

**Stony Brook University
The Graduate School**

Doctoral Defense Announcement

Abstract

Ultrafast Coherent Control Spectroscopy

By

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Coherent control is currently a very active area of research in physics. The goal of coherent is to prepare molecules in specific quantum states with shaped ultrafast laser pulses. Recently, there are increasing applications of coherent control towards cellular imaging. Coherent control spectroscopy has been demonstrated to selectively excite different fluorescent probes. As compared to using a tunable laser to separately excite tissues labeled with different fluorophores, coherent control allows selective imaging of fluorophores with a single broadband light source and pseudo-simultaneous imaging of different tissues by switching pulses shapes rapidly. It is especially beneficial for distinguishing broadband fluorophores with similar two-photon absorption cross-sections, e.g. for free and enzyme-bound nicotinamide adenine dinucleotide (NADH). In this thesis, we discriminate between samples containing either free NADH or enzyme-bound NADH solutions with a π phase jump at a given frequency within the excitation bandwidth. This parameter scan is sensitive to as low as 3% of binding. The same idea can be generalized to other two-photon fluorescence systems, and a close-loop feedback control approach should allow even wider application.

We also develop the two-dimensional Fourier transform spectroscopy in the deep UV (262 nm). The 2D Fourier transform spectroscopy is the optical analog of NMR. It has been widely used to study molecular structures and energy transfer with a sequence of phase-locked laser pulses. In recent years, because of increasing harmful solar radiation caused by ozone layer depletion, there is a growing interest in studying DNA excited state dynamics under UV radiation. We compare 2D spectroscopy measurements in the deep UV for adenine and uracil in solution. Both molecules show excited state absorption on short timescales and ground state bleach extending for over 1 ps. While the 2D spectrum for uracil shows changes in the center of gravity during the first few hundred femtoseconds, the center of gravity of the 2D spectrum for adenine does not show similar changes.

Date: January 31, 2012

Time: 9:00 am

Place: Physics Building, Room S-141

Program: Physics

Dissertation Advisor: Thomas Weinacht