A bi-axial quantum state that controls molecular collisions like a double-slit interferometer

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**Supplementary Materials**

**Calculation of the scattering image from scattering matrix elements**

As discussed in the main text, the complex amplitudes of the outgoing partial waves used to construct the scattering images are calculated using the scattering matrix elements. Here, we briefly describe the procedure used to calculate these amplitudes. For a more in depth discussion of these calculations, please refer to our earlier publication on HD-He scattering, which goes into this procedure in great detail.1

As described in the main text, the measured state-resolved differential scattering cross section is given by:

 (S1)

where the scattering amplitude may be expressed as:

 (S2)

Here,  is the state-to-state reaction amplitude within a unit solid angle defined by in the center-of-mass frame. In the remainder of the text, we will only show the *j*, *m* quantum numbers for simplicity.

In the scattering process, the input and outgoing states can be further expanded in terms of the partial waves with different orbital angular momentum. Following Arthurs and Dalgarno,2 for atom-diatom scattering, the reaction amplitude *q* can be expressed as,

 (S3)

Here,  is the scattering matrix element which gives the amplitude of the outgoing state with internal angular momentum quantum number *jf* and orbital angular momentum quantum number *lf* resulting from the scattering of the incoming state with angular momentum *j*, *l*. *J* gives the total angular momentum quantum number, which is conserved in a collision. represents the Clebsch-Gordan coefficient for the coupling of the three angular momentum vectors ***j*** (magnitude *j* and projection along the quantization axis *m­*), ***l***, and ***J***.  gives the spherical harmonic with angular momentum *lf* and projection on the Z axis *mlf*.

To calculate reaction amplitude *q*, we need to know the scattering matrix elements and participating incoming orbitals. Although this information is commonly obtained through theoretical calculations, in this work we have used the results of our previous HD-He collision experiments.1 As we detail in our earlier publication, because we conducted those experiments at relatively low collision energy (0-5 K), the only input partial waves that contribute to the collision process are those with angular momentum *l* ≤ 2. By fitting the experimental scattering angular distributions for the HSARP, VSARP, and XSARP orientations described in the main text, we concluded that only four scattering matrix elements contribute significantly to the scattering process. The complex values of these scattering matrix elements as determined from our fitting procedure are given in Table S1 below. For a comprehensive explanation of how these partial waves were determined, please refer to our earlier publication.1

Table S1. Scattering matrix elements for the four partial waves involved in the collision process as determined from our earlier experimental work.1

|  |  |
| --- | --- |
| Scattering matrix element | Fitted values |
|  | 1 |
|  | 0.39 *e*–0.92*i* |
|  | 0.31 *e*–0.06*i* |
|  | 0.14 *e*0.14*i* |

We note that in the experimentally determined scattering matrix elements shown in Table S1, we have found only incoming orbitals with *l* = 0, 2. Because of conservation of angular momentum, these input orbitals will only be able to produce the outgoing orbitals *lf* = 0, 2, 4. This dramatically reduces the number of terms involved the sum in Eq. (S3). The sum is additionally reduced due to the conservation of the Z component of the orbital angular momentum, denoted by quantum number *ml*and *mlf* in the input and output channels, respectively. Because we choose to analyze the collision process with the quantization axis along the relative velocity direction, *ml* is fixed at 0. Additionally, scattering into the (*v* = 1, *jf* = 0) fixes the projection of the internal angular momentum of the outgoing state *mf* = 0. Because the Z component of the angular momentum is also conserved, the Z component of the outgoing orbital angular momentum must be equal to the Z component of the incoming internal angular momentum: *mlf* = *m*. Therefore, the outgoing orbital states are limited to *lf* = 0, 2, 4, *mlf* = *m*.

In the analysis we present in this paper, the incoming and outgoing states internal states of the HD molecule are precisely defined. As detailed in the main text, we specifically consider three different HD molecular orientations, which we call HSARP, VSARP, and XSARP. These states are all prepared using the Stark-induced adiabatic Raman passage (SARP) process to select single or specified superpositions of orientational *m* states within the (*v* = 1, *j* = 2) energy eigenstate of the HD molecule:



 (S4)



Using resonance-enhanced multiphoton ionization (REMPI), the outgoing state (*v* = 1, *jf* = 0) is state-selectively detected as well. With the well-defined internal states in both the input and output channels further limit the number of terms involved in the sum Eq. (S3).

Writing out Eqs. (S1) and (S2) for the three specific input orientations:



 and (S5)



In Eq. (S5), we have dropped the *v* and *EC* indices because they are not changing during the collision process. Taking into account the well-defined input and output internal states, as well as the limited number of partial waves involved, we can now explicitly write out expressions for the three orientations in terms of the scattering matrix elements shown in Table S1:



 and (S6)



From these equations, the scattering angular distributions can be calculated as functions of the scattering angles (*θ*, *φ*).

To produce the images shown in the main text, we needed to express the scattering angle (*θ*, *φ*) in terms of the (*x*, *y*) coordinates in the image plane. A brief schematic representation of the geometry of the conversion is shown below in Fig. S1. Through energy conservation, the velocity of the HD molecules after scattering *u*HD can be precisely calculated. The time-of-flight *τ*TOF of is an experimentally measured quantity determined by the speed of the scattered particle as well as the geometry of the mass spectrometric detector. Therefore, for any particular experimental geometry, the total flight distance of the HD molecules *r*0 is known. To be more general, we have not explicitly defined the scale of the images shown in the main text, so they do not depend on the specific geometry of our apparatus. Using this defined quantity and the geometric relations shown in Fig. S1, the scattering angle (*θ*, *φ*) can be converted to (*x*, y) coordinates using the following two expressions:



 (S7)

Using these two equations to transform the coordinates allows us to use the expressions in Eq. (S6) to produce the scattering images shown in the main text.



Figure S1. A brief schematic for converting scattering angle (*θ*, *φ*) to the (*x*, *y*) coordinate on the image plane.

**References**

1. Perreault, W. E., Mukherjee, N. & Zare, R. N. HD (*v* = 1, *j* = 2, *m*) orientation controls HD-He rotationally inelastic scattering near 1 K. *J. Chem. Phys.* **150**, 174301 (2019).

2. Arthurs, A. M. & Dalgarno, A. The theory of scattering by a rigid rotator. *Proc. R. Soc. A* **256**, 540–551 (1960).