EPJ Web of Conferences **41**, 12009 (2013)
DOI: 10.1051/epjconf/20134112009

Ultrafast-Laser-Induced Backward Stimulated Raman Scattering for Tracing Atmospheric Gases

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Abstract. By combining tunable broadband pulse generation with nonlinear spectral compression, we demonstrate a prototype scheme for highly selective coherent standoff sensing of air molecules and discuss its coupling to the recently demonstrated backward atmospheric lasing.

Many applications in environmental science and security call for standoff chemical identification of airborne pollutants by their signature molecular vibrations. Raman LIDARs, relying on the observation of incoherent wide-angle scattering, have limitations in their practicability. The success of heterodyne detection as a cornerstone concept of the existing coherent LIDAR approaches motivate the development of optical remote sensing schemes where the coherence would be used to provide a highly directional backward optical signal through stimulated Raman scattering, thus enhancing the sensitivity and selectivity of standoff detection. Coherently-enhanced Raman scattering processes, such as Coherent Anti-Stokes Raman Scattering (CARS) and Stimulated Raman Scattering (SRS), open the possibility of recording signals carried in a laser-like beam. Generation of a backward phase-matched Raman signal is simple in the presence of reflective or diffuse objects capable of bouncing the laser beam back. However, an observer who sends out a laser pulse to interrogate an optically transparent atmosphere has to find an alternative way of initiating a backward-propagating laser beam and to solve the problem of phase matching. The recent experimental demonstration of a remotely pumped atmospheric backward laser using O_2 (λ=845 nm) [1] or N_2 (λ=337 and 357 nm) [2] has sparked proposals for coherent standoff Raman spectroscopies [3].

In this work we experimentally and theoretically investigate a prototype for SRS in air which will be ultimately based on the 337-nm atmospheric N_2 laser (Fig.1a). The interim model (Fig.1b) is based on a solid-state ultrafast laser system from which we derive a narrowband third harmonic Stokes pulse at ~340 nm, imitating the N_2 laser emission, and a synchronized tunable narrowband

pump pulse. As model systems we study responses from atmospheric nitrogen and oxygen as well as methane in a variable pressure cell. The chosen SRS approach [4] provides several advantages over CARS such as: inherent phase matching enabling a backward propagating geometry; absence of non-resonant background due to electronic contributions; and linear scaling of the signal with the number of probed molecular oscillators, which makes it feasible to detect a low-concentration specimen.

The conceptual scheme presented in Fig.1(a) requires the combination of a remotely pumped atmospheric laser (Stokes beam) with an additional high-energy tunable laser beam sent in the forward direction (pump beam). When the frequency difference between pump and Stokes beams matches a vibrational transition of the molecule to be detected, then the backward propagating Stokes experiences stimulated Raman gain (SRG), enabling sensitive molecular fingerprinting. The experimental challenge resides in the difficulty to detect the weak SRG sitting on top of the intense and possibly noisy Stokes pulse.

The SRS standoff model system in Fig.1(b) is based on a 400-μJ, 200-fs, 10-kHz 1030-nm diode-pumped Ytterbium CPA system (Pharos, Light Conversion Ltd.) The CPA laser drives a femtosecond optical parametric amplifier (OPA) producing tunable μJ pulses in the 600—700 nm range and generates the third harmonic from the laser output at ~340 nm via frequency doubling followed by sum-frequency generation. In both pump and Stokes arms of our SRS setup we employ the technique of second harmonic (SH) spectral compression [5] whereby the group delay mismatch of the interacting pulses is selected to shrink the fundamental frequency spectrum into a narrowband SH line (see Fig. 1(c)). This process maintains high efficiency because pairs of spectral components equally detuned from the designated centre SH frequency are phase-matched for sum-frequency generation.

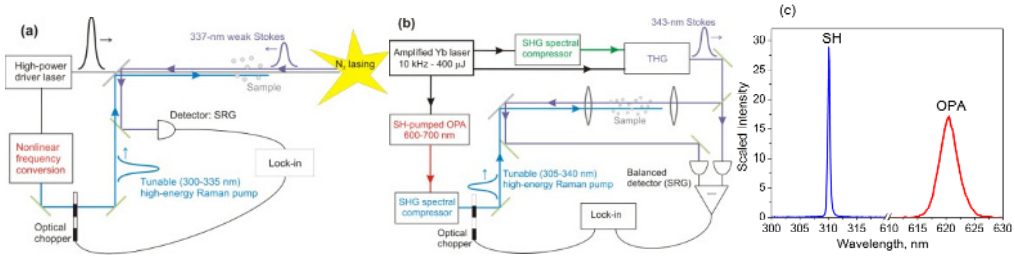

Fig. 1. (a) conceptual scheme of remote atmospheric sensing using backward SRS from an atmospheric laser; (b) setup of the proof-of-principle experiment; (c) spectra of the OPA output and generated SH; in order to highlight spectral brightness of the UV pulses the area under the curves are scaled to the energy of the pulses (7 μJ for the OPA output, and 1.6 μJ for SH).

The experimental results of backward SRS obtained for methane in a variable pressure cell and in air under normal conditions are summarized in Fig. 2(a)-(d). The narrowband third harmonic and frequency-doubled OPA spectra, in the case of methane (Fig. 2(a)-(b)) are separated by $\nu_R = 2920\ cm^{-1}$ detuning resonant to the C-H symmetric stretching in CH_4 [6]. In the case of measurements in air shown in Fig. 2(c)-(d) respective contributions of N_2 ($\nu_R = 2330\ cm^{-1}$) and O_2 ($\nu_R = 1555 cm^{-1}$) were recorded.

Although picosecond pump and Stokes pulses possess substantially broader spectra (corresponding convolution of pump and Stokes spectra is in the order of 37 cm^{-1} (Fig. 2(a)) as compared to the bandwidth of Raman response (<2 cm^{-1} in the case of CH_4 [6]), due to the fact that the spectral width of both pulses is comparable, our setup has rather high sensitivity. As it can be seen from Fig. 2(b), CH_4 at concentrations as low as 0.04 bar is detectable. In the case of measurements in air both major contributing parts, N_2 and O_2, were detected in counter-propagating geometry in a single scan with an integration time of 1 s (Fig. 2(c)). Since both pump and Stokes pulses are relatively short (with cross-correlation being in the order of 1 ps which, in the case of counter-propagating geometry, corresponds to 0.15 mm overlap in space), this measurement also allows evaluation of the size of interaction region at certain focusing conditions. Finally, the measurements of the intensity dependence of the SRG response in N_2 (Fig. (2d)) at given focusing

conditions gives a possibility of estimating the required pump pulse energy in the case of experiments with backward probe from N_2 lasing.

As a conclusion, these results are extremely promising in view of real-world stand-off sensing based on backward SRS from an atmospheric laser. A much stronger SRG can be expected in such case, since the 1-2 ns pulse duration of the atmospheric emission would result in an increased interaction length of 15÷30 cm. Moreover, the energy of the tunable Raman pump can be boosted by several orders of magnitude using an optical parametric chirped pulse amplifier, allowing a corresponding linear scaling of the SRG signal.

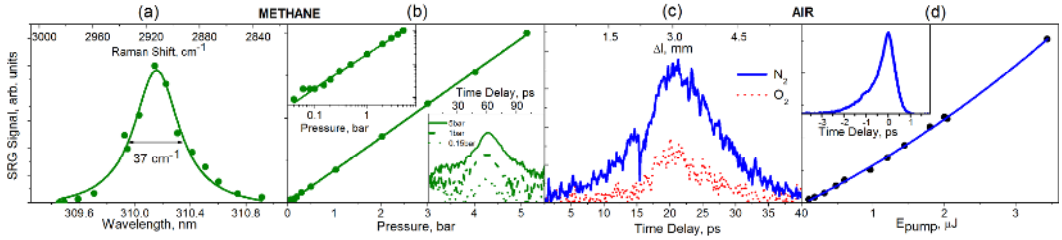

Fig. 2: (a) spectrum of the backward SRG signal of CH_4 (dots) measured at pressure of 5 bar; solid line is a guide for the eye; the actual FWHM is determined by the convolution of pump and Stokes spectra; (b) backward SRG signal as function of pressure measured in CH_4, displaying the expected linear concentration dependence; in order to highlight the dependence at low pressures in the top inset the signal is plotted in a log-log scale; in the bottom inset the SRG signal for three different pressures (indicated in the panel) is plotted as a function of the delay between the pump and Stokes pulses; (c) backward SRG signal as a function of delay between the pump and Stokes pulses measured in N_2 and O_2 in air under normal conditions in counter-propagating geometry; since the time overlap between the pump and Stokes pulses is less than 1 ps (half of that measured in co-propagating geometry (inset in the Fig.2d)), the signal is an indicative of convolution of confocal parameters of the pump and probe beams (Δl top axis); (d) dependence of the forward SRG signal on pump energy in atmospheric N_2 in co-propagating geometry; the solid line represents exponential fit to the experimental data; in the inset the forward SRG signal as a function of delay between the pump and Stokes pulses is presented; the signal is measured in N_2 in air under normal conditions in co-propagating geometry and represents cross-correlation between the pump and Stokes pulses.

References

[1] A. Dogariu, J.B. Michael, M.O. Scully, R.B. Miles, "High-gain backward lasing in air", Science **331**, 442–445 (2011).

[2] D. Kartashov, S. Ališauskas, A. Pugzlys, A. Baltuška, M. Shneider, and A. Zheltikov, "Free-space nitrogen laser from a mid-infrared filament," in *Research in Optical Sciences*, OSA Technical Digest (Optical Society of America, 2012), paper HW3C.2.

[3] P.R. Hemmer, R.B. Miles, P. Polynkin, T. Siebert, A.V. Sokolov, P. Sprangle, M.O. Scully, "Standoff spectroscopy via remote generation of a backward-propagating laser beam", Proc. Natl. Acad. Sci. USA **108**, 3130-3134 (2011).

[4] M. Maier, W. Kaiser, J.A. Giordmaine, "Backward stimulated Raman scattering", Phys. Rev. 177, 580-599 (1969).

[5] M. A. Marangoni, D. Brida, M. Quintavalle, G. Cirmi, F. M. Pigozzo, C. Manzoni, F. Baronio, A. D. Capobianco, G. Cerullo, "Narrow-bandwidth picosecond pulses by spectral compression of femtosecond pulses in second-order nonlinear crystals," Opt. Express **15**, 8884-8891 (2007).

[6] S. Brunsgaard Hansen, R. W. Berg, and E. H. Stenby, "Raman spectroscopic studies of methane–ethane mixtures as a function of pressure,"Appl. Spectrosc. **55**, 745-749 (2001).