

## 1.6 MICRON FIBER LASER SOURCE FOR CH<sub>4</sub> GAS LEAK DETECTION

Nicolas Cézard\*, Philippe Benoit, Guillaume Canat<sup>1</sup>

<sup>1</sup>ONERA – The French Aerospace Lab – F-91761 Palaiseau, France,

\*Email: nicolas.cezard@onera.fr

### ABSTRACT

We report on the development of a new pulsed fiber laser source at 1645.5 nm, based on stimulated Raman amplification. This laser source is intended to be used in a future lidar system, dedicated to methane gas leak monitoring in the vicinity of industrial facilities. In this paper we discuss reasons for choosing the 1645.5 nm wavelength, and then we present the two-stage amplification architecture of our fiber laser source under development. Recent experimental results are provided and perspectives are drawn.

### 1. INTRODUCTION

Methane is an important atmospheric greenhouse gas, as well as a major energy source. Methane gas leaks in industrial plants can cause human injuries, environmental concerns, and financial losses. It is then of primary interest to develop remote sensing methods to characterize methane gas leaks in the vicinity of industrial facilities, typically at a few hundred meters, up to 1 km range.

Integrated-Path Differential Absorption Lidars (IP-DIAL) for CH<sub>4</sub> detection at various ranges have been demonstrated in the past in the 1.6 μm band, using semiconductor lasers [1] or solid-state Optical Parametric Oscillators (OPO) [2-4]. An OPO-based spaceborne IPDA lidar is currently under development [5]. Range-Resolved systems (RR-DIAL) have also been developed using OPOs in the 1.6 μm band or 3.3 μm band [6-7]. These systems are efficient but complex and cumbersome (trucks).

In this work, we consider another approach and wish to investigate CH<sub>4</sub> IP-DIAL and RR-DIAL solutions based on silica-fiber-based components for both lidar source and lidar receiver. The main advantages of silica-fiber-based

technologies are (i) high potential for lidar compactness, robustness and portability relying on telecom-type components (ii) design versatility for shaping spectral and temporal pulse properties and (iii) possibility of using fiber coherent detection for simultaneous or sequential range-resolved wind and CH<sub>4</sub> measurements with a single lidar. This last property is particularly attractive in the context of gas leak monitoring, since evaluating the leak rate implies to measure not only the gas concentration but also the gas speed [8].

To our knowledge, so far fiber laser based detection of CH<sub>4</sub> has been reported only in short-range (≈10m) IP-DIAL arrangement [9]. The fiber laser used Raman stimulated amplification and delivered 1.2 W cw radiation at 1651 nm. To address range-resolved lidar measurement, our objective is to address pulsed operation (typ. 100 ns) and reach high peak powers.

### 2. WAVELENGTH SELECTION FOR CH<sub>4</sub> DETECTION WITH A FIBER LIDAR

Though it is well-known that silica fibers do not transmit light beyond 2.4 μm, it is interesting to first consider the wavelength selection problem on a more general basis.

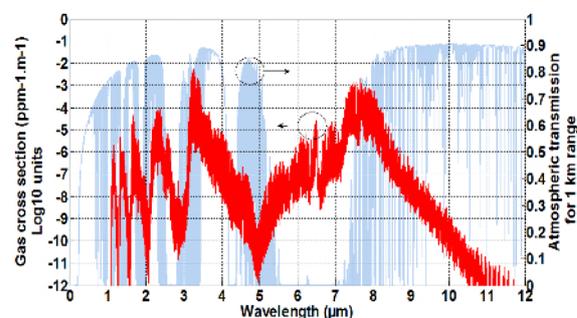
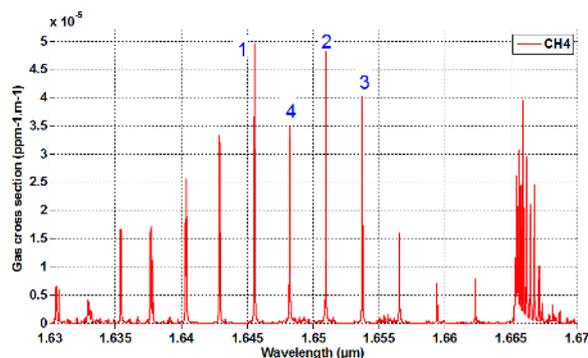


Figure 1: Absorption cross section of CH<sub>4</sub> across the optical 1-12 μm spectrum (red), and atmospheric transparency for 1 km propagation (blue). Both are calculated from Hitran 2012 database.

The four spectral bands that feature highest CH<sub>4</sub> absorption cross-sections in the 1-12 μm wavelength range are (from highest to lowest cross-section) 3.3 μm, 8 μm, 2.4 μm, and 1.6 μm (see fig.1). For 1 km lidar range, choosing a high-transparency atmospheric window is necessary. At 3.3 μm, CH<sub>4</sub> cross-section is so high (close to 5.10<sup>-3</sup> ppm<sup>-1</sup>.m<sup>-1</sup>) that the natural background methane concentration (~1.8 ppm) would completely extinct the laser light after a few hundred meters propagation. The 8 μm window can be discarded because it is extremely challenging for current high power laser systems. Consequently, near-infrared bands at 2.4 μm and 1.6 μm, with moderate absorption cross-sections close to 5.10<sup>-5</sup> ppm<sup>-1</sup>.m<sup>-1</sup>, are the best suited bands for CH<sub>4</sub> leaks monitoring at 1 km range. Choosing silica-based fiber technology further excludes the 2.3-2.4 μm band, which stands at the very edge of the silica transparency. Therefore, the 1.6 μm band chosen for this work is optimal both for technical reasons (fiber transparency) and with respect to the present application (kilometer range lidar).

A close-up of fig.1 around the 1.63-1.67 μm band shows four high cross-section transitions at 1645.54 nm, 1650.96 nm, 1653.72 nm, and 1648.23 nm (figure 2). All these transitions are actually made of multiple fundamental lines that merge into single ones under the atmospheric pressure broadening effect. Two important criteria must be evaluated in order to choose the best transition to work with: the water-vapor interference level and the cross-section temperature sensitivity.



**Figure 2: Absorption cross section of CH<sub>4</sub>: close-up on the 1.63-1.67 μm range. Four candidate transitions with high cross-sections are numbered.**

A calculation of the water vapor optical depth for 1 km range in a worse-case extremely humid atmosphere (30 000 ppm of water vapor) reveals that (i) transitions 2, 3 and 4 are well isolated (more than two linewidths apart) from the first significant H<sub>2</sub>O neighboring line and consequently shall not be disturbed (ii) transition 1 is separated by about one linewidth from the first significant H<sub>2</sub>O line. It is therefore a little more sensitive to H<sub>2</sub>O interference, but only on the short-wavelength side of the transition, and with a small expected impact since the optical depth of this H<sub>2</sub>O line reaches only 0.1 after 1 km in this very high humidity scenario.

The Relative Systematic Error (RSE) expected to occur on gas concentration retrieval because of temperature misreading can be evaluated by the following formula, derived for lorentzian lines with negligible OFF-line cross-section:

$$RSE = \frac{d\sigma}{\sigma} = G_1(v_{ON}) \frac{dT}{T} \quad \text{with}$$

$$G_1(v_{ON}, T) = \left[ (1 + x_{TIPS}) - \frac{100hc\Delta E}{kT} - n_{air} \frac{1 - [(v_{ON} - v_c) / \gamma_L]^2}{1 + [(v_{ON} - v_c) / \gamma_L]^2} \right]$$

Where  $\sigma$  is the gas cross section (ppm<sup>-1</sup>.m<sup>-1</sup>), T the temperature,  $x_{TIPS}$  the power exponent of the molecule Total Internal Partition Sum function ( $x_{TIPS} = 1.54$  for CH<sub>4</sub> at 296 K, as computed from a power function fit of the TIPS data provided by HITRAN 2012 database),  $\Delta E$  the fundamental energy level of the absorption transition in cm<sup>-1</sup>,  $n_{air}$  the power exponent of the transition intensity temperature dependence,  $v_c$  the central wavenumber in cm<sup>-1</sup> and  $\gamma_L$  the line HWHM in cm<sup>-1</sup>. However, here the lineshapes are not strictly lorentzians, because each line is made of merged multiplets. Therefore, temperature sensitivities have also been assessed using a line-by-line atmospheric propagation code at temperature T=296K and T+1K, and by calculating:

$$G_2 = T \cdot [\sigma(T+1) - \sigma(T)] / \sigma(T).$$

A significant discrepancy between G<sub>1</sub> and G<sub>2</sub> is found only for line 1, as shown in Table 1. For line 1, a 3K error (dT/T=1% at 296 K) turns into approximately 1.2% RSE for CH<sub>4</sub> concentration retrieval when the gas is probed on line center. Lines 2,3 and 4 are a little more sensitive than

line 1. For all lines, sensitivities are currently considered acceptable values for our purpose.

In conclusion, because it exhibits both the highest cross-section and the lowest temperature sensitivity, we have chosen to begin with the 1645.54 nm transition. Water-vapor interference is expected to remain very small, especially if the Off-line wavelength is chosen along the long-wavelength side of the line center.

Line N°	$\Delta E$ (cm <sup>-1</sup> )	G <sub>1</sub> (v <sub>c</sub> ,296K)	G <sub>2</sub> (v <sub>c</sub> ,296K)
1	220	0.8	1.2
2	105	1.3	1.3
3	63	1.5	1.4
4	157	1.1	1.2

**Table 1:** Spectroscopic parameters and temperature sensitivity coefficients for the four selected lines. Coefficients  $n_{\text{air}}=0.7$  and  $x_{\text{TIPS}}=1.54$  are the same for all lines. G<sub>1</sub> (lorentzian model) and G<sub>2</sub> (line-by-line calculation) are given here for  $v_{\text{ON}}=v_c$

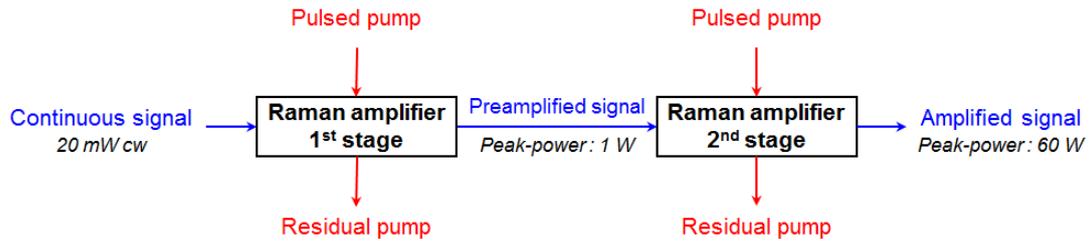
### 3. FIBER LASER SOURCE AT 1645.5 nm

Radiation at 1645 nm cannot be efficiently amplified in conventional Erbium-doped silica fibers, because the Erbium gain band stretches from 1530 nm up to 1620 nm only. Instead, our fiber laser design relies on Raman stimulated amplification. When a powerful laser pump beam propagates into a silica fiber, it interacts non-linearly with the silica matrix and experiences a 13 THz down-frequency (spontaneous) Raman shift. If the pump wavelength is 1535 nm, the Raman shift leads to the desired 1645 nm wavelength. Consequently, if a low-power seed at 1645 nm (signal wavelength) and a high-power beam at 1535 nm (pump wavelength) propagate into the fiber at the same time, stimulated Raman amplification can occur and transfers energy from the 1535 nm pump to the 1645 nm signal. In practice, the Raman gain is spectrally quite large and pump wavelength within 10-15 THz from the signal wavelength are acceptable (13 THz being the optimum).

Raman amplifiers are known for their simplicity and efficiency. The main drawback is the Raman gain weakness (only  $8 \cdot 10^{-14}$  m/W), which implies using a long fiber length and/or a high power pump to provide high signal amplification. Because long fiber lengths are required, the pump wavelength is quickly prone to experience Stimulated Brillouin Scattering (SBS), which limits the usable pump power. SBS mitigation strategies are therefore required to increase the pump power. Moreover, in order to generate high signal power at 1645 nm for lidar application, SBS mitigation techniques must also be used for the signal wavelength.

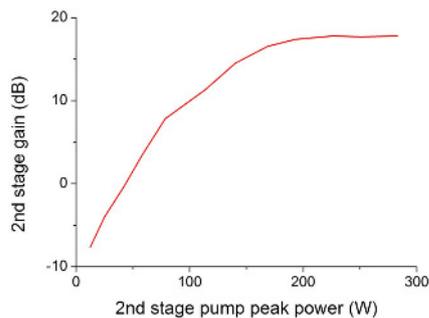
The fiber laser architecture is shown on figure 3. It is a two-stage MOFPA (Master Oscillator Fiber Power amplifier). The seeding master oscillator is provided by a DFB (Distributed FeedBack) laser diode tuned at 1645.54 nm (typically 20 mW of cw power). In the first amplifier stage, the 1645 nm cw seed and a pulsed high peak-power pump at 1563 nm are injected in co-propagation into a highly non-linear fiber (HNLF) through a fiber multiplexer. The pump is spectrally broadened to mitigate SBS at 1563 nm and to enable increased pump power. At the exit, the signal power at 1645 nm typically reaches 1W peak-power (+17 dB gain) with 80 ns pulses at 20 kHz. This could be improved in the future by optimizing the pump wavelength (1563 nm is far from the optimal pump wavelength).

In the second Raman amplifier stage, a high peak-power 1545 nm broad linewidth pump laser is coupled in a second HNLF in contra-propagation with the 1645 nm signal. Using a broadband pump laser allows here again mitigating SBS detrimental effects, and increasing the pump power (through two successive Erbium-Doped fiber amplifiers). A drawback is that this configuration also transfers and amplifies the pump intensity noise toward signal intensity noise. However, in lidar systems, signal intensity noise can usually be compensated for by pulse-to-pulse power reference measurements if needed.



**Figure 3 :** Architecture of the fiber laser source with two amplification stages.

We have recently obtained 60 W peak power at 1645 nm at the 2<sup>nd</sup> stage exit (+18 dB) with 30 ns pulses (signal pulses experience time compression during the amplification process) at 20 kHz (see figure 4). These are encouraging results, though they call for many further investigations. So far the gain is believed to be mainly limited by an observed 2<sup>nd</sup>-stage pump power depletion induced by spontaneous Raman emission at superior orders. This effect must then be properly investigated. Pulses duration also need to be reshaped in the first and second stage in order to obtain the desired 100-ns duration at 2<sup>nd</sup> stage exit. Finally, the 1645 nm signal spectral width will have to be measured carefully, since frequency chirp effects are expected to occur (due to the optical Kerr effect that takes place in the HNLF along with Raman scattering).



**Figure 4 :** Gain of the second Raman amplifier vs. pump peak power. The gain is ‘negative’ at low pump power because the amplifier has its own internal losses.

#### 4. CONCLUSIONS

A 60 W peak-power fiber laser source relying on Raman stimulated amplification at 1645.5 nm has been reported for the first time to our knowledge. This wavelength has been identified

as a good choice (high - but not too high - cross-section, low temperature sensitivity, low water-vapor interference level) for future lidar remote sensing of CH<sub>4</sub> gas leaks up to 1 km range. The obtained power may already allow performing IP-DIAL or short-range RR-DIAL measurements of methane. Another perspective is to keep improving the laser power and optimize the spectral and temporal parameters to allow both CH<sub>4</sub> and wind measurements to be made with a single lidar system.

#### ACKNOWLEDGEMENT

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