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Ph.D. DISSERTATION DEFENSE

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Date:	Thursday, December 8th
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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 Dr. Henry Du, Department of Chemical Engineering and Materials Science, School of Engineering & Science Dr. Francesco Narducci, Naval Postgraduate School, Monterey, California

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, Fractional Stimulated Raman Adiabatic Passage (F-STIRAP), which can be used to create a maximally coherent superposition of two energy levels is presented. It is demonstrated that the Fractional-STIRAP can also be used to create any arbitrary coherence. A theory for degenerate mirror-less lasing is also presented where a population inversion is created in Rubidium 87 atom to achieve positive gain using two linearly polarized pulses orthogonal to each other.