Absolute frequency measurements of CH_3I and CH_4rovibrational transitions by a frequency-comb-referenced optical parametric oscillator

I. Ricciardi[a], S. Mosca[a],V. Di Sarno[a], P. Maddaloni[a], M. Parisi[a], L. Santamaria[a],
M. De Rosa[a],J.-J. Zondy[b], and P. De Natale[c]

[a] CNR, Istituto Nazionale di Ottica,Napoli (Italy),E-mail: maurizio.derosa@ino.it
[b] LNE-CNAM, Paris (France)
[c]CNR, Istituto Nazionale di Ottica, Firenze (Italy)

Optical parametric oscillators (OPOs) combine high power, single mode emission, tunability, and wide spectral coverage from visible to mid infrared (MIR). All these characteristics make cw OPOs attractive for atomic and molecular spectroscopy. In the last years, OPOs proved to be suitable for high-resolution and accurate spectroscopic applications [1,2], demonstrating sub-Doppler resolution in the mid-infrared. OPOs emitting in the MIR have been combined with visible and NIR optical frequency comb (OFC) synthesizers, providing high-precision and absolute frequency determination outsidethe specific OFC covered range.

We present two different configurations for the measurement of absolute frequencies of MIRrovibrational transitions with uncertainty in the 1-50 kHz range. Both schemes are based on a singly-resonant OPO, whose idler mode in the MIR (2.7-4.2µm tunability range) probes sub-Doppler transition profiles in a saturation spectroscopy configuration.The absolute frequency of the OPO idler radiation is determined by comparison of both the OPO pump and signal frequencies to a self-referenced OFC synthesizer based on an amplified mode-locked Er:doped fibre laser, emitting an octave spanning the 1-2 µm wavelength interval. Repetition rate and offset frequency are stabilized against a 10 MHz BVA quartz, locked to a Rb-clock, referenced to the Cs primary standard via global positioning system (GPS).

In a first scheme, the pump laser can be scanned over 70 MHz across the absorption feature, while maintaining the laser locked to the OFC, enabling acquisition of the sub-Doppler absorption profile. In this way, we are able to record the whole absorption lineshapesof the six hyperfine components of the CH_3I transitions, assigning an absolute frequency scale. Then, a fitting of the recorded profilesprovidesthe relative frequencies of the hyperfine components.

In a second scheme, while the signal is frequency offset locked to the nearest tooth of the OFC, the idler frequency is locked to the transition center by actively controlling the pump frequency. Then, the comb repetition rateand the beat frequencies between pump (signal) and its nearest comb tooth are counted for about 200 s, resulting in a final typicalstatistical uncertainty of ~1 kHz (~10^{-11} of relative uncertainty). For this case, we selectedν_3 band transitions ofmethane, as they have been recently measured with comparable uncertainty [3,4].Systematic errors still limit the reproducibility of the measurements to ~10 kHz and are currently under investigation.

[1] I. Ricciardi et al.,*Optics Express***2012***20*, 9178.
[2] I. Ricciardi et al., *Molecular Physics*, **2012***110*, 2103.
[3] S. Okubo et al., *Optics Express*, **2011***19*, 23878.
[4] M. Abe et la., *J. Opt. Soc. Am. B***2013***30*, 1027.