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13. ABSTRACT (Maximum 200 words) The purpose of this research is to adapt the latest forefront research in the area of quantum optics to address the more challenging technical problems needed to support the warfighter in today's rapidly changing rules of engagement, and homeland security in the face of new threats. To do this we will align technology push in areas where we have unique expertise with applications pull in several key areas of national need.				
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Quantum Optics Initiative

Final Report

ONR project N00014-03-1-0639

Introduction:

The Texas Engineering Experiment Station and Texas A&M University, partnering with Princeton University, Purdue University, Rice University, Baylor University, the University of North Texas, and The University of Texas at Austin have developed a world class center for quantum optical sciences and engineering as an integral part of our strong quantum physics and telecommunications efforts. Utilizing new understandings of physics, breakthroughs are being made on the detection of trace gases and biological molecules, extending the limits of CARS spectroscopy, Bose-Einstein condensates, new radiation sources, and photonics. This has led to applications in national security, such as laser anthrax detectors, telecommunications, and other new areas such as extension of current optical techniques in the semiconductor industry.

The following pages summarize our accomplishments sorted by investigation institution:

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2. Purdue University Page 8
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5. University of North Texas Page 44
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1. Texas A&M and Princeton Universities

Report on femtosecond CARS for detection of Anthrax spores

As part of the Anthrax Project, the femtosecond Coherent Anti-stokes Raman Scattering (CARS) experiments performed at Texas A&M and Princeton Universities have been focused on demonstrating the possibility and the benefits of performing CARS measurements in the ultraviolet (UV). We have successfully demonstrated femtosecond UV-CARS signals from DPA, we have demonstrated that all-UV experiments are possible and provide resonant enhancement in DPA over the visible CARS signals, and we have demonstrated collinear UV-CARS signals in DPA. Furthermore, we have successfully demonstrated using the UV-CARS setup that we are able to discriminate between DPA and a series of confusers that could provide signal in the same spectral range. We have developed a method of extracting the information specific to dipicolinic acid, and used this method towards developing a ROC curve for the specificity of this detection method. We were able to clearly pick dipicolinic acid from not only tens of other chemical compounds, but we have also proved that our method is powerful enough to discriminate between isomers of DPA. In particular, we could clearly see the difference between the 2,6-DPA present in the spores, and 3,5-DPA, a variant of DPA with the same chemical composition.

1. Collinear All-UV CARS experiments.

We have previously reported the successful demonstration of an all-UV femtosecond CARS experiment in calcite. This work has been presented at conferences [1,2] and is published in Ref. [3]. However, the absence of signal in the all-resonant UV-CARS experiment in DPA, both in liquid and solid form, raised questions about the feasibility of a UV-CARS experiment in DPA altogether. There were concerns about ionizing the molecules, destroying the DPA molecules, etc., when irradiated with UV light. Our goal was to prove that UV-CARS is possible and, moreover, beneficial for detection of DPA. Furthermore, for the purpose of remote detection, we have designed a collinear setup for the UV-CARS experiment, as shown in Fig. 1.

We have performed several experiments to prove that we can measure anti-Stokes signals from DPA molecules by pumping in the visible and probing in the UV, also by pumping in the UV for better excitation of the vibrational modes, and probing in the visible for better discrimination of the signal from the other beams, and we have could always got a signal as long as we were just below the resonance. In Fig. 1, however, we show an all-UV setup, where all the beams are in the ultraviolet. The pump and Stokes beams are provided by an optical parametric amplifier (OPA) and the second harmonic of the amplified Ti:Sapphire system, while the probe beam is provided by a non-collinear

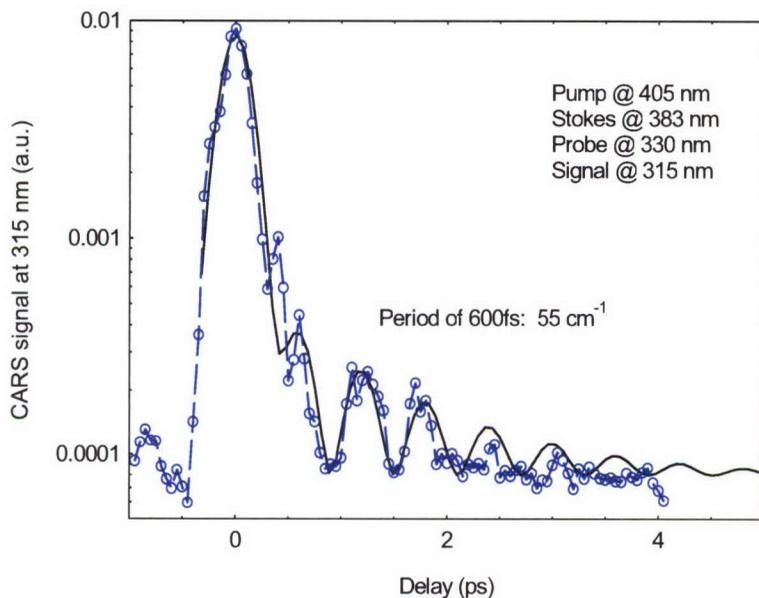


Fig. 3. Collinear all-UV femtosecond CARS signal in DPA.

We can clearly see in Fig. 3 the sub-picosecond coherence decay, and the interference effect obtained because of excitation of both 1435 and 1383 cm^{-1} ground state vibrational levels of the DPA molecule (shown in the inset in Fig. 1). Indeed, the period of 600fs corresponds to 55 cm^{-1} energy difference, close to the expected 51 cm^{-1} . As we will show later, this feature will prove very useful for the specificity of this detection method.

2. UV resonant enhancement.

By performing several femtosecond CARS experiments both in the visible and in the UV, we can experimentally verify the predicted resonant enhancement expected when the excitation and/or probe beams move closer to resonance. We compared the signals obtained in the UV and visible for calcite and DPA, and obtained the following:

- For calcite, with the bandgap of 8eV , moving from visible to UV provides a calculated (theoretical) enhancement for the generated signal photons per incident probe beam of 3.5. By measuring the conversion efficiency in our CARS experiments we obtain a measured enhancement of 3.
- For DPA, with a bandgap of 4.25eV , the theoretical prediction for the resonant enhancement is 10, because we are closer to the resonance. Comparing the normalized CARS signals between the UV and visible experiments, we obtain a measured resonant enhancement of 13.

We have hence been able to demonstrate resonant enhancement, which is going to be even stronger if the laser frequencies are even closer to the resonance.

3. Specificity. Distinguishing DPA from confusers.

In order to use the femtosecond CARS to detect the presence of DPA molecules, one needs to be able to distinguish between the signals obtained from DPA, and the CARS signals obtained from other molecules (we will call them “confusers”). In order to develop a ROC curve, we chose as a measuring parameter the strength of the temporal

beating shown in Fig. 3, obtained when the two ground state vibrational levels 51 cm^{-2} apart are excited simultaneously. We used UV for the pump and Stokes beams, and visible for the probe beam and the anti-Stokes signal. We employed a BOX-CARS geometry for the purpose of testing, because it gives us a better dynamic range.

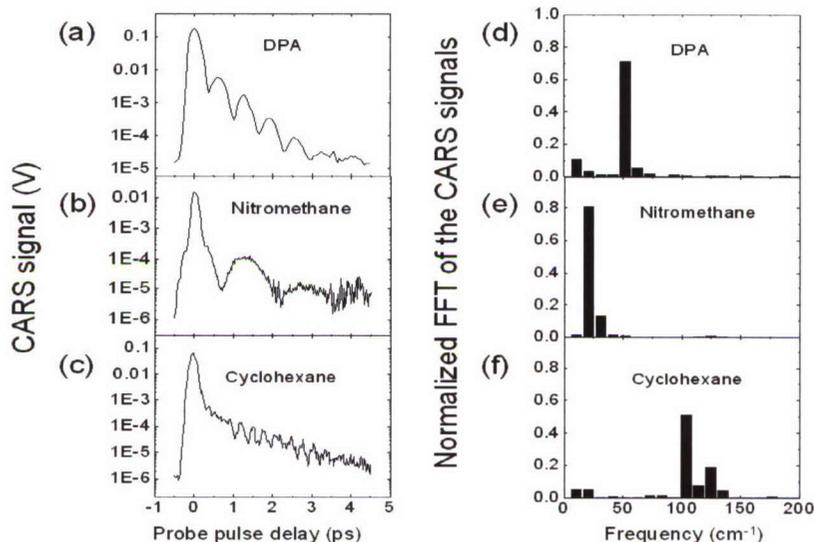


Fig. 4. CARS signals at 1400 cm^{-1} (a-c) and their Fourier transforms (d-f) for DPA, nitromethane, and cyclohexane (from top to bottom).

In Fig. 4 we chose DPA and two of the confuser molecules. As we can see, the signals from DPA (a), nitromethane (b), and cyclohexane (c) look different in the time domain, the temporal beatings have different frequencies. This makes obvious the choice for the specificity parameter (S), which is the area around the $51(\pm 3)\text{ cm}^{-1}$ frequency in the Fourier plane (right in Fig. 4), normalized to the total area of the FFT pattern. In other words, we are looking for the molecule that gives us the most beating signal between two vibrational levels 51 cm^{-1} apart.

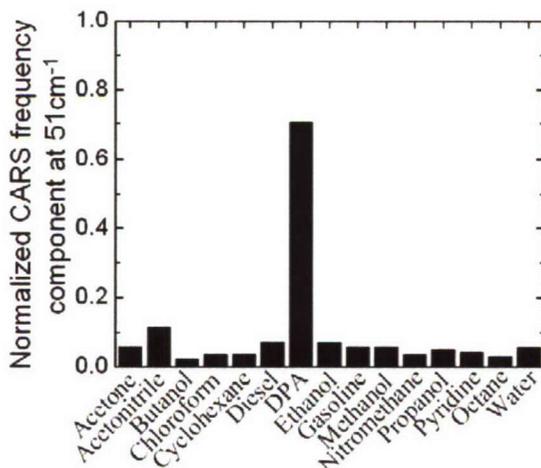


Fig. 5. The amount of beating at 51 cm^{-1} (S-parameter) shown in the CARS data, defined as the ratio between the area at $51(\pm 3)\text{ cm}^{-1}$ and the rest of the FFT data.

In Fig. 5 we plot the S-parameter for various molecules, and we can see that the dipicolinic acid molecules stand out very clearly. This graph shows that the femtosecond CARS signal is unique for DPA, and we can certainly distinguish the DPA molecules from the confusers. These results were presented at conference [4], and published in Ref. [5].

4. Discrimination between 2,6-DPA and 3,5-DPA

To further prove that we can specifically target the dipicolinic acid from spores, we have also compared the CARS signals from two samples of DPA isomers. The chemical structures for the two DPA variants are shown in Fig. 6.

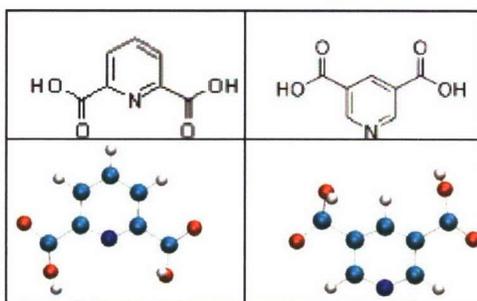


Fig. 6. Chemical structures for 2,6-pyridinedicarboxylic acid (left) and 3,5-pyridinedicarboxylic acid (right).

As shown in Fig. 7(left), the CARS signals obtained from the DPA molecules show strong temporal modulation, indicating the excitation of two (or more) vibrational modes.

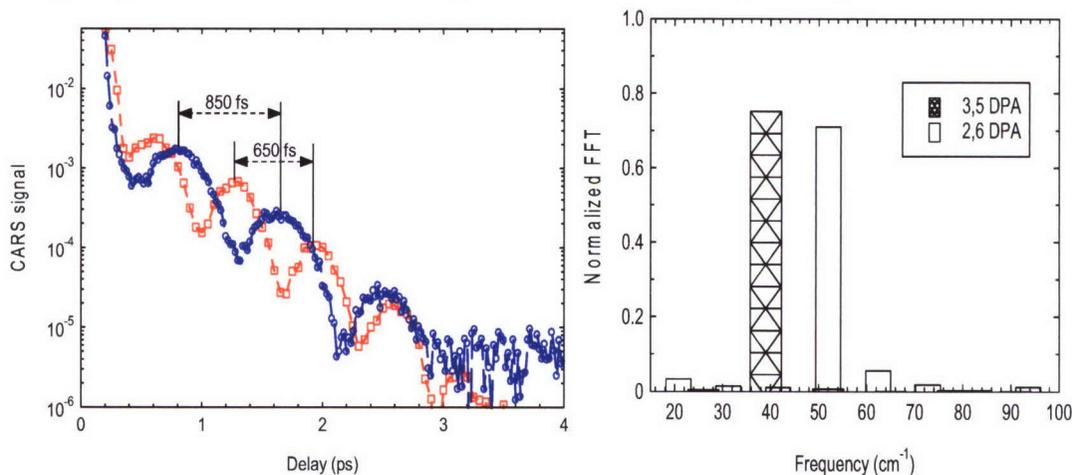


Fig. 7. Left: CARS signals at 1400 cm⁻¹ as function of probe delay for 2,6-DPA (squares) and 3,5-DPA (circles) solutions in H₂O/KOH. The signals are normalized to the peak of the four-wave-mixing signal. Right: Fourier transforms of the time-delayed CARS data for 3,5-pyridinedicarboxylic acid (shaded) and 2,6-pyridinedicarboxylic acid (empty).

Given the large dynamic range we can clearly see several modulation periods (even with a coherence lifetime of only 500 fs), and we can also see the difference between the 2,6-DPA and 3,5-DPA molecules. The 2,6-DPA molecule shows a 650 fs modulation period (squares in Fig. 7, left), corresponding to the 51 cm⁻¹ frequency difference between the

two competing modes, and the 3,5-DPA molecule shows an 850 fs modulation period (circles in Fig. 7, left), corresponding to a 38 cm^{-1} frequency difference.

On the right graph in Fig. 7 we show the actual measured frequencies, obtained by performing Fourier transformations on the temporal data, as explained above. The Fourier transforms peak at 51 cm^{-1} and 38 cm^{-1} for the 2,6-DPA and 3,5-DPA, respectively. These results were published in Ref.[6]

It is encouraging (and some may say “surprising”) that we can distinguish frequency shifts on the order of 10 cm^{-1} using laser pulses with spectra ten times broader. Figs. 4, 5, and 7, however, clearly demonstrate that the time-delayed CARS is a powerful spectroscopic tool, able to distinguish with high precision between very similar molecules.

5. UV pulse shaping.

Concomitantly with the UV-CARS measurements described above, we have actively pursued the idea of shaping the pump pulses, in order to achieve maximum coherent excitation of the ground state vibration(s). This is one of the main goals for us as part of FAST-CARS project, and we expect to see signal improvement using shaped pulses when we are close to the resonance.

We have successfully built an UV-pulse shaper, and we have tested several pulse shapes for the 400 nm pump beam. By applying a modulated RF signal onto the AOM crystal, we change the amplitude and phase independently, and we can design pulse shapes with spectral and temporal features that suit our experiment.

In Fig. 9 we show the experimental setup of the pulse shaper using an acousto-optical modulator (AOM) as the active element that alters the phase and/or amplitude of individual frequency components of a broadband pulse. We use a genetic algorithm, which lets the computer decide the best pulse shape in order to optimize the signal.

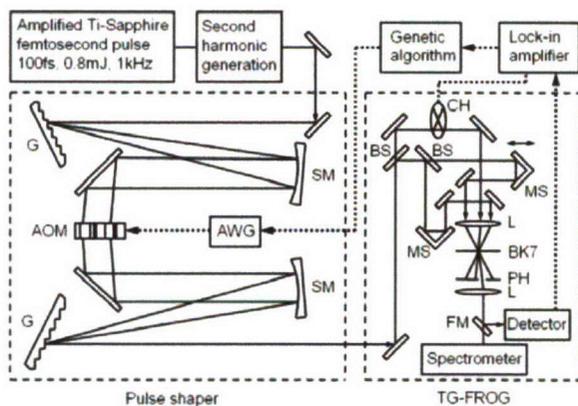


Fig. 9. Schematic of acousto-optical modulator (AOM) based pulse shaper experiment.

In Fig. 10 we show how we apply the UV pulse shaper to CARS. Using a box-CARS setup in the UV, we maximize the anti-Stokes CARS signal using the genetic algorithm described in Fig. 9.

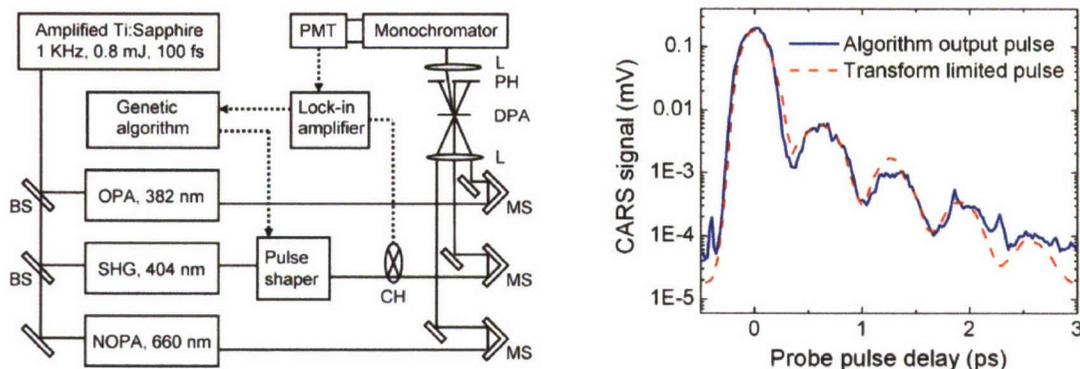


Fig. 10. (left) Schematic diagram of the femtosecond CARS experiment employing the adaptive pulse shaper. (right) CARS experiment on 300 mM dipicolinic acid solution using a Stokes pulse with the optimized spectral phase (solid line), as compared to the same experiment using a transform limited Stokes pulse (dashed line). DPA: dipicolinic acid solution; NOPA: noncollinear OPA; OPA: optical parametric amplifier; PMT: photomultiplier tube; SHG: second harmonic generation.

We can see from the temporal CARS data shown in Fig. 10 (right) that the signal recorded (solid line) reaches the maximum theoretical signal (dotted line) calculated for a transform limited pulse.

The UV-pulse shaper for CARS results were presented at a conference [7], and published in Ref. [8].

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2. Purdue University

1. Detection Limits for NO ERE-CARS

We made significant strides in lowering the detection limit for NO using ERE-CARS compared to our previous work [1]. This was accomplished in large part by

improving on the previous experimental apparatus. The present experimental apparatus for the ERE CARS measurements is shown in Figure 1. The major improvement over the previous system is that we use a Spectra-Physics Nd:YAG laser, purchased at Purdue University, that is injection-seeded so as to produce single-longitudinal-mode (SLM) laser output in a reliable fashion; the Continuum Nd:YAG laser used in the previous measurements was injection-seeded but did not produce SLM output in a reliable fashion. We also used a High-Finesse wave meter to measure the wavelengths of all beams in the experiment. This greatly aided us in identifying and probing particular spectral transitions rapidly and with great accuracy.

We devoted significant effort in this project to trying to lower the detection limit for our measurements. We achieved a detection limit of approximately 5 ppm for NO in a buffer gas of N₂ at room temperature and pressure. ERE-CARS spectra as a function of concentration are shown in Figure 2. As is evident from Figure 2, we can clearly see the NO ERE-CARS signal at a concentration level of 5 ppm, but the resonant signal is modulated by the nonresonant background signal. We also studied saturation effects for both the Raman transition and the ultraviolet probe transition in great detail, and modeling of these results is currently in progress.

2. Theoretical Analysis and Model Development

We have made significant progress in modeling the ERE-CARS process and increasing the accuracy of our calculation of the electronic resonance enhancement factor for specific ERE-CARS transitions for the NO molecule. The NO spectra are modeled both by using a perturbative (low laser intensity) analysis, and by using a nonperturbative modeling approach in which the time-dependent density matrix equations for the process are directly integrated.

At this point the perturbative ERE-CARS code is at a much more advanced state of development and is the primary code that we use to model the experimental data. The Sandia CARS code [2] was modified for the ERE-CARS calculations and the NO spectral data was obtained from the spectroscopic database code LIFBASE [3] and from previous high-resolution NO CARS measurements [4,5]. The ERE-CARS code is used to model spectra generated from both UV probe scans and Stokes scans. The ERE-

CARS code at this stage of development does not include saturation, which is a very important effect in our measurements, especially for atmospheric pressure and below. The effects of saturation are accounted for to some extent by performing calculations with variable laser frequency widths, especially for the ultraviolet probe beam.

A computer code based on direct integration of the time-dependent density matrix equations has been developed to model the ERE-CARS process. The density matrix approach allows us to account for saturation effects in a rigorous manner, but the computer code takes much longer to run than the modified Sandia CARS code. We are currently still testing this computer code and will be modeling saturated spectra that we have acquired in future work.

3. ERE-CARS Spectroscopy of Acetylene

The ERE-CARS spectroscopy of acetylene (C_2H_2) was also investigated using the same experimental system as for the NO measurements. The motivation for studying the acetylene molecule is that it is a polyatomic molecule with complicated electronic structure characteristic of molecule such as DPA. The Raman C-C stretch of C_2H_2 has a frequency of 1974 cm^{-1} , and has broad electronic resonance absorption features below 230 nm. Thus it is easy to switch between the NO and C_2H_2 ERE-CARS experiments simply by changing the Stokes dye laser wavelength from 591 nm for NO to 594 nm for C_2H_2 .

The detection limit for ERE-CARS detection of C_2H_2 was found to be approximately 25 ppm in a buffer gas of N_2 at room temperature and pressure. ERE-CARS spectra are shown as a function of C_2H_2 concentration in Figure 3; these spectra were recorded by scanning the Stokes dye laser beam. The low detection limit was surprising because the electronic bands of C_2H_2 below 230 nm are essentially structureless and we found no significant dependence of the ERE-CARS Stokes spectrum on the wavelength of the ultraviolet probe beam. However, the low detection limit that we achieved indicates that the electronic resonance enhancement factor is very significant. We also investigated the pressure dependence of the ERE-CARS Stokes signal, as shown in Figure 4. The signal increases dramatically with increasing pressure, again indicating that the resonance enhancement is a diffuse process and is not dependent

on tuning to a particular rotation-vibration transition in the electronic spectrum, as is the case with NO.

4. Benefits of the Seed Funding

The seed funding that we have received has been very helpful for student support. Waruna D. Kulatilaka has been supported from this grant and recently defended his PhD thesis. Waruna will be staying on for a short time as a postdoctoral research associate at Purdue. In the late fall he will begin a postdoctoral research appointment at Sandia National Laboratories in Livermore, California.

The seed funding has also been very helpful in support of our efforts to obtain Phase II STTR funding from the Air Force for measurements of NO in high-pressure flames. We were recently asked to write a Phase II proposal to perform these measurements.

5. List of Conference Presentations and Invited Seminars

1. R. P. Lucht, "New Optical Sensors and Diagnostics for Detection of Minor Species in Reacting Flows," presented at the Department of Mechanical Engineering, Pennsylvania State University, State College, PA, November 10, 2005. (Invited Seminar)
2. R. P. Lucht, "Electronic-Resonance-Enhanced CARS for Species Measurement: Detection Limits and Saturation Effects," 36th Winter Colloquium on the Physics of Quantum Electronics, Snowbird, Utah, January 2-6, 2006. (Invited Presentation)
3. R. P. Lucht, "Polarization Spectroscopy and Electronic-Resonance-Enhanced CARS for Quantitative Measurements in Reacting Flows," Pittsburgh Conference on Analytical Chemistry, 2006 Bomem-Michelson Award Symposium, "Spectroscopic Diagnostics: From Photolysis Products to Recycled Plastic," Orlando, FL, March 15, 2006. (Invited Presentation)
4. R. P. Lucht, "Recent Advances in Electronic-Resonance-Enhanced CARS for Minor Species Measurement," Princeton-TAMU Molecular Spectroscopy Symposium, Princeton, NJ, May 19, 2006. (Invited Presentation)

6. Papers Resulting from the Funding

Several papers are in preparation for submission to archival journals. These include:

1. J. P. Kuehner, W. D. Kulatilaka, S. Naik, N. Chai, R. P. Lucht, S. Roy, J. R. Gord, and M. O. Scully, "Theoretical and Experimental Investigation of Electronic-

Resonance-Enhanced Coherent Anti-Stokes Raman Scattering Spectroscopy of Nitric Oxide,” in preparation for submission to the Journal of Chemical Physics.

2. N. Chai, W. D. Kulatilaka, S. Naik, N. M. Laurendeau, and R. P. Lucht, “Electronic-Resonance-Enhanced Coherent Anti-Stokes Raman Scattering Spectroscopy of Acetylene,” in preparation for submission to the Journal of Chemical Physics.

6. References

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7. Figures

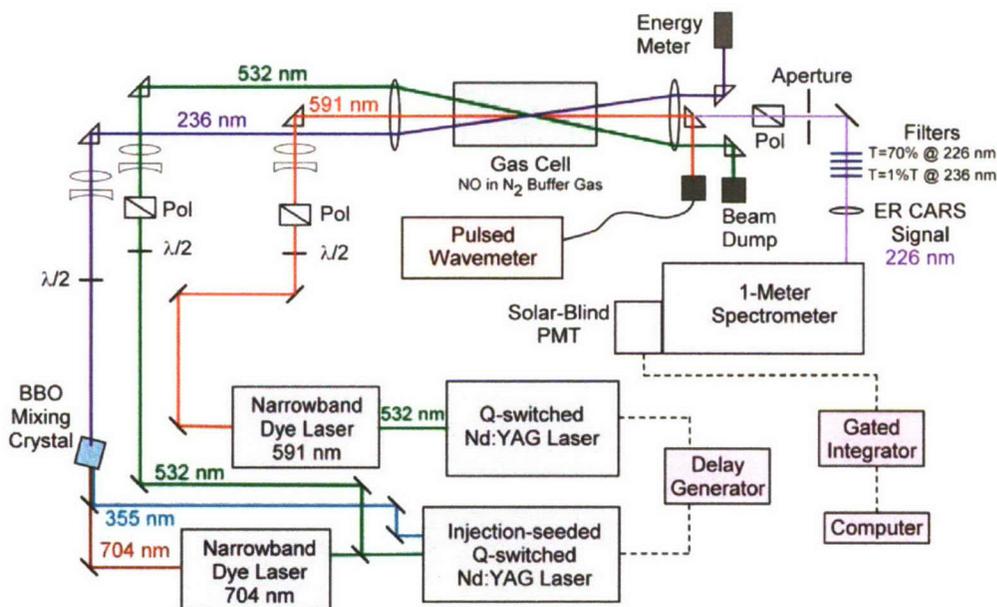


Figure 1. Experimental system for the NO ERE-CARS experiments.

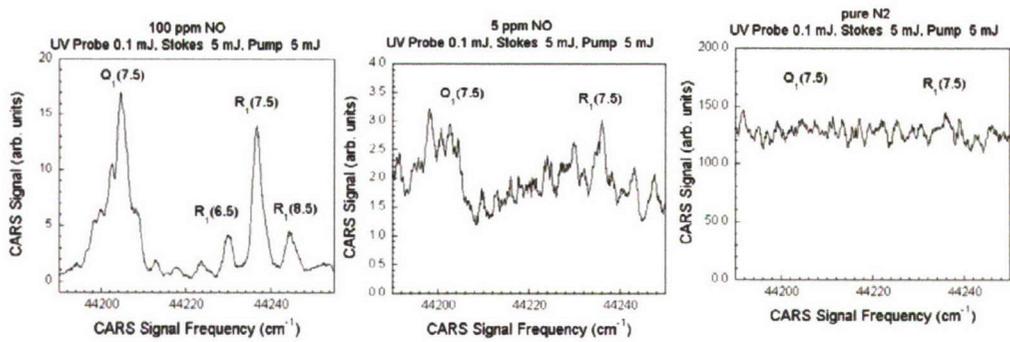


Figure 2. ERE-CARS spectra of NO at concentration levels of 100, 5, and 0 ppm.

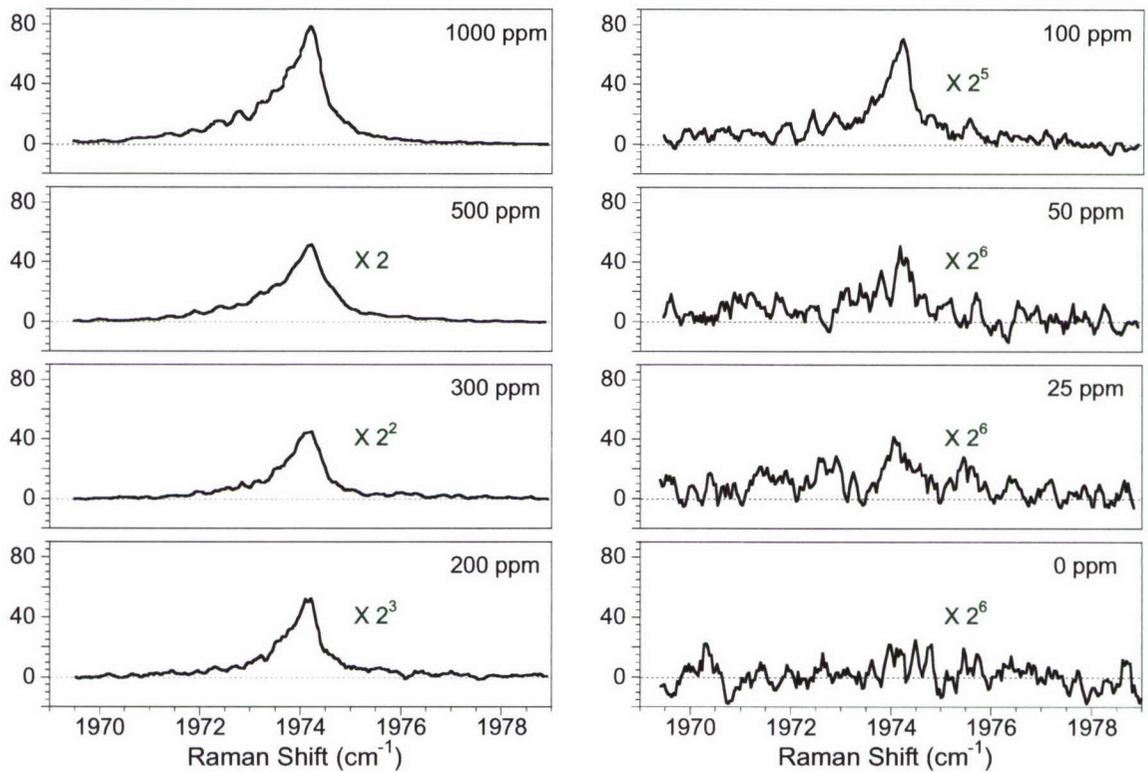


Figure 3. ERE-CARS spectra of acetylene at concentration levels ranging from 1000 to 0 ppm.

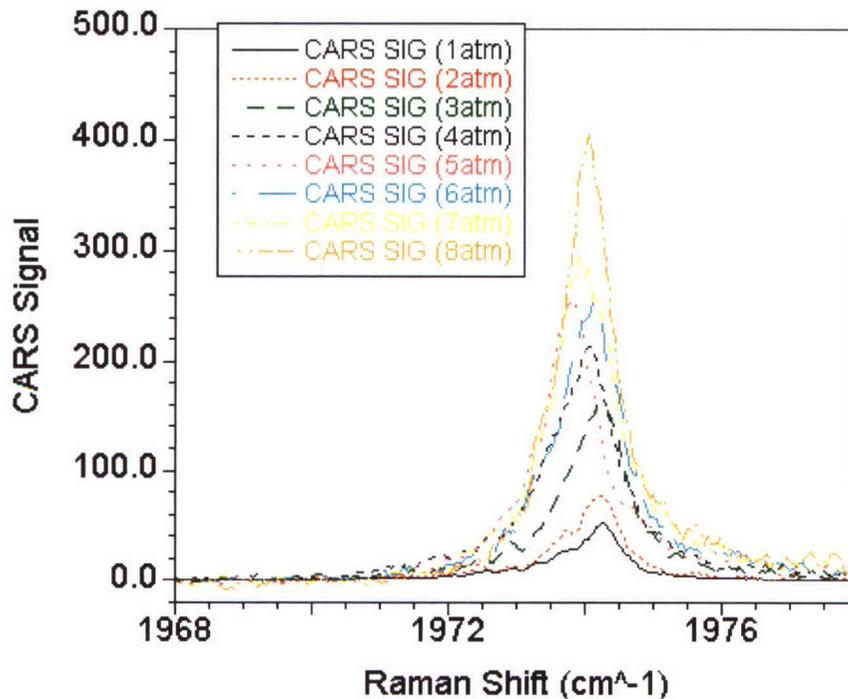


Figure 4. Pressure scaling of the ERE-CARS spectrum of acetylene.

3. Rice University

Molecular Bose-Einstein Condensates – R. G. Hulet

The goal of this work was to associate ultracold atoms into molecules and to create a molecular Bose-Einstein condensate (BEC). Important issues in this work concern the coherence of the association process, as coherently making and breaking of chemical bonds constitutes a new form of chemistry, and the formation of diatomic molecules from constituent atoms that are fermions. In that case, the molecules are analogous to Cooper pairs of electrons which underlie superconductivity. A Bose-Einstein condensate of these molecules should form a superfluid. Depending on the interaction strength between the constituent atoms, the superfluid may be best described as a condensate of composite bosons, or in the weakly coupled limit, as a Fermi or BCS superfluid. We have achieved the goal of creating a Bose-Einstein condensate (BEC) of molecules by using a magnetically-tuned collisional resonance, and we have explored the many-body nature of the crossover from the molecular BEC regime to the BCS regime of correlated Cooper pairs.

Funding of this work came from two ONR grants, and specific activities and findings are separately described for the two periods.

Phase 1 (March 15, 2003 to December 31, 2004):

Our main activity during this period was to investigate the formation of molecules using a magnetically-tuned collisional resonance, known as a Feshbach resonance. At a certain magnetic field, 525 G in the case of ${}^6\text{Li}$, the kinetic energy of a pair of colliding atoms can be brought into resonance with a bound vibrational state of the diatomic molecule. By sweeping the magnetic field through this resonance, a trapped gas of free atoms may be converted into bound molecules. Although these molecules are in their electronic ground state, they are vibrationally excited and would be expected to decay via inelastic collisions. We discovered, quite unexpectedly, that molecular lifetime in the trap was exceptionally long, nearly 1 s. Although the mechanism responsible for stabilizing the molecules against decay is not completely understood, we believe that the Pauli Exclusion Principle somehow inhibits the necessary three-body collisions. The long lifetime, however, enables further cooling of the molecules into the superfluid state.

Phase 2 (January 1, 2004 to April 4, 2006):

We cooled the molecules by evaporation from their confining optical trap. By lowering the intensity of the laser beam forming the optical trap, hot molecules could evaporate, leaving the remainder with an overall lower kinetic energy per particle. We succeeded in producing a condensate with nearly 100k molecules, as shown in Fig. 1. A Bose condensed gas of molecules, cooled to nearly absolute zero temperature, is the ideal system to explore the BEC-BCS crossover. The Feshbach resonance was employed again, this time to serve as a tuning knob for the interaction strength between the constituent atoms in the molecules/pairs. By varying the magnetic field, the gas could be tuned from the BEC regime of tightly bound molecules all the way to the BCS regime of weakly correlated Cooper pairs. We explored the microscopic nature of pairing, throughout this crossover regime, by using a molecular spectroscopic probe. By exposing the molecules/pairs to a laser beam resonant with an electronically-excited molecular state and measuring the rate of photoexcitation, it was possible to extract a quantitative measure the contribution of the Feshbach molecule to the quantum state of the molecules/pairs. This enables one to answer detailed questions about the pairs, including their spatial size. Furthermore, with this method it was possible to extract the order parameter of the superfluid and to compare with theory.

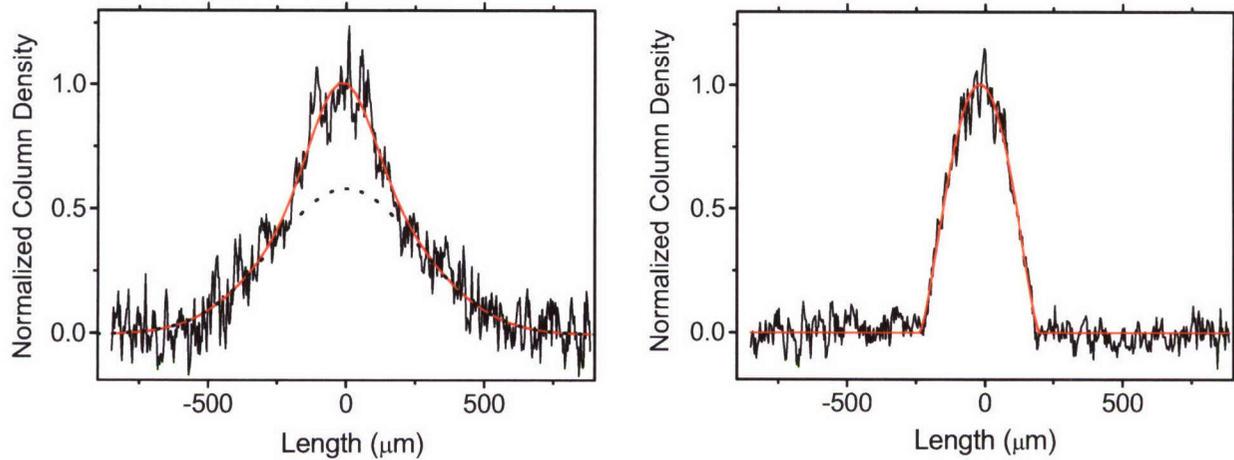


Figure 1. Absorption image profiles of a molecular Bose-Einstein condensate confined to an optical trap. The image on the left corresponds to partial evaporation in the optical trap. The broad pedestal comes from the thermal molecules, while the central peak is due to the condensate. The image on the right corresponds to full evaporation to a nearly pure molecular BEC. The red lines are fits to Bose-Einstein distributions.

Pairs of ultracold fermions normally consist of one spin-up and one spin-down particle, and in the usual theory of superconductivity, therefore, equal numbers of each spin component are required. We explored the consequences of violating this equality, by creating a gas with an unbalanced spin population. Although there has been an extraordinarily large theory effort on this problem for the past 40 years, there have been few experimental studies because of the fundamental difficulty of producing a magnetized (more spin-up electrons than spin-down) superconductors. With ultracold atoms, however, such an imbalance is straightforward to achieve. We found that the gas will phase separate into a superfluid core with equal numbers of spin-up and spin-down atoms, and with the excess unpaired spin-up atoms forming a shell surrounding the superfluid core (see Fig. 2). Moreover, the superfluid core can deform in order to maximize the numbers of pairs. There are several possible explanations for this deformation,

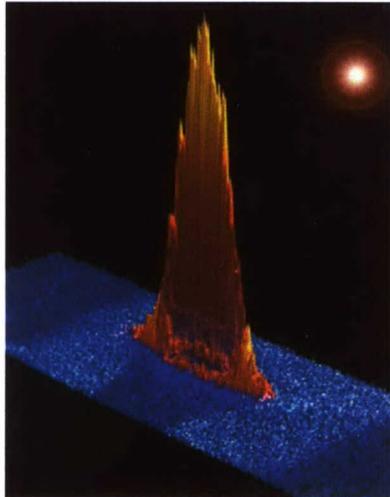


Figure 2. The figure is a false-color rendering of a two-dimensional image of a phase separated cloud of lithium-6 atoms. The tall, semi-transparent central region consists of fully paired atoms, one spin-up atom for every spin-down. This central core is believed to be a Fermi superfluid, in analogy with a superconductor. The opaque peaks on either side, as well as the faint ring around the bottom, are the excess unpaired spin-up atoms that have been expelled from the central core.

but one possibility is a new exotic form of superfluidity, called a deformed Fermi surface. We continue to explore the nature of this unbalanced system as it contains an abundance of new, previously unexplored physics.

This grant provided the funds necessary to hire Dr. Michael Jack, a post-doctoral researcher who is an expert in theoretical quantum optics. Dr. Jack provided the theoretical basis needed to analyze the experiment probing the microscopic nature of pairing using a molecular probe. By contributing to the success of the experiment to convert ultracold atom pairs into molecules, this grant helped us to obtain an ONR single-investigator grant, as well as a continuation of our NSF funding.

Resulting Publications

Guthrie B. Partridge, Wenhui Li, Ramsey I. Kamar, Yean-an Liao, and Randall G. Hulet, “Pairing and Phase Separation in a Polarized Fermi Gas”, *Science* **311**, 503 (2006); published online 22 December 2005 (10.1126/science.1122876).

G. B. Partridge, K. E. Strecker, R. I. Kamar, M. W. Jack, and R. G. Hulet, “Molecular Probe of Pairing in the BEC/BCS Crossover”, *Physical Review Letters* **95**, 020404 (2005).

K. E. Strecker, G. B. Partridge, R. I. Kamar, and R. G. Hulet, “Conversion of an Atomic Fermi Gas to a Molecular Bose-Einstein Condensate”, in *Atomic Physics 19, Proceedings of the Nineteenth International Conference on Atomic Physics*, L. G. Marcassa, K. Helmerson, and V. S. Bagnato, eds. (American Institute of Physics Conference Proceedings, vol. 770, New York, 2005).

K. E. Strecker, G. B. Partridge, A. G. Truscott, and R. G. Hulet, "Tunable interactions in ultracold Bose gases", *Advances in Space Research* **35**, 78 (2005).

Randall G. Hulet, "Atomic Fermi gases", in the 2004 McGraw-Hill Yearbook of Science & Technology, pp. 19-21 (McGraw-Hill, 2004).

Kevin E. Strecker, Guthrie B. Partridge, and Randall G. Hulet, "Conversion of an Atomic Fermi Gas to a Long-Lived Molecular Bose Gas", *Physical Review Letters* **91**, 080406 (2003).

Invited Presentations

International Symposium on Quantum Fluids and Solids, Kyoto (8/06).

Gordon Research Conference on Correlated Electron Systems, Mount Holyoke College, South Hadley, MA (6/06).

Nordita and the Niels Bohr Institute, Workshop on Strong Correlations in Fermi Systems, Copenhagen (6/06).

Los Alamos National Laboratory Quantum Information Research Institute Seminar (3/06).

APS March Meeting, Baltimore (3/06).

Banff Cold Atom Meeting, Banff, Canada (2/06).

Physics Colloquium, University of Houston, Houston (1/06).

Aspen Center for Physics Workshop on Strong Correlations in Ultracold Fermi Systems, Aspen, CO (1/06).

Physics Colloquium, University of California, San Diego (1/06).

Physics Colloquium, Oklahoma State University, Stillwater, OK (10/05).

Rice University Continuing Education Course on Einstein, Houston (10/05).

BEC Workshop, Princeton, NJ (10/05).

Physics Colloquium, Caltech, Pasadena, CA (10/05).

World Year of Physics Symposium, Fermilab, Batavia, IL (10/05).

EuroConference on Ultracold Gases and their Applications, San Feliu, Spain (9/05).

Physics Colloquium, University of Connecticut, Storrs, CT (9/05).

Workshop on New Developments in Quantum Gases, University of Washington, Seattle (8/05).

Workshop on Macroscopic Quantum Phenomena, Nanjing University, Nanjing, China (6/05).

Workshop on Macroscopic Quantum Phenomena, Institute of Physics, Beijing, China (6/05).

Aspen Center for Physics Workshop on Ultracold Trapped Atomic Gases, Aspen, CO (5/05).

APS Division of Atomic, Molecular, and Optical Physics Annual Meeting, Lincoln, NE (5/05).

Center for Nonlinear Studies International Conference, Santa Fe, NM (5/05).

Workshop on Strongly Interacting Quantum Gases, Ohio State University, Columbus, OH (4/05).

Physics Colloquium, Valparaiso University, Valparaiso, IN (3/05).

Public Lecture, Valparaiso University, Valparaiso, IN (3/05).

Physics Colloquium, Georgia Tech University, Atlanta, GA (3/05).

Deutsche Physikalische Gesellschaft Annual Meeting, Berlin (3/05).

Workshop on Solitons in BEC's, Almagro, Spain (2/05).

Banff Cold Atom Meeting, Banff, Canada (2/05).

Seminar, Harvard University, Cambridge, MA (10/04).

Physics Colloquium, University of California, Berkeley, CA (9/04).

International Conference on Atomic Physics, Rio de Janeiro, Brazil (7/04).

Boulder Condensed Matter Summer School, Boulder, CO (7/04).

Tutorial, APS Division of Atomic, Molecular, and Optical Physics Annual Meeting, Tucson, AZ (5/04).

Kavli Institute of Theoretical Physics Quantum Gas Conference, University of California, Santa Barbara, CA (5/04).

Physics Colloquium, Northwestern University, Chicago, IL (5/04).

NASA Annual Workshop on Fundamental Physics, Solvang, CA (4/04).

TAMU Award # R82180 FINAL REPORT (03/15/03-04/04/06)

“Monitoring of Environmentally Relevant Trace Gas Species using Quantum Cascade Lasers”

Frank K. Tittel and Robert F. Curl
Rice University

A. Proposed research

The objective of the research has been to develop compact, rugged trace gas sensors based upon the combination of cavity enhanced and quartz enhanced photo-acoustic laser spectroscopy with infrared semiconductor laser sources. Such highly sensitive, selective and real-time trace gas monitors have potential applications in a variety of fields such as environmental analysis, industrial chemical analysis process control, medical diagnostics and homeland security.

B. Methodology

Primarily we have focused in the 3 year period on 2 versatile, sensitive trace gas detection techniques:

Cavity enhanced absorption spectroscopy

A gas analyzer based on a continuous-wave mid-IR quantum cascade laser operating at $\sim 5.2 \mu\text{m}$ and on off-axis integrated cavity output spectroscopy (ICOS) was developed in 2004 to measure NO concentrations in human breath. A compact sample cell, 5.3 cm in length and with a volume of $\sim 80 \text{ cm}^3$, which is suitable for on-line and off-line measurements during a single breath cycle, was designed and tested. A noise-equivalent (signal-to-noise ratio of 1) sensitivity of 10 ppbv of NO was achieved. The combination of ICOS with wavelength modulation resulted in a 2-ppbv noise-equivalent sensitivity. The total data acquisition and averaging time was 15 s in both cases.

In 2005 a nitric oxide sensor based on a thermoelectrically cooled, cw DFB QCL laser operating at $5.45 \mu\text{m}$ (1835 cm^{-1}) and off-axis ICOS combined with a wavelength-modulation technique was demonstrated to determine NO concentrations at the sub-ppbv levels that are essential for a number of applications, such as medical diagnostics (specifically in detecting NO in exhaled human breath) and environmental monitoring. The sensor employs a 50-cm-long high-finesse optical cavity that provides an effective path length of 700 m. A noise equivalent minimum detection limit of 0.7 ppbv with a 1-s observation time was achieved.

Quartz enhanced photoacoustic spectroscopic techniques using an interband cascade lasers

A novel approach to photoacoustic detection of trace gases utilizing a quartz tuning fork (QTF) as a sharply resonant acoustic transducer was first reported by us in 2002. Advantages of the technique called quartz-enhanced photoacoustic spectroscopy (QEPAS) compared to conventional resonant photoacoustic spectroscopy include QEPAS

sensor immunity to environmental acoustic noise, a simple absorption detection module design, and its capability to analyze small gas samples, down to 1 mm³ in volume. A continuous-wave mid-infrared distributed feedback interband cascade laser was utilized to detect and quantify formaldehyde (H₂CO) using QEPAS. H₂CO is of particular interest since it is a hazardous and carcinogenic substance that is released from chemical binders present in numerous manufactured items and hence its presence in the environment cannot be avoided. The Occupational Safety and Health Administration (OSHA) has issued general industrial standards with an upper limit of 0.75 ppm for long term exposure (8 hours time-weighted average) and 2 ppm for short term exposure (15 minutes). NASA has also established spacecraft maximum allowable concentration levels for crew exposure to H₂CO for extended periods of time. H₂CO has been identified as a potential biomarker in breath analysis of human subjects. For example, in exhaled breath from breast cancer patients concentration levels of 1.2 ppmv were observed. Furthermore, H₂CO is an important and reactive component present in all regions of the atmosphere arising from the oxidation of biogenic and anthropogenic hydrocarbons. Tropospheric H₂CO concentration measurements provide a means of validating photochemical model predictions concerning hydrocarbon oxidation that are used to advance ozone chemistry. The sensor architecture is depicted in Fig 1. The laser was operated at liquid-nitrogen temperatures and provided a single-mode output power of up to 12 mW at 3.53 μm (2832.5 cm⁻¹). The noise equivalent (1σ) detection sensitivity of the sensor was measured to be 1.1×10⁻⁸ cm⁻¹W/Hz^{-1/2} for H₂CO in ambient air, which corresponds to a detection limit of 0.28 ppmv for a 1 second sensor time constant and 4.6mW laser power delivered to the absorption detection module.

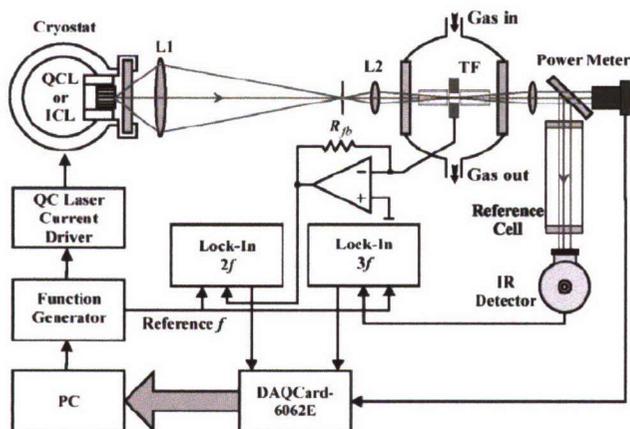


Fig. 1 Schematic of a QCL or ICL based QEPAS sensor platform.

C. New Funding

1. “Quantum cascade laser photoacoustic sensor for chemical warfare agent detection”, Pacific Northwest National Laboratory – PNNL; (\$ 210,000), 01/15/05-09/30/07

2. “Mid infrared technologies for the health and environment (MIRTHE)”, National Science Foundation – NSF; (\$ 86,000), 07/01/06-06/30/07,

D. Graduate Students

C. Roller (Ph.D May 2005), now with Ekips Technologies
S. So (M.Sc. May 2005), now in the Rice Ph.D program
M. McCurdy (in the Rice Ph.D/MD BCM programs)
R. Lewicki (in Ph.D program)
T. Mosely (M.Sc. May 2005)

E. List of Conferences (March 15, 2003-April 4, 2006)

- “Tunable Quantum Cascade Laser Spectrometer for Ethylene Monitoring,” D. Weidmann, A. A. Kosterev, **R. F. Curl**, M. P. Fraser, and **F. K. Tittel**, 87th OSA Annual Meeting, Tucson, AZ (October 5-9, 2003).
- “Quantum Cascade Laser Based Sensor for Carbonyl Sulfide,” C. Roller, A. Kosterev, D. Weidmann, **R. F. Curl**, **F. K. Tittel**, K. Uehara, and C. Gmachl, 87th OSA Annual Meeting, Tucson, AZ (October 5-9, 2003).
- “Off-Axis Integrated Cavity Output Spectroscopy for Nitric Oxide Detection in Human Breath Using a Quantum Cascade Laser,” Y. A. Bakhirkin, A. A. Kosterev, **R. F. Curl**, **F. K. Tittel**, and M. Allen, 87th OSA Annual Meeting, Tucson, AZ (October 5-9, 2003).
- “Applications of Pulsed Quantum Cascade Lasers to NO Monitoring in Industrial Exhaust Gases,” A. A. Kosterev, C. Roller, and **F. K. Tittel**, IEEE Sensors 2003, Toronto, Canada (October 22-24, 2003)
- “Chemical Sensors Based on Quantum Cascade Lasers,” **F. K. Tittel**, A. A. Kosterev, Y. A. Bakhirkin, D. Weidmann, and **R. F. Curl**, IEEE Sensors 2003, Toronto, Canada (October 22-24, 2003).
- “Current applications of quantum cascade laser in trace gas analysis and chemical sensing”, (INVITED) **F.K. Tittel**, Y. Bakhirkin, **R.F. Curl**, A. A. Kosterev, M. McCurdy, C. Roller, S. So, D. Weidmann, and G. Wysocki, International Workshop on Quantum Cascade Lasers, Sevilla, Spain (January 4-8, 2004).
- “Advanced detection techniques of trace gas monitoring for human space flight,”(INVITED) A. A. Kosterev, Y. Bakhirkin, and **F.K. Tittel**, Habitation 2004 International Conference, Orlando, FL (January 4-7, 2004).
- “Mid-Infrared Quantum Cascade Laser Sensors for Gas Analysis and Medical Diagnostics,”(INVITED) **F. K. Tittel**, A. A. Kosterev, Y. A. Bakhirkin, C. Roller, D. Weidmann, and R. F. Curl, Photonics West 2004, San Jose, CA (January 25-29, 2004).

- “Recent advances in quartz-enhanced gas-phase photoacoustic spectroscopy,” A. A. Kosterev and **F.K. Tittel**, OSA Topical Meeting on Laser Applications to Chemical and Environmental Analysis, (LACEA), Annapolis, MD, (February 9-11, 2004).
- “Off-axis integrated Cavity Output Spectroscopy for Nitric Oxide Detection in Human Breath Using a Quantum Cascade Laser”, M. R. McCurdy, Y. A. Bakhirkin, C. Roller, A. A. Kosterev, **R.F. Curl**, **F. K. Tittel**, OSA Topical Meeting on Laser Applications to Chemical and Environmental Analysis, (LACEA), Annapolis, MD, (February 9-11, 2004).
- “Off-axis integrated Cavity Output Spectroscopy for Nitric Oxide Detection in Human Breath Using a Quantum Cascade Laser”, M. R. McCurdy, Y. A. Bakhirkin, C. Roller, A. A. Kosterev, **R.F. Curl**, **F. K. Tittel**, 21st Annual Houston Conference on Biomedical Engineering Research (HSEMB), Houston, TX, (February 12-13 2004).
- Quantum Cascade Laser Spectrometer for Trace-Gas Detection of Exhaled Carbonyl Sulfide” M. McCurdy, G. Wysocki, C. Roller, S. So, D. Weidmann, **R.F. Curl**, **F.K. Tittel**, R. Bag and M.C. Paraguya, 21st Annual Houston Conference on Biomedical Engineering Research (HSEMB), Houston, TX, (February 12-13 2004).
- “Off-axis Integrated Cavity Output Spectroscopy for Nitric Oxide Detection in Human Breath Using a Quantum Cascade Laser”, M. McCurdy, Y. Bakhirkin, A. Kosterev, **R.F. Curl**, M.G. Allen, and **F. K. Tittel**, Am. Acad. of Allergy, Asthma and Immunology (AAAAI) Meeting, San Francisco, CA, (March 19-23, 2004).
- “Compact Quantum Cascade Laser Based $^{13}\text{CO}_2/^{12}\text{CO}_2$ Isotopic Ratiometer for Field Measurements of Volcanic Gases”, **F.K. Tittel**, D. Weidmann, C. Roller, **R.F. Curl**, and K. Uehara, International Conference on Optical Diagnostics and Monitoring (OPDIMON), Naples, Italy, (March 21-26, 2004).
- “Gas sensing with 9 μm thermoelectrically-cooled quasi-cw distributed feedback quantum cascade laser”, D. Weidmann, A. A. Kosterev, **F. K. Tittel**, T. Aellen, M. Beck, S. Blaser, D. Hofstetter, and J. Faist, Conference on Lasers and Electro-Optics/International Quantum Electronics Conference (CLEO/IQEC 2004), San Francisco, CA (May 16-21, 2004).
- “Quantum cascade laser spectrometer for trace-gas detection of exhaled carbonyl sulfide,” G. Wysocki, Stephen So, M. McCurdy, C. Roller, D. Weidmann, P. Frantz, **R. F. Curl**, and **F. K. Tittel**, Conference on Lasers and Electro-Optics/International Quantum Electronics Conference (CLEO/IQEC 2004), San Francisco, CA (May 16-21, 2004).
- “Multi-species QEPAS based gas sensing using a wavelength-programmable diode laser source,” A.A. Kosterev, D. Weidmann, and **F. K. Tittel**, N. Ryan and D. McDonald, Conference on Lasers and Electro-Optics/International Quantum Electronics Conference (CLEO/IQEC 2004), San Francisco, CA (May 16-21, 2004).
- “Wavelength modulation off-axis integrated cavity output spectroscopy for biogenic NO detection in human breath,” Y. A. Bakhirkin, A. A. Kosterev, C. Roller, **R. F. Curl**, and **F. K. Tittel**, Conference on Lasers and Electro-Optics/International Quantum Electronics Conference (CLEO/IQEC 2004), San Francisco, CA (May 16-21, 2004).

- “Applications of quantum cascade lasers in trace gas analysis and chemical sensing”, (INVITED), **F.K. Tittel**, Y. Bakhirkin, **R.F. Curl**, A.A. Kosterev, S. So, D. Weidmann, G. Wysocki, 6th International Conference on Mid-Infrared Optoelectronics Materials and Devices (MIOMD-VI), St. Petersburg, Russia, (June 28-July 1, 2004).
- “Development of compact quantum cascade laser based trace gas sensors” S.G So, G. Wysocki, D. Weidmann, J. Patrick Frantz, A.A Kosterev, **R.F Curl** and **F.K. Tittel**, 6th International Conference on Mid-Infrared Optoelectronics Materials and Devices (MIOMD-VI) , St. Petersburg, Russia, (June 28-July 1, 2004).
- “Development of optical trace gas monitoring technology for NASA human space flight,”(INVITED), J. H. Miller, A. R. Awtry, B. McAndrew, **F. K. Tittel**, A. A. Kosterev, R. Q. Yang, C. J. Hill, C. M. Wong, B. Yang, and G. Bearman, 34th International Conference on Environmental Systems (ICES 2004), Colorado Springs, CO, (July 19-22, 2004).
- “Quartz-enhanced photoacoustic spectroscopy with semiconductor lasers: the road to ultracompact trace gas sensor “, A.A. Kosterev, Y. A. Bakhirkin, and **F.K. Tittel**, Novosibirsk, Russia, 4th International Symposium on Modern Problems of Laser Physics, (MPLP 2004), Novosibirsk, Russia, (Aug. 22-27, 2004)
- “Industrial applications of pulsed quantum cascade laser analyzer for trace-gas monitoring”, G. Wysocki, A. A. Kosterev, Stephen So, and **F. K. Tittel**, XV Intl. Symposium on Gas Flow Chemical Lasers and High Power Laser Conference (GCL/HPL 2004), Prague, Czech Republic, (Aug. 30-Sept. 3, 2004).
- “Compact Quantum Cascade Laser Based $^{13}\text{CO}_2/^{12}\text{CO}_2$ Isotopic Ratiometer for Field Measurements of Volcanic Gases”, **F.K. Tittel**, D. Weidmann, C. Roller, and **R.F. Curl**, (INVITED), IAEA Intl. Workshop on Stable Isotope ratio measurements by means of infrared spectrometry (SIRIS 2004), Vienna, Austria (Sept. 6-8, 2004).
- “Development of quantum cascade laser based sensor technology for trace gas monitoring applications”, **F.K. Tittel**, Yu. Bakhirkin, **R.F. Curl**, A.A. Kosterev, S. So, D. Weidmann, and G. Wysocki, 5th Quantum Cascade Laser Workshop, Fraunhofer Institute for Physical Measurement Techniques, Freiburg, Germany, (Sept. 23-24, 2004)
- “Exhaled human breath analysis by means of quantum cascade laser based gas sensors”, G. Wysocki, M. McCurdy, S. So, C. Roller, and **F. K. Tittel**, Breath Gas Analysis for Medical Diagnostics, Dornbirn, Austria, (Sept. 23-26, 2004).
- “Recent advances in quartz-enhanced gas-phase photoacoustic spectroscopy”, **F. K. Tittel**, A.A. Kosterev, and Y. Bakhirkin, 3rd IEEE Intl. Conference on Sensors, Vienna, Austria (Oct. 24-27, 2004).
- “Interband Cascade Laser based formaldehyde sensor using Quartz-enhanced Photoacoustic Spectroscopy”, Y.A. Bakhirkin, M. Horstjann, A. A. Kosterev, and **F.K. Tittel**, Optoelectronics 2005, Photonics West, San Jose, CA (Jan. 22-27 2005).

“Room-Temperature Continuous-Wave Quantum Cascade Laser based Nitric Oxide Measurements using Integrated Cavity Output Spectroscopy”, Y. A. Bakhirkin, and **F. K. Tittel**, SPIE Optics East Conferences: Infrared to terahertz technologies for health and the environment (LS6010), Boston (October 23-26, 2005)

F. List of Publications (March 15, 2003 – April 4, 2006)

1. Yu. A. Bakhirkin, A. A. Kosterev, C. Roller, **R. F. Curl, and F. K. Tittel**, “Mid-Infrared Quantum Cascade Laser based Off-Axis Integrated Cavity Output Spectroscopy for Biogenic NO Detection,” *Applied Optics* **43**, 2257-2266, (April 2004).
2. A.A. Kosterev, Y.A. Bakhirkin, **F.K. Tittel**, S. Blaser, Y. Bonetti, and L. Hvozdar, “Photoacoustic phase shift as a chemically selective spectroscopic parameter”, *Applied Physics B*, (Rapid Communications), **78**, 673-676, (April 2004).
3. D. Weidmann, A.A. Kosterev, **F.K. Tittel**, N. Ryan, D. McDonald, “Application of widely electrically tunable diode laser to chemical gas sensing with quartz-enhanced photoacoustic spectroscopy”, *Optics Letters* **29**, 1837-1839 (August 2004).
4. G. Wysocki, M. McCurdy, S. So, D. Weidmann, C. Roller, **R.F. Curl, and F.K. Tittel**, “Pulsed quantum cascade laser based sensor for trace-gas detection of carbonyl sulfide”, *Applied Optics* **43**, 6040-6046, (2004).
5. A. A. Kosterev and **F.K. Tittel**, “Ammonia detection using quartz-enhanced photoacoustic spectroscopy with a near-IR telecommunication diode laser”, *Applied Optics* **43**, 6213-6217, (2004).
6. M. Horstjann, Y. A. Bakhirkin, A. A. Kosterev, **R. F. Curl, and F. K. Tittel**, “Formaldehyde sensor using interband cascade laser based quartz-enhanced photoacoustic spectroscopy”, *Applied Physics B*, **79**, 799-803 (2004).
7. D. Weidmann, **F.K. Tittel**, T. Aellen, M. Beck, D. Hofstetter, J. Faist, and S. Blaser, “Mid infrared trace gas sensing with a quasi-continuous wave Peltier-cooled distributive feedback quantum cascade laser”, *Applied Physics B*, **79**, 907-913, (2004).
8. A. A. Kosterev, Y. A. Bakhirkin and **F. K. Tittel**, “Ultrasensitive gas detection by quartz-enhanced photoacoustic spectroscopy in the fundamental molecular absorption bands region” *Applied Physics B* **80**, 133-138, (2005).
9. D. Weidmann, G. Wysocki, C. Oppenheimer, **F.K. Tittel**, “Development of a compact quantum cascade laser spectrometer for field measurement of $^{13}\text{CO}_2/^{12}\text{CO}_2$ ”, *Applied Physics B* **80**, 255-260, (2005).
10. A. A. Kosterev, **F.K. Tittel**, D. Serebryakov, A. Malinovsky and A. Morozov, “Applications of Quartz Tuning Fork in Spectroscopic Gas Sensing”, *Journal of Scientific Instruments Review* **76**, 043105 (2005).

11. Yu. A. Bakirkin, A. A. Kosterev, **R. Curl**, **F. K. Tittel**, D. A. Yarekha, L. Hvozdar, M. Giovannini, and J. Faist, “Sub-ppbv Nitric Oxide Concentration Measurements using CW Room-Temperature Quantum Cascade Laser based Integrated Cavity Spectroscopy”, *Applied Physics B* **82**, 149-154, (2006).

TAMU Award # R82180 FINAL REPORT (04/05/04-04/04/06)

“Quantum Optics Initiative”

Frank K. Tittel and Robert F. Curl

Rice University

A. Proposed research

The proposed research effort was focused on the development and demonstration of a broadly tunable external cavity quantum cascade laser (EC-QCL) source centered at 5.2 microns.

The proposed technical objectives for such an external cavity EC-QCL system included the following:

1. Tuning range: 100 cm^{-1} or greater,
2. Output power: $\sim 10\text{ mW}$, average,
3. Laser linewidth: $< \sim 0.1\text{ cm}^{-1}$,
4. Non-cryogenic operation: near room temperature to -30C with thermoelectric cooling.

B. Methodology

A novel EC-QCL architecture was designed and implemented. The instrument employed a piezo-activated cavity mode tracking system for modehop free operation. The mode-tracking system provides independent control of the EC length and diffraction grating angle. The flexibility of this arrangement allows the instrument to be used with other QC lasers at other wavelengths without changing the EC configuration. The system performance and spectroscopic application capability was demonstrated with a QCL gain medium operating at $\sim 5.2\text{ }\mu\text{m}$.

The optical configuration of the EC-QCL is schematically shown in Fig. 1. The Littrow type EC arrangement consist of three main elements: the QCL chip, a beam collimating lens (24 mm-diameter $f/0.6$ germanium aspheric lens with a $3\text{--}12\text{ }\mu\text{m}$ AR coating), and a diffraction grating. The first order diffraction from the grating reflected with an efficiency of $\sim 95\%$ provides the laser feedback, while the zeroth order reflection provides the output laser beam. The grating, GR, is mounted on the moving platform, which allows independent control of the EC length and grating angle. The platform position is controlled by a piezoactuated linear translation stage as well as a rotary stage, which is equipped with a motorized coarse angle control and piezo-actuated fine control. The performance of the EC-QCL exhibits coarse single mode tuning over 35 cm^{-1} and a continuous mode-hop free fine tuning range of $\sim 1.2\text{ cm}^{-1}$ as depicted in Fig. 2. In the

future it will be possible to extend the mode hop free tuning range to $> 200 \text{ cm}^{-1}$ with room temperature, cw QCLs in the spectral range from 4.3 to 12 μm . The overall spectrometer system performance was demonstrated by means of direct absorption spectroscopic measurements of NO and H_2O at reduced pressures. The main limitations in the scanning speed of the actual prototype system result from the vibrational resonances of the mechanical construction, which can be addressed in future EC QCL designs.

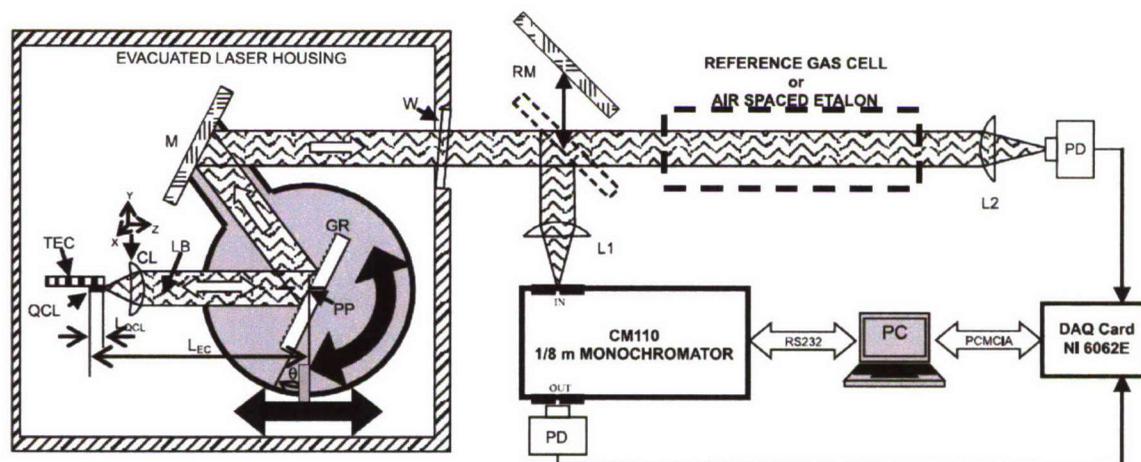


Fig. 1 Schematic diagram of the EC QCL laser and the associated measurement system. QCL – quantum cascade laser, TEC – thermoelectric cooler, CL – collimating lens (1" diameter, $f/0.6$, Ge AR-coated 3–12 μm) mounted on a motorized 3D translation stage, LB – laser beam, GR – diffraction grating (150 gr/mm blazed for 5.4 μm), PP – pivot point of the rotational movement, M – mirror (mounted on the same platform with GR), W – CaF2 window (thickness 4 mm, tilted $\square 5^\circ$), RM – removable mirror, PD – photodetector (Hg-Cd-Zn-Te, TE-cooled, Vigo Systems, PDI-2TE-6), L1, L2 – ZnSe lenses

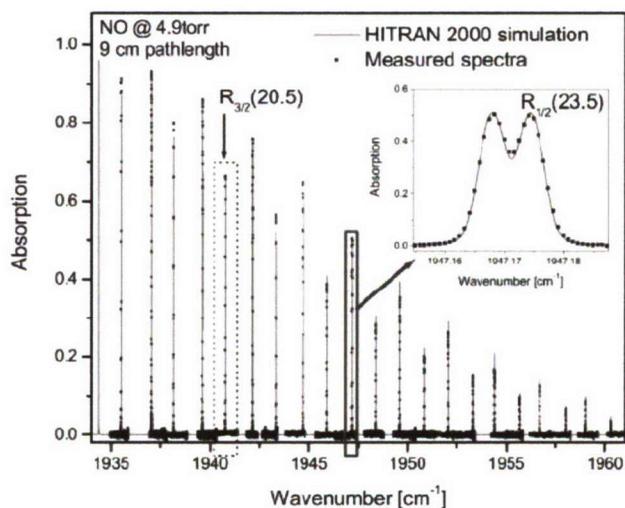


Fig. 2 Nitric oxide absorption spectra measured at different diffraction grating angles of the external cavity quantum cascade laser. The narrow laser linewidth allows resolving two spectral peaks separated by $\sim 0.006 \text{ cm}^{-1}$ (see inset)

C. Research funding

1. BROADLY TUNABLE INFRARED QUANTUM CASCADE LASER TECHNOLOGY FOR REMOTE SENSING , Department of Energy (DoE); (\$ 50,000), 07/01/06-03/31/07
2. LOW COST INTEGRATED IR QUARTZ ENHANCED PHOTOACOUSTIC GAS SENSOR, National Science Foundation STTR; (\$ 50,000), 01/01/06-12/31/06
3. MID INFRARED TECHNOLOGIES FOR THE HEALTH AND ENVIRONMENT (MIRTHE), National Science Foundation; (\$ 86,000), 07/01/06-06/30/07

D. Graduate Students

C. Roller (Ph.D May 2005), now with Ekips Technologies
 S. So (M.Sc May 2005), now in the Rice Ph.D program
 M. McCurdy (in the Rice Ph.D/MD BCM programs)
 R. Lewicki (in Ph.D program)
 T. Mosely (M.Sc May 2005)

E. List of Conferences (March 15, 2003-April 4, 2006)

- “Recent advances in quartz-enhanced gas-phase photoacoustic spectroscopy”, **F. K. Tittel**, A.A. Kosterev, and Y. Bakhirkin, 3rd IEEE Intl. Conference on Sensors, Vienna, Austria (Oct. 24-27, 2004).
- “Recent advances of trace gas sensors based on diode and quantum cascade lasers”, **F.K. Tittel**, A.A. Kosterev, Yu. Bakhirkin, G. Wysocki, C. Roller, S. So, D. Weidmann, and **R.F. Curl**, 2004 Annual IEEE LEOS Meeting, Rio Mar, Puerto Rico, (Nov. 7-11, 2004).
- “Sensitive trace gas detection with quantum cascade lasers”, G. Wysocki, Y. A. Bakhirkin, A. A. Kosterev, S. So, D. Weidmann, and **F. K. Tittel**, IEEE/LEOS Seminar, Wroclaw, Poland (Dec. 22, 2004)
- "Quantum cascade laser based trace-gas sensors for human breath analysis", G. Wysocki, Matt McCurdy, Stephen So, Chad Roller, and **F.K. Tittel**, 35th Winter Colloquium on The Physics of Quantum Electronics Snowbird, Utah, (January 2-6, 2005)
- “Recent developments of quantum cascade laser based trace gas sensor technology: opportunities and challenges”, **F.K. Tittel**, Y. Bakhirkin, **R.F. Curl**, A.A. Kosterev, S. So, D. Weidmann, G. Wysocki, Optoelectronics 2005, Photonics West, San Jose, CA (Jan 22-27, 2005).
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- “Ultra sensitive gas detection by quartz enhanced photoacoustic spectroscopy”, **F.K. Tittel**, A.A. Kosterev, Yu. Bakhirkin, G. Wysocki, S. So and **R.F. Curl**, Gordon Research Conference on Photoacoustic and Photothermal Phenomena, Trieste, Italy (June 26-July 1, 2005)
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“Widely Tunable External Cavity Quantum Cascade Laser for High Resolution Spectroscopy and Trace-gas Detection”, G. Wysocki, **R. F. Curl, F. K. Tittel**, R. Maulini, J. M. Bulliard, J. Faist, SPIE Optics East Conferences: Infrared to terahertz technologies for health and the environment (LS6010), Boston (October 23-26, 2005)

“Semiconductor laser based trace gas sensor technology recent advances and applications”, **F.K. Tittel**, A.A. Kosterev, Y. Bakhirkin, G. Wysocki, S.G. So, and **R.F. Curl**, Mid-Infrared Coherent Sources (MICS) 2005, Barcelona, Spain (November 6-11, 2005)

“Widely Tunable Mode-hop Free External Cavity Quantum Cascade Laser for High Resolution Spectroscopic Applications”, G. Wysocki and **F.K. Tittel**, Mid-Infrared Coherent Sources (MICS) 2005, Barcelona, Spain (November 6-11, 2005)

“Semiconductor laser based trace gas sensor technology: advances and opportunities”, **F. K. Tittel**, Y. Bakhirkin, A. Kosterev, M. McCurdy, S. So and G. Wysocki, 36th Winter Colloquium on the Physics of Quantum Electronics Snowbird, Utah, (January 2-6, 2006)

“Development of Optical Trace Gas Monitoring Technology for NASA Human Space Flight” J. Houston Miller, Brendan McAdrew, Anatoliy A. Kosterev , Y. A. Bakhirkin, Gerard Wysocki, **F. K. Tittel** and Rui Q. Yang, HABITATION 2006, Orlando, FL (February 5-8, 2006)

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“Trace Humidity Sensor based on Quartz-Enhanced Photoacoustic Spectroscopy” , A. Kosterev, **F. K. Tittel**, T. S. Knittel, A. Cowie, and J. D. Tate, Laser Application to

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“Detection of formaldehyde using off-axis integrated cavity output spectroscopy with an interband cascade laser“, J. H. Miller Y. A. Bakhirkin, T. Ajtai, **F.K. Tittel**, C.J. Hill, B. Yang, and R. Q. Yang, Laser Application to Chemical Security and Environmental Analysis (LACSEA 2005), Incline Village, NV (February 5-9, 2006)

F. List of Publications (April 5, 2004 – April 4, 2006)

1. G. Wysocki, **R. F. Curl**, **F. K. Tittel**, R. Maulini, J. M. Bulliard, J. Faist, “Widely Tunable Mode-hop Free External Cavity Quantum Cascade Laser for High Resolution Spectroscopic Applications”, Applied Physics B **81**, 769-777 (2005)
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3. **F.K. Tittel**, Y. Bakhirkin, A. Kosterev and G. Wysocki, “Recent advances in trace gas detection using quantum and interband cascade lasers”, The Review of Laser Engineering **34**, 275-282 (2006)
4. J. H. Miller, Y. A. Bakhirkin, T. Ajtai, and **Frank K. Tittel**, “Detection Of Formaldehyde Using Off-Axis Integrated Cavity Output Spectroscopy With An Interband Cascade Laser” to appear in Applied Physics B (July 2006)
5. A. A. Kosterev, T. S. Mosely, and **F. K. Tittel**, “Impact of humidity on quartz enhanced photoacoustic spectroscopy based detection of HCN, to appear in Applied Physics B (July 2006)
6. S. G. So, G. Wysocki, J. P. Frantz, **F. K. Tittel**, “Development of Digital Signal Processor controlled Quantum Cascade Laser based Trace Gas Sensor Technology”, to appear in IEEE Sensors Journal, (October 2006)

4. Baylor University

Principal Investigator: Carlos E. Manzanares

“Thermal Lensing and Thermal Grating Spectroscopy in Cryogenic Solutions”

In this proposal the thermal lens technique was used for the first time to study vibrational overtone absorptions of hydrocarbons dissolved in cryogenic liquids. In particular, the C-H ($\Delta v = 6$) overtone spectra of hydrocarbons dissolved in liquefied Ar, Xe, Kr, and N₂. Using CH₄-Ar and CH₄-N₂ solutions; a complete analysis of the thermal lens intensities was made as a function of the concentration of CH₄, solvent (Ar or N₂), limits of detection, and time dependent profiles of the signals for single and dual beam

experiments. Experimental results were used to test theoretical models of thermal lens intensities. The overtone spectra of alkanes: ethane, propane, butane, and isobutane were obtained, as well as the spectra of the alkenes: ethylene, propylene, isobutylene, cis-2-butene, and trans-2-butene in cryogenic solutions between 80 K and 140 K. We were able to obtain signals for concentrations as low as 1×10^{-4} mole fraction. The limit of detection of the thermal lens technique was estimated around 10^{-5} - 10^{-6} mole fraction.

We are now planning experiments using another opto-thermal technique (thermal grating) to study unimolecular chemical reactions of alkenes in cryogenic liquids. The studies will allow us to obtain the enthalpy of reaction (ΔH), the change in molecular volume (ΔV) from reactants to products, the thermal diffusion coefficient (D_{thermal}), and the molecular diffusion coefficients (D_i) of reactants and products. Initial experiments involve the ultraviolet and visible photochemistry of cyclobutene in liquefied ethane, methane, argon, and nitrogen.

Graduate Student Support: The following students have received their Ph.D. degrees with partial support from the Quantum Optics Initiative:

<u>Name</u>	<u>Year</u>	<u>Dissertation</u>
Dr. Craig J. Moehnke	Aug-2005	"Low temperature Phase Shift Cavity Ring Down: Applications to High Vibrational Overtones"
Dr. Alfredo Lopez-Calvo	Aug-2006	"Vibrational Spectroscopy in Cryogenic Solutions: Application of Thermal Lensing and Fourier Transform Techniques to the Study of Molecular C-H Overtone Transitions"
Dr. Juan G. Navea	Aug-2006	"Studies on the Application of Laser Photo-ionization of Molecular Beams for the Generation of Ionic Clusters"

Publications: The following papers have been published acknowledging partial support from the Quantum Optics Initiative:

1. J. G. Navea, A. Lopez-Calvo, C. E. Manzanares, "Thermal Lens Spectroscopy in Liquid Argon solutions: ($\Delta v = 6$) C-H Vibrational Overtone Absorption of Methane", J. Phys. Chem. A, 110, 1594 -1599 (2006).

2. C. J. Moehnke, E. K. Lewis, A. Lopez-Calvo, and C.E. Manzanares, "Phase shift cavity ring down at low temperatures: vibration-rotation overtone absorption of H-D ($\Delta v = 4$) at 297 and 105 K", Chem. Phys. Lett., 418, 576-580 (2006).
3. E. K. Lewis, C. J. Moehnke, and C. E. Manzanares "Phase Shift Cavity Ring Down and FT-VIS Measurements of C-H ($\Delta v = 5$) Vibrational Overtone Absorptions", Chem. Phys. Lett., 394, 25-31 (2004).
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5. A. Lopez-Calvo, C. E. Manzanares, "Thermal Lens Spectroscopy in Cryogenic Solutions: Analysis and Comparison of Intensities in CH₄-N₂ and CH₄-Ar Liquid Solutions", J. Phys. Chem. A, (submitted) 2006.

Principal Investigator: Kenneth T. Park

"Investigation of Heteroepitaxial Molecular Thin Films"

I. Scientific Background

Ultra thin organic films are of great technological interest. The properties can be tailored to the desired specifications both by limitless engineering of organic molecules and by precisely controlling thickness and sequence in an atomically clean environment. The promise of novel characteristics and properties unique to a molecule-metal interface has stimulated significant research activities in the past decades on various thin organic films for the potential use as functional components in displays, sensors, electronic, photovoltaic, and nonlinear optical devices [1-3].

The great diversity of interfacial phenomena involving ultra thin organic films further offers an opportunity to explore chemistry and physics of systems with reduced dimensions. The nature of bonding, the binding process of a molecule, alteration in both the molecule and the surface electronic structures, and their effect on subsequent adsorption must be understood before the two-dimensional motion and the organization of adsorbates are addressed. Knowledge of molecular motion and binding with step edges and other surface defects is also essential as they play a crucial role, influencing adsorption, desorption, reaction kinetics, and long-range ordering of molecular overlayers.

Objectives of the Research Program

This research has focused on a specific aspect of the intermolecular and the molecule-surface interaction, exemplified in metallo-phthalocyanines (MPc, M = Co, Ni, Cu) on a "5x20" Au(001) surface. The specific goals were (1) to determine the nature of bonding and the role of surface states in MPc-metal interfaces and (2) to establish a quantitative and predictive picture of the effects of step edges on MPc molecules at the initial adsorption stage.

Summary of Research Activity

During the summer of 2003, we achieved the deposition and thickness control of ultra-thin NiPc and CoPc films with sub-monolayer precision. The *in situ* grown films were investigated using the high resolution ultraviolet photoemission spectroscopy (UPS) and low energy electron diffraction (LEED) at National Synchrotron Light Source, Brookhaven National Laboratory (BNL). During the winter of 2003, heteroepitaxial MPc molecules were observed using a variable temperature scanning tunneling microscopy (STM) in the Center for Functional Nanomaterials (CFN) at Brookhaven National Laboratory (BNL).

Ultrathin layers of NiPc were formed onto a "5 x 20" Au(001) at room temperature by systematically increasing the total deposition time from 45 to 270 seconds [4]. The nominal thicknesses of deposited films were calculated through the Au 4f_{7/2} core level intensity attenuation and estimated to be 1.4, 3.0, 5.6, 7.3, and 11.8 Å, corresponding to the total depositions of 45, 90, 135, 180, and 270 seconds, respectively. Since MPc molecules are known to adsorb in its flat-lying geometry on Au(001)_{HEX} as well as on many other single crystal surfaces, the estimated values correspond to 0.4, 0.9, 1.6, 2.1, and 3.5 ML, where 1 monolayer (ML) is defined as 3.4 Å, the approximate interlayer spacing [5].

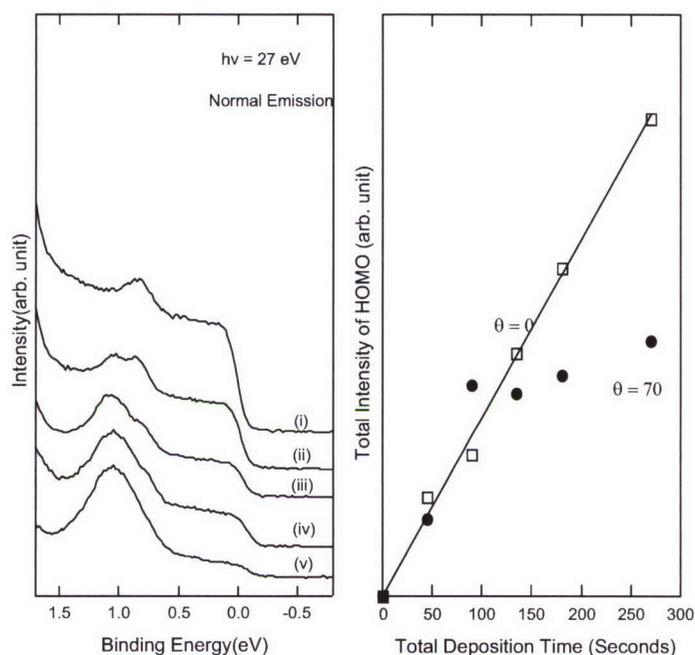


Fig. 1 (Left) The UPS spectra in the valence band region, taken with $h\nu = 27$ eV and photoelectrons measured along $\theta = 0^\circ$ from a reconstructed Au(001) (topmost EDC), after depositing NiPc for total (i) 45, (ii) 90, (iii) 135, (iv) 180, and (v) 270 seconds. (Right) The intensity of HOMO measured as a function of the total deposition time is shown for $\theta = 0^\circ$ and $\theta = 70^\circ$.

After the first 45 seconds of NiPc evaporation onto the "5 x 20" Au(001), the highest occupied molecular orbital (HOMO) of NiPc appears at the binding energy of 0.84 eV from the Fermi level (EDC (i) in Fig. 1). The HOMO is believed to be entirely of phthalocyanine character in A_{1u} symmetry of the D_{4h} group [6]. The full-width-at-half-maximum (FWHM) of the HOMO is *ca* 186 meV is comparable to that of CuPc molecules in gas phase [7]. The deposited NiPc overlayer results in no new LEED spots. Only faint streaks at 1/5th order spots are visible with the significantly increased background intensity, suggesting the presence of perhaps partially ordered NiPc islands. After 90 seconds of the total deposition (0.9 ML), a new component at 1.06 eV appears side-by-side with the lower binding energy peak (0.84 eV) from the first deposition. The doublet can be fitted with two components at the minimum. A simple Gaussian fitting reveals that the higher binding energy component has its FWHM of 253 meV, which is only slightly broader than that of the lower energy component (186 meV).

The surface exhibits a distinctive LEED pattern resulting from a highly ordered NiPc overlayer. The surface unit mesh vectors of the NiPc overlayer are azimuthally aligned with $\langle 110 \rangle$ and $\langle 1\bar{1}0 \rangle$ of Au(001). The size of the square unit mesh is estimated approximately 12.8 \AA by 12.8 \AA as the (10) spots from NiPc overlayer coincide with the 1/5th order spots from the substrate within experimental error. The measured unit cell size of NiPc is well within the range of the reported values for the heteroepitaxial CuPc on Au(001) [8,9] and Au(111) [10]. Further deposition of NiPc results in the increase in the intensity for the higher binding energy component. The binding energy component gradually shifts from 1.06 eV to 1.13 eV, and at the same time the FWHM continues to broaden from 253 meV to 397 meV.

In order to get further insight into the nature of the HOMO peak shift by 0.22 eV at the very early stage of thin film formation, other MO positions of NiPc at ultra-thin coverages are examined. The difference VB spectra clearly reveal that both the $6e_g$ MO at 2.5 eV (not shown) and the $2a_{1u}$ HOMO broaden and shift toward higher binding energy with increasing coverage [4]. Such behavior on other lower-lying MOs can be also recognized although much larger FWHMs of the experimental peaks (perhaps representing closely spaced MOs) make it subtler. The uniform energy shift observed from more than one MO with very different orbital characteristics, indicates that the interaction between NiPc and Au(001)_{HEX} at the very initial adsorption is not chemisorptive but rather electrostatic, for example, due to the variation in the local electronic potential perhaps due to the initial adsorption at step edges, which NiPc molecules experience for sub-monolayer coverages.

The variation in the local electronic environments can have a significant effect on adsorption even if the bonding is physisorption-type. For Au(111) surface, which the hexagonally reconstructed Au(001) closely resembles, it is well known that the occupied Shockley surface states are located at 0.4 eV below the Fermi level [11]. Shockley surface states are spatially extended and these *sp* states can have increased overlap in charge density with the adsorbate. Bertel has proposed that they can contribute to the repulsive part of physisorption potential well and weaken the physisorption strength when occupied [12]. Above step edges, the density of occupied surface states decreases while it increases near lower edges of the steps. Therefore, physisorbed adatoms or

molecules such as MPc may have a higher affinity for binding at upper step edges, compared to terraces as well as lower step edges because of the decrease in the occupied surface state density. The stronger bonding can bring the molecule closer to the surface for more efficient screening by the conduction electrons hence the observed binding energy shift toward the Fermi level.

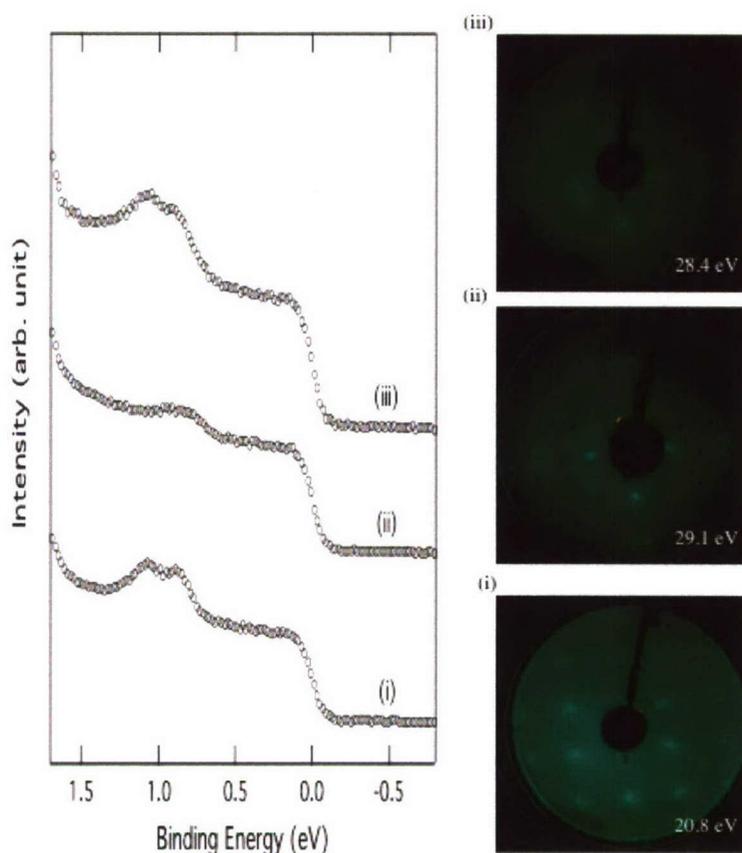


Fig. 2 (Left) The photoemission EDCs in the region of the NiPc HOMO peak, taken along surface normal, with $h\nu = 27$ eV: (i) after 90 seconds of NiPc deposition on a reconstructed Au(001) (≈ 0.9 ML NiPc), (ii) after heating NiPc/Au(001) up to 500 °C for 5 minutes, and (iii) after additional 45 seconds of NiPc on the NiPc/Au(001) (≈ 1 ML NiPc). (Right) The LEED images corresponding with the experimental conditions for EDCs (i), (ii), and (iii) are also shown.

This model also predicts that a larger adsorption enthalpy for MPc should be expected from the initial adsorption. 0.8 ML of heteroepitaxial NiPc was first formed (EDC (i) of Fig. 2) and then heated up to $T = 500$ °C - well beyond the typical sublimation temperature - for 5 minutes. The intensity of the HOMO peak decreases to about 25 % of the initial value (EDC (ii) of Fig. 2). Clearly, the HOMO peak is entirely

of the lower binding energy component, and no high binding energy component is observed after heating [4]. The LEED pattern further shows fractional spots with doublets, characteristic to a "5x20" reconstructed Au(001), with a higher diffused background intensity but no LEED spots from NiPc, suggesting the presence of only disordered NiPc. The remaining NiPc after heating is believed to be mostly molecular, evidenced in the difference valence band spectrum after heating, which reveals the same MO features observed during the earlier "layer-by-layer" deposition experiment.

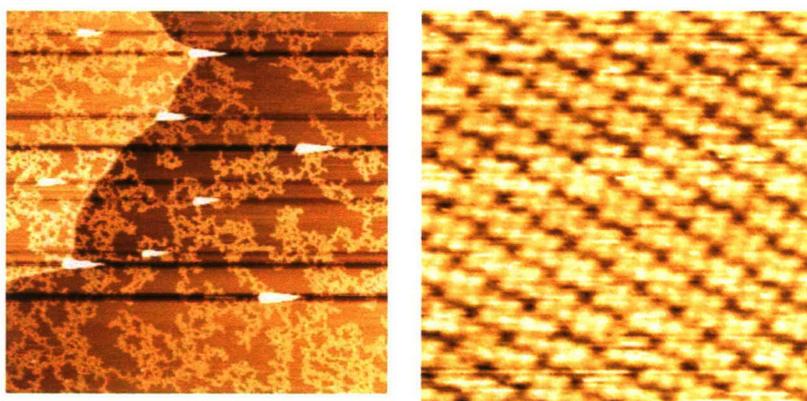


Fig. 3 (left) STM Image (400 nm x 400 nm) of CuPc on Au(111) after heating the heteroepitaxial layers up to 700 K for 30 minutes. $V = -1.54$ V and $I = 0.032$ nA; Fig. 4 (right) STM Image (14.6 nm x 14.6 nm) of heteroepitaxial CuPc multilayers formed on Au(111). $V = -1.37$ V and $I = 0.03$ nA

Our recent temperature-dependent STM study of CuPc/Au(111) [13] confirms that the residual CuPc adsorbates after heating up to 700 K for 30 minutes are most likely molecular, evidenced by

the distinctive "four-lobed" shape of MPc and moreover that the remnants are disordered in "dendritic" formation (Fig. 3). Such an uncharacteristically strong bond also has been reported for other organic molecules on a metal surface. On Cu(111), most benzene molecules desorb at 250 K, but some bind more strongly on step edges and other defect sites, resulting in the high temperature tail in the temperature-programmed desorption data [14]. For several endcapped oligothiophenes, and quaterthiophene ($\alpha 4T$) on Ag(111), the desorption peak of the monolayer at a higher temperature is clearly distinguishable from that of multilayers [15], albeit a covalent-type bonding between the

π orbitals and the Ag 5s states has been suggested. In our experiments, after depositing NiPc onto the surface for 45 seconds, the total intensity of the HOMO grows nearly equal to that of the initially deposited NiPc with the high binding energy component clearly present (EDC (iii) of Fig. 4). The binding energies of the doublet are almost identical to those measured before heating (EDC (i) of Fig. 4) as well as other MOs gleaned from the difference spectrum (Fig. 5). Intriguingly, the LEED shows no spots other than those from the substrate, indicating that the overlayer still lacks the long-range order.

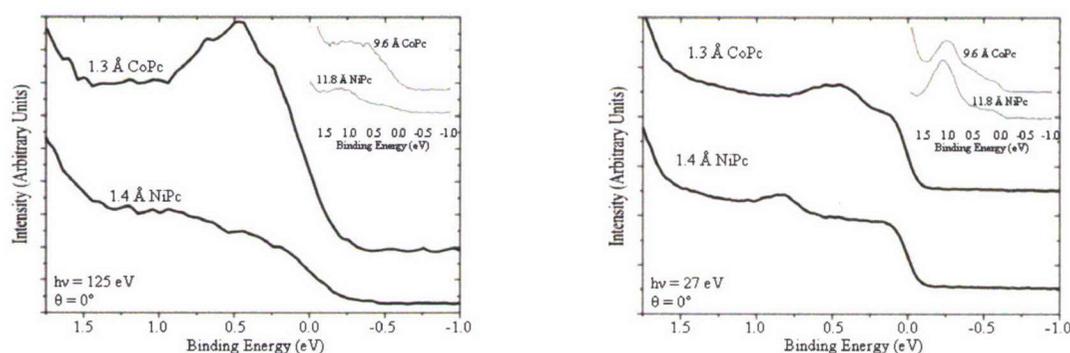


Fig. 5 (Top) The photoemission EDCs in the region of the HOMO peak for thin NiPc and CoPc on a reconstructed Au(001) with $h\nu = 27$ eV. (Bottom) The photoemission EDCs with $h\nu = 125$ eV for the same NiPc- and CoPc- covered surface as in Top. In the inset, the EDCs for thicker NiPc and CoPc are also shown.

The interaction of CuPc with a metal surface is much like that of NiPc detailed in this report. The interaction can be characterized as physisorption-type. Both form highly ordered, heteroepitaxial layers with the close-packed square lattice at room temperature, from about a monolayer and beyond (Fig. 4). On contrary, CoPc at room temperature forms only one-dimensional chains, and domains are relatively oriented at 15° from each other [9,16]. The contrast between CoPc and other MPc ($M = \text{Ni, Cu}$) further manifests in the electronic structure of the interfacial molecules. The interfacial CoPc molecules show a large density of states at 0.5 eV, much closer to the Fermi level than 0.84 eV measured for interfacial NiPc (Fig. 5). Furthermore, the peak is much more pronounced in the photoemission spectra using the photon energy 125 eV than in those using 27 eV.

Because the photoionization cross section for the d sub-shell is much larger than that of either s or p shells at 125 eV [9], the observed interfacial states near the Fermi level may be indicative of a significant d orbital contribution. One possible explanation is that a partially occupied $13 a_{1g}$ (mostly Co d_{z^2}) [6] hybridizes with the surface states in a chemisorptive bonding and perhaps related to the orbital mediated tunneling [17]. Understanding the exact nature of the interaction between CoPc and Au is currently under progress.

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The following graduate students have been involved in this project:

- Mr. Trinity Ellis
- Mr. Kedar Manandhar
- Mr. Aroshan Jayasinghe

Conference Presentations:

- "Low Energy Electron Diffraction and Ultraviolet Photoelectron Spectroscopy Studies of Metallo-Phthalocyanine Thin Films," American Physical Society March Meeting, Montreal, Canada, March 22-26, 2004. Oral Presentation by Ken T. Park
- **Invited** "Molecular Interaction and Ordering in Metallo-phthalocyanine Thin Films," Oak Ridge National Laboratory, Oak Ridge, TN, February 27, 2004. Oral Presentation by Ken T. Park
- "Heteroepitaxial Metallo-phthalocyanine (M = Co, Ni, Cu) Thin Films on a "5x20" Reconstructed Au(001)," Strategic Partnership for Research In Nanotechnology (SPRING), Austin, TX. August 26, 2003. Poster Presentation by Trinity Ellis.

The following manuscripts have been either submitted or prepared for publication:

- [4] "Interactions of Nickel Phthalocyanine Molecules on "5x20" Au(001) During the Initial Formation of Heteroepitaxy," Ken T. Park, Trinity Ellis, Steve L. Hulbert, Marc Ulrich, and Jack E. Rowe, submitted to *Chem. Phys. Lett.* (2004).
- [17] "Metallo-Phthalocyanine (MPc, M = Co, Ni) Thin Films: A UPS and LEED Study," T. Ellis, K.T. Park, S.L. Hulbert, M.D. Ulrich, and J.E. Rowe, submitted to *J. Appl. Phys.* (200).
- [13] "Variable Temperature STM study of Heteroepitaxial Copper Phthalocyanine," T. Ellis, K. Manandhar, K. T. Park, Z. Song, T. Cai, and J. Hrbek, in preparation.

Principal Investigator: Dwight P. Russell

1. Electron Irradiation of Alkali Halides:

This work will continue my studies of the effects of relatively low energy (50 eV –5000 eV) electron beam irradiation on alkali halides. My work will focus upon two aspects. First the formation of alkali clusters of the surface of an alkali halide. Temperature programmed desorption studies show that these follow a Volmer-Weber type desorption law with fractional order desorption. No work of this type has been done for the other alkali halides. This provides a field of study ready for development.

2. Chemical Reactivity of Irradiated Surfaces.

As a feature of the research discussed above, we have shown that the alkali metal clusters on the surface form reactive site. In particular, we showed that hydrogen adsorbs only to the alkali rich regions of the surface. I hope to explore this property by exposing other gases to the surface and study the nature of the deposited material. An example would be disilane (Si₂H₆). Exposure of disilane to the irradiated alkali halide is expected to deposit silicon on the surface. This would allow for patterning of the deposited material since it would deposit only on the alkali rich region. The nature of the deposited material, crystalline, polycrystalline or amorphous, pure or compound is unknown.

The following students have completed degrees with partial support from the Quantum Optics Initiative:

Du Fei Master in Physics completed 2006
Zhang, Zhiwen Master's in Physics completed 2006

Presentations supported in part by the Quantum Optics Initiative:

Invited - 'Electron Irradiation Effects on Alkali-Halides,' ,Trinity University, SanAntonio, April 22, 2003.

Contributed –'Quantum Beats in Thin Foil and Surface Scattering of 10keV-30keV H⁺ ions' Joint TSAPS, TSAAPT, SPS 2004 Spring Meeting, Tarlton State University, Stephenville Tx, Apr 1-3, 2004

Publications supported in part by the Quantum Optics Initiative:

'Role of Surface Dimer and Gas Phase Excitation Models in Electron Stimulated Desorption of Ions from Sodium Chloride(100) Crystals,' D. P. Russell, W. Durrer, Rad. Eff and Def in Sol, 160 (2005) 151-154.

Principal Investigator: Walter Wilcox
“The Effect of Unruh Radiation on Spin State”

This research employed a model of randomly varying B field with a characteristic correlation time of L/c (L^3 being volume of a box with the particle in it) to model the effect of unruh radiation on a accelerating electron's spin state. The electron starts out in a pure state which becomes mixed as it goes into equilibrium with the unruh temp. To leading order in the relaxation time the distribution over the mixture of states was described by a heat equation on the spin space (the surface of a sphere in 4D) with a real time dependent diffusion coefficient, where the density matrix was an integral over that distribution times the outer product of that state with itself. In flat space then a minimum amount of mixing was found to occur when the system had different incoming and outgoing velocity, which was given by an expression of this form: $P' = P * \exp(-k(Y-1)) + 1 - \exp(-k(Y-1))$ where P is the incoming density matrix and P' is the outgoing, and Y is the relativistic gamma factor of the velocity difference between in and out asymptotic states, and k is a constant.

5. University of North Texas (Donald H. Kobe)

In this report we describe in Section 1 the research performed. Section 2 gives the benefits the grant has produced. Section 3 lists the conferences attended and titles of talks given on the basis of this grant. Section 4 gives a list of papers produced on the basis of this grant. In Section 5 I give some closing remarks.

1. Research Performed

In this research I proposed to investigate whether Bohmian mechanics and standard quantum mechanics are equivalent. The goals I wished to accomplish were the following. (1) To generalize Bohm's approach to include a coupling of a charged particle to a classical electromagnetic field described by vector and scalar potentials, and verify the gauge invariance of the theory. (2) To extend the Bohm approach many particles in configuration space. (3) To examine the Aharonov-Bohm effect in which a shielded solenoid containing magnetic flux behind the two slits can shift the Young's interference pattern within its unshifted envelope. (4) To investigate the transition from Bohmian quantum theory to classical mechanics by taking the limit as Planck's constant goes to zero.

I accomplished goals (1) and (2), but did not address (3) and (4). In the course of doing the research I realized that a more realistic approach was to focus on a particular problem that could be addressed by both Bohmian mechanics and standard quantum mechanics. I chose to examine the question of quantum chaos from the Bohmian viewpoint as an alternative to many different approaches using standard quantum mechanics. The Bohmian formulation enables quantum chaos to be treated in a way analogous to classical chaos, since the Bohmian formulation gives quantum trajectories, which do not exist in standard quantum mechanics. The simplest model that exhibits classical chaos is the kicked rotor. Together with a graduate student we examined the classical kicked rotor to develop numerical techniques and to study momentum diffusion in more detail than had previously been done.

Then we treated the quantum kicked rotor. Momentum diffusion for the quantum kicked rotor can be treated from both the viewpoint of standard quantum mechanics and

the Bohmian formulation. At first we obtained different results, but then we realized that when the quantum potential was added to the Bohmian kinetic energy the results agreed. We realized that the quantum potential is really part of the quantum kinetic energy and that the total quantum kinetic energy is required to calculate the momentum diffusion.

We then calculated the Lyapunov exponents that are characteristic of chaos. In standard quantum mechanics there are no Lyapunov exponents because there are no trajectories. The Bohmian formulation has the advantage that it does have quantum trajectories from which Lyapunov exponents can be calculated. We first calculated the Lyapunov exponents for the classical kicked rotor in order to compare them with the quantum Lyapunov exponents, which we calculated for the case of quantum resonance. The quantum Lyapunov exponents are similar to the classical ones, but showed only remnants of the islands of stability that the classical exponents exhibit.

This research has opened up many new problems in quantum chaos. One of the most pressing problems for future research is to investigate the classical limit of the Bohmian approach to quantum chaos as Planck's constant goes to zero. When this limit is taken, does the quantum chaos go smoothly over to the corresponding classical chaos? This question can be answered by investigating the behavior of the quantum Lyapunov exponents or by examining the corresponding Poincare sections in this limit. Since the Bohmian formulation of quantum theory is based on the Schroedinger equation, it should be completely equivalent to standard quantum mechanics. However, it can be used to investigate quantum chaos because it gives approach that is not available in standard quantum mechanics. There are many different methods for treating quantum chaos in standard quantum mechanics to which it can be compared.

2. Benefits Produced

This grant provided partial support for a Ph.D. graduate student Yindong (Richard) Zheng during his research on this project. He was supported on this grant as a research assistant for two summers and part time for a spring and fall semester. . In the summer of 2005 he was awarded a Ph.D. from the University of North Texas based on

his dissertation entitled “Quantum and Classical Chaos and Momentum Diffusion in the Kicked Rotor.” In this dissertation he applied the Bohmian approach to quantum mechanics to treat chaos and momentum diffusion in the kicked rotor in a way similar to the classical approach. One result was to show that the quantum potential is a fictitious potential in the sense that it is part of the quantum kinetic energy. Another result was to find quantum Lyapunov exponents for the quantum kicked rotor.

The grant also supported me with salary for one summer to work on this project.

Except for partial support from the University of North Texas in the form of a Teaching Assistantship for Yindong Zheng and my regular salary, no additional funding was obtained.

3. Conferences Attended and Talks Given

The research performed under this grant resulted in six contributed talks given at meetings of the Texas Section of the American Physical Society:

- 1) Donald H. Kobe, University of North Texas, “Lagrangian Densities and Principle of Least Action in Quantum Mechanics,” APS Presentation at the Annual Spring Meeting of the Texas Sections of the APS and AAPT, Angelo State University, San Angelo, Texas, 23-25 March 2006. Abstract published online at: <http://meetings.aps.org/Meeting/TSS06/Event/50313>
- 2) Donald H. Kobe, University of North Texas, “Bohmian Interpretation of Quantum Mechanics and Ehrenfest’s Theorem for Energy,” APS Presentation at the Annual Fall Meeting of the Texas Sections of the APS and AAPT, University of Houston, Houston, Texas, 20-22 October 2005. Abstract published online at <http://meetings.aps.org/Meeting/TSF05/Event/50667>
- 3) Yindong Zheng and Donald H. Kobe, University of North Texas, “Quantum Potential and Chaos of the Kicked Rotor,” APS Presentation at the Annual Joint Spring Meeting of the Texas Sections of the APS and AAPT, Stephen F. Austin State University, Nacogdoches, Texas, 3-5 March 2005. Abstract published online at: <http://meetings.aps.org/Meeting/TSS05/Event/31038>

- 4) Yindong Zheng and Donald H. Kobe, "Anomalous Momentum Diffusion in the Classical Kicked Rotor," APS Presentation at the Annual Fall Meeting of the Texas Sections of the APS and AAPT, Baylor University, Waco, Texas, 7-9 October 2004. Abstract published online at:
<http://www.aps.org/meet/TSF04/baps/abs/S80001.html>

- 5) Yindong Zheng and Donald H. Kobe, University of North Texas, "Trajectory Simulation of the Kicked Rotor and its Lyapunov Exponents," APS Presentation at the Annual Spring Meeting of the Texas Sections of the APS and AAPT, Tarleton State University, Stephenville, Texas, 1-3 April 2004. Abstract online at:
<http://www.math.tarleton.edu/physics/Conference/2004%20Joint%20Spring%20Meeting%20Program%206.doc>

- 6) Y.-D. Zheng and D. H. Kobe, [de Broglie-Bohm Approach to Quantum Mechanics Applied to Quantum Chaos: The Kicked Rotor](#), APS Presentation, Annual Fall Meeting of the Texas Sections of the APS and AAPT, Texas Tech University, Lubbock, Texas, 23-25 October 2003. Abstract online at:
<http://flux.aps.org/meetings/YR03/TSF03/baps/abs/S40005.html>

In addition, I attended the Quantum Optics Workshop Sponsored by ONR/DARPA/TAMU at Grand Targhee, Alta, Wyoming, July .7-11, 2003 with partial travel support from this grant.

4. List of Papers Published, Pending, and in Preparation

As a result of this grant two papers have been published, one paper is to be published, two papers are submitted for publication, and two more are in preparation.

Papers published:

1. Yindong Zheng and Donald H. Kobe, "Numerical solution of classical kicked rotor and local Lyapunov exponents," Phys. Lett. A **334**, 306-311 (2005).
2. Yindong Zheng and Donald H. Kobe, "Anomalous momentum diffusion in the classical kicked rotor," Chaos, Solitons and Fractals **28**, 395-402 (2006).

Paper to be published:

3. Yindong Zheng and Donald H. Kobe, "Momentum diffusion of the quantum kicked rotor: Comparison of Bohmian and standard quantum mechanics," Chaos, Solitons and Fractals (to be published 2006).

Papers submitted:

4. Yindong Zheng and Donald H. Kobe, "Bohmian Quantum Mechanical and Classical Lyapunov Exponents for Kicked Rotor," (submitted)
5. Donald H. Kobe, "Quantum Power in de Broglie-Bohm Theory," (submitted).

Papers in preparation:

6. Donald H. Kobe, "de Broglie-Bohm Theory for a Many-Particle Quantum System in an Electromagnetic Field."
7. Donald H. Kobe, "Lagrangian Density in Terms of Hamiltonian Operator in Quantum Mechanics and its Applications."

5. Summary Remarks

This grant has enabled me to investigate quantum chaos from the Bohmian approach to quantum mechanics and compare it with classical chaos. In general, the Bohmian approach to quantum mechanics is essentially equivalent to standard quantum mechanics in its results. In the area of quantum chaos it has a distinct advantage in that quantum chaos can be treated in a way similar to classical chaos because of the existence of quantum trajectories. This approach to quantum chaos has much potential. It can be compared with the many different approaches to quantum chaos based on standard quantum mechanics. There are many more systems than the kicked rotor that can be investigated. The scaled Planck's constant is a parameter that can be varied. In particular, the classical limit is obtained as it goes to zero, but an

open question is whether the quantum chaos goes smoothly over to the classical chaos.

Classical and quantum chaos is a subject that gives insight into a realm of nature that has previously been considered too complex for mathematical analysis. History has shown that as we begin to understand new phenomena, many new applications will be found:

6. University of Texas at Austin

A.) Radiation Matter Interaction

R. M. Walser, L. Reichl

This research explores a new approach for enhancing the interaction of quasi-bound states in optically confined systems, with optically excited states in atoms, molecules, quantum dots, and nanocrystals. Quasi-bound states are formed when the amplitude of the fields around a local scatterer decay sharply away from it resulting in strong photon confinement. In the new approach, a small dipole ($d/l < 0.001$) excited by the dominant mode in a high Q open resonator, is resonantly coupled to its image field via a “hall of mirrors” effect. The solution to this resonant multiple scattering problem, intractable by electromagnetic boundary value techniques, requires Maxwell’s equations to be converted to a self-consistent Lippmann-Schwinger equation. The theory predicts that the near fields of the mesoscopic object are strongly confined and enhanced compared to the average field distribution. The resonant enhancement would greatly increase the detectivity of quantum optic radiation coupled to the dipole via its near field, but prior to our research, had not been experimentally observed.

The electromagnetic scattering theory adopts procedures from quantum scattering theory, but the physics of the problem are classical and the interaction is linear. Thus, its validity could be tested, for convenience, at scaled microwave frequencies. In the experiments

small magnetic and electric dipoles were introduced into open strip line cavities resonating in TEM_{0on} eigenmodes with Qc's of $\sim 10^3$. Near field enhancements of $\sim 10^5$ were observed when the dipoles were positioned to optimize the resonant scattering. The field enhancements were calculated from the dipole induced reductions in the reflection coefficients of 4 different resonators at harmonic resonances in the 100 MHz to 3 GHz frequency range. If this approach can be used to scale near field resonances to THz, IR, and optical frequencies, radiative emissions from molecular scale quantum optic systems could be readily detected using dipoles with dimensions of 10-100 nm ($d/l \approx 0.001$).

Another goal of this research is to understand the internal dynamics of complex molecules which interact with radiation pulses. Radiation pulses can have significant effect on the internal dynamics of complex molecules. Although STIRAP (stimulated Raman adiabatic passage) and CARS (coherent anti-Stokes Raman spectroscopy) have become important tools for coherently controlling and changing the vibrational and electronic states of entire molecular populations, traditional models of STIRAP generally view the molecular target system as a simplified multilevel system. We are looking at the effect on STIRAP of the actual nonlinear dynamics that occurs when the laser pulses interact with molecular systems. Laser pulses can induce chaos and this can strongly affect the response of the molecule to the radiation field. In addition, the internal dynamics of an unperturbed molecule may itself be exhibiting the manifestations of chaos and simple pictures of the molecular level structure are likely not valid. We have studied the effect of chaos on STIRAP for a simple anharmonic system and find unexpected chaos-induced transitions [3]. We are currently studying the effect of chaos on STIRAP for the rotational states of molecules and we are beginning to study STIRAP on molecular systems for which experimentally determined Hamiltonians for vibrational dynamics now exist.

B.) Bose-Einstein Condensates

D. Heinzen, M. Raizen.

We have been investigating a quantum phase transition of a dilute, ultracold gas of ^{87}Rb

atoms. In order to do this, we superimposed an “optical lattice potential” on an ^{87}Rb Bose-Einstein condensate with a temperature of about 150 nanoKelvin. We found that when the strength of the lattice potential U reached a certain critical value U_c , a quantum phase transition from a superfluid state to a Mott insulator state occurred. In the Mott insulator state, the number fluctuations of the atoms are strongly suppressed, so that nearly every lattice site contains exactly the same number of atoms. We have shown that we can create Mott insulator states with $n = 1, 2$ or 3 atoms per site. We have studied the momentum distributions of the atoms released from the Mott insulator phase, and found that the momentum spread exhibits a sharp increase for values of U just greater than U_c . Finally, we have realized a coherent and reversible conversion from atoms to molecules in the gas. In particular, we have found that we can create a gas with a central zone containing 40% of the atoms, which coherently oscillates between an atomic and molecular form.

We have also succeeded in making a 1-D condensate in rubidium, together with single-atom addressability. This system can be used for realization of a quantum tweezer for atoms which could be the basis for a quantum computer. It can also be used to study quantum phase transitions and quantum chaos.

C.) Open Quantum Systems

C.K. Shih, L. Reichl, C. G. Sudarshan

We are working on optical quantum information processing in semiconductor nanostructures. In this project, we use laser spectroscopy to control the quantum states in semiconductor quantum dots. In particular, we will use the discrete electronic states formed in quantum dots as the basis for quantum bits. Ultra-fast laser pulses on or near resonance are used to coherently manipulate the probability distribution and the relative quantum phase between two energy levels. Moreover, we will be developing a high- Q optical cavity to allow us to couple the excitonic states and the cavity modes. More specifically, we would like to demonstrate strong coupling cavity QED in a solid state

system.

We have continued our work on the dynamics of quantum networks formed from GaAs/AlGaAs based ballistic electron waveguides. Flying qubits can be formed from pairs of waveguide leads. We have shown that Bell states can be generated with such devices. We have also shown that quantum networks formed from multiple leads can be used to add simple numbers. We are continuing work on the entanglement dynamics of such systems.

We investigated several aspects of the dynamics of measured and open quantum systems with emphasis on the Quantum Zeno effect using the funds from TAMU. The appearance of the Quantum Zeno effect and the possibility of the so called 'anti-Zeno' effect in a quantum system interacting with a measuring device was modeled and studied both theoretically and numerically. We also looked at the Zeno and anti-Zeno effects in three level quantum systems that are similar to systems that have recently been studied experimentally. We also studied the dynamics of open quantum systems described by master equations of the Kossakowski-Lindblad type over the past year. The origin of the spin-statistics connection and its extension to higher space dimensions was a third line of research we pursued during this period.

D.) Cosmology

S. Weinberg, L. Reichl

We have continued our work on understanding the observed fluctuations in the cosmological photon distribution in terms of quantum fluctuations in other fields at the time, of inflation.

We have studied the nonlinear dynamics and stability properties of a cosmological model of spatially homogeneous coupled gravity and matter fields using methods of classical mechanics. We find that the system exhibits regions of chaos and dramatic changes in

structural stability as the strength of the coupling between the fields is varied. Numerical simulations suggest that Hamiltonian systems with structure appropriate for describing matter-gravity interaction constitute a new class of nonlinear systems with very unusual and rich dynamics.

E.) Granular Media

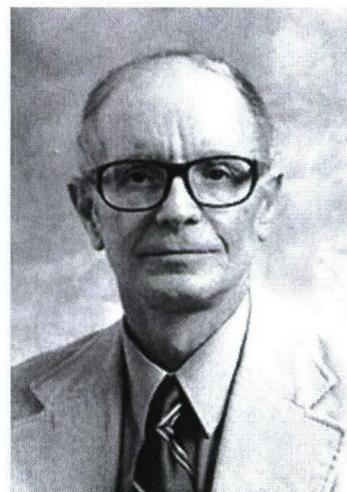
H. L. Swinney

Wave patterns with long-range order form spontaneously in diverse optical, fluid, chemical, plasma, solid, and other systems driven far from equilibrium. We are studying coherent waves and the effect of noise on the waves formed in granular media. Granular media are collections of particles that dissipate energy in collisions. In our study, layers particles are oscillated vertically to maintain a far from equilibrium granular gas. The experiments and particle simulations reveal, surprisingly, that noise plays a role in a granular system similar to that in continuum systems. We are also studying the formation of craters by the impact of a projectile in a granular medium. Our measurements and molecular dynamics simulations show that the time required for a projectile to stop is independent of its energy at impact. This highly non-intuitive result will help in understanding craters formed by explosives and by meteorite impacts on planets and the moon.

★ Scully, Marlan O.

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ISI Rating: Highly Cited
ISI Assigned Category: Physics
ISI Indexed Name: SCULLY MO SCULLY



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Date of Birth:
 City, State/Province,
 Country of Birth:
 Citizenship(s):
 Language(s):

Education

Date	Degree	Institution	Subject
1966	Ph.D.	Yale University, New Haven, CT	Physics
1963	M.S.	Yale University, New Haven, CT	Physics
1962	Nuclear Engineering Program	Rensselaer Polytechnic Institute, Troy, NY	
1961	B.S.	University of Wyoming, Laramie, WY	Engineering Physics
1959	A.S.	Casper College, Casper, WY	Engineering

Appointments/Affiliations

Dates	Title	Organization	City, State/Province	Country
2001- Present	Director	Texas A&M University, Institute for Quantum Studies	College Station, TX	United States

2000- Present	Distinguished Research Chair	Texas Engineering Experiment Station	College Station, TX	United States
1999- Present	Professor of Electrical Engineering	Texas A&M University	College Station, TX	United States
1996- Present	Burgess Distinguished Professor	Texas A&M University	College Station, TX	United States
1995- Present	Director	Texas A&M University, Center for Theoretical Physics	College Station, TX	United States
1992-1996	Professor of Physics	Texas A&M University	College Station, TX	United States
1980- Present	Auswertiges Wissenschaftliches Mitglied	Max-Planck-Institut für Quantenoptik	Garching	Germany
1980-1992	Distinguished Professor of Physics	University of New Mexico	Albuquerque, NM	United States
1980-1992	Director	University of New Mexico, Center for Advanced Studies	Albuquerque, NM	United States
1979-1980	Director	University of Arizona, Theory Division ARL	Tucson, AZ	United States
1969-1980	Professor of Physics and Optical Sciences	University of Arizona	Tucson, AZ	United States
1969-1971	Associate Professor	Massachusetts Institute of Technology	Cambridge, MA	United States
1967-1969	Assistant Professor	Massachusetts Institute of Technology	Cambridge, MA	United States
1961-1962	Instructor	Yale University	New Haven, CT	United States
1961-1962	Physicist	General Electric		United States
1956-1959	Laboratory Technician	Mobil Oil Company		United States
	Adjunct Professor	Texas Tech University	Lubbock, TX	United States
	Adjunct Professor	University of Arizona	Tucson, AZ	United States
	Adjunct Professor	University of New Mexico	Albuquerque, NM	United States
	Consultant	Los Alamos Scientific Laboratory	Los Alamos, NM	United States
	Consultant	U.S. Air Force	Washington, DC	United States
	Consultant	Litton Industries	Woodland Hills, CA	United States
	Consultant	Rocketdyne		United States
	Consultant	Rockwell International		United States
	Consultant	Marin-Marietta	Daytona Beach, FL	United States
	Consultant	BDM		United States
	Consultant	Dynalectric Corporation	Washington, DC	United States
	Consultant	Dayton Research Institute	Dayton OH	United States
	Consultant	La Jolla Institute	La Jolla, CA	United States

Consultant	Science Applications, Inc.	Poolesville, MD	United States
Consultant	U.S. Army	Washington, DC	United States
Advisor	Los Alamos National Laboratory, Physics Division	Los Alamos, NM	United States
Advisor	High Energy Laser Review Board		United States
Advisor	Air Force Office of Scientific Research	Washington, DC	United States
Advisor	National Research Council	Washington, DC	United States
Advisor	Joint Council on Quantum Electronics		United States
Advisor	International Committee on Optics		United States
Advisor	Rice University	Houston, TX	United States
Advisor	North Texas State University	Denton, TX	United States
Advisor	United States Air Force	Washington, DC	United States
Advisor	United States Army	Washington, DC	United States
Advisor	University of Texas, El Paso	El Paso, TX	United States
Advisor	University of Wyoming	Laramie, WY	United States
Adjunct Professor	University of Colorado		United States
Adjunct Professor	Italian Institute of Optics	Firenze	Italy
Adjunct Professor	University of Texas at Dallas	Dallas, TX	United States

Honors/Awards

Date	Name	Description
1983-1986	Member, Scientific Advisory Board	Santa Fe Institute
1966	Class Marshall	Yale University
	Member	National Academy of Sciences
	Member	Max Planck Society
	Adolph E. Lomb Medal	Optical Society of America
	Elliott Cresson Medal	Franklin Institute
	EG&G Medal	Society of Optical and Quantum Electronics
	Charles H. Townes Award	Optical Society of America
	Senior Faculty Award	Alexander von Humboldt Foundation
	Fellow	Alfred P. Sloan Foundation
	Fellow	John Simon Guggenheim Foundation
	Elected External Member	Max Planck Society
	Elected Director-at-Large	Optical Society of America
	Elected Member	Academia Europea
	Fellow	American Association for the Advancement of Science
	Fellow	Optical Society of America
	Outstanding Science Communicator Award	Sigma Xi
	Distinguished Research Achievement	

Award	Texas A&M University
Outstanding Research Award	University of New Mexico
Eminent Scholar	State of New Mexico
Research Excellence Award	University of New Mexico
Distinguished Alumnus Award	University of Wyoming
Distinguished Alumnus Award	Casper College
Distinguished Lecturer	Texas Tech; Waterloo University; University of Colorado
Member	Sigma Xi
Honorary Member	All-University
Physics Honorary President	Sigma Pi Sigma
Graduate Fellowship	National Science Foundation
RPI Scholarship	General Electric
Walker Merit Scholarship	

Memberships

Dates	Organization	Position
	McGraw-Hill Encyclopedia of Science and Technology	Member, Editorial Board
	Physics of Quantum Electronics Series	Member, Editorial Board
	Optics Communications	Member, Editorial Board
	OSA Publication Committee	Member, Editorial Board

Research Interests

Field of Expertise &
Research Interests:

Research Funding/Grants

Dates	Grant #	Grant Title	Organization	Grant Amount	Primary Investigator
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Publications

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