{n_2}(\vec{r}_2) V[\vec{r}_1, \vec{r}_2, \vec{R}(t)] \Phi_{m_1}(\vec{r}_1)\Phi_{m_2}(\vec{r}_2)\} . \tag{4}$$

When Eq. (3) is solved exactly for the probability of a transition from initial state 1 to final state 2, $P_{12}(\rho, \phi) = |a(+\infty)|^2$, for a given ρ and ϕ, the full two-state excitation cross section is given by

$$Q(1,2) = \int_0^\infty \rho\, d\rho \int_0^{2\pi} d\phi\, P_{12}(\rho, \phi) . \tag{5}$$

If the back coupling V_{12} is ignored, one can easily derive the distorted Born impact-parameter cross section[5]

$$\sigma_{DB}(1,2) = \int_0^\infty \rho\, d\rho \int_0^{2\pi} d\phi \left| \int_{-\infty}^{+\infty} V_{21}[\vec{R}(t)] \exp[i\alpha(t)/\hbar]\, dt \right|^2 , \tag{6}$$

$$\text{with} \qquad \alpha(t) = \int_0^t \{E_2 + V_{22}[\vec{R}(\tau)] - E_1 - V_{11}[\vec{R}(\tau)]\} d\tau \ ;$$

ignoring V_{11}, V_{22}, and V_{12} leads to the Born impact-parameter cross section[5]

$$\sigma_B(1,2) = \int_0^\infty \rho\, d\rho \int_0^{2\pi} d\phi \left| \int_{-\infty}^{+\infty} V_{21}[\vec{R}(t)] \exp[it(E_2 - E_1)/\hbar]\, dt \right|^2 . \tag{7}$$

The coupling-matrix elements

$$V_{n,m}[\vec{R}(t)] = V^{1s1s}_{2lm2l'm'}[\vec{R}(t)] = \sum_L C_L V_L[\vec{R}(t)]\, D(ll'L, mm'M) Y_{LM}(\hat{R}) , \tag{8}$$

are given in Table II of an earlier paper.[7]

The necessary distortion matrix elements, evaluated by means of techniques previously discussed,[7,10] are given below

$$V^{1s1s}_{1s1s}(\vec{R}) = e^{-2R}(-4R^2 - 18R + 15 + 24R)^{-1}/24 ,$$

$$V^{2s2s}_{2s2s}(\vec{R}) = e^{-R}(7!\,2^5 R^{-1} + 104\,895 + 24\,255R + 25\,305R^2 - 2940R^3 - 420R^4 - 21R^5 - 9R^6)/7!\,2^5 , \tag{9a}$$

$$V^{2s2p_m}_{2s2p_m}(\vec{R}) = e^{-R}(7!\,2^7 R^{-1} + 427\,140 + 104\,580R + 51\,660R^2 - 10\,080R^3 - 1344R^4 - 140R^5 - 12R^6)/7!\,2^7$$
$$+ C_{0,m} Y_{20}(\hat{R})(\pi/5)^{\frac{1}{2}} e^{-R}(-1302R^2 - 1302R^3 - 177R^4 - 23R^5 - 3R^6)/7!\,2^3 , \tag{9b}$$

and

$$V^{2p_m 2p_{m'}}_{2p_m 2p_{m'}}(\vec{R}) = e^{-R}(7!\,2^5 R^{-1} + 109\,935 + 29\,295R + 945R^2 - 2\,104R^3 - 630R^4 - 21R^5 - R^6)/7!\,2^5$$
$$+ C^1_{m,m'} e^{-R}(2835 + 2835R + 1155R^2 + 210R^3 - 7R^5 - R^6)/5\times 7!\,2^3$$
$$+ 16(\pi/5)^{\frac{1}{2}} Y_{20}(\hat{R})[C^2_{m,m'} e^{-R}(-1302R^2 - 1302R^3 - 171R^4 - 17R^5 - R^6)/7!\,2^6$$
$$+ C^1_{m,m'} e^{-R}(-105R^2 - 105R^3 - 45R^4 - 10R^5 - R^6)/7\times 7!\,2^5$$
$$+ C^1_{m,m'} \pi^{\frac{1}{2}} 192 Y_{40}(\hat{R})[105R^{-5} + e^{-R}(-105\times 7!\,2^7 R^{-5} - 105\times 7!\,2^7 R^{-4} - 105\times 7!\,2^6 R^{-3}$$

$$-35\times 7!\,2^{6}R^{-2}-35\times 7!\,2^{4}R^{-1}-564\,480-94\,080R-13\,440R^{2}-1680R^{3}-185R^{4}$$
$$-17R^{5}-R^{6})/7!\,2^{7}]/35\;, \qquad (9c)$$

where $C_{0,0}=C_{0,0}{}^{1}=C_{0,0}{}^{2}=1$, $C_{0,\pm 1}=C_{0,\pm 1}{}^{1}=C_{\pm 1,\pm 1}{}^{2}=C_{\pm 1,\mp 1}{}^{2}=-\frac{1}{2}$, and $C_{\pm 1,\pm 1}{}^{1}=C_{\pm 1,\mp 1}{}^{1}=C_{0,\pm 1}{}^{2}=\frac{1}{4}$.

III. RESULTS AND DISCUSSION

The Born and two-state impact-parameter cross sections are calculated for the collisions

$$\mathrm{H}(1s)+\mathrm{H}(1s)\rightarrow \mathrm{H}(2s,2p_0,2p_\pm)+\mathrm{H}(2s,2p_0,2p_\pm)$$

over an incident energy range 6.25 keV $\leq E \leq$ 3.0 MeV. The cross sections for total $2p$ excitation of the projectile and the contributions from individual m sublevels are presented in Tables I and II, for which they have been summed over the final-state m sublevels of the target.

Within the accuracy of the computation (error $<1.0\%$), the distorted Born and the two-state cross sections agree so that back coupling has a negligible effect on H-H double-excitation cross sections.

For double-excitation collisions, the interaction potentials vary from a short-range exponential dependence for $V_{2s2s}^{1s1s}(\vec{R})$ to a long-range R^{-3} dependence for

$$V_{2p_m 2p_m}^{1s1s}(\vec{R})\;.$$

Correspondingly, the calculations show that the percentage contributions to the total cross sections from $\rho<2.0$ decrease in the order $2s2s > 2p_m 2s > 2p_m 2p$, and Tables I and II show that the effects of distortion at a given velocity decrease in the same order. Distortion decreases all cross sections at low velocities and shifts all maxima to higher velocities. For velocities beyond the maxima, $Q(2s2s)$ and $Q(2p_\pm 2l)$ approach the Born values from below, while $Q(2p_0 2l)$ approaches them from above. The effects of distortion for velocities beyond the maxima are much greater for $Q(2s2s)$ than for $Q(2p2l)$ because of cancellation between the m sublevels. The results of distortion are quite similar to those found for single-excitation collisions by Bates[11] with H^+-H, and Flannery[8] with H-H.

Cross sections are also calculated on the assumption that the quantization axis of the atoms was $\vec{R}(t)$–the rotating axis approximation – but even at $v=0.5$ a.u., there is serious disagreement with the cross sections calculated from the exact interaction potential.

Vainshtein, Presnyakov, and Sobel'man[12] have suggested that an approximation that replaces

TABLE I. Cross sections for $\mathrm{H}(1s)+\mathrm{H}(1s)\rightarrow \mathrm{H}(2s,2p_0,2p_\pm)+\mathrm{H}(2s)$.

v (a.u.)	$Q(2s2s)$[a]	$\sigma(2s2s)$[b]	$Q(2p2s)$[a]	$\sigma(2p2s)$[b]	$Q(2p_0 2s)$[c]	$\sigma(2p_0 2s)$[d]	$Q(2p_\pm 2s)$[c]	$\sigma(2p_\pm 2s)$[d]
0.5	8.57^{-5}[e]	2.29^{-3}	2.58^{-4}	1.85^{-3}	2.28^{-4}	1.54^{-3}	1.52^{-5}	1.54^{-4}
0.7	5.09^{-4}	8.04^{-3}	1.51^{-3}	1.15^{-2}	1.04^{-3}	8.83^{-3}	2.36^{-4}	1.31^{-3}
1.0	2.96^{-3}	1.08^{-2}	1.17^{-2}	2.55^{-2}	7.99^{-3}	1.70^{-2}	1.87^{-3}	4.25^{-3}
1.25	4.36^{-3}	9.16^{-3}	1.93^{-2}	2.73^{-2}	1.24^{-2}	1.58^{-2}	3.46^{-3}	5.74^{-3}
1.50	4.46^{-3}	7.11^{-3}	2.10^{-2}	2.46^{-2}	1.23^{-2}	1.23^{-2}	4.36^{-3}	6.13^{-3}
1.75	4.00^{-3}	5.48^{-3}	1.94^{-2}	2.07^{-2}	1.02^{-2}	8.90^{-3}	4.60^{-3}	5.90^{-3}
2.0	3.43^{-3}	4.29^{-3}	1.69^{-2}	1.72^{-2}	7.92^{-3}	6.35^{-3}	4.47^{-3}	5.40^{-3}
2.5	2.45^{-3}	2.79^{-3}	1.21^{-2}	1.19^{-2}	4.52^{-3}	3.29^{-3}	3.80^{-3}	4.28^{-3}
3.0	1.79^{-3}	1.95^{-3}	8.77^{-3}	8.49^{-3}	2.61^{-3}	1.81^{-3}	3.08^{-3}	3.34^{-3}
5.0	6.85^{-4}	7.03^{-4}	3.22^{-3}	3.14^{-3}	4.39^{-4}	2.84^{-4}	1.39^{-3}	1.43^{-3}
6.0	4.80^{-4}	4.88^{-4}	2.22^{-3}	2.20^{-3}	2.21^{-4}	1.41^{-4}	1.00^{-3}	1.03^{-3}
9.0	2.15^{-4}	2.17^{-4}	9.86^{-4}	9.75^{-4}	4.63^{-5}	2.91^{-5}	4.70^{-4}	4.73^{-4}
11.0	1.45^{-4}	1.45^{-4}	6.57^{-4}	6.53^{-4}	2.11^{-5}	1.32^{-5}	3.18^{-4}	3.20^{-4}

[a] Two-state cross sections in units of a_0^2 for the excitation of a particular l level summed over all m sublevels of the projectile and target.

[b] Born cross section in units of a_0^2 for the excitation of a particular l level summed over all m sublevels of the projectile and target.

[c] Two-state cross section in units of a_0^2 for the excitation of a particular m sublevel of the projectile summed over all m sublevels of the target.

[d] Born cross section in units of a_0^2 for the excitation of a particular m sublevel of the projectile summed over all m sublevels of the target.

[e] The exponents give the power of 10 by which each entry must be multiplied.

TABLE II. Cross sections for $H(1s)+H(1s)\rightarrow H(2p_m)+H(2p_{m'})$.

v (a.u.)	$Q(2p2p)$ [a]	$\sigma(2p2p)$ [b]	$Q(2p_0 2p)$ [c]	$\sigma(2p_0 2p)$ [d]	$Q(2p_\pm 2p)$ [c]	$\sigma(2p_\pm 2p)$ [d]
0.5	9.56^{-4} [e]	2.29^{-3}	8.42^{-4}	1.63^{-3}	5.68^{-5}	3.30^{-4}
0.7	4.99^{-3}	1.77^{-2}	3.11^{-3}	1.42^{-2}	9.43^{-4}	1.74^{-3}
1.0	4.27^{-2}	6.81^{-2}	2.91^{-2}	4.95^{-2}	6.80^{-3}	9.29^{-3}
1.25	8.02^{-2}	9.76^{-2}	5.42^{-2}	6.44^{-2}	1.30^{-2}	1.66^{-2}
1.50	9.76^{-2}	1.11^{-1}	6.24^{-2}	6.70^{-2}	1.76^{-2}	2.18^{-2}
1.75	1.00^{-1}	1.09^{-1}	5.94^{-2}	5.98^{-2}	2.03^{-2}	2.44^{-2}
2.00	9.45^{-2}	1.02^{-1}	5.15^{-2}	5.11^{-2}	2.15^{-2}	2.52^{-2}
2.50	7.77^{-2}	8.23^{-2}	3.57^{-2}	3.47^{-2}	2.10^{-2}	2.38^{-2}
3.00	6.17^{-2}	6.50^{-2}	2.39^{-2}	2.34^{-2}	1.89^{-2}	2.08^{-2}
5.00	2.74^{-2}	2.83^{-2}	5.97^{-3}	5.90^{-3}	1.07^{-2}	1.12^{-2}
6.00	1.98^{-2}	2.03^{-2}	3.42^{-3}	3.38^{-3}	8.19^{-3}	8.45^{-3}
9.00	9.18^{-3}	9.30^{-3}	8.99^{-4}	8.96^{-4}	4.14^{-3}	4.20^{-3}
11.00	6.23^{-3}	6.26^{-3}	4.47^{-4}	4.44^{-4}	2.89^{-3}	2.91^{-3}

[a] Two-state cross section in units of a_0^2 summed over all m sublevels of projectile and target.

[b] Born cross section in units of a_0^2 summed over all m sublevels of projectile and target.

[c] Two-state cross section in units of a_0^2 for a particular m sublevel of the projectile summed over all m sublevels of the target.

[d] Born cross section in units of a_0^2 for a particular m sublevel of the projectile summed over all m sublevels of the target.

[e] The exponents give the power of 10 by which each entry must be multiplied.

$\alpha(t)$ in Eq. (6) by

$$\{\alpha(t)^2+4V_{12}[\vec{R}(t)]^2\}^{1/2} ,$$

is an improvement on the conventional distorted Born. However, since Eq. (6) and the full two-state approximation give very similar results, the suggested modification[12] is in this instance no better than the distorted Born approximation.

The polarizations of emitted radiation are calculated by means of the formula of Percival and Seaton[13]:

$$P(2p)=300\left[\frac{Q(2p_0,\Sigma)-Q(2p_\pm,\Sigma)}{7Q(2p_0,\Sigma)+11Q(2p_\pm,\Sigma)}\right] , \quad (10)$$

where Σ implies summation of the cross section over all final states of the target. The results, which include the excitation of the target to $2s$ and $2p_m$ states, are compared in Table III with those calculated by Flannery,[9] which assume that the target is not excited. The addition of the double-excitation cross sections causes $P(2p)$ to remain small but positive for 25 keV $\leq E \leq$ 100 keV, because

$$\sum_{l,m} Q(2p_0,2lm) > \sum_{l,m} Q(2p_+,2lm)$$

sufficiently to compensate for $Q(2p_0,1s)$ being less than $Q(2p_+,1s)$. This behavior is in harmony with the experimental results of Dose, Gunz, and Meyer[14] for He, Ne, and Ar targets. It appears that the high-energy ($E \geq 25$ keV) behavior of $P(2p)$ is controlled by double-excitation collisions, as has been suggested by Dose.[15]

TABLE III. Polarization of radiation emitted by the excited projectile $H(2p_m)$, with and without excitation of the target.

v (a.u.)	P_{single} [a]	P_{total} [b]
0.2	+34.5	+34.5
0.3	+28.2	+28.2
0.4	+24.4	+24.4
0.5	+20.1	+20.1
0.6	+15.2	+15.2
0.8	+6.6	+6.6
1.0	−0.4	+4.9
1.2	−5.8	+5.6
1.4	−10.0	+6.8
1.6	−13.1	+6.7
1.8	−15.6	+5.8
2.0	−17.4	+4.4

[a] Percentage polarization determined by Flannery (see Ref. 9) considering single excitations only.

[b] Percentage polarization including double-excitation collisions that raise the target atom $H(1s)$ to $H(2s$ or $2p_m)$.

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