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Direct observation of product-pair correlations in rotationally inelastic collisions of ND$_3$ with D$_2$

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We present a combined experimental and theoretical study of state-to-state inelastic scattering of ND$_3$(j$^D_k = 1_1$) with D$_2$ (j = 0, 1, 2, 3) molecules at collision energies around 800 cm$^{-1}$. Using a crossed molecular beam apparatus which employs the combination of Stark deceleration and velocity map imaging, we observe the correlated rotational excitations of both collision partners. For D$_2$, both elastic ($\Delta j_{D_2} = 0$), inelastic excitation ($j = 0 \rightarrow j = 2$) and inelastic de-excitation ($j = 2 \rightarrow j = 0$) processes are observed. For a number of final ND$_3$ states, inelastic channels in which D$_2$ is rotationally excited or de-excited appear surprisingly strong. The experimental results are in excellent agreement with the predictions from quantum scattering calculations which are based on an ab initio ND$_3$–D$_2$ potential energy surface.

1 Introduction

Obtaining a fundamental understanding of intermolecular interactions is an important objective in physical chemistry. In the past decades, the study of rotational energy transfer in molecular collisional processes has helped us tremendously to probe and understand molecular interactions. These experimental studies have been pivotal to test and develop theory, in order to calculate accurate potential energy surfaces (PESs) and to perform quantum scattering calculations using these PESs. Studies of collision energy transfer find applications in various research areas. In astrophysics, for example, the estimation of molecular abundances in the interstellar medium (ISM) from spectral line data requires collisional rate coefficients of various molecules with the most abundant interstellar species such as He, H and H$_2$.\(^{16}\)

Compared to atom–molecule systems, the study of molecule–molecule systems is much less mature, and our understanding of bi-molecular collisions is rather limited, although significant progress was made recently.\(^{17,18}\) In bi-molecular collisions, energy transfer between both collision partners can occur, resulting in much more complex collision dynamics compared to atom–molecule collisions. The measurement of rotational energy transfer that occurs in both partners simultaneously, referred to as rotational product-pairs, would yield valuable information on bimolecular scattering processes, and can help us to verify theoretical calculations at an extremely high level.

Experimentally, the velocity map imaging (VMI) technique\(^{19}\) offers the opportunity to probe product-pairs directly. The idea is that if one can accurately measure the internal state and kinetic energy of one molecular collision partner, one can infer the simultaneous excitation of the unobserved collision partner from a change in kinetic energy of the detected molecule. This method has been pioneered and successfully used in molecular reactive collision experiments,\(^{20-31}\) in which vibrational product-pairs in the reaction products were probed. For rotationally inelastic scattering, however, the direct observation of rotational product-pairs is challenging, as the typical rotational energy spacing in molecules is rather small, and high image resolutions are required to resolve individual rotational channels in the scattering images.

Recently, using Stark-decelerated molecular beams, Gao et al. reported the first direct observation of product-pairs for inelastic collisions between NO radicals and O$_2$ molecules, at collision energies of 160 cm$^{-1}$\(^{32}\) and 480 cm$^{-1}$.\(^{33}\) The NO and O$_2$ molecules have rather similar rotational spacings, and for given final states of NO a series of inelastic transitions in O$_2$ could be observed. For systems in which the collision partners have very different rotational spacings, such as NO–D$_2$, the observation of product pairs has to date been elusive, even at collision energies as high as 720 cm$^{-1}$\(^{34}\).

Here, we present the first direct observation of rotational product-pairs for collisions between molecules with very different rotational constants. Using a crossed beam apparatus combining
Stark deceleration and velocity map imaging, we study energy transfer processes in collisions between ND$_3$ and D$_2$ molecules at collision energies around 800 cm$^{-1}$. Compared to NO-O$_2$, the ND$_3$-D$_2$ system is very different, both from a theoretical and experimental perspective. In contrast to NO, the ND$_3$ molecule has a significant dipole moment, and is a non-linear polyatomic whose rotational levels are characterized by both the rotational quantum number $j$, and the projection quantum number $k$. Experimentally, the state-selective ionization detection of ND$_3$ imparts significant recoil energy to the ions, resulting in a significant blurring of the images that may overshadow product-pair structures. Despite this disadvantage, for selected final states of ND$_3$, we clearly observe rotational product-pairs, pertaining to both inelastic excitation ($j = 0 \rightarrow 2$) and de-excitation ($j = 2 \rightarrow 0$) processes in D$_2$. The measured angular and radial distributions of the scattering images are in excellent agreement with the distributions obtained from simulated images which are based on theoretically predicted integral (ICS) and differential cross sections (DCSs).

Rotationally inelastic collisions involving ammonia has attracted considerable interest since the discovery of ammonia in the interstellar medium in 1968 by the observation of emissions from the inversion transitions within the rotational levels $|j_k\rangle = |1_1\rangle$ and $|2_2\rangle$. Observed inversion transitions, in combination with models that take collision-induced population exchange into account, are frequently used to probe the temperature of molecular clouds. Rotational energy transfer in collisions of ammonia with various collision partners has been studied by several groups, both with and without selection of the inversion symmetry in the initial state.

### 2 Experimental and theoretical methods

#### 2.1 Experimental set-up

The experiments are performed in a crossed molecular beam apparatus that is schematically shown in Fig. 1, and that has been described in detail before. A molecular beam of ND$_3$ seeded in different carrier gases (Ar, Kr) at a typical pressure of 1 bar is formed using a Nijmegen Pulsed Valve (NPV) and loaded into a 2.6 meter long Stark decelerator. After exiting the decelerator, the packet is scattered with a pulsed beam of neat D$_2$ at an intersection angle of 90$^\circ$. The beam of D$_2$ is produced using a commercially available pulsed valve (Jordan Inc.), and has a mean velocity of 2300 m s$^{-1}$. In the beam crossing area, the ND$_3$ molecules are state-selectively ionized by a 2 + 1 Resonance Enhanced Multi Photon Ionization (REMPI) scheme via the $B \leftrightarrow X$ transition using a tunable pulsed dye laser, and subsequently detected by a VMI spectrometer. The VMI detector is calibrated using the method described by Onvlee et al.

The REMPI process imparts about 20 m s$^{-1}$ recoil velocity to the ions, resulting in a reduction of image resolution.

In the experiments, we use the Stark decelerator in two different modes of operation. In the first, we use Ar as a carrier gas and operate the decelerator to guide the ND$_3$ through the decelerator at a constant speed of 610 m s$^{-1}$. This mode of operation yields the highest particle density for the ND$_3$ packet, but also the largest velocity spread and corresponding lowest image resolutions. In the second, we use Kr as a carrier gas and operate the Stark decelerator to slow a packet of ND$_3$ to a final velocity of 306 m s$^{-1}$. The deceleration process reduces the particle density, but improves the velocity spread of the packet and the resolution in the scattering images. We therefore refer to these modes below as high density mode and high resolution mode, respectively. The collision energy is very similar in both modes and amounts to 800 cm$^{-1}$ and 750 cm$^{-1}$ for the high density and high resolution modes, respectively.

The ground state of ND$_3$ has a pyramidal ($C_{3v}$) equilibrium geometry. Due to the “umbrella” vibrational mode and the double-minimum potential along the inversion coordinate, all the rotational levels of ND$_3$ split into doublets where the upper and lower components have negative and positive parity, respectively. The rotational levels of ND$_3$ are denoted by $j_k$ where $j$ represents the angular momentum and $k$ is the projection of $j$ on the $C_3$ axis. The rotational energy level diagrams of ND$_3$ and D$_2$ are shown in Fig. 2. Only the rotational levels involved in this experiment are shown. The D$_2$ molecule has a symmetric wave function under permutations of the two identical nuclei, deuterons, having a nuclear spin $I = 1$. As a result, D$_2$ molecules exist in two different nuclear spin configurations, namely ortho ($I_{\text{total}} = 0$ or 2) and para ($I_{\text{total}} = 1$) where $I_{\text{total}}$ denotes the total nuclear spin which can take the values 0, 1 or 2. ortho and para-D$_2$ correspond to rotational states for which the rotational wave function is symmetric ($J_{\text{total}} = \text{even}$) and anti-symmetric ($J_{\text{total}} = \text{odd}$) under permutation of the nuclei, respectively.

Before the collision, more than 99.5% of ND$_3$ resides in the upper inversion level of the $1_1$ state ($1_1^-$). As discussed by Tkáč et al., the ND$_3$ molecule exists in three nuclear spin modifications. Since the selected $1_1^-$ level has E symmetry, only final states which also have E symmetry can be observed in our experiments. Since we used normal D$_2$ in this experiment, various initial states of D$_2$ are populated in the molecular beam. The populations in these states were probed using REMPI at room temperature (see Fig. 3), from which we can
compare the radial and angular distributions of the scattering intensity in both the experimental and simulated images. The details about the simulation and analysis methods are described elsewhere\textsuperscript{32,33} and will not be discussed here. The advantage of this procedure is that we do not have to rely on extensive fitting procedures to extract cross sections from our images, which can be prone to error and misinterpretation of the results. The disadvantage of the method is that we do not directly compare measured cross sections with the calculated ones.

2.2 Quantum scattering calculations for ND\textsubscript{3}–D\textsubscript{2}

Quantum scattering calculations were performed on the five-dimensional NH\textsubscript{3}–H\textsubscript{2} potential energy surface (PES) of Maret \textit{et al.}\textsuperscript{55} The PES depends on the coordinates $R$ (the length of the vector $\mathbf{R}$ connecting the NH\textsubscript{3} and H\textsubscript{2} centers of mass), $\theta_1$ (the angle between $R$ and the $C_3$ axis of NH\textsubscript{3}), $\phi_1$ (the angle of rotation of this vector around the $C_3$ axis), and ($\theta_2$, $\phi_2$), which are the polar and azimuthal angles used to describe the orientation of H\textsubscript{2} relative to NH\textsubscript{3}. Both molecules are thus assumed to be rigid rotors.

The PES was constructed\textsuperscript{55} by computing the energy for 89 000 nuclear geometries with the coupled-cluster method with single, double, and perturbative triple excitations [CCSD(T)] and the aug-cc-pVDZ basis set. These energies were calibrated using a complete basis set extrapolation procedure based on a set of 29 000 points calculated with the larger aug-cc-pVTZ basis set. The resulting PES has an estimated accuracy of about 1 cm\textsuperscript{-1}. The accuracy of the PES was previously assessed in molecular beam\textsuperscript{50} and spectroscopic\textsuperscript{56} experiments. At each distance $R$, the energy was expanded in terms of a set of angular functions as

$$V(R, \theta_1, \phi_1, \theta_2, \phi_2) = \sum_{l_1 \mu_1 l_2 \mu_2} v_{l_1 \mu_1 l_2 \mu_2}(R)\delta_{l_1, l_2}(\theta_1, \phi_1, \theta_2, \phi_2),$$

where the angular functions $v_{l_1 \mu_1 l_2 \mu_2}$ are given in ref. 57. The radial functions were fitted using cubic splines and extrapolated to large $R$ with the appropriate $R^{-n}$ behaviour.

We assume here that the ND\textsubscript{3}–D\textsubscript{2} system can be described with the same PES as NH\textsubscript{3}–H\textsubscript{2}. This PES was expressed in different coordinates, however, to take into account the shift in the position of the center of mass of ND\textsubscript{3} with respect to NH\textsubscript{3}, as discussed in ref. 58. This transformation affects the coordinates $R$ and $\theta_1$, and the PES was re-expanded according to eqn (1) in terms of the new coordinates $(R', \theta_1', \phi_1, \theta_2, \phi_2)$. The expansion coefficients $v_{l_1 \mu_1 l_2 \mu_2}(R')$ were computed up to $l_1 = 11$ and $l_2 = 4$.

Quantum scattering calculations of integral and differential cross sections for ND\textsubscript{3}–D\textsubscript{2} collisions were carried out with an in-house scattering code\textsuperscript{59} that implements the close-coupling method for symmetric top – linear molecule collisions.\textsuperscript{60} State-to-state cross sections were computed at two collision energies (750 cm\textsuperscript{-1} and 800 cm\textsuperscript{-1}) for the initial states ND\textsubscript{3}(1,1) + D\textsubscript{2}(j), where $j = 0, 1, 2, 3$. The rotational basis for the close-coupling calculations consisted of all \textit{para} rotational states of ND\textsubscript{3} with $j \leq 10$, all \textit{ortho} states with $j \leq 4$ of D\textsubscript{2} for collisions of ND\textsubscript{3} with D\textsubscript{2}(0) or D\textsubscript{2}(2), all \textit{para} states with $j \leq 3$ of D\textsubscript{2} for collisions estimate that before the collision about 37%, 28%, 30%, and 5% of the D\textsubscript{2} molecules reside in the $j = 0, 1, 2,$ and 3 levels, respectively. The minor initial population in $j = 3$ is not relevant for the experiments reported here, and will be further neglected. In collisions, population transfer between $\omega$-D\textsubscript{2} and $\rho$-D\textsubscript{2} is forbidden, such that only $j_{D_2} = \text{even} \rightarrow j_{D_2} = \text{even}$ and $j_{D_2} = \text{odd} \rightarrow f_{D_2} = \text{odd}$ transitions are allowed.

The experimental results are compared with cross sections predicted by theory. For this, we simulate scattering images based on the cross sections from the quantum scattering calculations and the accurately known experimental conditions. The blurring due to ion recoil imparted by the ND\textsubscript{3} detection is taken into account in these simulations. We then directly
with $D_2(1)$, and all para states with $j \leq 5$ of $D_2$ for collisions with $D_2(3)$. The rotational constant of $D_2$ was taken as $B = 30.443 \text{ cm}^{-1}$, while the rotational constants of $ND_3$ were taken as $A = 5.1432 \text{ cm}^{-1}$ and $C = 3.1015 \text{ cm}^{-1}$. The inversion splitting between in the ground umbrella vibrational state was 0.0530 cm$^{-1}$.

Since the PES does not describe the inversion of $ND_3$, the umbrella motion was treated with a two-state model in which the ground inversion-tunnelling states are taken as linear combinations of the two rigid equilibrium states. This model was shown to be in excellent agreement with results obtained by treating the umbrella motion of ammonia explicitly for the scattering of NH$_3$ with rare gas atoms.\footnote{ Vide infra }

The calculations were performed on a grid of 115 values of $R'$ in the range 3.5–25 $a_0$. Partial waves with total angular momenta up to $J = 70$ were considered, and the convergence of the cross sections was checked with respect to all the parameters discussed above.

### 3 Results and discussion

#### 3.1 High density mode

A broad range of final states can experimentally be probed when the Stark decelerator is operated in the high density mode. Fig. 4 shows a collection of experimental scattering images for inelastic collisions in which ND$_3$ molecules are excited into both inversion components of the final states $2_1^1$, $2_2^1$, $3_1^1$, $3_2^1$, $4_1^1$ and $4_2^1$. In most of the images, up to three concentric rings can be seen which correspond to different rotational transitions of $D_2$ during the collision. The most intense ring, which coincides with the masked beam spot,\footnote{ Vide infra } corresponds to the elastic $D_2$ channels, \textit{i.e.}, in these collisions the $D_2$ molecules remain in their initial quantum state. The three elastic $j = 0 \rightarrow 0$, $j = 1 \rightarrow 1$ and $j = 2 \rightarrow 2$ channels all have the same ring radius and are superimposed in the image. At slightly smaller radii, a clear inelastic channel is visible that is peaked in a narrow window of scattering angles around forward scattering. This channel corresponds to inelastic $j = 0 \rightarrow 2$ rotational excitation transitions in $D_2$. At slightly larger radii, a third inelastic channel is visible, pertaining to rotational $j = 2 \rightarrow 0$ de-excitation collisions. Both assignments are confirmed from the radial scattering distributions \textit{vide infra} in combination with the rotational energy level structure of $D_2$. Note that no indications are found for inelastic $j = 1 \rightarrow 3$ rotational excitation transitions, although a significant initial population in $j = 1$ is present and the transition energy is well within the collision energy of the experiment.

#### 3.2 High resolution mode

In order to take a closer look at the three individual scattering channels, selected images were measured again using the Stark decelerator in high resolution mode. The upper and lower inversion component of the final state $3_1^1$ were selected for this, as this scattering channel has a relatively high integral cross section. The experimental scattering images are shown on the left side of Fig. 6. Indeed, the three scattering channels are observed with a slightly better resolution compared to the images shown in Fig. 4, although the improvement in resolution is only marginal. This is the consequence of the image blurring due to the ion recoil velocity imparted by the REMPI detection process, which dominates the image resolution in the present experiments irrespective of the velocity spreads of the incoming beams.

#### 3.3 Comparison with theory

To compare the experimental results with theory, scattering images are simulated based on the kinematics of the experiment and the cross sections derived from the scattering calculations. The resulting simulated scattering images are shown in Fig. 5, and for the two high-resolution images in Fig. 6. To facilitate a more quantitative comparison, the radial and angular distributions are extracted from both the experimental and simulated images, and shown in Fig. 7 and 8, respectively. The distributions of the high-resolution images from Fig. 6 are shown in Fig. 9 and 10.

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**Fig. 4** Experimental scattering images for ND$_3$–D$_2$ collisions at a collision energy of 800 cm$^{-1}$. The final states of ND$_3$ are probed and labelled in each image. All images are presented such that the relative velocity vector is oriented horizontally and forward scattering appears on the righthand side of each image. A small section of the images near forward scattering is masked due to imperfect state selection of the initial ND$_3$ packet.
In the radial distributions, the positions at which the elastic, inelastic $j = 0 \rightarrow 2$ excitation as well as the inelastic $j = 2 \rightarrow 0$ de-excitation transitions are expected based on the kinematics of the experiment are indicated by vertical dashed lines. For those states where a clear inelastic excitation or de-excitation channel for $D_2$ was not observed, the corresponding vertical traces were omitted.

It is seen that near-perfect agreement between experiment and simulations is obtained for almost all images, both regarding the radial and the angular distributions. The angular distributions, as well as the intensity ratios between elastic and inelastic $D_2$ channels for given final states of $ND_3$ are well reproduced by the simulations. This indicates that the scattering calculations capture the scattering dynamics well.

Some interesting trends can be observed in the images. First, most scattering channels display a predominantly forward scattered behaviour, with the exception of $2_2^+, 3_2^-$ and $4_2^+$ final states that have a significant component toward side-scattered angles. Secondly, the inelastic $D_2$ channels have surprisingly large intensities compared to the corresponding elastic channels, considering the relatively large amount of rotational energy that is transferred in an inelastic event. It should be noted here that, since we always cut out the beamspots at the forward direction in our analysis, the scattering signals (mostly the $j = 2 \rightarrow 0$ de-excitation channel of $D_2$) which overlap with the beamspots are also cut out. That is the reason why...
in some scattering channels of ND$_3$, we could not observe the accompanying de-excitation channel of D$_2$.

In comparison with our previous NO–D$_2$ inelastic scattering results,$^{34}$ D$_2$ shows much stronger rotational (de-)excitation when it collides with ND$_3$. This can be explained by the role of the dipole and quadrupole moments of ND$_3$, which are larger than those of NO by about an order of magnitude. Fig. 11 shows the radial expansion coefficients $v_{l_1l_2l_3}(R)$ of the ND$_3$–D$_2$ interaction potential. The dominant anisotropic terms over a large range of $R$-values are the 1023 and 2024 terms that correspond to the electrostatic interactions between the dipole and quadrupole of ND$_3$ and the quadrupole of D$_2$. These interactions, which decay as $R^{-4}$ and $R^{-5}$, are also dominant in the long range, because all other terms decay as $R^{-n}$ with $n \geq 6$.

Fig. 12 and 13 show integral cross sections (ICSs) calculated with different terms included in the potential. They illustrate that these electrostatic interaction terms largely determine the cross sections of the $j = 0 \rightarrow 2$ excitation and $j = 2 \rightarrow 0$ de-excitation transitions in D$_2$. In other words, the dipole–quadrupole and quadrupole–quadrupole interactions play a dominant role in the rotational (de-)excitation of D$_2$ when it collides with ND$_3$.

Another striking observation, illustrated in Fig. 8, is that the $j = 0 \rightarrow 2$ and $j = 2 \rightarrow 0$ (de-)excitation transitions of D$_2$ occur prominently for final $j_k$ states of ND$_3$ with $k = 1$, while they are much weaker for final states with $k = 2$. Also this can be explained by considering the anisotropic expansion of the potential shown in Fig. 11. The initial $j_k = 1_1^-$ state of ND$_3$ is coupled to final states with $k = 1$ by all terms in the potential with $\mu_1 = 0$, which include the dominant 1023 and 2024 electrostatic terms mentioned in the preceding paragraph. Final states with $k = 2$ are coupled to the initial $k = 1$ state by the terms with $\mu_1 = 3$. This can be understood by realizing that $k$ and $\mu_1$ represent the absolute values of the actual $k$ and $\mu_1$ quantum numbers. Terms with $\mu_1 = \pm 3$ couple states with $k = 1$ to states with $k = \pm 2$. Since the anisotropic interaction terms

Fig. 8  Angular distributions for the experimental scattering images in Fig. 4 (exp), and the corresponding simulated images from Fig. 5 (sim). The final states of ND$_3$ and rotational transitions of D$_2$ are labelled. For those final ND$_3$ states where a clear inelastic excitation or de-excitation channel for D$_2$ was not observed, the corresponding panels were left empty.
with $\mu_1 = 3$ are much smaller than the terms with $\mu_1 = 0$, see Fig. 11, this explains the more prominent (de-)excitation of D$_2$ accompanying transitions to the ND$_3$ final states with $k = 1$. 

**Fig. 9** Radial distributions for the experimental and simulated scattering images in Fig. 6. The different rotational transitions of D$_2$ are indicated by the dashed lines.

**Fig. 10** Angular distributions for the experimental and simulated scattering images in Fig. 6. The different rotational transitions of D$_2$ are labelled on the vertical axis on the right.

**Fig. 11** Radial expansion coefficients $v_{l_1m_1l_2}(R)$ of the ND$_3$–D$_2$ interaction potential. The curve labelled 0000 is the isotropic interaction potential, the other curves are the different anisotropic contributions.

**Fig. 12** Comparison of calculated ICSs for D$_2$ ($j = 0$) → D$_2$ ($j = 2$). Three different potentials were used in the calculations [gray: full PES, red: isotropic + dipole–quadrupole (1 0 2 3) terms, blue: isotropic + dipole–quadrupole + quadrupole–quadrupole (2 0 2 4) terms]. The final states of ND$_3$ are labelled below the bars.

**Fig. 13** Comparison of calculated ICSs for D$_2$ ($j = 2$) → D$_2$ ($j = 0$). For the labelling, see Fig. 12.
4 Conclusion

We presented high-resolution measurements of rotational product-pairs for inelastic collisions between state-selected and velocity controlled ND₃ molecules and D₂ molecules at collision energies around 800 cm⁻¹. Many final rotational states of ND₃ were studied. For a number of ND₃ final states we found interesting trends in the pair-correlated state-to-state cross sections: not only collisions elastic in D₂ were observed, but also collisions in which D₂ is excited or de-excited with ΔJ = ±2. The experimental results are compared to quantum scattering calculations using an ab initio ND₃–D₂ potential energy surface. Excellent agreement between experiment and theory is found throughout, indicating that the potential energy surface is accurate and the scattering calculations capture pair-correlated states of ND₃ were studied. For a number of ND₃ final states instance, which would constitute a further stringent test for oscillations in the angular scattering distributions, for the scattering images. This prohibits the observation of diffraction large recoil energy to the ions, thereby significantly blurring cross sections: not only collisions elastic in D₂ were observed, we found interesting trends in the pair-correlated state-to-state collision energies around 800 cm⁻¹/C₀.

Conflicts of interest

The authors declare no competing financial interest.

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