Using computer controlled ultrafast laser pulses to control matter

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Ultrashort laser pulses, because of their bandwidth and their peak intensity are able to impart energy through nonlinear optical processes at essentially any wavelength in the electromagnetic spectrum. Controlling the amount of energy delivered is achieved through pulse shaping and an approach known as multiphoton intrapulse interference (MII). MII takes advantage of nonlinear optical interference and provides a robust platform for controlling molecular excitation through the introduction of specific phase functions on the laser pulse [1].

The first step in ultrashort laser pulse experiments (less than two optical cycles in duration) is to measure and eliminate spectral phase distortions which are inherent to femtosecond lasers. Given the sensitivity of the laser to temperature, humidity and the optical components in the path, pulse compression usually requires expert manual tweaking and patience. In an effort to eliminate the tedious and imprecise manual tweaking, our group developed multiphoton intrapulse interference phase scan (MIIPS), a method based on a programmable pulse shaper that measures and compresses ultrashort <5fs pulses at the focal plane of the experimental setup (see Fig. 1) [2].

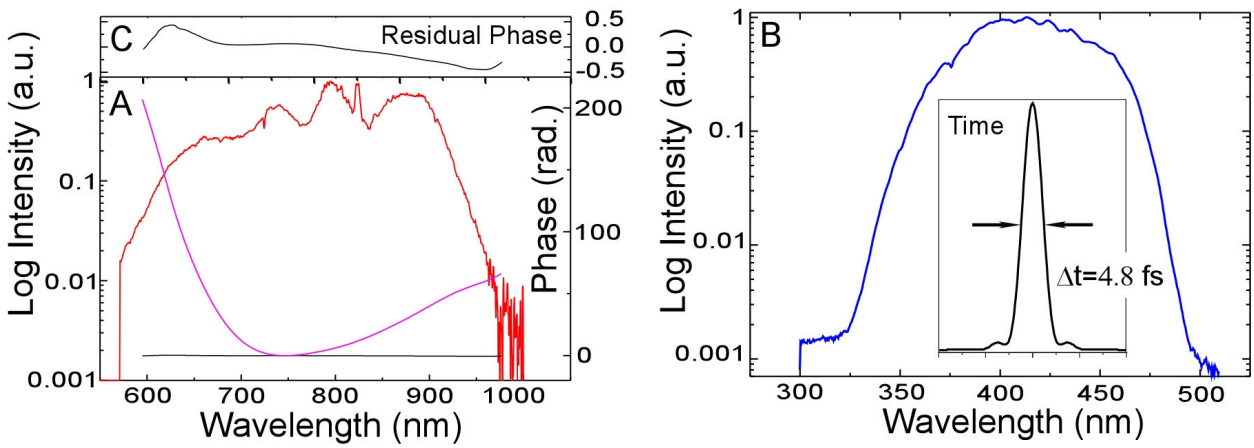

Fig. 1. Output from the hollow core fiber (a) spectrum and phase measured by MIIPS, top inset is the residual phase after compression. (b) Second harmonic spectrum after compression and autocorrelation of the output pulses.

The systematic use of MII to control two-photon excitation processes has allowed significant improvements in nonlinear optical biomedical imaging [3]. In addition, MII has been used to achieve vibrational mode selective excitation in large molecules in gas, liquid and solid states; this ability greatly improves coherent anti-Stokes Raman scattering methods for standoff detection (see Fig. 2) [4]. The ability of femtosecond lasers to activate molecules on a timescale faster than nuclear motion is being used in our laboratory to revolutionize biological mass spectrometry with important implications for the fields of proteomics and metabolomics [5]. Experimental results for all these breakthrough applications of ultrashort shaped pulses will be presented.

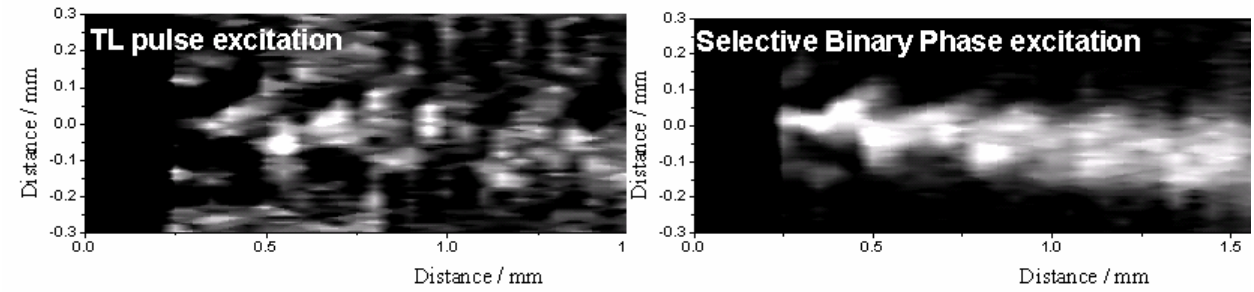

Fig. 2. CARS imaging of a micro-jet of CO_2 gas in air. (left) using TL pluses one is not able to detect the CO_2 molecules. (right) By introducing a phase modulation that breaks the pulse into sub-pulses with a time delay matching the symmetric stretch of CO_2 one is able to achieve selective vibrational mode excitation and the integrated CARS signal increases, permitting one to image the CO_2 jet. The turbulence observed is expected because the nozzle used has a rectangular shape.

The Dantus Research Group has experience in supercontinuum pulse compression (from PCF fibers and from hollow core fibers). We also have experience in alignment, and selective vibrational mode excitation. We routinely work with small molecules as well as peptides in solution or in the gas phase (using ion trap mass spectrometers). We look forward to collaborations where we can use ultrashort pulses to prepare the initial state of the system to be probed by x-rays.

References

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