Coherent control of physicochemical processes

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Summary

This talk addresses a number of instances where coherent control methods can help advance research projects using short pulse x-ray sources. First, coherent control, in the form of phase modulation, has been shown to be a powerful method in the optimization of laser sources that generate XUV and X-ray pulses.[1] Similarly, optimization of the pump source can be used to optimize other types of x-ray sources that depend on a femtosecond laser pump. Phase and amplitude optimization of a laser source can be useful in pump (x-ray) probe experiments. In those experiments the pump laser initiates the dynamics and the x-ray absorption or scattering probes the temporal evolution of the system. In these experiments, it is important to excite as many molecules as possible by the pump pulse to enhance the time-dependent signal. If intense transform limited (TL) pulses are used as the pump, numerous undesired nonlinear processes such as multiphoton excitation and photoionization occur. Coherent control can be used to limit the extent and nature of the detrimental nonlinear processes that take place, while maximizing the excitation of the sample molecules to the desired state. This optimization would be ideal for experiments aimed at visualizing protein dynamics following laser excitation.

Experiments from the Dantus laboratory with phase shaped pulse to optimize sample excitation will illustrate how coherent control can be used to optimize laser molecule interactions. The principles of multiphoton intrapulse interference (MII) will be introduced.[2,3] This understanding is essential for establishing how phase modulation can be used to control excitation of large molecules in condensed phase. Most of the experiments presented will demonstrate control and selectivity in two-photon transitions.[4,5] In these cases one can visualize how the frequencies within the bandwidth of a femtosecond pulse can constructively or destructively interfere as they combine to induce a two-photon transition. By phase shaping the laser pulse, one can control the nature of the interference and thus gain selectivity. MII will be shown to be useful in controlling two vs. three photon transitions and for selective two-photon microscopy.[3]

Recently, there has been a great deal of excitement about the use of Genetic Algorithms (GA) coupled with phase and amplitude pulse shapers for controlling laser-molecule interactions.[6] In this talk we will discuss some of the practical parameters of pulse shaping. Most importantly, we will first discuss if amplitude control is important or not. Then we will address how much control over phase is required, keeping in mind that continuous control over the phase of all frequencies in the pulse leads to an infinite number of different shaped pulses. A GA that samples only a small fraction of an infinite search space is not likely to find optimal solutions. Therefore, all optimization approaches must make a compromise to reduce the number of frequencies and phases to be explored. Some of the ingredients that lead to the highest efficiency will be discussed. In particular, the advantages of binary phase shaping will be discussed and illustrated with experiments on selective two-photon excitation.[7] Binary phase modulated femtosecond pulses, where each pixel in the shaper introduces a phase retardation of zero or π, can selectively excite one type of probe molecule, leaving the others in their ground state. The optimization of the phase mask is derived from knowledge of the absorption spectrum of the probe molecules and the characteristics of the laser pulse based on the multiphoton intrapulse interference (MII) theory. The success of binary phase modulation will be demonstrated with results from the Dantus group on functional imaging through biological tissue.[8,9]

Finally, the direct characterization of x-ray pulses will be discussed. The ideal method would be one that is similar to a method developed in the Dantus group for characterization and compensation of phase distortion of femtosecond pulses. The method, multiphoton intrapulse interference phase scan (MIIPS) is advantageous because it is a single beam technique—it is not based on interferometry.[10] The phase is retrieved directly from the intrapulse interference created by a calibrated reference phase that is introduced using a pulse shaper. Once the phase

distortions in the pulse are measured, the same pulse shaper used for characterization introduces a phase that counteracts the distortions to obtain TL pulses. The operation of this method will be demonstrated with sub 11 fs pulses. The requirements needed to implement the MIIPS method using x-ray pulses will be discussed.

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