Simple and phase-stabilized setup for heterodyne anti-Stokes Raman scattering microscopy

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Abstract

Coherent anti-Stokes Raman scattering (CARS) is a powerful tool due to its chemical selectivity and label-free nature. However, simultaneously to the resonant CARS process also a nonresonant four-wave mixing process can take place within the sample due to the nonresonant part of the third-order nonlinear susceptibility. The resulting nonresonant signal can be quite strong, and, therefore does lead to a distortion of the Raman lineshape as well as a reduction of image contrast and sensitivity [1].

Heterodyne CARS (hCARS) can be used to separately measure the different contributions to the CARS signal by interferometrically superimposing the CARS signal with a local oscillator (LO). By choosing the relative phase between the CARS signal and the LO, the real and imaginary parts of the susceptibility, containing the nonresonant and resonant signal, respectively, can be measured separately, allowing for background-free detection. In addition, the weak CARS signal is amplified with increasing LO power by the according heterodyne gain [2,3].

We substantially simplified the setup for hCARS by using only a single light source [4] that provides the pump and Stokes as well as the LO pulses. Due to the source-internal generation the LO exhibits a fixed phase relation with respect to the pump and Stokes pulses and shows an up to four times higher power compared to previous implementations [2].

The suppression of nonresonant signal by using hCARS is demonstrated for a sample consisting of oil and PMMA (Fig. 1(a)-(b)) or PS (Fig. 1(c)-(d)) beads, respectively. For the hCARS images the imaginary contribution of the nonlinear susceptibility was measured individually, containing only resonant information, thus leading to a contrast enhancement by a factor of 3.6 and 1.8 compared to standard CARS. Additionally, an active phase stabilization scheme was implemented which enables long-term hCARS measurements for more than 1.5 hours (Fig. 1(e)) even under outside-laboratory conditions.

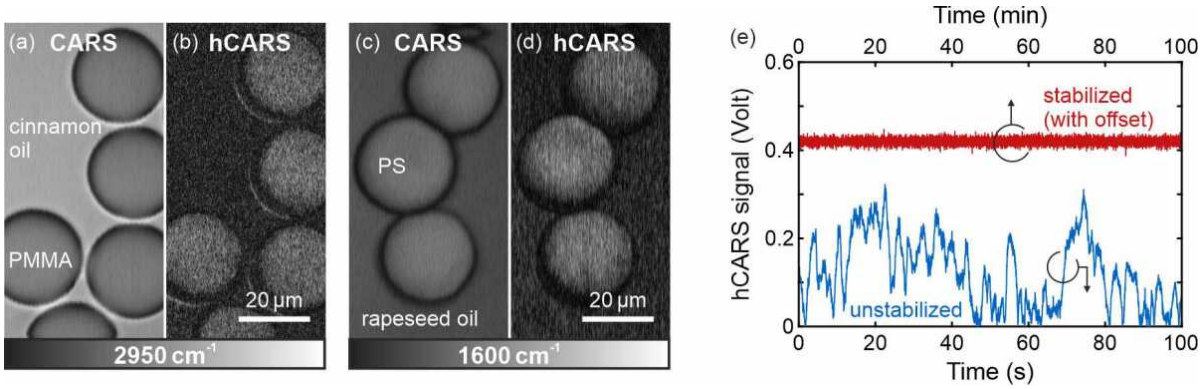

Fig. 1.: (a)-(b) PMMA beads and oil, (c)-(d) PS beads and oil, showing the suppression of the nonresonant signal of the oil by using heterodyne CARS (hCARS). All images have 256 x 512 pixels and were acquired with a pixel dwell time of 12 µs. (e) hCARS signal with the stabilization mechanism switched off (blue) and on (red).

References

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