Enhanced contrast CARS for a single molecule imaging

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Nonlinear optics has been at the heart of modern imaging techniques that require noninvasive measurements with high spectral selectivity. Coherent anti-Stokes Raman scattering belongs to one of the frontiers of molecular microscopy with broad applications in biology and medicine. There has been a demand for the advancement of CARS microscopy to distinguish between related species having vibrational frequency differences within a fraction of a wave number and to register a coherent signal from a single molecule. That is where pulse shaping technology may prove useful allowing active phase-amplitude control to maximize CARS signal. We have developed a method for ultrafast pulse shaping in CARS that allows enhancing CARS signal by three orders of magnitude and suppressing the background to result in a measurement sensitive enough to observe single molecules. The method creates the maximum virational coherence in a predetermined molecular fragment by making use of the chirp variation of the pump and Stokes pulse [1], Fig.1. The method was patented [2] and the proposed pulse shaping was realized experimentally, Fig.2. We have analyzed the impact of spontaneous decay and collisional dephasing on controllability, [3], and showed that the proposed method allows to sustain high coherence in the desired vibration when implemented in the form of two chirped pulse trains with the pulse repetition rate close to spontaneous decay rate. Also, we investigated the impact of the coupling between vibrational modes on controllability of selective excitation of Raman active vibrational modes, [4]. The method may be implemented to detect the high energy phosphorus-oxygen bond in the adenosine triphosphate (ATP) known as the "energy currency" of the cell.

References


