NASA-FAR (grant NAG3-1974) ANNUAL PROGRESS REPORT:
*Development of Synchronously Scanned OPO CARS as a New Probe for Hostile Environments*
Year 2: 10/1/97 - 9/30/98

INTERIM
IN-25
361057

Peter C. Chen
Spelman College Chemistry Department
350 Spelman Ln. Box 307
Atlanta, GA 30314
Ph: 404-223-7667
Fax: 404-223-1449
email: pchen@spelman.edu

Summary: During the second year of this project, we have focused on three new developments. The first has been the accidental discovery of a degenerate peak at the $0cm^{-1}$ position in the coherent Raman spectrum that is not due to incoherent Rayleigh scattering. The second is the incorporation of a second OPO to provide additional capability to the instrument. The third is the design of a new system that could perform single-shot CARS using a solid state broad-band OPO. The project continues to focus on student research training at the undergraduate level, and three students have worked on the project through this grant. The outcome of this work has been 1 publication published during this period, 2 publications submitted (by invitation), and 7 presentations given or invited. During this period, one additional grant was funded. The details on each of these items is given below.

Degenerate Peak

During our studies on the use of SSOPO CARS for low frequency Raman spectroscopy, we accidentally discovered a peak at the $0cm^{-1}$ position that was not due to incoherent Rayleigh scattering. This peak disappeared when any of the three input laser beams were blocked. Furthermore, the linewidth of the peak was similar to that of the OPO (~0.2 $cm^{-1}$), while that of the incoherent Rayleigh scattering should have a linewidth comparable to the monochromator (~$1cm^{-1}$). Spectra subtraction was used to remove any Rayleigh scattering, and the resulting sharp peak at $0cm^{-1}$ remained and was similar in intensity to that of the rotational peaks from nitrogen and oxygen in air at 1atm.

The size of the peak varied, making quantitative measurements difficult. We attributed this irreproducibility to interference from the injection seeder of the Nd:YAG because the wavelength of the OPO idler beam matched the wavelength of the Nd:YAG fundamental (1064nm) at the position of the $0cm^{-1}$ peak. We solved this problem by introducing a second OPO into the system to replace the Nd:YAG fundamental, which was being used for the second of the three input beams. This modification solved the problem, and allowed us to quantitatively measure the size of the degenerate peak relative to that of the CARS rotational peaks and nonresonant background.

Based upon work carried out by others in this field, we now attribute this degenerate peak to stimulated Brillouin scattering (i.e., electrostriction). Crim and coworkers at U. Wisconsin discovered that electrostriction plays the major role in atmospheric pressure air for two-color Laser Induced Grating Spectroscopy (LIGS).

The significance of this work is that we have discovered a way to combine LIGS with CARS using a single technique. LIGS has the advantage of being incredibly sensitive, but 1) is difficult to quantitate in a reproducible way, and 2) may be somewhat susceptible to interference from other effects. CARS, although not as sensitive, has a very high signal-to-interference ratio and can be highly reproducible for quantitative measurements. The combination of these two spectroscopies into a single technique could provide some advantages. We are currently preparing to submit this work for publication, and are planning some additional experiments.

Additional OPO

The introduction of a second OPO into the instrument has helped us to solve interference problems described in the previous section, while also providing additional flexibility for some electronic enhancement studies. In our previous setup, we could not change the frequency of the second beam. Therefore, the output beam was always fixed at 532nm. With this new setup, we can set the wavelength of the output beam so that it can achieve additional electronic enhancement. We are currently running such experiments. To the best of our knowledge, we are the first to study the use of two separate OPO systems for a CARS experiment.

Broadband OPO

We are currently working with Laservision, a company that makes OPOs, to build a new OPO system that has a broad bandwidth. If this project is successful, we will have developed a fully solid-state system that can take single shot CARS spectra of the entire vibration region. One possible application of such a device would be to obtain point-by-point single shot Raman spectra. Each spectrum would be sufficiently broad to give both vibrational and rotational information on all major species and temperature. We are currently in the testing stage of this new OPO system.

Undergraduate Research Assistants - This grant has supported three undergraduate research students to work in my laboratory this year. Alice Turner has just completed her sophomore year as a chemistry major. Her work focuses on the electronic enhancement that can be achieved using this technique by incorporating a second OPO into the instrument. Kanika Benton is going into her sophomore year as a chemistry major and has focused on using this technique to obtain Raman spectra in the very low frequency region. Brandi Johnson is also a rising sophomore chemistry major who is working on using the technique to combine the benefits of CARS with DFWM.

Presentations (given, submitted, or accepted) during the second year period:

1) Chen, P. C., "Use of an OPO for CARS and Single Wavelength Detection Spectroscopy," FACSS '98, Austin, TX, invited speaker for October 13, 1998.

2) Chen, P. C., "Single Wavelength Detection Spectroscopy," Ohio University Chemistry Department Seminar Speaker, Athens, OH, May 13, 1998.

3) Chen, P. C., "Development of Synchronously Scanned OPO CARS As a New Probe for Hostile Environments," NASA HBCU Research Conference, Cleveland, OH, April 8, 1998.

4) Chen, P. C., "Recent Developments using Single Wavelength Detection Spectroscopy," The Pittsburgh Conference '98, New Orleans, LA, Mar. 2, 1998.

5) Chen, P. C., "Nonlinear Laser Spectroscopy Using an Optical Parametric Oscillator," Pomona College Chemistry Department Seminar Speaker, Claremont, CA, Jan. 26, 1998.

6) Chen, P. C., "Single Wavelength Detection Spectroscopy," University of Vermont Chemistry Department Seminar Speaker, Burlington, VT, Jan. 22, 1998.

7) Chen, P. C., "SSOPO CARS: Making Coherent Raman Spectroscopy Practical and More Powerful," 1997 Pacific Conference on Chemistry and Spectroscopy, Irvine, CA, October 22, 1997.

Publications (published, submitted, or accepted) during the second year period:

1) Chen, P. C. and Pinnix, M., "Low-Frequency Raman Spectra using Synchronously Scanned Optical Parametric Oscillator CARS," *Applied Spectroscopy* 51, 1678-1681, 1997.

2) Chen, P. C., "Nonlinear Raman Spectroscopy, Instruments," to appear in the *Encyclopedia of Spectroscopy and Spectrometry*, edited by J. Lindon, G. Tranter, and J. Holmes, Academic Press Ltd., London, 1999.

3) Chen, P. C., "Single Wavelength Detection Spectroscopy for gas phase Raman spectroscopy," submitted to the Internet Journal of Vibrational Spectroscopy, 1998.

New grants funded during the second year period

"A Discovery-based approach to Interdisciplinary Science using Lasers," NSF CCD program, January 1998.