

H + D₂ Reaction Dynamics in the Limit of Low Product Recoil Energy

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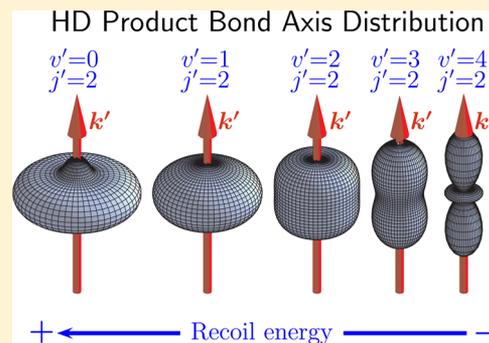
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ABSTRACT: Both experiment and theory recently showed that the H + D₂($v = 0, j = 0$) → HD($v' = 4, j'$) + D reactions at a collision energy of 1.97 eV display a seemingly anomalous HD product angular distribution that moves in the backward direction as the value of j' increases and the corresponding energy available for product recoil decreases. This behavior was attributed to the presence of a centrifugal barrier along the reaction path. Here, we show, using fully quantum mechanical calculations, that for low recoil energies, the collision mechanism is nearly independent of the HD internal state and the HD product becomes aligned, with its rotational angular momentum j' pointing perpendicular to the recoil momentum k' . As the kinetic energy to overcome this barrier becomes limited, the three atoms adopt a nearly collinear configuration in the transition-state region to permit reaction, which strongly polarizes the resulting HD product. These results are expected to be general for any chemical reaction in the low recoil energy limit.

SECTION: Kinetics and Dynamics



The development of reaction dynamics^{1,2} and the understanding of the mechanism of H + H₂ elementary collisions³ have unfolded in parallel and can be studied as a single unit. This does not mean, however, that our knowledge about the H₃ system is complete. Quite the contrary, its study continues rendering unexpected results^{4,5} whose applicability extends to other elementary collision processes. The interest in the H + H₂ reaction is exemplified by the recent work of Jankunas et al.,⁶ where the analysis of the theoretical and experimental differential cross sections (DCSs) for the H + D₂($v = 0, j \leq 2$) → HD($v' = 4, j'$) + D state-to-state reactions at a collision energy $E_{\text{coll}} = 1.97$ eV leads to conclusions that seemed to contradict common knowledge in reaction dynamics. In particular, it was found that the angular distributions shifted into the backward region as the rotational excitation of the HD products increased, apparently defying the well-established rule that correlates head-on (glancing) direct collisions, characterized by small (large) values of the impact parameter, backward (sideways) scattering, and low (high) rotational excitation of the products. Such apparently anomalous behavior was explained in terms of a centrifugal barrier, located in the reaction path, whose influence becomes more significant as the recoil energy of the products tends to zero, which is always the case for large enough values of j' . As a consequence of the presence of the barrier, the small and medium values of the total angular momentum, J , dominate the collisions, leading to formation of rotationally excited HD($v' = 4, j'$) molecules, while the larger J values, responsible for the forward and sideways scattering, become ineffective when it comes to forming such molecules in those quantum states. This same effect is expected to be observed for any other direct

bimolecular reaction where most of the energy available for the reaction accumulates in the products' internal degrees of freedom.

Even for the simplest collision processes, there are four vectorial quantities whose relative arrangement must be accounted for if one wishes to understand how the collisions take place. These vectors are k and k' , the approach and the recoil directions, and the rotational angular momenta, j and j' , of the reactant and product molecules, respectively. Among all of the possible vectorial correlations⁷ involving these properties, the differential cross section (DCS) is the simplest one as it just considers the connection between k and k' .

The aim of the present work is to use quantum mechanical calculations to ascertain the mechanism of the state-to-state



collisions at $E_{\text{coll}} = 1.97$ eV when the recoil energy of the products becomes small, which are the same conditions under which the experiments were carried out by Jankunas et al.⁶ To this end, we consider the $k-k'-j'$ correlation that describes the polarization of the products and makes possible the determination of the spatial distribution adopted by j' and r' , the HD internuclear axis direction.

The first step when considering directional effects is to define a suitable frame of reference. In the present work, all of the directional information will be referred to that frame where the

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z axis is parallel to k' , the x - z plane is the scattering plane (i.e., the plane defined by k and k'), and the y axis is parallel to the cross product $k \times k'$. The angle formed by the vectors k and k' is the scattering angle, θ , which has the value of 0 to 180° for forward to backward scattering, respectively.

In a quantum mechanical context, the k - k' - j' correlation is expressed in terms of a set of numbers called polarization moments⁸⁻¹⁰ that contain information about the alignment or orientation of j' with respect to certain spatial directions. Such information may refer to collisions leading to formation of products at a well-defined value of the scattering angle θ , in which case, the moments are called polarization-dependent differential cross sections^{11,12} (PDDCSs), or, alternatively, may be averaged by integrating over the angle θ , in which case, the moments are termed polarization parameters^{9,11} (PPs). Among the whole set of moments, we focus on the (2,0) one, which measures the alignment of j' with respect to the product's recoil direction k' . In the aforementioned frame of reference, positive (negative) values of the (2,0) moment correspond to an alignment of the rotational angular momentum j' parallel (perpendicular) to k' . The converse applies if, instead of j' , one considers the arrangement adopted by the HD internuclear axis vector r' . Notice that the frame that we are using, with the z axis along the k' vector, differs from the usual one in which k is the vector that defines that axis, in which case, the (2,0) moment measures the alignment along the initial relative velocity.^{9,12}

The determination of the quantum mechanical values of the PPs and PDDCSs implies that the scattering matrix¹³ corresponding to the process under study has been previously calculated. The elements of this matrix, which are complex numbers, contain all of the information concerning the collisions, including that necessary to evaluate the PDDCSs and PPs, and are labeled as $S_{\nu j \Omega, \nu' j' \Omega'}^{J E}$, where E is the total energy and J indicates the total angular momentum, which coincides with the orbital angular momentum of the reactants, l , for rotationless collision partners. Calculations have been carried out using the time-independent quantum scattering hyperspherical coordinate method by Skouteris et al.¹³ with the potential energy surface (PES) by Boothroyd et al.¹⁴ at $E_{\text{coll}} = 1.97$ eV for the $\text{H} + \text{D}_2(\nu = 0, j = 0)$ reactive collisions. The S matrix was determined for every value of J until convergence was achieved ($J_{\text{max}} = 40$). The asymptotic state of reactants and products is given by $\nu j \Omega$ and $\nu' j' \Omega'$, respectively, with Ω (Ω') denoting the helicity of the reactants (products), that is, the projection of j (j') on the approach (recoil) direction of the reactants (products). The results obtained at energies slightly different from this one (as those spanned by the experiment in ref 6), are essentially the same. The following equations express the PDDCS(2,0) and PP(2,0) moments as a function of the scattering matrix elements for initial rotational state $j = 0$

$$\text{PDDCS}(2, 0) = \sum_{\Omega'} |\bar{f}_{\Omega', 0}(\theta)|^2 \langle j' \Omega' 2 0 | j' \Omega' \rangle \quad (2)$$

and

$$\text{PP}(2, 0) = \frac{2}{C} \sum_J \sum_{\Omega'} (2J + 1) |S_{\nu j \Omega, \nu' j' \Omega'}^{J E}|^2 \langle j' \Omega' 2 0 | j' \Omega' \rangle \quad (3)$$

where $\langle \dots \rangle$ indicate Clebsch–Gordan coefficients⁸ and the quantum numbers $\nu j \Omega$ that define the reagent state have been

substituted for by 0 as it corresponds to the process under study, eq 1. The scattering amplitude $\bar{f}_{\Omega', 0}(\theta)$ is given by

$$\bar{f}_{\Omega', 0}(\theta) = \frac{\sum_J (2J + 1) d_{\Omega', 0}^J(\theta) S_{\nu j \Omega, \nu' j' \Omega'}^{J E}}{C^{1/2}} \quad (4)$$

Here, $d_{\Omega', 0}^J(\theta)$ is a reduced rotation matrix element⁸, and C is a normalization factor

$$C = 2 \sum_{J \Omega'} (2J + 1) |S_{\nu j \Omega, \nu' j' \Omega'}^{J E}|^2 \quad (5)$$

Although the value of the PDDCS(2,0) depends on the coherences between the different partial waves, this is not the case for the polarization parameter PP(2,0), where the partial waves and the Ω' values contribute incoherently to the polarization moment. Please notice that eq 3 with the Clebsch–Gordan coefficient $\langle j' \Omega' 0 0 | j' \Omega' \rangle$ takes the value 1, which is the PP(0,0).¹⁰

Figure 1 presents the polarization parameters PP(2,0) for the different vibrational and rotational levels of the HD molecule.

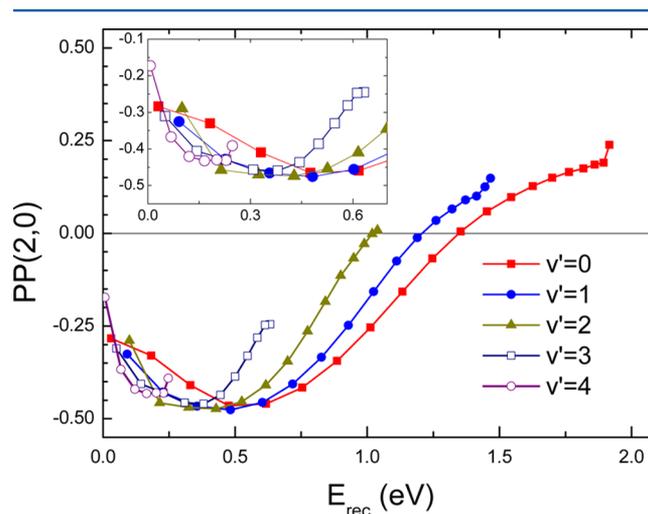


Figure 1. State-to-state PP(2,0) moment for the $\text{H} + \text{D}_2(\nu = 0, j = 0) \rightarrow \text{HD}(\nu', j') + \text{D}$ reactive collisions at $E_{\text{coll}} = 1.97$ eV as a function of the recoil energy, E_{rec} , of the products. For each vibrational progression, the point located to the right of the curve corresponds to $j' = 1$, and the rotational level increases in moving to the left. The inset shows an enlargement of the region of low recoil energies where the different vibrational progressions overlap. Note that these PPs refer to the alignment of j' along k' .

The moments are plotted as a function of the recoil energy, E_{rec} , of the products. Please notice that the initial point of each vibrational progression, located on the right extreme of the curve, corresponds to $j' = 1$ and not to $j' = 0$ as it is meaningless to define the PP(2,0) moment for rotationless products. Accordingly, as we move toward smaller recoil energies, the HD rotational level increases. The polarization parameters possess a well-defined range of values whose limits depend on j' .¹⁵ In principle, it would be necessary to refer the values of the moments to such limits when comparing results for different final rotational levels, as is the case here. However, such modification of the polarization parameters would barely change Figure 1 and will not be implemented in order to keep the discussion of the results as simple as possible.

The moments belonging to the $\nu' = 0$ manifold display a change from positive to negative values as the recoil energy

decreases. Such a change indicates that, while formation of products in low rotational states leads to alignments with some weak preference of j' parallel to the products' recoil direction, when the products become rotationally excited, j' tends to be strongly aligned perpendicular to k' . As ν' increases, the corresponding curves shift toward smaller values of E_{rec} while the positive values of the alignment gradually disappear ($\nu' \geq 2$). At low enough recoil energies, the polarization moments of the different plots converge around $\text{PP}(2,0) \simeq -0.5$. Finally, for even smaller values of E_{rec} the values of the polarization parameter start increasing and approach zero. This behavior will be explained later.

The confluence of the PPs experienced by the different vibrational manifolds at low recoil energies suggests that the reaction mechanism becomes independent of the internal state of the products under these conditions and that it is mainly determined by the same centrifugal barrier that justified the anomalous observations of Jankunas et al.⁶ The value of $\text{PP}(2,0)$ around which the convergence takes place ($\simeq -0.5$) supports this idea as it corresponds to a very strong polarization of j' perpendicular to k' , while r' and k' tend to be parallel. This means that, regardless of their internal state, in the exit channel, the three atoms adopt a nearly collinear arrangement (r' parallel to k') that resembles the geometry of the transition state and the minimum-energy path of the PES. This geometry makes it easier to surmount the barrier and to reach the HD product valley when the amount of kinetic energy available for such a process is small. The drift toward zero experienced by the (2,0) polarization moments as $E_{\text{rec}} \rightarrow 0$ may seem difficult to reconcile with this interpretation. However, its origin is easy to understand when one realizes that, in the limit of zero recoil energy, only those collisions characterized by a final orbital angular momentum quantum number (l') equal to zero will give rise to product formation and that the channel would be closed. This determines the value that can be adopted by the polarization parameter because, in the absence of contributions from $l' > 0$ collisions, it can be easily shown (and analytically proven¹⁶) that j' can have any direction with respect to k' so that the $\text{PP}(2,0)$ moment vanishes. It is important to make clear that this cancellation takes place progressively as the number of l' values relevant for the reaction decreases and that the consequences of approaching the $l' = 0$ regime begin to appear far before reaching it. These consequences include not only the drift toward zero experienced by the polarization parameters but also the reduction of J values that contribute to the reaction and the subsequent shift of the differential cross sections observed by Jankunas et al.⁶

Consideration of the stereodynamics as a function of the scattering angle confirms that at low recoil energies, the mechanism becomes nearly independent of the final state. This is exemplified in Figure 2, where the renormalized PDDCS-(2,0) moments, $\rho_0^{(2)}(\theta)$, for different final states corresponding to the same recoil energies ($\simeq 0.10$ eV, top panel, and $\simeq 0.20$ eV, bottom panel) are presented. Renormalization, which is achieved by dividing the polarization differential cross section by the angular distribution corresponding to the state-to-state process under consideration, renders the directional information contained in the PDDCSs independent of the amount of flux scattered in each direction, so that their values for different θ angles can be meaningfully compared. No line for $\nu' = 0$ appears in the upper panel as the possible rotational states are characterized by recoil energies too far from 0.1 eV. In good accordance with the discussion of the former paragraphs, and

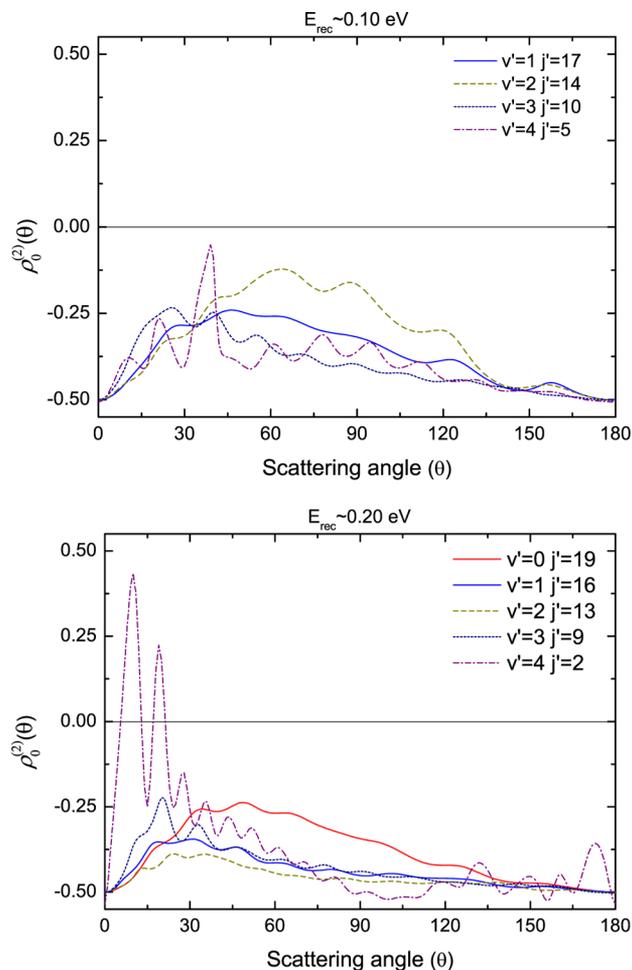


Figure 2. Renormalized PDDCS(2,0), $\rho_0^{(2)}(\theta)$, for those $\text{H} + \text{D}_2(\nu = 0, j = 0) \rightarrow \text{HD}(\nu', j') + \text{D}$ state-to-state processes at $E_{\text{coll}} = 1.97$ eV characterized by values of the recoil energy at around 0.10 (top panel) and 0.20 eV (bottom panel).

regardless of the final state, the profiles of the $\rho_0^{(2)}$ in Figure 2 are strikingly similar and correspond to a very significant alignment perpendicular to the recoil velocity, thus denoting a collinear configuration of the products in the exit valley. The only exception to this trend, which takes place for forward scattering into the $\text{HD}(\nu' = 4, j' = 2)$ state (bottom panel), is relatively unimportant as the amount of flux scattered in this angular region is small (see Figure 3). This picture would be basically unchanged if we had chosen any other recoil energy above 0.05 eV and compressed in the interval where the polarization parameters converge. Below this energy, however, the number of product partial waves starts decreasing, and the reaction becomes progressively dominated by the lowest l' values.

So far, our attention focused on the (2,0) moment that measures the alignment of j' with respect to k' . Although, in the present case, this turns out to be the most relevant piece of information when it comes to characterizing the mechanism of the title reaction, other polarization moments offer additional information¹⁰ about the alignment or orientation of j' with respect to certain spatial directions. A compact and simple visualization of the information contained in the complete set of moments is provided by stereodynamical r portraits.^{17,18} These are three-dimensional distributions that represent the spatial arrangement of r' , expressed through the polar (θ, ν) and

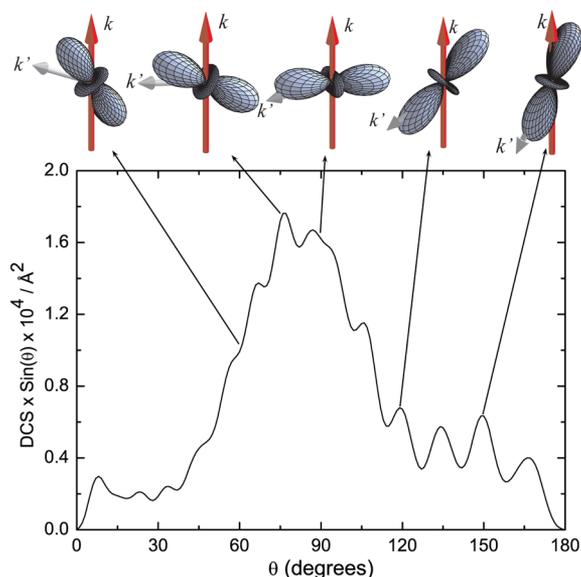


Figure 3. Differential cross section for the $\text{H} + \text{D}_2(v=0, j=0) \rightarrow \text{HD}(v'=4, j'=2) + \text{D}$ reactive collisions at $E_{\text{coll}} = 1.97$ eV. The portraits represent the internuclear axis distribution of the product molecule at selected values of the scattering angle. Note that irrespective of the scattering angle, the distribution of r' tends to lie along the recoil direction, k' .

azimuthal ($\phi_{r'}$) angles that define the position of r' in the frame of reference, corresponding to the whole set of moments ($\rho_q^{(k)}(\theta)$) that are being analyzed. Their expression is given by

$$P(\theta_{r'}, \phi_{r'} | \theta) = \sum_{k=0}^{2j'} \sum_{q=-k}^k \frac{2k+1}{4k} \rho_q^{(k)}(\theta) \langle j' 0 k 0 | j' 0 \rangle C_{kq}^*(\theta_{r'}, \phi_{r'}) \quad (6)$$

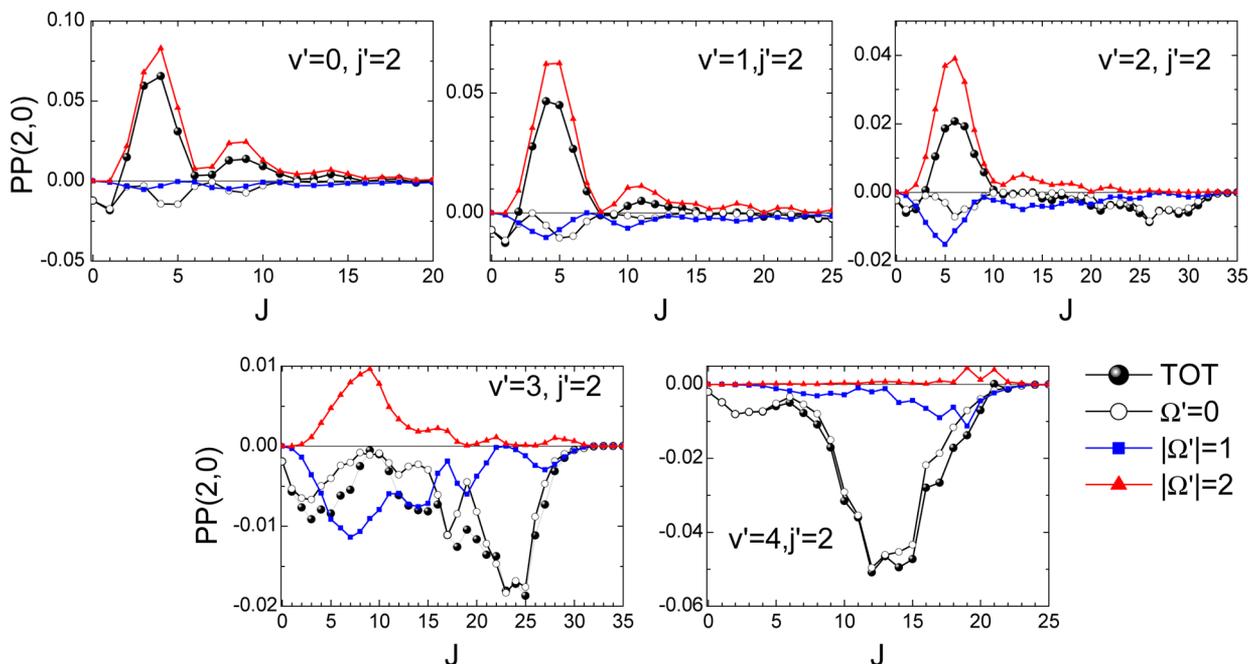


Figure 4. Decomposition of $\text{PP}(2,0)$ into $|\Omega'|$ and J contributions (see eq 3) for $\text{H} + \text{D}_2(v=0, j=0) \rightarrow \text{HD}(v'=0-4, j'=2) + \text{D}$ reactive collisions at $E_{\text{coll}} = 1.97$ eV.

Please notice that, because renormalized PDDCSs, $\rho_q^{(k)}(\theta)$, are used to generate the portraits, the shape and the information provided by these distributions will refer to reactive collisions at well-defined values of the scattering angle θ . It is worth pointing out that the stereodynamical portraits given by eq 6 are purely quantum mechanical distributions that do not resort to any approximation whatsoever. Their usefulness is exemplified in Figure 3, where the differential cross section for the $\text{H} + \text{D}_2(v=0, j=0) \rightarrow \text{HD}(v'=4, j'=2) + \text{D}$ reaction and the stereodynamical portraits for selected values of θ are presented. The tilting displayed by the r' distributions increases as the scattering angle moves from backward to forward so that, to a very large extent, the axis of the portrait follows k' , the recoil direction of the products. A similar result is obtained for the other internal states considered in Figure 2, which confirms the nearly collinear arrangement adopted by the three atoms in the exit valley when E_{rec} is small.

By studying collisions where the final rotational state is fixed, instead of considering a fixed recoil energy, we analyze how the reaction stereodynamics becomes determined as the kinetic energy of the products diminishes progressively for successive vibrational states. In view of the weak dependence on the scattering angle displayed by the $\text{PDDCS}(2,0)$, this goal can be easily achieved by means of its integrated form, that is, using the $\text{PP}(2,0)$ moment. In addition, this polarization parameter can be expressed as an incoherent sum of contributions from different partial waves and $|\Omega'|$ values. Figure 4 presents the decomposition of the J dependence of this moment into the contributions of the various $|\Omega'|$ for the $\text{H} + \text{D}_2(v=0, j=0) \rightarrow \text{HD}(v'=0-4, j'=2) + \text{D}$ reactions, that is, the representation of the different terms in eq 3 for the various J and Ω' values. For this set of processes, E_{rec} decreases as the vibrational excitation of the products increases. From the sign of the Clebsch–Gordan coefficients in eq 3, whose values for $j'=2$ are $\langle 2 0 2 0 | 2 0 \rangle = -0.534$, $\langle 2 1 2 0 | 2 1 \rangle = -0.267$ and $\langle 2 2 2 0 | 2 2 \rangle = +0.534$, $|\Omega'| = 2$ collisions add positively to the value of

the moment, and their $|\Omega'| = 0$ and 1 counterparts contribute negatively, in such a way that the sign and modulus of the overall PP(2,0) parameter is determined by the contributions of the different $|\Omega'|$ summed over all of the partial waves necessary to converge the calculations. For these rovibrational states, the number of J values necessary to ensure convergence increases steadily from $\nu' = 0$ to 3 but suddenly drops for $\nu' = 4$, where the centrifugal barrier hinders the reaction for collisions characterized by large values of J . When the vibrational excitation of the products increases, the relative importance of the different $|\Omega'|$ values change drastically. The $|\Omega'| = 2$ contribution, corresponding to an alignment of j' somewhat parallel to k' (PP(2,0) > 0), dominates the PP(2,0) moment when the products are formed in the $\nu' = 0, j' = 2$ state. As ν' increases and E_{rec} becomes smaller, the sign of the polarization parameter changes, and the $|\Omega'| = 2$ contribution becomes progressively less significant, while its role as the main source of alignment is replaced by $|\Omega'| = 0$ and, to a lesser extent, $|\Omega'| = 1$, in a clear attempt to compensate for the lack of kinetic energy by adopting geometries that facilitate the passage over the barrier.

In summary, we have used quantum mechanical calculations to show how the centrifugal barrier behind the anomalous differential cross sections measured by Jankunas et al.⁶ for the $\text{H} + \text{D}_2 \rightarrow \text{HD}(\nu' = 4, j') + \text{D}$ reactions does not only affect the directions in which the products are scattered but also the polarization of their rotational angular momentum. As E_{rec} decreases, the alignment of the HD internuclear axis with respect to the products' recoil direction becomes nearly independent of the final state, so that the products adopt an arrangement where the three atoms tend to lie along a straight line defined by k' . This configuration, which resembles the transition state of the PES, facilitates the system to overcome the barrier and to access the products' valley. Finally, we stress that the spatial effects described throughout this work are expected to be of interest for the $\text{H} + \text{H}_2$ collisions not only at $E_{\text{coll}} = 1.97$ eV but over a large range of collision energies. Moreover, the validity of the conclusions will extend to other elementary processes whenever E_{rec} becomes small as compared to the height of the reaction barrier.

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Notes

The authors declare no competing financial interest.

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