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Analysis of spectral coverage and resolution performance in spectral focusing CARS spectroscopy

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Abstract: An open-code simulation tool for spectral focusing Coherent Anti-Stokes Raman Scattering (SF-CARS) spectroscopy has been developed for the analysis of spectral coverage and resolution performance. The main limitation has been found due to high-order dispersion and nonlinearities in laser sources.

1. Introduction

Coherent anti-Stokes Raman (CARS) microscopy and spectroscopy is an emerging technique in which two narrowband pulses (pump and stokes beams) are typically used [1]. The interaction of the pulses generates a beating frequency signal resonant to a Raman molecular vibration. An interesting approach where broadband femtosecond laser sources can be used for the same purpose is by chirping the pulses, referred to as the spectral focusing (SF) technique [2]. Equally chirped pump and Stokes beams can lead to a constant Instantaneous Frequency Difference (IFD), described as the difference in frequency between the two pulses $\Omega_R = \omega_P - \omega_S$, that can excite a certain Raman mode at frequency Ω_R. The IFD and therefore the excited Raman line can be chosen by the relative temporal delay between the pump and the Stokes pulses. By changing the time delay between the pulses, a complete CARS spectrum can be recorded [3]. In CARS spectroscopy, in order to identify and map multiple molecules it is advantageous to use spectrally broad sources and therefore broad spectral coverages $\Delta\Omega$. In SF-CARS, an optimized chirp along with fast a delay scanning device, are able to achieve the desired resolution and high-speed acquisition without compromising spectral coverage. The spectral resolution is defined by the ability to distinguish two closely locating resonances which can be approximated to the bandwidth of the IFD and thus to the narrowness of the interaction bandwidth of the pump and Stokes beams $\Delta\Omega_R$. As explained in [3], the best spectral resolution is achieved when the pulses are chirp-matched, that is when the chirp rate is made equal for both the pump and Stokes pulses. This condition enables the minimum bandwidth of the IFD signal. Also, it has been demonstrated that using pump and Stokes pulses with larger bandwidths results in an enhancement of the spectral resolution for the case of chirp-matching [2,3]. Thus, in SF-CARS spectroscopy the spectral resolution is limited both by the duration of the chirped pump and Stokes pulses and by how precisely the chirp rates are matched. Furthermore, the two central wavelengths of the pump and Stokes beams need to be chosen carefully to excite the relevant spectral region both in the fingerprint region (between 400 and 1600 cm^{-1}), and excitation of C-H stretching vibrations (2800-3200 cm^{-1}) for specific applications. Multiple excitation sources such as Er-doped fiber lasers, Ti:sapphire or sub-nanosecond microchip lasers have been implemented in the last decades along with SC generation in nonlinear fibers such as photonic crystal fibers (PCF) or high nonlinear fibers (HNLF) [4,5]. These schemes have been able to measure spectral resolutions as low as 15 cm^{-1} in the fingerprint region and 30 cm^{-1} for the lipid region [6]. However, there still exists some limitations related to spectral coverage and spectral resolution due to effects such as, misaligned chirps of the input pulses, higher order dispersion, and nonlinear effects due to the employment of different type of fibers in the experimental setups. These effects have been found to be difficult to compensate or eliminate at high input powers, thus limiting the spectral resolution and spectral coverage [5-7].

In this work we present a detailed simulation tool developed in MATLAB® to simulate spectral focusing technique for CARS applications in order to understand the limitations of the spectral resolution and spectral coverage for a specific laser source. To do that we study the relationship between the bandwidth of the input pump and Stokes beams, their corresponding chirp and the influence of high-order dispersion and nonlinear parameters. Overall, this work offers a simple method to find the best spectral resolution and spectral coverage for each spectroscopy scheme and thus to enable further improvements of the system.

2. Theory and Simulation Results

In coherent Raman spectroscopy, the overall bandwidth $\Delta\omega_R = \sqrt{\Delta\omega_p^2 + \Delta\omega_S^2}$ for the Raman excitation can be calculated from the bandwidth of the nonlinear interaction of the pump and Stokes electric fields (Raman active vibration) $I_R \approx \tilde{E}_p \tilde{E}_S^*$. Therefore, the spectral resolution converted to wavenumbers can be calculated as [8],

$$\Delta\omega_R = \frac{2Ln2}{\pi}\sqrt{2\left(\frac{1}{\tau_p^2}+\frac{1}{\tau_s^2}\right)} \quad (1)$$

Where τ_p and τ_s correspond to the chirped pulse widths of the pump and Stokes beams, respectively. Experimental setups based on chirping devices such as gratings or prisms generate the temporal pulse broadening by introducing a certain amount of Group Delay Dispersion (GDD). However, these devices not only apply GDD but also Third-Order Dispersion (TOD) and higher-order dispersion coefficients, thus decreasing the spectral resolution. Moreover, due to the TOD, the time to frequency mapping of the pulse (see Fig1(a)) is no longer linear, which implies that some spectral disturbances appear in the Raman spectrum during the scanning process of the Raman frequencies [8]. An example of this has been simulated in a home-built MATLAB code by applying a generalized femtosecond laser source of 100MHz with two different initial chirped Gaussian pulses at 1µm (pump) and 1.55 µm (Stokes) with a pulse width of 150fs and 1ps, respectively. These input pulses are generated from established pulse propagation simulation using split-step Fourier transform technique. A maximum energy of 10nJ for the pump and 1nJ for the Stokes has been used for this simulation. The code analyzes the FROG (Frequency Resolved Optical Gating) traces for both pulses in order to compare and check their corresponding chirp parameter (Fig.1(a)) and the intensity distribution of the Raman interaction for every time-delay (Fig.1(b)). From these figures the total IFD value for each time delay can be obtained. This gives information on the spectral coverage, in this case measured from 1000 to 4200 cm^{-1} (marked in Fig1(b) with red lines) and thus extending both for the fingerprint and lipid region. The corresponding spectral resolution can be described by the bandwidth of the interaction (measured about 25cm^{-1} for this scheme). As shown in Fig.1(a) and (b), the influence of high-order dispersion introduces nonlinearities in the frequency to time mapping and thus a broadening in the Raman intensity distribution, yielding a worsening of the spectral coverage and resolution.

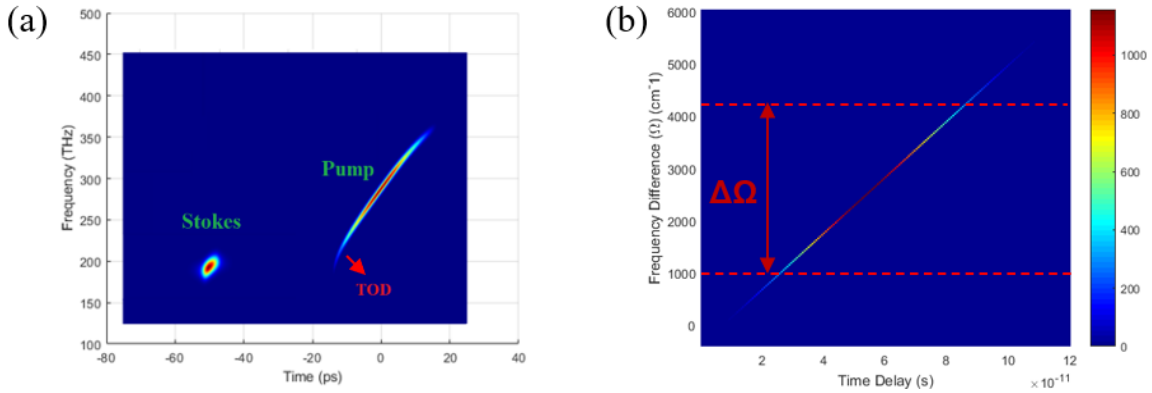

Fig. 1. (a) Frequency-time mapping of the two chirped pulses: pump (right) at 1µm with 150 fs duration and 260 nm spectral bandwidth and Stokes (left) at 1.55 µm with 1ps duration and 40 nm spectral bandwidth. (b) Intensity distribution of the Raman interaction for each time-delay and spectral coverage.

In conclusion, a complete Raman Response simulation for SF-CARS spectroscopy has been developed. The simulation code takes pulse input from any type of laser source (bandwidth, chirp, dispersion, etc.) in order to evaluate the performance of the system. This tool will offer the possibility to optimize Raman spectroscopy setups in all sorts of pumping and readout conditions, offering the ability to find the best possible implementation in SF-CARS spectroscopy.

3. References

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