**Ultrafast –femtosecond-time-resolved chemistry**

In this experiment we will introduce the basic pulsed-laser tools for time-resolved studies of femtosecond dynamics in processes of paramount interest in chemistry. The femtosecond time scale includes some slower electronic processes and covers most of molecular nuclear dynamic processes, such as various rovibrations, which also extend into picosecond and longer scale region.

Femtosecond pulses can be used directly as agents or as spectroscopic monitoring tools for photo-excited charge carriers, dynamics of solvation, etc. just to mentation a few. Of particular interest in very fast edge of this scale (going to the atto-scale) are interactions between coherently excited electronic states in fast EM fields.

We will start with experiment first and do the analysis later. The key experiment done here is a basic pulse-pulse (auto)correlation experiment, which measures physical observables of electromagnetic femtosecond scale pulse(s). We will use a new laser LIBRA which operates at ~1kHz, and produces femtosecond pulses in 800nm region at or less than 100fs.

During the lecture and workshop we will develop methods for modeling fast pulses and describe their interaction with some molecular objects. Some Mathematica application templates for analysis of correlation functions will be used for that. We will also learn more about the femto-lasers we use in our experiment (TiS) and molecular physics/photonics of electromagnetic filed interactions.

*After becoming familiar with the time measurements tools (experiment below), you will participate in a workshop where you will write a proposal for a project that applies the so called pump-probe experimental architecture. The ultrafast pump-probe method is based on the detection of the laser pulse probe or its effects. For example we can use a well-defined pulse to excite a molecular system into a specific electronic state and then several femtoseconds later probe that state with another pulse, well defined for a process that involves that excited state, say for example its ionization. Note that probe may or may not be energetically or impulse intensity softer than the pump probe. We can vary the time delay between the pump and probe and see how it changes the system response. As we did in other experiments, like other pulse (scattering) response measurements -say DLS, we can invoke help of very small-change-sensitive detection techniques based on auto- or cross- correlations. As always function transform or some learning (AI) techniques could help too.*

*When the pump probe project is approved we will construct an new experimental configuration for data acquisition and use a similar correlation method for time resolved measurement of a chemical process.*

Time measurement-The experimental steps include

– **SAFETY FIRRST:** **Optical glasses MUST be on all the time in the lab. In case of any accident with injuries to body call 911.** Safety procedures (including appropriate optical glassware with relevant OD>7 in the laser regions). Note that pulsed lasers compress high level of energy for a very very short time. These energies may be compared not only to chemical bonds but also to the energies present at astrophysical or radioactive-nuclear events. Damages are instantaneous and permanent. Consequently only small groups of students will work in a controlled environment. You will enter and use only the designated areas of the laboratory room 237 around the autocorrelation instrument setup. You must limit your movement so that you do not disturb the alignment nor cross the laser beam. You must keep your head always above the laser light planes.

-**INTRODUCTION TO INSTRUMENTATION:** You will firstly prepare and test alignments and properties of autocorrelation (AC) instrument using a less energetic HeNe laser in lab 104 or 301. You will also carefully examine the same prepared AC instrument using the femtosecond pulses. There will be a long ~2m black anodized 1 inch diameter aluminum tube that will bring the laser light to the optical table where the AC system is positioned. DO NOT move or walk close to that tube!

**EXPERIMENT: Your first goal is to project the details of the light paths on the drawing or image of the AC system. You mast name and identify all optical and other parts of the system.**

**You will describe in situ the operation of the system you see and take notes about it. Concepts like coherence, autocorrelation function, SHG, BBO, NDF OD, PD, etc. will be included in your notes.**

**You will describe the 4 major stapes of the beam alignment from the optical table where the femtosecond pulsed laser is positioned to the AC table, and the AC system alignment**. Note that optics for ultrafast spectroscopy is different from regular optics, it is very expensive and requires careful treatment (including special clean gloves). The optics is from a professional research system used at a National Lab. The experiment uses computer controlled micrometer precision driver. The core of AC alignment requires a lot of practice and skills, so please do not disturb it manually unless you have practiced the manual alignment. We will give you precise instructions *in situ* (*on the fly*). - Violation of any procedure steps could result in accidents and damages of you or instrument and you not being able to complete experiment (and could lead to lower grade) .

**You will become familiar with the detector and electronics, and the basics of the LaView program and interface (DAQ) that links the detector. You will also go through the procedure for correlation based time measurement.**

**Finally you will acquire data for the determination of the time of a pulse that will be constructed in the laser compressor specially for you.**

**After the experiment you will fit the results into a an autocorrelation function and calculate the time in femtoseconds.**

**Based on the workshop , labnotes, data and calculation you will complete a short report.**