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High-precision ozone spectroscopy with a tunable SI-traceable frequency-comb-based mid-infrared QCL

Ozone (O3) is an important molecule playing a pivotal role in Earth's atmosphere and climate chemistry through atmospheric oxidation. Accurate measurements of O3 molecular line frequencies and shapes over a wide spectral range through laser spectroscopy open the way to improve the determination of atmospheric ozone mixing ratios or abundance profiles in the atmosphere.
In 2018, we have reported the first frequency evaluation of four intense rovibrational transitions in the ν3 fundamental band of ozone between 1049 and 1052 cm-1 based on a free-running quantum cascade laser (QCL) [1]. The effect of foreign gas pressures has also been measured and these data have since been used as a calibration for ozone pressure shifts by air in the HITRAN 2020 database [2].
Recently, through collaboration with LPL at University Sorbonne Paris Nord and financial support from the LabEx FIRST-TF and the CNRS interdisciplinary program MITI 80, we have successfully developed a high-resolution mid-infrared (MIR) molecular spectrometer of atmospheric species, traceable to the primary frequency standards at LNE-SYRTE (Observatoire de Paris). It combines a widely tunable ultra-narrow QCL at 9.54 µm with a nonlinear OP-GaAs crystal for frequency up-conversion of the MIR laser wavelength to an optical frequency comb at 1550 nm using a comb at 1860 nm [3]. This way, the QCL emission is stabilized onto the 1550 nm frequency comb whose repetition rate is locked to the 100 MHz reference distributed by the REFIMEVE network. The connection to REFIMEVE is established by a local fiber between LERMA and LKB, to which we are connected via a local link at the campus of Sorbonne University. Part of the QCL beam passes through an 80 cm glass cell equipped with ZnSe windows for ozone absorption measurements.
The resulting QCL relative frequency stability has been evaluated to be presently below 10-10 at 1 s, but much higher stabilities can be achieved [3]. A continuous frequency tuning range over 200 MHz is also reported. We use the experimental apparatus to perform saturated absorption spectroscopy of ozone lines in the low-pressure regime around 2 mTorr and we aim to demonstrate a statistical uncertainty at the kHz-level on molecular transitions dedicated to atmospheric remote sensing applications and molecular databases [2].
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[2] I.E Gordon et al., "The Hitran 2020 Molecular Spectroscopic Database", J. Quant. Spectrosc. Radiat. Transfer 277, 107949 (2022).
[3] B. Argence et al., "Quantum cascade laser frequency stabilization at the sub-Hz level". Nature Phot. 9, 456, (2015)

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