PULSE SHAPING BASED ULTRA-BROADBANDWIDTH MULTIDIMENSIONAL SPECTROCOPICal METHODS

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09/17/2013
Final Report

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1. REPORT DATE (DD-MM-YYYY) 11-09-2013
2. REPORT TYPE Final Progress Report
3. DATES COVERED (From - To) 15-JUN 2010 to 14-JUN-2013

4. TITLE AND SUBTITLE PULSE SHAPING BASED ULTRA-BROADBANDWIDTH MULTIDIMENSIONAL SPECTROSCOPIC METHODS

5a. CONTRACT NUMBER FA9550-10-1-0312
5b. GRANT NUMBER
5c. PROGRAM ELEMENT NUMBER
5d. PROJECT NUMBER
5e. TASK NUMBER
5f. WORK UNIT NUMBER

6. AUTHOR(S) Marcos Dantus

7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)
MICHIGAN STATE UNIVERSITY
CONTRACT & GRANT ADM
301 ADMINISTRATION BLDG
EAST LANSING MI 48824-1046

8. PERFORMING ORGANIZATION REPORT NUMBER

9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)
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AF OFFICE OF SCIENTIFIC RESEARCH
875 NORTH RANDOLPH STREET, RM 3112
ARLINGTON VA 22203
SHARON B. TAYLOR 703-696-7340

10. SPONSOR/MONITOR'S ACRONYM(S)

11. SPONSOR/MONITOR'S REPORT NUMBER(S)

12. DISTRIBUTION/AVAILABILITY STATEMENT
Approved for public release; distribution unlimited.

13. SUPPLEMENTARY NOTES

14. ABSTRACT
Single-beam approaches to coherent anti-Stokes Raman scattering (CARS) have been developed to allow diagnostic capabilities to study molecular systems under extreme conditions such as those found in combustors. Unlike methods that combine beams with different wavelengths at different angles, the approaches developed use a single beam, thus simplifying implementation outside the research laboratory. Collaboration with scientists at the Air Force Research Laboratory (Wright-Patterson AFB) resulted in six publication and one manuscript pending submission. The first confocal images of flames providing species selective density and temperature were recorded.

15. SUBJECT TERMS
Laser spectroscopy, temperature and density measurements in flames.

16. SECURITY CLASSIFICATION OF:
a. REPORT U
b. ABSTRACT U
c. THIS PAGE U

17. LIMITATION OF ABSTRACT UU

18. NUMBER OF PAGES

19a. NAME OF RESPONSIBLE PERSON Marcos Dantus
19b. TELEPHONE NUMBER (Include area code) 517-355-9715 x314

Standard Form 298 (Rev. 8/98)
Prescribed by ANSI Std. 239.18
Adobe Professional 7.0
Objective: Develop a greatly simplified approach to coherent anti-Stokes Raman scattering (CARS), allowing us to extend its diagnostic capability to study molecular systems under extreme conditions. Unlike methods that combine beams with different wavelengths at different angles, our approach uses a single beam, thus simplifying measurements on difficult to reach locations such as within reactive flows. Collaborate with scientists at the Air Force Research Laboratory (Wright-Patterson AFB) where temperature and pressure measurements in reactive flows such as those of aircraft turbines are required.

Accomplishments:
1. Flame diagnostics working closely with AFRL scientists:
Accomplished confocal imaging of flames to provide species specific density and temperature information using single-beam CARS.

Experimental results displaying nitrogen percent distribution and temperature in different flames.
2. Refining single-beam temperature determination and calibration:

Figure 1. Experimental measurement of CARS vibration-rotational spectra for oxygen at room temperature (left) and high temperature (right). (red and blue) The rotational features are simulated based on a Boltzmann distribution of populations for each temperature. Notice that in addition to changes in the rotational distribution the vibrational hot-band is observed at high temperatures. The vibrational hot band provides additional temperature information based on its expected Boltzmann population. A temperature and pressure controlled cell from the AFRL has arrived at MSU, and we are using it to calibrate our method.

3. New probe methods (narrower, multiple, and broader) for improved signal and multiplexing.

Instead of using one broad-bandwidth pump and one narrow bandwidth probe, we decided to explore the use of multiple probe frequencies as shown in the next figure.

Fig. 2. (left) Single beam CARS spectrum of air (oxygen and nitrogen) obtained with a single frequency probe (see insert). (right) Single beam CARS spectrum of air, this time obtained with a probe beam that has four discrete frequencies. The four frequencies lead to four lines for each of the two species. While this complicates the spectrum, the pattern is easy to discern and provides higher signal to noise data.

3. Single-shot species selective single beam CARS.

The multiple probe wavelengths, when generated in a pulse shaper, can each receive a calibrated delay, and therefore can be used to provide the coherence dephasing time, a measure that is sensitive to temperature, directly in a single laser shot. This approach led to accurate temperature determination in a single laser shot and to the following publication.


In this paper we demonstrated a single-shot method for measuring the species-selective concentration and temperature. The advantages of this method are illustrated in the figures below.
Fig. 3 (left) The measurements are calibrated and show precision and accuracy of 25K. (Middle) The method is independent of changes in pressure/density. This is very important because nanosecond laser based methods are very sensitive to pressure/density changes because collisions occur within the time of the measurement. In this method the measurement is completed within a couple of picoseconds and therefore there is no influence from pressure/density. (right) Because the measurement is “ratiometric” it is independent of laser intensity fluctuations. In this figure the average power of the laser is changes from 20-140mW (a factor of 7) and yet the temperature variations registered are within 10% of the target temperature. We are presently using this approach to image a propane/air flame.

Results from this Grant:

Peer Reviewed Journal Articles:


Figure 2 (from the OPN article). Being able to image reacting flows requires selective excitation of desired Raman vibrational transitions. (left) Selective excitation of the Fermi diad in CO₂ associated with the symmetric stretch and two quanta of the bending motion. Note that selective excitation at different frequencies is capable of isolating single vibrational transitions. (right) Single beam CARS imaging of a turbulent flow of CO₂ molecules. The top image is obtained with transform limited pulses, the bottom image is obtained with selective excitation of the symmetric stretch.


Invited Talks:

“Phase Control of Laser-Matter Interactions and Applications” M. Dantus, Committee on Atomic, Molecular, and Optical Sciences, April 3-4, 2012, National Academies’ Keck Center –Washington, DC

“Pulse Shaping Based Ultra-Broadbandwidth Multidimensional Spectroscopic Methods,” M. Dantus AFOSR Contractors Meeting, January 5-6, 2012


“Imaging reactive flows and trace quantities of hazardous substances using single beam CARS” P.J. Wrzesinski, D. Pestov, V. V. Lozovoy, S. Roy, J.R. Gord, and M. Dantus, PQE Snowbird, Utah (Jan, 2011)


Contributed Presentations:


