LABORATOIRE DE PHYSIQUE DES LASERS

CNRS UMR7538 - UNIVERSITE PARIS 13 - INSTITUT GALILÉE

Thesis

Presented to obtain the degree of
"Docteur en Physique de l'Unversité PARIS 13" by

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Widely tunable and SI-traceable frequency-comb-stabilised mid-infrared quantum cascade laser: application to high precision spectroscopic measurements of polyatomic molecules

The thesis consists in developing a high-resolution mid-infrared spectrometer traceable to primary frequency standards and providing a unique combination of resolution, tunability, detection sensitivity and frequency control. A quantum cascade laser (QCL) emitting at 10.3 μm is phase locked to an optical frequency comb stabilized to a remote 1.55 μm ultra-stable reference developed at LNE-SYRTE, monitored against primary frequency standards and transferred to LPL via an active noise compensated fibre link. This results in a 0.1 Hz QCL linewidth, a stability below 10^{-15} at 1 s and an uncertainty on its absolute frequency below 4×10^{-14}. Moreover, the setup allows the QCL to be widely scanned over 1.4 GHz while maintaining the highest stabilities and precisions. This QCL was used to carry out saturated absorption spectroscopy of several molecules in a compact multipass cell. We demonstrated statistical uncertainty on line-center frequencies at the kHz level and sub-10 kHz systematic uncertainty. We have recorded several singular *K*-doublets and many rovibrational transitions of methanol, in particular weak transitions and weak doublets - unreported so far. Precise parameters modelling trioxane have been determined with only a few tens of rovibrational transitions recorded at unprecedented accuracy. The quadrupole hyperfine structure of an ammonia transition has been resolved for the first time. This setup constitutes a key element for the project aiming at the first observation of parity violation in molecules currently held at LPL, and, more generally, for various fields of physics, from atmospheric and interstellar physics to fundamental physics beyond the standard model.

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Key words: ultra-high resolution molecular spectroscopy, frequency metrology, mid-infrared, quantum cascade lasers, optical frequency comb, saturated absorption spectroscopy, ultra-stable lasers, laser stabilization, optical fibre links, frequency reference transfer, methanol, hyperfine structure

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