Stimulated Raman gas sensing by backward UV lasing from a femtosecond filament

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We perform a proof-of-principle demonstration of chemically specific standoff gas sensing, in which a coherent stimulated Raman signal is detected in the direction anti-collinear to a two-color laser excitation beam traversing the target volume. The proposed geometry is intrinsically free-space as it does not involve back-scattering (reflection) of the signal or excitation beams at or behind the target. A beam carrying an intense mid-IR femtosecond pulse and a parametrically generated picosecond UV Stokes pulse is fired in the forward direction. A femtosecond filament, produced by the intense mid-IR pulse, emits a backward propagating narrowband picosecond laser pulse at the 337-nm and 357-nm transitions of excited molecular nitrogen, thus supplying a counter-propagating Raman pump pulse. The scheme is linearly sensitive to species concentration and provides both transverse and longitudinal spatial resolution.

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Coherent enhancement of weak signals through phase matching is one of key strengths of nonlinear laser spectroscopy. In standoff spectroscopic sensing [1] the use of nonlinear coherent enhancement would permit to extract a highly directional species-selective signal without the trade-off between standoff distance and input laser power that occurs with incoherent techniques. Standoff optical sensing requires that the wave-vector of one or more impinging or generated laser fields is inverted, so that the beam propagates back towards the source and the detector, providing an optical signature of the target volume. In dense optical media strong nonlinearities enable artificial nonlinear mirrors, based, for example, on phase-conjugation schemes [2, 3] or on plasma reflection [4]. In low-density gaseous media, on the other hand, in the absence of back-scattering/-reflection at hard surfaces, it is extremely challenging to achieve wave-vector inversion, producing a coherent beam that retraces its way back to the laser. This difficulty is due to momentum conservation which strongly prohibits wave-vector reversal, while the optical nonlinearities in the gas phase are too weak to enable it.

To acquire a coherent standoff signal from a gas volume without the use of back-scattering/reflecting surfaces, one needs to provide a spatially-coherent back-propagating beam originating from the gas itself. The recent demonstrations of the oxygen atmospheric laser, emitting NIR pulses under deep UV excitation [5], and of high-energy UV lasing of nitrogen molecules in a filament induced by ultrashort mid-IR pulses [6], provide candidates for the role of temporally synchronized, backward propagating beams supporting a phase-matched scheme. In such mirrorless free-space or "air" lasers, excited molecules emit amplified stimulated emission (ASE) pulses at fixed wavelengths both in the forward and backward directions with respect to the excitation beam.

These recent breakthroughs in atmospheric laser sources strongly motivate the search for efficient nonlinear spectroscopy schemes, whereby the backward-propagating fixed-frequency atmospheric laser beam interacts with a forward-propagating frequency tunable beam, thus providing information on the chemical content of the atmosphere [7]. Coherent Raman spectroscopy, with its inherent chemical sensitivity, is an ideal approach in this respect, although the choice of the specific technique is severely limited by phase-matching issues. Coherent anti-Stokes Raman scattering (CARS), which is powerful in forward detection [8] and in the epi-detection mode in microscopy [9], in the case of counter-propagating beams suffers from a large wave-vector mismatch between the interacting waves, which strongly limits its efficiency [7, 10]. In laboratory experiments, this problem can be overcome by retro-reflection/scattering of a forward generated CARS signal [11-14], which however cannot be applied in a standoff detection arrangement. On the other hand, the stimulated Raman scattering (SRS) technique, detecting either the stimulated Raman gain (SRG) at the Stokes frequency or the stimulated Raman loss (SRL) at the pump frequency [15], is intrinsically phase-matched

and thus optimally lends itself to a counter-propagating geometry. Recently, we have performed a feasibility study of backward SRS signal detection from a gas volume pumped and probed by two counter-propagating narrowband UV pulses [16], steered into each other with mirrors in order to mimic a standoff detection geometry.

This work provides a proof-of-principle demonstration of gas sensing by an SRS process in a standoff geometry, where counter-propagating pump and Stokes pulses are generated from a pair of forward-directed laser beams. The backward propagating UV pump is provided by an atmospheric nitrogen laser triggered by an intense mid-IR femtosecond pulse. Interaction with a tunable forward propagating Stokes pulse, mediated by the sample gas, imprints on the pump pulse an SRL that is detected at the location of the laser source. This arrangement represents a significant conceptual step forward with respect to the model experiment in [16], where the counter-propagating beams were obtained by steering the pump and the Stokes beams into each other with mirrors. Here we report on a counter-propagating scheme that is intrinsically self-generated from a forward-directed beam, by a sequence of processes set off by a femtosecond filament. Moreover, our work provides the first demonstration, to our knowledge, of nonlinear-optical signal generation using a pulse from a filament-induced free-space laser.

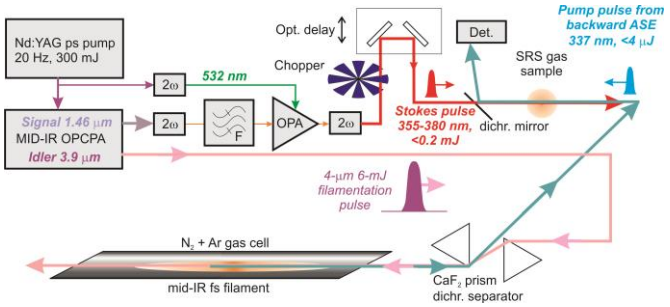

Fig.1. Experimental setup. The laser system emits two synchronized pulses in the forward direction: the tunable Stokes pulse and the intense mid-IR pulse for filamentation. The backward 337-nm beam is emitted by the filament. Detector Det. registers a SRL of the 337-nm pump beam. F, grating-based spectral filter for narrowing the 748-nm SH of the OPCPA signal wave. Because of the lack 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.

The schematic of the experiment is presented in Fig. 1. The laser source is a modified multi-mJ femtosecond (fs) optical parametric chirped pulse amplifier (OPCPA), reported previously [17], pumped by a picosecond (ps) Nd:YAG laser and seeded by the stretched output of an optical parametric amplifier (OPA), which in turn is pumped by a fs Yb:CaF_2 laser. The Raman pump arm is based on the approach reported in [6], whereby standoff lasing is obtained in nitrogen within a filament ignited by the mid-IR idler of the OPCPA. To drive the filament laser, the 3.9-μm OPCPA pulse with 6-mJ energy and 80-fs duration, at 20 Hz repetition rate, is focused by a f= 1 m lens into a 4-m-long gas cell with Brewster-angled CaF_2 windows, containing a mixture of N_2 and Ar. For an Ar pressure exceeding 3 bar, lasing is achieved simultaneously at two lines at 337 nm and 357 nm. For the optimal mixture (1 bar N_2 and 5 bar Ar), the measured total energy at both 337-nm and 357-nm lines is ≈3.5 μJ, corresponding to a 5×10^{-4} energy conversion efficiency from the mid-IR laser pulse to the UV radiation. Laser emission at the 337(357)-nm line is pulsed with ≈1(2)-ns duration, as determined by a fast photodiode [6]. The spectrum of the 337-nm line, shown in Fig. 2(a), was measured to have 0.035 nm (<2 cm^{-1}) width by a Czerny-Turner monochromator. The spatial beam profile of the laser has a roughly super-Gaussian shape, with divergence of ≈1.6 mrad [6]. Referring back to the prototype experiments described in [16], the parameters of the atmospheric nitrogen laser (pulse energy, pulse duration, spatial beam quality) are very well suited for the backward SRS sensing scheme.

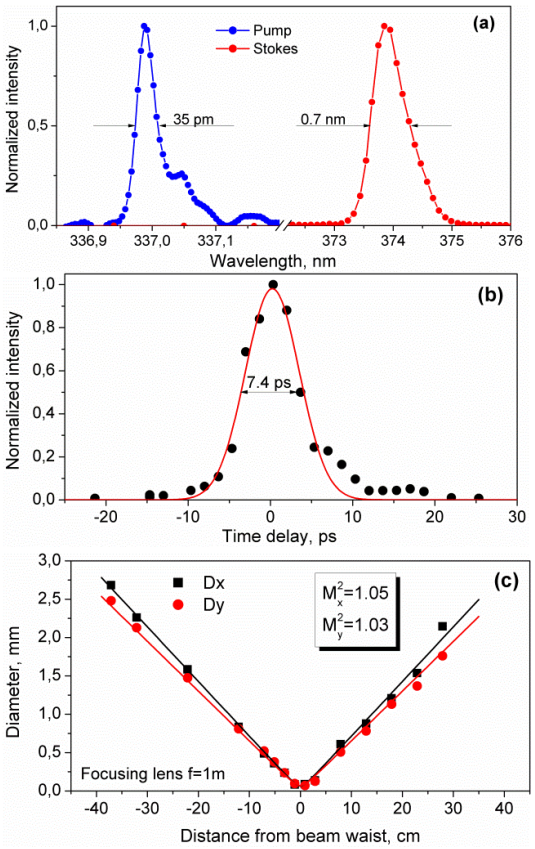

Fig.2. (a) Spectra of pump and Stokes pulses, as generated form the N_2 laser filament and from the FH of the OPCPA signal. (b) Cross-correlation of the Stokes UV pulse with a 200 fs NIR pulse. (c) Diameter of the backward propagating filament-induced pump pulse, as measured with a beam profiler after a 100-cm focal length lens.

The conceptual architecture of the remote sensing with SRS requires the combination of the backward propagating, remotely pumped atmospheric N_2 laser pulse (acting as a Raman pump) with a narrowband tunable Stokes pulse sent in the forward direction. When the pump-Stokes frequency detuning matches the Raman resonance of the molecule to be detected, the backward propagating Raman pump experiences SRL, enabling

molecular fingerprinting. For this experiment we chose the 337-nm wavelength of the atmospheric laser. The Stokes pulses were obtained by nonlinear frequency conversion starting from the same laser system that drives the mid-IR OPCPA, thus ensuring intrinsic synchronization with the atmospheric laser. The wavelength-tunable Stokes is derived as the fourth harmonic (FH) of the uncompressed OPCPA signal, with center wavelength of 1460 nm. The spectrum of the signal extends from 1420 nm to 1520 nm, which gives a tunability of the FH from 355 nm to 380 nm. This corresponds to a Raman shift, with respect to the 337-nm radiation, ranging from 1500 cm^{-1} to 3350 cm^{-1}. The second harmonic (SH) of the OPCPA signal pulse is first spectrally narrowed by the optical filter F (Fig. 1), consisting of a 1700 lines/cm transmission grating and a 3-mm wide slit at a distance of 1.5 m, resulting in pulses with 1.5 nm FWHM bandwidth and tunability from 710 to 760 nm. The SH pulses then undergo amplification up to 0.9 mJ in a two-stage OPA employing a 4-mm-long type I BBO and a 10-mm-long type I LBO crystal, which are both pumped by the 532-nm SH of the Nd:YAG laser. FH generation in a 15-mm-long BBO crystal finally leads to tunable UV Stokes pulses, with energy up to 0.2 mJ. Due to the spectral filtering of the SH seed, the bandwidth of the UV Stokes pulses is<0.5 nm FWHM (0.7 nm spectral width measured with a spectrometer having spectral resolution of 0.4 nm, see Fig. 2(a)). Cross-correlation with 200-fs pulses from the Yb:CaF_2 system reveals 7.4 ps Stokes pulse duration (Fig. 2(b)).

One challenge of the setup is the spatial separation of the forward propagating IR beam (the 3.9-µm filament igniter) from the backward propagating UV light (the filament laser). Since it was difficult to identify suitable beam splitters, we employed, as shown in Fig. 1, a pair of CaF_2 Brewster prisms, separated by a distance of 7 cm, as a dichroic beam separator. Because of the small distance between the prisms, temporal and spatial chirp introduced for the 3.9-µm pulses are negligible and do not affect filamentation and backward lasing. On the other hand, because of the different refractive index for 3.9 µm (n = 1.409) and 337 nm (n = 1.448) light, backward propagating UV pulses bypass the apex of the second prism. Furthermore, because of the narrow bandwidth of the UV light, angular dispersion due to propagation through a single prism is negligible.

Both backward propagating pump and forward propagating Stokes beams are focused in the gas sample cell with 10-cm focal length lenses. They were measured to have waist radii of w_{Stokes} = 35 µm (w_{pump} = 50 µm) and Rayleigh ranges of z_{Stokes} = 2.9 mm (z_{pump} = 5.5 mm). The waists of the pump and Stokes beams are different due to different beam diameters on the focusing lenses and to small differences in their divergences. Surprisingly, the M^2 of the ASE filament-assisted atmospheric laser, here measured for the first time to our knowledge (see Fig. 2(c)), is very similar to that of the frequency-quadrupled OPA and corresponds to a nearly diffraction-limited beam.

In the SRS experiment the Stokes beam was modulated at 10 Hz by a synchronized mechanical chopper. Due to the low repetition rate of the system and the energy fluctuations of the atmospheric laser, it was necessary to adopt a balanced scheme for the detection of the weak SRL signal [18]. A fraction of the atmospheric laser beam was sampled with a beam splitter before the gas cell (reference), while the beam transmitted through the cell was picked up by another dichroic mirror transparent to the Stokes radiation. Reference and pump pulses were sent into two equal large-area photodiodes with enhanced UV sensitivity, while an additional photodiode was used to monitor the Stokes pulse energy and correct for its fluctuations. Following the approach by Schriever *et al.* [18], pump and reference energies were precisely measured by a high speed digitizer.

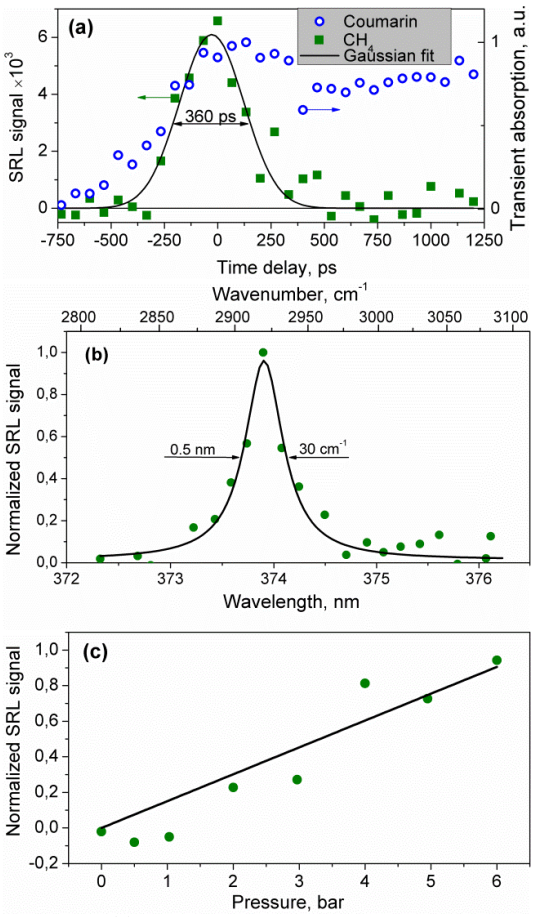

Fig.3. Fig. 3: (a) SRL in CH_4 at a pressure of 6 bar (solid rectangles) as a function of the pump-Stokes time delay. Data are accumulated over 1000 shots. The black solid line is a Gaussian fit to the experimental data. Open dots show the normalized transient absorption of Coumarin 500 probed at 337 nm when pumped by the 374-nm Stokes pulses. (b) SRL spectrum in CH_4 at a pressure of 6 bar. Dots represent experimental points obtained by averaging over 2000 shots, while the black solid line is a fit with a Lorentzian function. (c) SRL in CH_4 as a function of pressure.

In order to detect the backward SRS signal, it is necessary to provide spatial and temporal overlap and proper frequency detuning of the interacting pulses. To find the temporal overlap, we performed pump-probe on an ethyl-alcohol solution of Coumarin 500 dye in a 1-mm-thick quartz cell. The dye was chosen because it has strong ground-state absorption at 370 nm, excited state absorption at 337 nm and excited state lifetime of several

nanoseconds. Open circles in Fig. 3(a) show the transient absorption signal in Coumarin 500 as a function of the pump-probe delay. The pump-probe measurements allowed us to determine the exact temporal overlap between the pump and Stokes pulses as well as to optimize their spatial overlap.

Due to its large Raman cross section, we chose CH_4 as model target gas, with its CH stretching mode (ν_1) at 2917 cm^{-1}. This corresponds to a 374-nm Stokes wavelength. Figure 3(a) shows as solid squares the SRL signal in a gas cell filled with 6 bar of CH_4, as a function of the pump-Stokes delay. The maximum signal was measured to be 6×10^{-3}, which is a factor of 3 higher than in the experiments reported in [16]. By changing the pump-Stokes delay we precisely determined the duration of the N_2 laser pulse as 360 ps, which is consistent with a filament length, determined visually, between 10 and 20 cm.

Figure 3(b) shows the spectral response of the SRL signal as a function of the pump-Stokes frequency detuning. It exhibits the typical resonant-like behavior with a FWHM of 30 cm^{-1}, which is consistent with the convolution of the Stokes bandwidth with the spectral width of the CH-stretching vibrational manifold of methane. The sensitivity of the setup was then tested recording the SRL signal as a function of CH_4 pressure (Fig. 3(c)). On the one hand, the measurements revealed the linear concentration dependence that is characteristic of the SRS signal; on the other hand they clearly show a strong limitation to the sensitivity, with a lowest detectable CH_4 pressure of 2 bar only, corresponding to a SRL signal of 2×10^{-3}. Scaling down this limit clearly implies an upgrade of the laser system, in terms of increasing the Stokes pulse energy, for higher signals, and the repetition rate, for more efficient averaging.

In summary, we report what we believe to be the first observation of a coherent standoff signal from a gas medium, obtained via interaction of the medium with two frequency-detuned, counter-propagating pulses in a SRS regime. The backward propagating pulse is remotely generated through nitrogen lasing within a plasma filament induced by a femtosecond high-energy mid-IR pulse. This result represents a conceptual milestone, as it proves the feasibility to return to the observer a coherent Raman signal without any use of reflection or backward-directed excitation beams. However, the sensitivity of the technique in its present realization is still low. For such a strong Raman scatterer as methane the lowest detectable pressure was 2 bar. Down-scaling this limit requires an additional intense effort towards a substantial increase of the energy of the excitation pulse (Stokes in our configuration) and to a higher-repetition-rate laser source, leading to higher detection sensitivity. Both upgrades are technologically within reach. A second critical aspect is an upgrade of the system to make it compatible with the natural atmospheric environment, implying elimination of Ar as means of enhancing N_2 population inversion and taking into account additional noise sources, such as air turbulence and reduced signal collection efficiency. Such an upgrade requires, first, an increase in the peak power of the OPCPA system to provide pulses well above the critical power of self-focusing and, second, to prolong plasma lifetime in the filament afterglow [19, 20] which would enhance backward lasing. In the near future, we plan to develop a dedicated OPA for narrowband seed amplification which would enable to keep the amplified pulse duration close to that of the driving laser (70 ps).

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