

MID-INFRARED SATURATION SPECTROSCOPY FOR VIBRATIONAL FREQUENCY MEASUREMENTS AT THE SUB-KHZ LEVEL

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Molecular systems, owing to their numerous degrees of freedom, offer promising perspectives for improving tests of fundamental physics and precision measurements in general^{1,2,3,4,5}. In our group at Laboratoire de Physique des Laser, using an optical frequency comb, a 10 μm quantum cascade laser (QCL) is stabilized at the sub-Hz level to an ultra-stable near infrared reference signal operated at the French metrology institute. This signal is calibrated there to some of the best atomic clocks and transferred through a noise-compensated 26-km long fiber cable of the REFIMEVE infrastructure⁶. This results in a record relative frequency uncertainty of 10^{-14} and a 0.1 Hz QCL linewidth⁷.

We have used this ultra-stable and frequency controlled mid-infrared source to perform high resolution spectroscopy on various molecular species of fundamental, atmospheric and astrophysics interest such as methanol, ammonia or trioxane, with a few kilohertz uncertainties on line positions using sub-Doppler saturated absorption spectroscopy in a multipass-cell⁸.

Here, I will discuss our latest efforts in order to improve these spectroscopic measurements, by realizing cavity enhanced measurements. I will show results on methanol achieving a few hundreds of Hz uncertainties on molecular frequencies, I will then present investigations on various sources of noise currently limiting our signal-to-noise ratio and perspectives for improvements. These results enhance our ability to meet the needs for our ongoing efforts towards studying the variation of

¹ doi:10.1070/QEL16880, A. Cournol *et al.*, *Quantum Electronics* **49**, 288 (2019)

² doi:10.1021/acs.jpcllett.2c02434, M.R. Fiechter *et al.*, *J. Phys. Chem. Lett.* **13**, 42, (2022)

³ doi:10.1140/epjqt/s40507-022-00130-5, G. Barontini *et al.*, *Quantum Technol.* **9**, **12**, (2022)

⁴ doi:10.1103/PhysRevLett.127.043201, J. Lukusa Mudiayi *et al.*, *Phys. Rev. Lett.* **127**, 043201, (2021)

⁵ doi:10.1088/1681-7575/aaa790, J. Fischer *et al.*, *Metrologia*. **55**, R1 (2018)

⁶ www.refimeve.fr, REseau Fibré MÉtrologique à Vocation Européenne

⁷ doi:10.1038/nphoton.2015.93, B. Argence *et al.*, *Nature Photon.* **9**, 456-460 (2015)

⁸ doi:10.1364/OPTICA.6.000411, R. Santagata *et al.*, *Optica* **6**, 411 (2019)

fundamental constants³ or testing fundamental symmetries, such as the measurement of the tiny parity-violating energy difference between enantiomers of a chiral molecule^{1,2}, via precise molecular spectroscopy. Furthermore, it can contribute to the enrichment and refinement of the HITRAN database⁹, which can be highly beneficial for the fields of astronomy¹⁰ and atmospheric physics¹¹, providing valuable data for accurate modeling and analysis.

⁹[doi:10.1016/j.jqsrt.2021.107949](https://doi.org/10.1016/j.jqsrt.2021.107949), I. Gordon *et al.*, *J. Quant. Spectrosc. Radiat. Transfer* **277**, 107949 (2022)

¹⁰[doi:10.1038/326049a0](https://doi.org/10.1038/326049a0), W. Batrla *et al.*, *Nature* **326**, 49-51 (1987)

¹¹[doi:10.1029/98JD02747](https://doi.org/10.1029/98JD02747), D. Simpson *et al.*, *J. Geophys. Res.* **104** (D7), 8113-8152 (1999)