Broadband CARS Microscopy - Overview and Applications

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Several distinct characteristics make coherent anti-Stokes Raman scattering (CARS) microscopy potentially ideal for non-invasively imaging chemically complex systems such as complex materials and biological cells. These characteristics include high spatial resolution, high sensitivity, and label-free chemical specificity. Chemically sensitive imaging of complex samples, such as the intracellular medium will require broadband spectral sensitivity over the "fingerprint" frequency range of 500 – 1800 cm^{-1}. Broadband CARS microscopy presents the promise of great chemical resolving power, but Raman peaks of complex chemical systems are crowded and often weak in the fingerprint region, so can be obscured by noise associated with the nonresonant background (NRB), which normally accompanies the CARS signal.

I will discuss experimental and numerical methods of extracting the Raman-like signal component from a broadband CARS spectrum. Experimentally, we have demonstrated several approaches to suppress NRB. Each of these builds on a broadband CARS technique that we previously demonstrated[1]. The most successful of these methods utilize an efficient 3-color CARS generation mechanism[2] which allows simplified, single-shot approaches to background-free broadband CARS microscopy. One of these approaches takes advantage of the short temporal duration of a broadband pulse to allow complete suppression of the NRB through time-delayed detection without significant loss of resonant vibrational signal to an extent that is not possible for narrowband, picosecond CARS.[3] Another of these approaches relies on interferometric suppression of the background. In cases where NRB cannot be entirely eliminated from the signal, post-processing is desirable, and we have developed a rapid method for accomplishing this that is computationally not too intensive.

[1] Kee and Cicerone, Simple approach to one-laser, broadband coherent anti-Stokes Raman scattering microscopy Opt. Lett. 29, 2701 (2004).
[2] Y. J. Lee, Y. Liu, M. T. Cicerone, Characterization of three-color CARS in a two-pulse broadband CARS spectrum, Opt. Lett. 32, 3370 (2007).
[3] Y. J. Lee, M. T. Cicerone, Viarational dephasing time imaging by time-resolved broadband coherent anti-Stokes Raman scattering microscopy, Appl. Phys. Lett. 92, 041108 (2008).