Comparison of spectral focusing approaches in single-beam CARS

Ruan Viljoen[1], Dirk Spangenberg[2], Pieter Neethling[1], Alexander Heidt[2], Thomas Feurer[2], Erich Rohwer[1]

1- Laser Research Institute, Stellenbosch University

2- Institute of Applied Physics, Bern University

Single-beam coherent anti-Stokes Raman scattering (SB-CARS) spectroscopy is a special implementation of CARS which utilises a single broadband light source, as opposed to the up to three lasers found in a traditional implementation. This CARS configuration, in combination with spectral focusing, has been shown to be able to target and isolate chosen Raman transitions from broad spectra in applications such as microscopy and stand-off detection. The experimental setup combines our unique white light source with our novel pulse characterization technique to develop new improved methods for SB-CARS.

In this poster we highlight two spectral focusing pulse shaping strategies implemented in our custom setup. Spectral focusing allows for the targeting of chosen Raman transitions within the SB-CARS spectrum. These strategies use the introduction of known phase functions in the form of either quadratic phase functions or carefully tailored binary sequences. We show that the signal-to-background of spectra can be larger by a factor of three when using quadratic phase functions as compared to spectra obtained using binary sequences.

Single-beam coherent anti-Stokes Raman scattering

SB-CARS is a special geometric implementation of CARS, a third order non-linear optical process. Generally, to induce CARS in a molecule, three optical fields are needed. A probe (ω_p) and Stokes (ω_S) field together excite a molecule to an excited vibrational state. A third field, the probe (ω_{pr}), induces the emission of a fourth field, an anti-Stokes (ω_{as}) field. This process is shown in fig. 1.

Phase matching conditions, as illustrated in fig. 2, must be fulfilled. In the case of SB-CARS where the pump, Stokes, and probe fields are contained within a single broadband laser beam, the phase matching is fulfilled using a high numerical aperture microscope objective.

Further requirements for successful SB-CARS, is a single light source with sufficient bandwidth to excite a broad range of molecular excitations simultaneously. A simulation of such a broadband light source, shown in fig.3, with a bandwidth in the order of 100 nm is sufficient to cover a large excitation range. The calculated population amplitude of such an excitation spectrum, shows that a broad probing range is available up to around 3000 cm^{-1}, as seen in fig. 4.

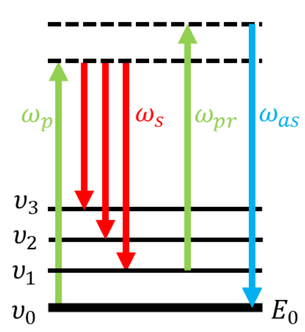

Fig. 1. Diagram showing pump, stokes and probe fields producing anti-Stokes field.

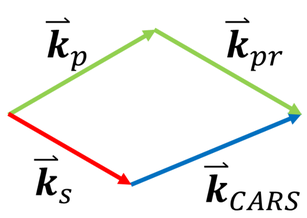

Fig. 2. Diagram illustrating phase matching condition for CARS.

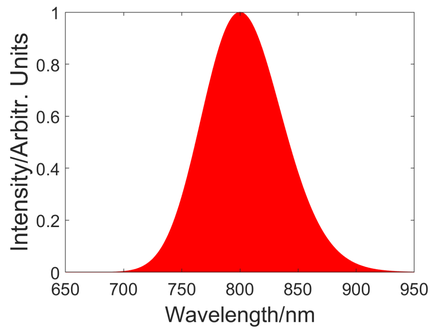

Fig. 3. Simulated broadband excitation spectrum.

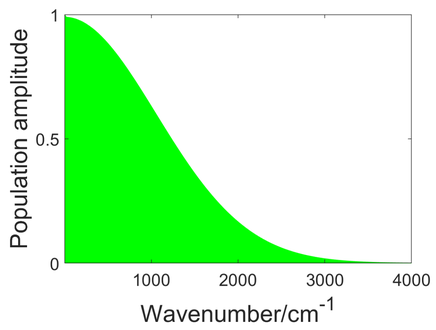

Fig. 4. Calculated population amplitude of simulated excitation spectrum.

Experimental setup

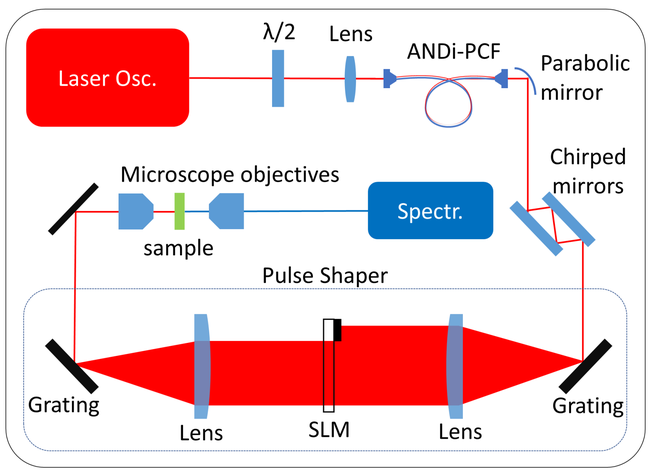

Fig. 5. Diagram showing pump, stokes and probe fields producing Anti-Stokes field.

In our implementation of SB-CARS (as described by fig. 5), a fs-oscillator is used to pump a polarisation maintaining all-normal dispersion photonic crystal fiber (PM-ANDi-PCF) to produce a supercontinuum with sufficient bandwidth (~100 nm) for SB-CARS spectral measurements. Laser pulses are produced at 80MHz and arrive at the sample plane with low pulse energies (0.69 nJ), allowing for fast acquisition of spectra with negligible energy deposited to samples.

Due to dispersion in the PCF, pulses have to be characterised and temporal dispersion is compensated for by using a spatial light modulator (SLM) in 4f-shaper geometry, as a pulse shaper.

Using the capabilities of the pulse shaper, two collinear pulse characterisation and compression techniques were used to compress the supercontinuum pulses. MIIPS, a widely used technique, performed poorly compared to our in-house developed method, i^{2}PIE [1]. This was illustrated when measuring the SB-CARS spectrum of p-xylene.

A measurement was performed using pulses compressed with MIIPS and another with i^{2}PIE and is shown in fig. 6. A signal-to-background increase of a factor 4 [2], was seen when comparing the spectrum obtained using i^{2}PIE compressed pulses to that from MIIPS.

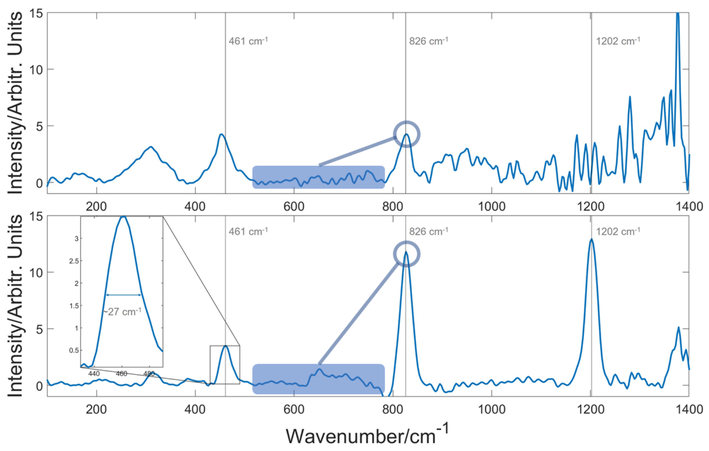

Fig. 6. Para-xylene SB-CARS spectrum obtained using pulses compressed with MIIPS (top) and i^{2}PIE (bottom).

Spectral focusing approaches

Spectral focusing is a technique utilised in CARS to 'focus' the entire excitation spectral bandwidth to excite only a single chosen transition.

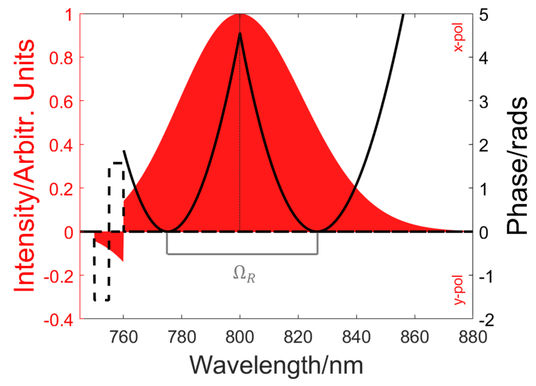

Fig. 7. Pulse shaping scheme for quadric phase spectral focusing.

In SB-CARS, because we have control of the spectral phase, this can be actioned by applying quadratic phase functions (QPF) to the pump part of the spectrum. A QPF is equivalent to a temporal chirp. Thus, dividing the pump in two and adding identical QPF to those sections, produce two temporally chirped pulses. The interference of these pulses produce pulse trains with a periodicity that match the chosen focused transition. In the spectral picture, however, this process bins the pump.

The addition of the identical QPFs creates spectral pairs that are always in phase with each other and out-of-phase with other spectral components. In order to tailor the QPFs for a particular transition, the turning points of the QPFs are positioned to be a 'spectral distance' away from each other that match the frequency of the chosen target transition (as shown in fig. 7). The extent of focusing can further we tailored by changing the steepness of these functions, where an increase of the steepness corresponds to an increase of the focusing 'strength'.

Another approach to SB-CARS spectral focusing uses low autocorrelating binary sequences (BPFs). Two identical BPFs similarly bins the pump and generates frequency pairs that are always in-phase with each other and out-of-phase with other spectral components. This shaping approach is shown in fig. 8. In this scenario the focusing strength depends on the bin number of the chosen sequence and the extent of low correlation between two BPFs. These properties can, however, not be tailor made and are unfortunately constrained by the physical SB-CARS setup and the existence of these sequences.

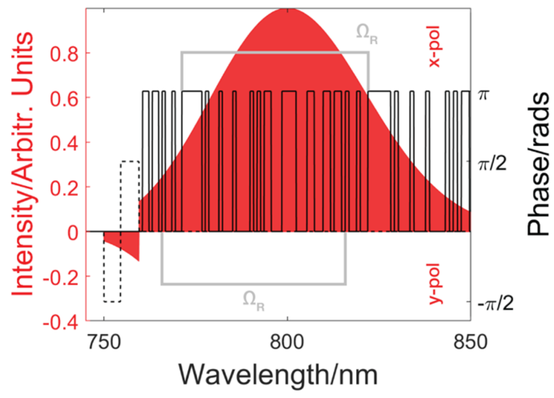

Fig. 8. Pulse shaping scheme for binary phase spectral focusing.

Spectral focusing measurements

Target frequency (cm^{-1})	S:B 10 000 fs^2	S:B 15 000 fs^2
461	4	13
826	41	68
1206	32	30

Spectral focusing with QPFs was used to target resonant transitions of para-xylene and the signal-to-background was quantified for different resonances and summarised in above table. In fig. 9 we show the measured spectral focused para-xylene spectra. It also shows that in increase in QPF steepness results in improved signal-to-background.

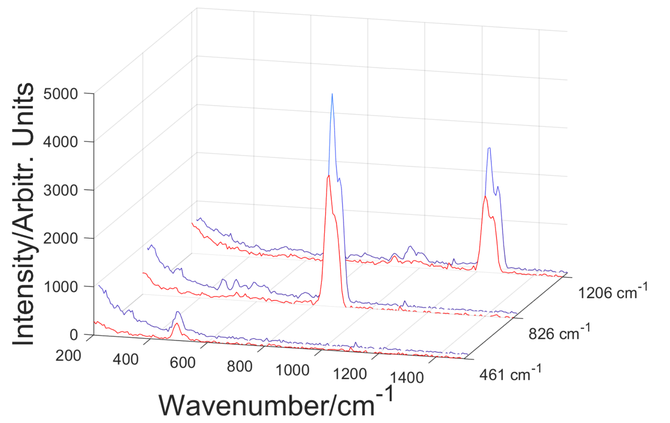

Fig. 9. SB-CARS spectra of para-xylene with supercontinuum shaped to focus chosen transitions. Steepness values of QPFs were adjusted from 10e3 fs^2 (red) to 15e3 fs^2 (blue).

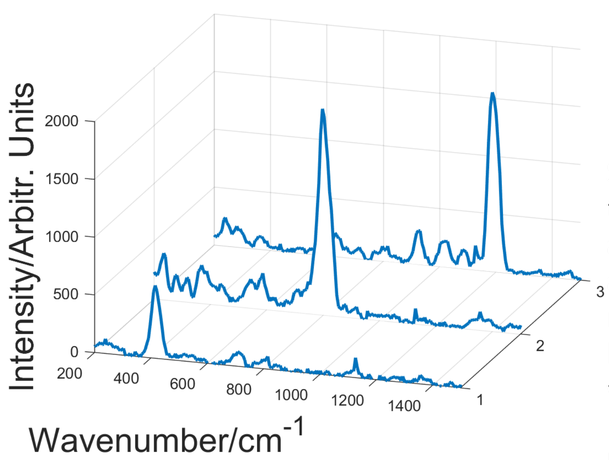

Fig. 10. SB-CARS spectra of para-xylene with supercontinuum shaped to focus chosen transitions using BPFs.

Target frequency (cm^{-1})	Bin number	S:B
461	41	18
826	79	23
1206	122	24

Spectral focusing with BPFs was also used to target chosen resonant transitions of par-xylene and is shown in fig. 10. Comparing S:B values (in above table) obtained with those from QPF measurements, show that an improvement can be gained by up do 3 times (comparing those values obtained for 800 cm^{-1}).

In practice it is clear that QPFs provide more tailoring flexibility, with comparable focusing strength to that of BPFs and should be the approach of choice.

[1] D.M. Spangenberg, E. Rohwer, M. Brügmann, and T. Feurer, "Extending time-domain ptychography to generalized phase-only transfer functions," Opt. Lett. **45**, 300 (2020).

[2] R. Viljoen *et al.*, "Implementation of temporal ptychography algorithm, i^{2}PIE, for improved single-beam coherent anti-Stokes Raman scattering measurements," *J. Opt. Soc. Am. B*, vol. 37, no. 11, p. A259, Nov. 2020

science & technology
Department:
Science and Technology
REPUBLIC OF SOUTH AFRICA

This work is based upon research supported by the South African Research Chair Initiative of the Department of Science and Technology and the National Research Foundation

Contact: rviljoen@sun.ac.za