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Final Technical Report:
Acquisition of a Femtosecond Optical Parametric Oscillator/Amplifier and
Pulse Diagnostics
AFOSR F49620-97-1-0116

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This equipment grant was intended to significantly enhance our experimental capabilities in femtosecond time resolved studies of molecular dynamics in condensed media. Two major improvements were the goals. Both have been successfully completed, and have generated results that have already been published. I describe them succinctly.

a) Since electronic recording speeds are limited to the GHz range, time resolved studies on femtosecond time scales invariably involve the use of two or more laser pulses, in which time steps are derived as a function of optical delay lines between pump and probe pulses. Quite clearly, to be able to follow the relevant science, the light sources should have the flexibility to tune to system resonances. As such, it is obvious that both pump and probe wavelengths should be tunable. Our femtosecond laser consisted of one tunable leg, one Optical Parametric Oscillator (OPO) pumped by a Ti:Sapphire laser. Under this grant, we developed a second one pumped by the same source, thus creating a two color, independently tunable, yet synchronized femtosecond spectrometer. While our initial intention was to purchase a second OPO, given the unsatisfactory specifications of commercially available units, we designed and constructed our own OPO. The system proved to be quite efficient, enabling us to use intensities as low as 500 $\mu\text{J}/\text{pulse}$ to pump both OPOs. A schematic of our design is presented in Fig. 1. We now boast of a two-color fs spectrometer with 80 fs time resolution (from cross correlation between the two arms) and independent tunability from 480 nm to 2400 nm in each arm. Data obtained with this system, measurements of nonadiabatic dynamics in solids, is already in press.¹

b) Crucial to all femtosecond measurements is the characterization of the laser pulses used in experiments, and in particular knowledge of the coherence of the pulses, i.e. the joint time-frequency distribution of the laser. Manipulation of the coherence of the laser pulse is one of the more important dimensions in fs spectroscopy, since it enables measurements of coherence of the evolution of a system under study and its coherent control. The methods of choice for characterization of source coherence are the techniques that come under the name Frequency Resolved Optical Gating (FROG).² We constructed our own FROG system, using the Kerr gate geometry, with a purchased CCD array detector as the main acquisition. We use commercially available software for pulse retrieval (FROG 1.5, Femtosecond Technologies, Inc.). This system is complete, and has already been used in advancing concepts of measurement and characterization of system coherence.³ In an important paper that is already in press, we show through experiment and theory that system coherences can be measured using chirped probe pulses, and show that through a time-frequency to a time-coordinate transformation, an isomorphism can be established between measurements of radiation coherence through cross correlation and system coherence through measurements of signal as a function of probe chirp. Analytic

expressions have been derived and tested out quite satisfactorily with experiment. A preprint of the paper that describes the set-up, the experiments, and the theory, is included in this report.

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FEMTOSECOND SPECTROMETER

