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14. ABSTRACT

We are reporting our progress and our ongoing effort in preparing quantum states of H2 molecules using Stark induced adiabatic Raman passage (SARP). The supplement award (Grant # W911NF1510192), made it possible to acquire the necessary small cap equipment including, a high voltage power supply, a dry scroll pump, a X-Y-Z vacuum manipulator, a high fidelity pulsed valve, a chiller (refrigerator) for cooling dyes and a few necessary optics, for example lens, half-wave retarder, high power dielectric mirrors and high power UV polarizers. These

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UU	UU	UU	UU		Richard Zare
					19b. TELEPHONE NUMBER
					650-723-3062

Report Title

Final Report: Reaction Dynamics Using a Coherent M-state Superposition Within a Single (v, J) Rovibrational Energy Eigenstate

ABSTRACT

We are reporting our progress and our ongoing effort in preparing quantum states of H2 molecules using Stark induced adiabatic Raman passage (SARP). The supplement award (Grant # W911NF1510192), made it possible to acquire the necessary small cap equipment including, a high voltage power supply, a dry scroll pump, a X-Y-Z vacuum manipulator, a high fidelity pulsed valve, a chiller (refrigerator) for cooling dyes and a few necessary optics, for example lens, half-wave retarder, high power dielectric mirrors and high power UV polarizers. These components were absolutely necessary for extending SARP in preparing new quantum states that can be potentially applied to study low energy collisions. Using SARP with a sequence of delayed pump and Stokes pulses we have not only been able to generate H2 (v=1, J=0,1,2,3) states with a significant population of the ground vibrational (v=0) state, recently we have been successful in pumping a HD molecule from the ground (v=0) to HD (v=4) state with a significant population of the initial ground HD (v=0) state. This high vibrational pumping using SARP is a major break-through, which opens many possibilities for coherently driven chemistry and ultracold chemistry.

Enter List of papers submitted or published that acknowledge ARO support from the start of the project to the date of this printing. List the papers, including journal references, in the following categories:

(a) Papers published in peer-reviewed journals (N/A for none)

Received Paper

TOTAL:

Number of Papers published in peer-reviewed journals:

(b) Papers published in non-peer-reviewed journals (N/A for none)

Received Paper

TOTAL:

Number of Papers published in non peer-reviewed journals:

(c) Presentations

Number of Presentations: 0.00

Non Peer-Reviewed Conference Proceeding publications (other than abstracts):

Received Paper

TOTAL:

Number of Non Peer-Reviewed Conference Proceeding publications (other than abstracts):

Peer-Reviewed Conference Proceeding publications (other than abstracts):

Received Paper

TOTAL:

Number of Peer-Reviewed Conference Proceeding publications (other than abstracts):

(d) Manuscripts

Received Paper

TOTAL:

Number of Manuscripts:

Books

Received Book

TOTAL:

Received Book Chapter

TOTAL:

Patents Submitted

Patents Awarded

Awards

Graduate Students

NAME	PERCENT_SUPPORTED
FTE Equivalent:	
Total Number:	

Names of Post Doctorates

NAME	PERCENT_SUPPORTED
FTE Equivalent:	
Total Number:	

Names of Faculty Supported

NAME	PERCENT_SUPPORTED
FTE Equivalent:	
Total Number:	

Names of Under Graduate students supported

NAME	PERCENT_SUPPORTED
FTE Equivalent:	
Total Number:	

Student Metrics

This section only applies to graduating undergraduates supported by this agreement in this reporting period

The number of undergraduates funded by this agreement who graduated during this period: 0.00

The number of undergraduates funded by this agreement who graduated during this period with a degree in science, mathematics, engineering, or technology fields:...... 0.00

The number of undergraduates funded by your agreement who graduated during this period and will continue to pursue a graduate or Ph.D. degree in science, mathematics, engineering, or technology fields:...... 0.00

Number of graduating undergraduates who achieved a 3.5 GPA to 4.0 (4.0 max scale):...... 0.00

Number of graduating undergraduates funded by a DoD funded Center of Excellence grant for Education, Research and Engineering:...... 0.00

The number of undergraduates funded by your agreement who graduated during this period and intend to work for the Department of Defense 0.00

The number of undergraduates funded by your agreement who graduated during this period and will receive scholarships or fellowships for further studies in science, mathematics, engineering or technology fields:...... 0.00

Names of Personnel receiving masters degrees

NAME

Total Number:

Names of personnel receiving PHDs

NAME

Total Number:

Names of other research staff

NAME	PERCENT_SUPPORTED

FTE Equivalent:
Total Number:

Sub Contractors (DD882)

Inventions (DD882)

Scientific Progress

To compare a benchmark collision experiment (D+H2?HD+H) with theory it is absolutely essential to prepare the target molecule (H_2) in a well-defined quantum state.   We are reporting our progress and our ongoing effort in preparing quantum states of H_2 molecules using Stark induced adiabatic Raman passage (SARP). The supplement award (Grant # W911NF1510192), made it possible to acquire the necessary small cap equipment including, a high voltage power supply, a dry scroll pump, a X-Y-Z vacuum manipulator, a high fidelity pulsed valve, a chiller (refrigerator) for cooling dyes and a few necessary optics, for example lens, half-wave retarder, high power dielectric mirrors and high power UV polarizers. These components were absolutely necessary for extending SARP in preparing new quantum states that can be potentially applied to study low energy collisions. Using SARP with a sequence of delayed pump and Stokes pulses we have not only been able to generate H_2 (v=1, J=0,1,2,3) states with a significant population of the ground vibrational (v=0) state, recently we have been successful in pumping a HD molecule from the ground (v=0) to HD (v=4) state with a significant population of the initial ground HD (v=0) state.   This high vibrational pumping using SARP is a major break-through, which opens many possibilities for coherently driven chemistry and ultracold chemistry.   In addition, highly vibrationally excited H_2 or HD is required to make a high intensity negative H^- beam, which is much needed in making fusion experiments.

Technology Transfer

FINAL REPORT

GRANT/CONTRACT TITLE:	Reaction Dynamics Using a Coherent M-state Superposition Within a Single (v, J) Rovibrational Energy Eigenstate
GRANT/CONTRACT NUMBER:	W911NF-15-1-0192 (Equipment Supplement)
PROGRAM MANAGER:	James K. Parker
REPORTING PERIOD:	05/15/15 – 05/14/16
PRINCIPAL INVESTIGATOR:	Richard N. Zare zare@stanford.edu 650-723-3062
INSTITUTION:	Stanford University Office of Sponsored Research 651 Serra Street Stanford, CA 94305-4125

Reaction Dynamics Using a Coherent *M*-state Superposition Within a Single (v, J) Rovibrational Energy Eigenstate

Nandini Mukherjee, William Edward Perreault and Richard N. Zare

To compare a benchmark collision experiment ($D+H_2 \rightarrow HD+H$) with theory it is essential to prepare the target molecule (H_2) in a well-defined quantum state. To prepare a target molecule in a desired rovibrational *M*-quantum state within the ground electronic surface we introduced a new coherent optical technique called the *Stark induced adiabatic Raman passage* (SARP). This research is currently funded by the ARO, DURIP and MURI. The past DURIP awards and a partial support from MURI have been utilized to build a high-vacuum reaction chamber and acquire the necessary single mode nanosecond laser sources which allowed the successful demonstration of SARP. Nearly the complete population of the ground (v=0, J=0) H_2 molecule was transferred to a vibrationally excited (v=1, J=0,2, M) state, thus reaching *the first milestone* of the proposed project. This work has been published: W. Dong, N. Mukherjee, and R. N. Zare, "Optical Preparation of H_2 Rovibrational Levels with Almost Complete Population Transfer," J. Chem. Phys. 139, 074204-1-6. (2013). Following this work SARP has achieved *the second important milestone* by preparing a target H_2 molecule in a coherent superposition of *M*-states within a single rovibrational (v=1, J=2) energy eigenstate. Specifically, we prepared superposition of M-states within a single rovibrational (v=1, J=2) energy eigenstate: $|\psi(t)\rangle = \sum_{M} C_M |v=1, J=2, M\rangle$ where, the complex coefficients of superposition C_M are controlled by mixing various polarizations of the pump and Stokes laser pulses. This work has also been published: *N. Mukherjee, W. Dong, and R. N. Zare, "Coherent Superposition of M-States in a Single Rovibrational Level of H_2 by Stark-Induced Adiabatic Raman Passage," J. Chem. Phys. 140, 074201 (2014).* In addition, using SARP with a sequence of delayed pump and Stokes laser pulses we were able to prepare the various rovibrational energy eigenstates H_2 (v=1, J=0,1,2,3) with a significant population of the ground vibrational (v=0) state.

Here we are reporting our progress and our ongoing effort in preparing highly vibrationally excited quantum states of H_2 molecules using Stark induced adiabatic Raman passage (SARP). This will allow us to study low temperature (<1 K) collision revealing the quantum character of a molecular encounter. For the first time, we have been successful in pumping a HD molecule from the ground (v=0) to HD (v=4) state with nearly complete population transferred from the initial ground HD (v=0) state. . This is achieved with a sequence of partially overlapping nanosecond pump (355 nm) and Stokes (680 nm) single-mode laser pulses of unequal intensities. By comparing our experimental data with our theoretical calculations we are able to draw two important conclusions: **(1) using SARP a large population (>10^{10} molecules per laser pulse) is prepared in the (v=4, J=0) level of HD, and (2) the polarizability $\alpha_{00,40}$ (□ 0.6 x 10^{-41} Cm^2V^{-1}) for the (v=0, J=0) to (v=4, J=0) Raman overtone transition is only about five times smaller than $\alpha_{00,10}$ for the (v=0, J=0) to (v=1, J=0) fundamental Raman transition.** This work has been published: Perreault William, N. Mukherjee, and R. N. Zare, J. Chem. Phys. **145**,154203 (2016)]. ***This***

pumping of highly vibrationally excited molecular energy eigenstate using SARP is a major break-through, which opens new avenues to study coherently controlled chemistry and ultracold chemistry. In addition, highly vibrationally excited H_2 or HD is required to make a high intensity negative H^- beam, which is much needed in making fusion experiments.

To reach the final goal, we have used the supplement award (Grant # W911NF1510192) to acquire the necessary small cap equipment including, a high voltage power supply, a dry scroll pump, a X-Y-Z vacuum manipulator, a high fidelity pulsed valve, a chiller (refrigerator) for cooling dyes and a few necessary optics, for example lens, half-wave retarder, high power dielectric mirrors and high power UV polarizers. These components were absolutely necessary for extending SARP in preparing highly vibrationally excited quantum states that can be potentially applied to study low energy collisions. The detail description of the equipment purchased using this award is described below.

Capital equipment purchased and how these pieces where vital to this project:

MDC Vacuum - REQ 3725501 - PO 60972342 - Translation Stage - 5,374.40
The vacuum translation stage was used to manipulate and align the molecular beam through a 0.5 mm skimmer within a high vacuum reaction chamber. Accurate positioning of the molecular beam through the skimmer is extremely important to access the coldest and most intense part of the molecular beam for quantum state preparation and for the study of collision.

Agilent Technologies - REQ 379701 - PO 61020376 - Scroll Vacuum Pump - 10,232.92
For the study of collision dynamics, maintaining high vacuum condition (10^{-7} Torr) in the detection area of the high vacuum reaction chamber is essential. The Scroll pump is used as the roughing pump for the molecular turbo pump. Moreover, the Scroll pump provides an oil free ambience essential for low noise imaging detector.

Coherent - REQ 3927333 - PO61189186 - Wavemaster Meter - 12,635.33
For coherent state preparation using Stark Induced Adiabatic Raman Passage (SARP) we need to use single mode frequency tunable laser sources. The frequency (wavelength) of these laser sources must be measured at all time during the state preparation. The Wavemaster-Meter needs to be hooked at all times to measure the laser frequency within an accuracy of 100 MHz.

LAMID - REQ 3771432 - PO 61020377 - Pulsed Valve - 14,147.22
Evan-Levi pulsed valve made by LAMID provides a way to make cold and highly intense molecular beam, which is essential for the study of coherent stereo dynamics. Just today we have found that this valve produces a highly rotationally cold molecular beam of HD, which has helped us to prepare nearly 10^{11} molecules in HD (v=4) level.