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RPPR Final Report

as of 10-Jun-2020

Agency Code:

Proposal Number: 72142CHRIP

Agreement Number: W911NF-18-1-0205

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Report Date: 29-Feb-2020

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Final Report for Period Beginning 05-Jun-2018 and Ending 30-Nov-2019

Title: Coherent Control of Molecular Scattering Using Stark-Induced Adiabatic Raman Passage

Begin Performance Period: 05-Jun-2018

End Performance Period: 30-Nov-2019

Report Term: 0-Other

Submitted By: Ph.D. Nandini Mukherjee

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Distribution Statement: 1-Approved for public release; distribution is unlimited.

STEM Degrees:

STEM Participants:

Major Goals: Major Goal:

In order to study cold collisions in the greatest possible detail, our objective is to prepare molecules in a single vibrationally excited quantum state with a precise rotational quantum number. By eliminating averaging over initial states, our goal is to understand the fundamental nature of microscopic forces that bind matter together. To study quantum controlled collisions, we plan to prepare diatomic molecules such as H₂, HD, HCl, CO, NO, as well as linear polyatomic molecules like C₂H₂ in high vibrational states using a coherent optical method called Stark-induced adiabatic Raman passage or SARP. SARP has been demonstrated by us to effectively prepare large populations of hydrogen molecules in a single rovibrationally excited (v, j) quantum state within the collision free ambience of a supersonically expanded molecular beam. The co-expansion of colliding partners in a single beam brings the collision temperature down to a few Kelvin and specifies the direction of their relative velocity. To overcome the reaction barrier in a cold collision we need to prepare highly vibrationally excited target states. Our objective is to understand at what level of vibrational stretch the constituent atoms become reactive enough to overcome the barrier.

Accomplishments: In order to study cold collisions in the greatest possible detail, our objective is to prepare molecules in a single vibrationally excited quantum state with a precise rotational quantum number. By preparing H₂ and other molecules in highly vibrationally excited quantum state, we like to investigate at what level of vibrational stretch do the constituent atoms become reactive enough to overcome the barrier in a cold collision. To be able to experimentally study a cold collision with reasonable Signal/Noise, we need to prepare a large ensemble in a desired target state. To prepare a scattering sensitive population in a highly vibrationally excited quantum state, we devised a coherent optical technique, known as Stark induced adiabatic Raman passage or SARP. SARP uses a pair of visible single frequency nanosecond laser pulses to adiabatically transfer a large population to a single vibrational rotational 'm' eigenstate within the ground electronic state of a molecule. The quantum number 'm' defines the projection of the angular momentum vector j on a suitable symmetry axis, thus defining a collision geometry.

We have utilized the DURIP equipment grant to acquire the essential laser systems to carry out the quantum state preparation using SARP. Using SARP earlier we have demonstrated preparation of a variety of rovibrational states of H₂ (v = 1, 2, 4, j = 0, 1, 2, 3) and its isotopologues, 1-3 achieving nearly complete population transfer from the

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initial to the target state. More recently we demonstrated, for the first time, complete population transfer from the ground ($v = 0, j = 0$) to the seventh vibrationally excited ($v = 7, j = 0$) level of H₂ molecules.⁴ A large ensemble of $\sim 10^9$ H₂ ($v = 7, j = 0$) molecules is prepared in the collision-free environment of a supersonic beam by transferring nearly the entire H₂ ($v = 0, j = 0$) ground-state population. The preparation of highly vibrationally excited H₂ molecules opens new opportunities to experimentally study the quantum effects in cold and ultracold chemistry.

References

1. Dong, W., Mukherjee, N. & Zare, R. N. Optical preparation of H₂ rovibrational levels with almost complete population transfer. J. Chem. Phys. 139, 074204 (2013).
2. Perreault, W. E., Mukherjee, N. & Zare, R. N. Preparation of a selected high vibrational energy level of isolated molecules. J. Chem. Phys. 145, 154203 (2016).
3. Perreault, W. E., Mukherjee, N. & Zare, R. N. Stark-induced adiabatic Raman passage examined through the preparation of D₂ ($v = 2, j = 0$) and D₂ ($v = 2, j = 2, m = 0$). J. Chem. Phys. 150, 0–10 (2019).
4. Perreault, W. E., Zhou, H., Mukherjee, N. & Zare, R. N. Harnessing the power of adiabatic curve crossing to populate the highly vibrationally excited H₂ ($v=7, j=0$) level. Phys. Rev. Lett., 124,163202 (2020).

Training Opportunities: One of our graduate students, William Edward Perreault successfully completed his Ph. D, and was listed as one of the finalists for the 2020 Justin Jankunas Doctoral Dissertation Award in Chemical Physics. A new graduate student, Haowen Zhou, has started his Ph. D work and has already contributed to this project.

Results Dissemination: Journal publication 2019-2020

1. W. E. Perreault, N. Mukherjee, and R. N. Zare, "Stark-induced adiabatic Raman passage examined through the preparation of D₂ ($v = 2, j = 0$) and D₂ ($v = 2, j = 2, m = 0$)", J. Chem. Phys. 150, 234201 (2019).
2. Perreault, W. E., Zhou, H., Mukherjee, N. & Zare, R. N. Harnessing the power of adiabatic curve crossing to populate the highly vibrationally excited H₂ ($v=7, j=0$) level. Phys. Rev. Lett., 124,163202 (2020).

Honors and Awards: William Edward Perreault has been awarded Ph.D doctoral degree by Stanford University.

Protocol Activity Status:

Technology Transfer: Nothing to Report

PARTICIPANTS:

Participant Type: PD/PI

Participant: Richard N. Zare

Person Months Worked: 1.00

Funding Support:

Project Contribution:

International Collaboration:

International Travel:

National Academy Member: Y

Other Collaborators:

Participant Type: Co-Investigator

Participant: Nandini Mukherjee

Person Months Worked: 1.00

Funding Support:

Project Contribution:

International Collaboration:

International Travel:

National Academy Member: N

Other Collaborators:

Participant Type: Postdoctoral (scholar, fellow or other postdoctoral position)

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Participant: William Edward Perreault
Person Months Worked: 1.00
Project Contribution:
International Collaboration:
International Travel:
National Academy Member: N
Other Collaborators:

Funding Support:

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Authors: 1. William E. Perreault, Nandini Mukherjee, and Richard N. Zare

Keywords: Quantum Controlled scattering, Scattering matrix, entangled state

Abstract: To investigate how molecular orientations affect low energy scattering, we have studied the rotational relaxation of HD ($v = 1, j = 2, m$) ? ($v' = 1, j' = 0$) by collision with ground-state He, where v, j , and m designate the vibrational, rotational, and magnetic quantum numbers, respectively. We experimentally probed different collision geometries by preparing three specific m sublevels, including an m entangled sublevel, belonging to a single rovibrational ($v = 1, j = 2$) energy level within the ground electronic state of HD using Stark-induced adiabatic Raman passage. Low collision energies (0-5 K) were achieved by co-expanding a 1:19 HD:He mixture in a highly collimated supersonic beam, which has defined the direction of the collision velocity and restricted the incoming orbital angular momentum states, defined by quantum number l , to $l = 2$. Partial wave analysis of experimental data shows that a single $l = 2$ input orbital dominates the scattered angular distribution, implying the pre

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Title: Quantum control of cold molecular collisions using stark-induced adiabatic Raman passage

Authors: William, E. Perreault

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as of 10-Jun-2020

Coherent Control of Molecular Scattering Using Stark-Induced Adiabatic Raman Passage

In order to study cold collisions in the greatest possible detail, our objective is to prepare molecules in a single vibrationally excited quantum state with a precise rotational quantum number. By preparing H₂ and other molecules in highly vibrationally excited quantum state, we like to investigate at what level of vibrational stretch do the constituent atoms become reactive enough to overcome the barrier in a cold collision. To be able to experimentally study a cold collision with reasonable Signal/Noise, we need to prepare a large ensemble in a desired target state. To prepare a scattering sensitive population in a highly vibrationally excited quantum state, we devised a coherent optical technique, known as Stark induced adiabatic Raman passage or SARP. SARP uses a pair of visible single frequency nanosecond laser pulses to adiabatically transfer a large population to a single vibrational rotational '*m*' eigenstate within the ground electronic state of a molecule. The quantum number '*m*' defines the projection of the angular momentum vector *j* on a suitable symmetry axis, thus defining a collision geometry.

We have utilized the DURIP equipment grant to acquire the essential laser systems to carry out the quantum state preparation using SARP. Using SARP earlier we have demonstrated preparation of a variety of rovibrational states of H₂ ($v = 1, 2, 4, j = 0, 1, 2, 3$) and its isotopologues,¹⁻³ achieving nearly complete population transfer from the initial to the target state. More recently we demonstrated, for the first time, complete population transfer from the ground ($v = 0, j = 0$) to the seventh vibrationally excited ($v = 7, j = 0$) level of H₂ molecules.⁴ A large ensemble of $\sim 10^9$ H₂ ($v = 7, j = 0$) molecules is prepared in the collision-free environment of a supersonic beam by transferring nearly the entire H₂ ($v = 0, j = 0$) ground-state population. The preparation of highly vibrationally excited H₂ molecules opens new opportunities to experimentally study the quantum effects in cold and ultracold chemistry.

References

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3. Perreault, W. E., Mukherjee, N. & Zare, R. N. Stark-induced adiabatic Raman passage examined through the preparation of D₂ ($v = 2, j = 0$) and D₂ ($v = 2, j = 2, m = 0$). *J. Chem. Phys.* **150**, 0–10 (2019).
4. Perreault, W. E., Zhou, H., Mukherjee, N. & Zare, R. N. Harnessing the power of adiabatic curve crossing to populate the highly vibrationally excited H₂ ($v = 7, j = 0$) level. *Phys. Rev. Lett.*, **124**, 163202 (2020).