Targeted single-beam CARS using phase-and-polarization shaping

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A single-beam geometry implementation of a Coherent Anti-Stokes Raman Scattering (CARS) setup [1], is capable of producing well resolved Raman spectra and can be used for CARS microscopy applications. Single-beam CARS (SB-CARS) is an affordable approach to performing CARS measurements, as laboratories with existing femtosecond laser infrastructure can be retrofitted for SB-CARS measurements.

In our implementation (fig. 1.a), an 80 MHz femtosecond laser oscillator, 1 W average power, is used to pump a polarization maintaining all-normal dispersion photonic crystal fibre (ANDi PCF) which produces stable, coherent supercontinuum pulses allowing for the probing of a broad spectral region. Due to dispersion in the PCF, these pulses are temporally broad and require compression for optimal use in non-linear applications. A ptychography based collinear pulse characterisation technique, i^{2}PIE [2], using an SLM in a 4f shaper, is capable of compressing pulses with a complicated spectral phase, in the sample plane. We recently showed a 4 times improvement of signal-to-background in SB-CARS measurements and a 6.5 times improvement in integrated intensity of the SB-CARS response when using i^{2}PIE to compress the supercontinuum pulses, as compared to the standard alternative technique (multiphoton intrapulse interference phase scan, MIIPS) [3]. These optimally compressed pulses, in the sample plane, play a vital role in the success of SB-CARS shaping strategies.

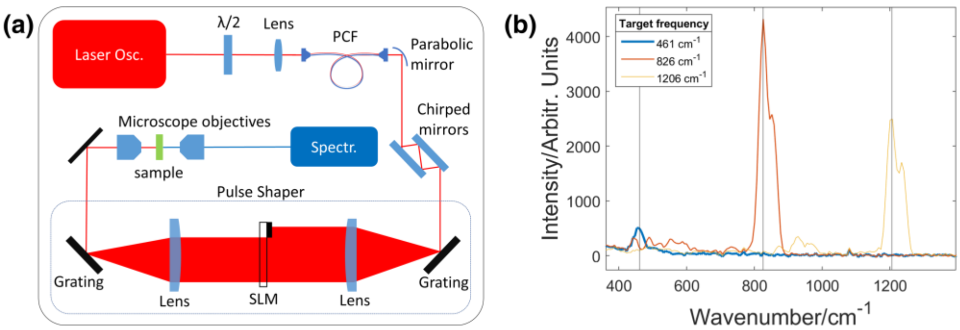

Fig. 1 Schematic representation of SB-CARS setup in (a) and resultant CARS spectra of para-xylene using quadratic phase functions to target chosen resonant transitions in (b).

CARS measurements using a broadband source produce a broad, non-resolved CARS spectrum, where the contributions from non-linear processes dominate the resonant CARS response and thus obscure the information rich resonant CARS spectrum. Central to many of the solutions to this challenge [4–6] is the use of a spatial light modulator (SLM) to actively shape the excitation source and extract the resonant CARS spectrum. The SLM thus has a dual purpose in the setup which allows for collinear pulse compression/characterisation as well as the implementation of SB-CARS shaping strategies.

Phase-and-polarization control of the supercontinuum enables direct and simultaneous measurement of a broad excitation range [4]. We were able to modify this existing approach to also target specific resonant transitions in the CARS spectrum, by tailoring and applying quadratic phase functions to the supercontinuum. As shown in fig. 1.b, only the chosen resonant transitions of para-xylene are present in the respective spectra while other vibrations are suppressed. This approach together with the low average power and high repetition rate laser source has application in microscopy. Additionally, the additive nature of spectral phase, allows us to shape the supercontinuum and measure the temporal evolution of resonant peaks for decoherence studies with a compact and affordable SB-CARS setup.

References

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