High-Spectral-Resolution Stimulated Raman Spectroscopy with Amplified fs Pulse Bursts

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Abstract: We propose a new approach to a species-selective SRS gas sensing combining the advantages of strong signal levels and the ease of optical frequency conversion, obtainable with broadband fs pulses, with the high frequency resolution characteristic of narrowband sub-ns pulses. Spectra are mapped by electronic phase control of the amplified burst.

Four-wave mixing techniques based on stimulated Raman scattering (SRS) play a very prominent role in resonant nonlinear spectroscopy, used in environmental and biomedical sensing, by providing complementary information to linear optical absorption spectroscopy through addressing Raman-active (and thus optically-inactive) vibrational and rotational transitions. Unlike in the linear spectroscopy case, the detected signal strength of coherent anti-Stokes emission (CARS), stimulated Raman pump loss (SRL) and Stokes gain (SRG) scale nonlinearly and depend on a product of input field intensities, which greatly favors the use of intense femtosecond pulses for signal scaling. Given the natural pressure-dependent broadening of Raman transition lines, an ideal detection scheme in the simplest two-color case (SRG or SRL) would involve a pair of temporally coincident narrowband transform-limited pulses with durations in the 2—100-ps range and their center frequencies detuned by Raman transition (vibrational) frequency. As a rule, the frequency-detuned pulse replica of the pump pulse is generated in a highly inefficient two-stage ns OPO and the SRS spectra are recorded by tuning the OPO output optical frequency. The past two decades saw the development of a number of approaches, where the advantages of intense broadband pulses for optical frequency conversion [1] were harnessed while attempting not to wash out the spectral resolution. One elegant solution to the latter problem is implemented in the so-called spectral focusing [2], whereby two broadband pulses with detuned center frequencies are strongly chirped with exactly parallel and strictly linear chirps such that the difference between their carrier frequencies is constant at every instance within the temporal overlap of their envelopes. Spectral scanning is then achieved by mechanically changing the pathlength difference between the chirped pulses. In practice, frequency resolution of the spectral focusing scheme suffers from imperfect dispersion management (chirp linearization) [3].

In this contribution, we introduce and justify numerically an alternative fs pulse-burst technique, summarized in Fig. 1. Compared to a spectral focusing technique based on a pair of chirped pulses, the burst approach requires neither precise dispersion management nor optical delay scanning with movable parts.

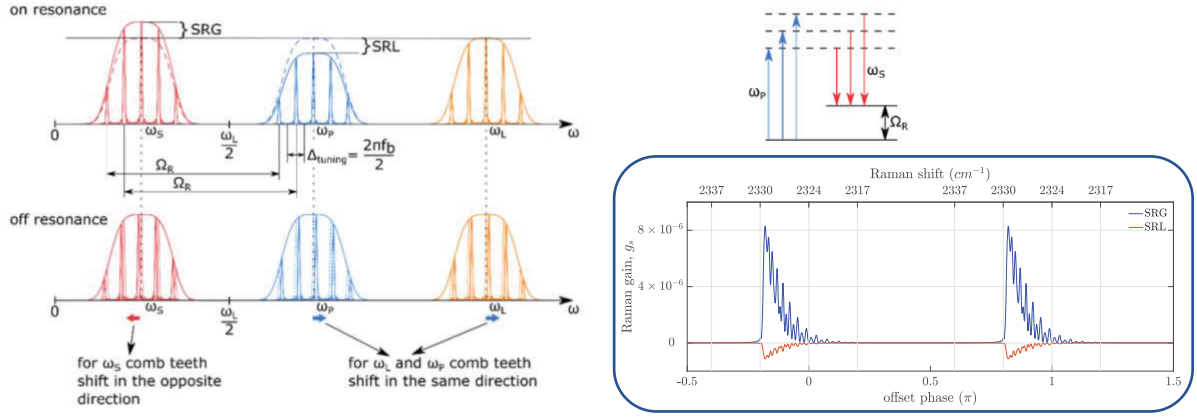

Figure 1: Stimulated Raman scattering using two femtosecond pulse bursts, generated by optical parametric amplification, with their center frequencies detuned by the Raman resonance frequency.

The main technical prerequisite for implementing the method in Fig. 1 has been recently realized in our group [4] and is based on a programmable generation of amplified fs pulse bursts in a regenerative amplifier (RA) operated in the Vernier mode relative to the cavity length of the master oscillator (MO). As experimentally shown in [4], spectral interference of the pulses in the generated bursts leads to a comb-like structure with a 1 THz ≈ 33.4 cm^{-1} intermodal

distance if the detuning between the RA and MO roundtrips $\Delta\tau = |L_{RA} - L_{MO}|/c$ = 1 ps. The THz-spaced pseudo-modes have the spectral width determined by the inverse duration of the burst, $(\Delta\tau N)^{-1}$, with N being the number of pulses in the burst, and are filled with a very dense, MHz-interval-frequency comb structure that is irrelevant for our study. The peak frequencies of the pseudo-modes, corresponding to constructive spectral interference, are programmed, alongside the individual amplitudes of the pulses forming the burst, by use of an AOM placed between the MO and RA. We note that the aspect of chirp management becomes irrelevant for the spectral narrowing that is achieved in this case purely through interference. Correspondingly, a white-light-seeded OPA driven by such a pulse burst will behave exactly like an OPA driven by a single isolated fs laser. As the result, an independent signal-idler pulse pair will originate from each laser pulse in the burst and similar pseudo-combs will arise under the respective spectral envelopes (Fig. 1) through the spectral interference of signal and idler pulse bursts. We further note that due to phase conjugation in a properly designed OPA [5], the phase control of the fundamental laser burst will cause spectral modes to shift in the opposite directions for the signal and idler pseudo-combs. Therefore, as shown in Fig.1, resonant SRS conditions can be simultaneously fulfilled or simultaneously missed by multiple pseudo-modes depending on the input phase of the pulses loaded into the RA. The free-space frequency scanning range corresponds to half the intermodal spacing, i.e. to ~17.2 cm^{-1} under the assumed conditions, which is adequate to resolve a complete vibrational manifold structure of simple molecules (e.g. N_2) when the signal pulse burst is taken as an SRS pump wave and the idler burst – as a Stokes wave, respectively. The periodic jump to the next set of resonant pump-Stokes mode pairs is shown for atmospheric N_2 in the inset to Fig. 1 and indicates the easy phase-to-frequency calibration procedure.

The critical point to validate this technique is to prove that the broadband SRS signal, arising from the interaction with each respective pair of fs signal and idler pulses in the time-coincident bursts, will nevertheless retain the expected spectral resolution given by $(\Delta\tau N)^{-1}$. Using N_2 as a model system, we present exhaustive numerical simulations, Figure 2, based on the solution of a time-dependent nonlinear Schrödinger equation that compare the performance of i) various bursts with conventional two-pulse cases: ii) transform-limited narrowband pulse pair and iii) chirped pulse pair under ideal spectral focusing conditions.

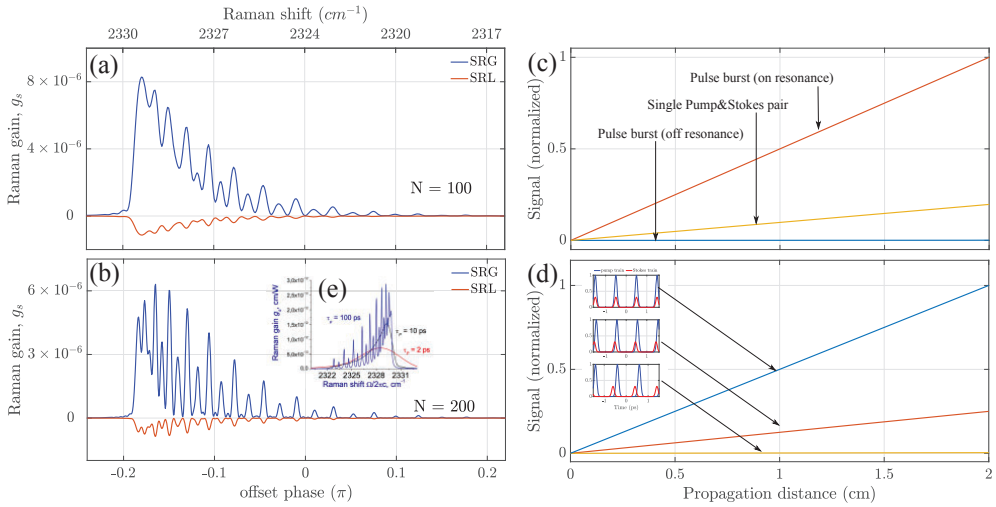

Figure 2: Numerical simulation of burst-driven SRL/SRG in N_2 at atmospheric pressure. a, b) Signal dependence on the burst phase for N = 100 and N = 200. c) Scaling of SRG signal for bi-color burst driving (0.1-ps pulse duration, 1-ps interpulse interval) and bi-color pulse driving when the burst duration ΔτN and the ps driver pulse durations are equal and the respective pump and Stokes energies are also identical for both cases. Note favorable scaling for the case of bursts which is due the due to higher peak intensity. d) Suppression of SRS signal when the pump and Stokes bursts are not time-coincident. e) Spectral resolution of a conventional bi-color SRS scan for a pulsed pump and cw Stokes wave as a function of TL duration of the pump pulse.

References

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