Mirrorless Backward SRS in Free-Space Gas Driven by Filament-Initiated UV Laser

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Abstract: Stimulated Raman scattering combined with ASE lasing from nitrogen molecules in a femtosecond filament is shown to provide a highly directional chemical-bond-sensitive coherent readout in the direction opposite to that of the driver laser beams.
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1. Introduction

Generation of a backward propagating coherent emission is of crucial importance to standoff gas and atmospheric sensing as a way to enable coherent signal enhancement. In contrast to incoherent light scattering techniques, the use of coherent enhancement permits to transmit a species-selective signal with high directionality, thereby allowing reduction of the input laser power and an increase of the standoff distance. In wave-mixing schemes, phase-matching in the backward direction requires, in the general case, a backward-directed input laser beam, which is a challenge for free-space standoff detection techniques that are not relying on surface reflections. The recent demonstration of backward lasing from oxygen gas under pulsed deep UV excitation [1,2] and from nitrogen gas in a femtosecond mid-IR filament [3] suggest possibilities for realizing a backward-phase-matched scheme in air.

In this contribution we report on what we believe to be the first demonstration of a backward phase-matched coherent Raman detection scheme in a gas where one of the input waves is provided by a standoff filament-initiated nitrogen laser emitting backward UV pulses at 337 nm.

2. Experimental results

The concept scheme of the experiment is shown in Fig. 1. The laser source is a modified mid-IR multi-mJ 20-Hz optical chirped parametric amplifier (OPCPA) reported previously [4]. The driving laser emits two pulses: an intense 3.9-μm femtosecond pulse to induce a filament in a gas cell filled with a N_2 and Ar mixture and a tunable UV picosecond pulse serving as a Stokes pulse for stimulated Raman scattering (SRS). The standoff lasing initiated by mid-IR filamentation provides a fixed-wavelength picosecond pump pulse at 337 nm that propagates in the anti-collinear direction with respect to the Stokes pulse. The Stokes arm is augmented to the OPCPA setup using residual pump energy from the picosecond Nd:YAG laser and the broadband signal wave centered at 1460 nm, as schematically depicted in Fig. 2. The 1460-nm pulse is frequency-doubled, frequency narrowed using a diffraction-grating-based spectral filter and parametrically amplified using 532-nm pulses from the second harmonic of the Nd:YAG laser. The spectrum of the signal pulse of the OPCPA extends between 1420 and 1520 nm which corresponds to the tunability range of the fourth harmonic between 355 and 380 nm. This covers the range of Raman shift between 1500 and 3350 cm^{-1} with respect to the 337-nm pump pulse generated by the filament laser.

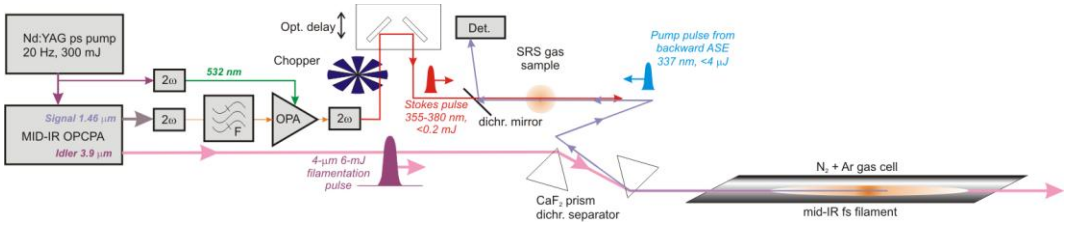

Fig. 1 Experimental setup. Note that the laser system emits two synchronized pulses in the forward direction: the tunable Stokes pulse and the intense mid-IR pulse for filamentation. The backward 337- nm is emitted by the filament. Detector *Det.* registers a Stimulated Raman Loss of the 337-nm pump beam. F, grating-based spectral filter for narrowing the 748-nm second harmonic of the signal wave. Because of the absence of a UV/mid-IR dichroic mirror capable of handling high-intensity mid-IR, we were forced to depart from a truly free-space geometry by introducing a pair of closely spaced CaF_2 prisms.

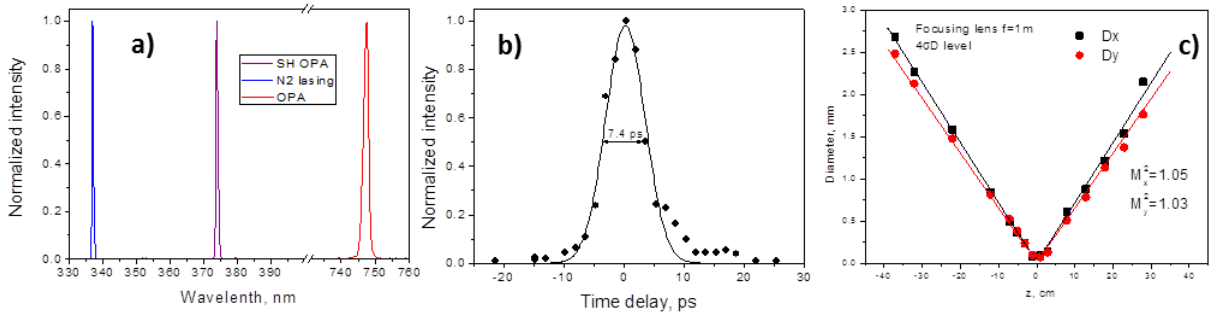

Fig. 2 Performance summary of the Stokes and pump pulses used in the SRS experiment. (a) measured spectra of the output of the NIR OPA seeded with frequency doubled signal pulses of the mid-IR OPA, spectrum of the Stokes pulse resulting from an additional frequency doubling of the NIR OPA output. (b) cross-correlation of the 374-nm Stokes pulses with 200-fs 1030-nm reference pulses from an Yb:CaF_2 seed amplifier. (c) M^2 measurement of the free-space-generated 337-nm beam indicating a near diffraction-limited divergence.

Fig. 2 summarizes the properties of the pump and Stokes pulses. Fig. 2a shows the spectrum of the filament-induced backward 337-nm pulses (measured FWHM ~0.35 nm), the spectrum of the OPA tunable around 747.8 nm and its second harmonic near 374 nm (measured FWHM ~0.7 nm). Fig. 2b presents the cross-correlation trace between a 200-fs pulse and the Stokes pulse. Fig. 2c gives the results of an M^2 measurement of the backward ASE pulse which indicate that the backward emitted beam has a nearly diffraction limited divergence.

The target gas used in this experiment was methane that exhibited reliably measurable stimulated Raman loss (SRL) on the pump beam peaking at the detuning frequency of 2920 cm^{-1}. The results of standoff methane detection via SRL are summarized in Fig. 3 which shows dependencies of the SRL signal on pump—Stokes delay(Fig.3a), frequency detuning (Fig.3b) and gas concentration (Fig. 3c). At the present level, the sensitivity of the method is very low, requiring the use of a target with an unrealistically high concentration. The lowest detectable pressure of methane is 1 bar, corresponding to a SRL signal of 10^{-3}. Also the ASE pump laser, although truly free-space in terms of the pumping and lasing scheme, is nevertheless confined to a long gas cell required to raise the gas concentration to the level compatible for mid-IR filamentation with the currently available pulse energy at 3.9 μm. In addition, developing a high damage threshold mid-IR –UV dichroic beamsplitter required to merge the UV and the filamenting beam is a challenge. The interim solution was the use of a prism-based dichroic separator that was a further compromise of free space geometry. These obstacles should be removed with the expected growth of the mid-IR few-cycle pulse peak power that should enable in the near future filamentation in ambient air at the atmospheric pressure allowing optimal use of the high ponderomotive energy of the mid-IR pulse in the intensity-clamping regime of a filament.

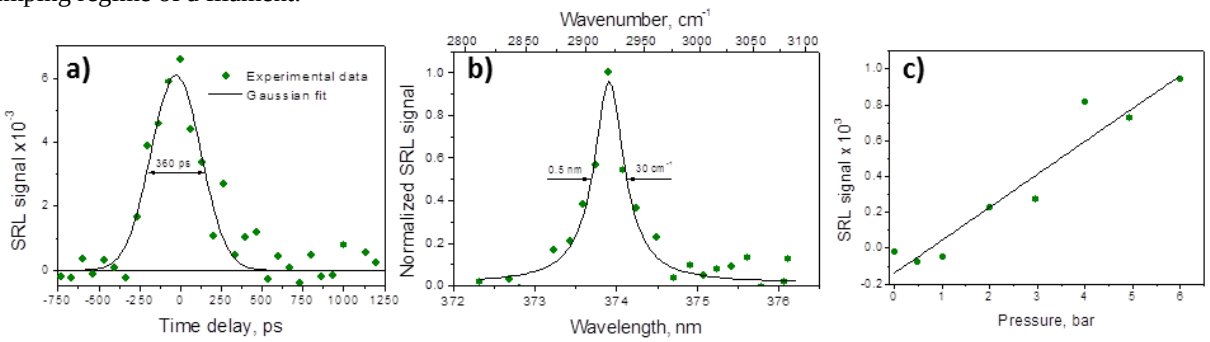

Fig. 3 (a) Dependence of SRL signal in CH_4 (at 6 bar pressure) on the delay between the Stokes and pump pulses. (b) SRL spectrum measured in CH_4 at 6 bar; Black solid line shows a Lorentzian function fit. (c) SRL signal dependence on the CH_4 concentration.

In conclusion, we have realized the first successful demonstration of a backward coherent enhancement of Raman signal using filament initiated standoff source of a coherent, backward propagating emission.

[1] A. Dogariu, J.B. Michael, M.O. Scully, R.B. Miles, "High-gain backward lasing in air", Science 331, 442–445 (2011).
[2] A.J. Traverso, R. Sanchez-Gonzalez, L. Yuan, K. Wang, D.V. Voronine,b, A.M. Zheltikov, Y. Rostovtsev, V. A. Sautenkov, A.V. Sokolov, S.W. North, and M.O. Scully, "Coherence brightened laser source for atmospheric remote sensing", Proc. Natl. Acad. Sci. USA 109, 15185–15190 (2012).
[3] D. Kartashov, S. Alisauskas, G. Andriukaitis, A. Pugzlys, M. Shneider, A. Zheltikov, S.L. Chin and A Baltuska, "Free-space nitrogen gas laser driven by a femtosecond filament", Phys. Rev A. 86, 033831 (2012).
[4] G. Andriukaitis, T. Balčiūnas, S. Ališauskas, A. Pugžlys, A. Baltuška,T. Popmintchev, M.-C. Chen, M.M. Murnane, H.C. Kapteyn, "90-GW Peak-Power Few-Cycle Mid-IR Pulses from an Optical Parametric Amplifier", Opt. Lett. 36, 2755 (2011).