Ph.D. DISSERTATION DEFENSE

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Degree: Doctor of Philosophy
School/Department: Schaefer School of Engineering & Science / Department of Physics
Date: Thursday, December 8th
Time/Location: 11:00 am (EST) / Babbio Center, Room 310
Title: Chirped pulse control of quantum coherence in atoms and molecules: A semiclassical theory with applications for detection and sensing

Chairperson: Dr. Svetlana Malinovskaya, Department of Physics, School of Engineering & Science

Committee Members: Dr. Ting Yu, Department of Physics, School of Engineering & Science
Dr. Xiafeng Qian, Department of Physics, School of Engineering & Science
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ABSTRACT

Coherent Anti-Stokes Raman Spectroscopy (CARS) is one of the leading non-linear optics techniques with variety of applications in physics, chemistry, and biology. In this work, a chirped-pulse quantum control scheme applicable to Coherent Anti-Stokes Raman Scattering spectroscopy, named as C-CARS, is developed aimed at maximizing the vibrational coherence in molecules. The scheme implies chirping of three incoming pulses in the four-wave mixing process of CARS, the pump, the Stokes and the probe, to fulfill the conditions of adiabatic passage. The scheme is derived in the framework of rotating wave approximation and adiabatic elimination of excited state manifold simplifying the four-level model system into a “super-effective” two level system. The robustness, spectral selectivity and adiabatic nature of this method are helpful in improving the existing methods of CARS spectroscopy for sensing, imaging, and detection. It is demonstrated that the selectivity in excitation of vibrational degrees of freedom can be controlled by carefully choosing the spectral chirp rate of the pulses.

A semiclassical theory based on the C-CARS scheme is then developed which maximizes the coherence and generates an optimum anti-Stokes signal from a cloud of molecules at kilometer scale. The theory is based on the solution of the coupled Maxwell's and the Liouville von Neumann equations and focuses on the quantum effects induced in the target molecules by the control pulse trains. The propagation effects of pulses are taken into account and the built-up of the anti-Stokes
signal which may be used as a molecular signature in the backward CARS signal is demonstrated numerically. A deep learning technique, using Convolutional Neural Networks (CNN), is implemented to characterize the control pulses and evaluate time-dependent phase characteristics from them. The effect of decoherence induced by spontaneous decay and collisional dephasing is also examined.

Another adiabatic process, the Fractional Stimulated Raman Adiabatic Passage (F-STIRAP) is presented, which was used to create a maximally coherent superposition in a multi-level system aiming remote detection. We demonstrated that by changing the ratio of amplitudes of the pump and the Stokes fields, it is possible to generate an arbitrary coherent superposition state using F-STIRAP.

We considered a degenerate multi-level system of hyperfine states in Rubidium 87 to analyze a phenomenon of mirrorless lasing. Using strong linearly polarized pump field, a population inversion is created between the ground and excited degenerate states. A seeding probe pulse induced lasing in the medium, resulting in the gain of electric field having the same mode as the probe field.