Highly sensitive coherent anti-Stokes Raman microscopy with shaped femtosecond pulses

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With Coherent anti-Stokes Raman scattering (CARS) microscopy and its ability to reflect the characteristic vibrational spectrum, three-dimensional images can be taken from unstained samples with chemical contrast. Due to the interference of CARS emission, the signal has a dependence on the squared number of Raman scatterers. While beneficial for majority chemical species, this poses difficulties for the detection of low concentrations. However, interfering the CARS signal with an auxiliary local oscillator (LO) field allows a more sensitive heterodyne detection with linear concentration dependence. The drawback commonly is a complex experimental setup, requiring that several laser beams be overlaid with wavelength precision.

To achieve all this in a very simple and robust setup, we have developed single-beam heterodyne CARS [1]. In a single-beam CARS-scheme [2, 3], all CARS frequencies are provided within the spectrum of a broadband laser pulse with femtosecond pulse shaping ensuring spectral resolving power. Without complicating the single-beam setup in any way, we have introduced interferometric detection by not only controlling the CARS signal generation with the pulse shaper, but by also using it to generate a phase-controlled local oscillator field at the CARS signal wavelengths. Amplification factors of more than 5000 have been already demonstrated.

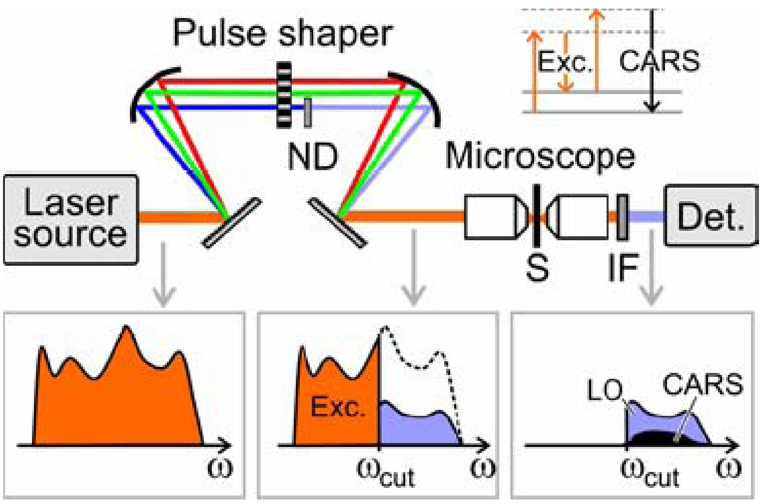

Fig. 1 Single-beam heterodyne CARS: Schematic of the experimental setup. A broadband laser source is used to provide all necessary excitation frequency components for CARS in a single beam, which is sent through a femtosecond pulse shaper to tailor a coherently controlled excitation (Exc.) field and an independent phase-managed local oscillator (LO).

Apart from the well-known sources for ultrashort pulses, it would be also very attractive to have a compact broadband excitation based on a standard 100fs - Ti:Sa-oscillator source for these nonlinear optical techniques. This can be achieved by nonlinear broadening of the laser spectrum in a microstructured fibre into supercontinuum and subsequent compression. [4] Such spectrally broad excitation pulses are however difficult to tame and require a precise phase management in order to compensate for inevitable temporal distortions in the microscope setup. To achieve this, the femtosecond pulses can be characterized and shaped *in situ* in the microscope with a simplified SPIDER scheme. In this shaper-assisted collinear (SAC-) SPIDER scheme [5], a femtosecond pulse shaper eliminates the need of a complicated interferometer setup and performs all characterization, compression and tailoring of complex pulses at the same time.

With the application of coherent control techniques on linear optical processes single-beam heterodyne CARS microscopy together with shaped white light continuum and compression by SACSPIDER, all done by a single shaper setup, offers a most robust and experimentally compact approach for sensitive interferometric detection, and will be a valuable tool to gain insight into the microscopic composition of a broad variety of samples.

References:

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