Shaperless single-pulse CARS for micro-spectroscopy and fast modulated temporal pulse splitting for selective excitation

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We demonstrate two novel schemes for single-pulse, micro-spectroscopic CARS measurements based on spectral shaping and selective excitation. Firstly, we show the ability to obtain spectral resolution an order of magnitude better than the pulse bandwidth using a single amplitude and phase shaping element. Specifically, we employ a resonant photonic crystal slab (RPSC) filter in lieu of a commonly used 4-f shaping apparatus. The RPCS is a double grating waveguide structure which is comprised of multiple layers, and when a beam illuminates the RPCS at a given angle, a narrow spectral range is on resonance with the grating waveguide. As a result no light is transmitted through the RPCS at the resonant wavelengths, and the RPCS acts as a narrowband notch filter in transmission. The notch (probe) spectral location is controlled by the RPCS angular orientation. The spectral feature is also observed in the CARS four-wave mixing spectrum and located at frequencies blue-shifted from the probe by the vibration frequency, allowing for retrieval of the CARS spectra. We show the ability of this method in micro-spectroscopy to identify perfluorodecalin in water (Fig 1) and the skeletal mode of starch within a potato cell (Fig 2).

In another study we investigate the ability to rapidly modulate pulse shapes to selectively excite different materials and simultaneously resolve them in the frequency domain using a spectrum analyzer. It has been shown that temporal splitting of pulses can be used to selectively excite vibration levels depending on the pulse separation in time. Since different separation times will result in local minima and maxima in the CARS spectra of particular chemical, we created a phase mask to modulate the excitation of two chemicals. To do this we define a phase pattern on a 2-D SLM of four specific pulse separation times such that that spatial pattern creates a periodic selectivity for the different chemicals at different spatial frequencies. Therefore, when a beam is rapidly scanned across the SLM, the chemicals are selectively excited at different harmonics of the fundamental scan frequency. This method of selectively exciting chemicals at different frequencies allows for simultaneous and fast detection in the frequency domain with an extinction coefficient of greater than 10.