THz-Comb-Assisted Molecular Spectroscopy

L. Consolino[1], S. Bartalini[1], A. Taschin[2], P. Bartolini[2], M.S. Vitiello[1,3], H. Beere[4], D. Ritchie[4], A. Tredicucci[3], P. Cancio Pastor[1], R. Torre[2], P. De Natale[1]

1. *INO, Istituto Nazionale di Ottica – CNR, Largo E. Fermi 6, Firenze I-50125, Italy.*
2. *LENS, European Laboratory for NonLinear Spectroscopy, Via N. Carrara 1, Sesto Fiorentino (FI) I-50019, Italy.*
3. *NEST, Istituto Nanoscienze – CNR and Scuola Normale Superiore, Piazza San Silvestro 12, Pisa I-56127, Italy.*
4. *Cavendish Laboratory, University of Cambridge, J J Thomson Avenue, Cambridge CB3 0HE, UK.*

Thanks to the recent progresses in photonics and nanotechnology, Terahertz (THz) radiation is finding increasing use in a wide range of applications. Since their first demonstration, THz Quantum Cascade Lasers (QCLs) have been the most important sources for a large range of applications and also for fundamental research. An important characteristic of THz QCLs is the very low intrinsic linewidth of their emission [1], at a level of tens of Hz, and this further qualifies them as ideal metrological sources. Moreover thanks to recent improvements in THz technology, robust and versatile metrological frequency references are available [2-4], enabling absolute measurements of THz frequencies.

We report on the still missing breakthrough of combining these metrology tools to classical schemes of optical molecular spectroscopy, allowing the determination of the absolute frequency of the investigated transition. As reported in [4], our approach to THz metrology consists of a free space THz Frequency Comb Synthetizer (FCS), while the CW THz source is a QCL operating at about 2.5 THz. A small fraction of the QCL radiation is withdrew from the main beam and used for the optical beating between the QCL and one tooth of the frequency comb. The beatnote signal is acquired on a real time FFT spectrum analyser. The remaining part of the QCL radiation is sent through a spectroscopy cell, in a direct absorption spectroscopy scheme.

A first technique to retrieve the absolute frequency of the molecular transition consists (once known the order N of the tooth involved in the beating process) in recording synchronously the frequency of the beatnote and the absorption signal, while scanning the laser current – and therefore its frequency - across the molecular transition.

Fig. 1a shows the absorption profile acquired by a pyroelectric detector, its fit with a Voigt functions and the corresponding residuals. Fig. 2b shows some statistics on the measurements performed with this technique. A preliminar value obtained for the line center, 2553831.6 (1.0) MHz, is compared with the value calculated for this transition in [5]. These measurements need to be refined and accurately analysed. At the moment the biggest source of uncertainty is the lack of an absolute metrological reference as, for example, a Rb-GPS (Global Positioning System) disciplined 10-MHz quartz oscillator. Once the experimental apparatus will be upgraded, a statistical analysis of the obtained values will be performed to reduce the experimental error towards the level of the estimated error of the calculated value, thus allowing an experimental validation and refinement of the molecular model.

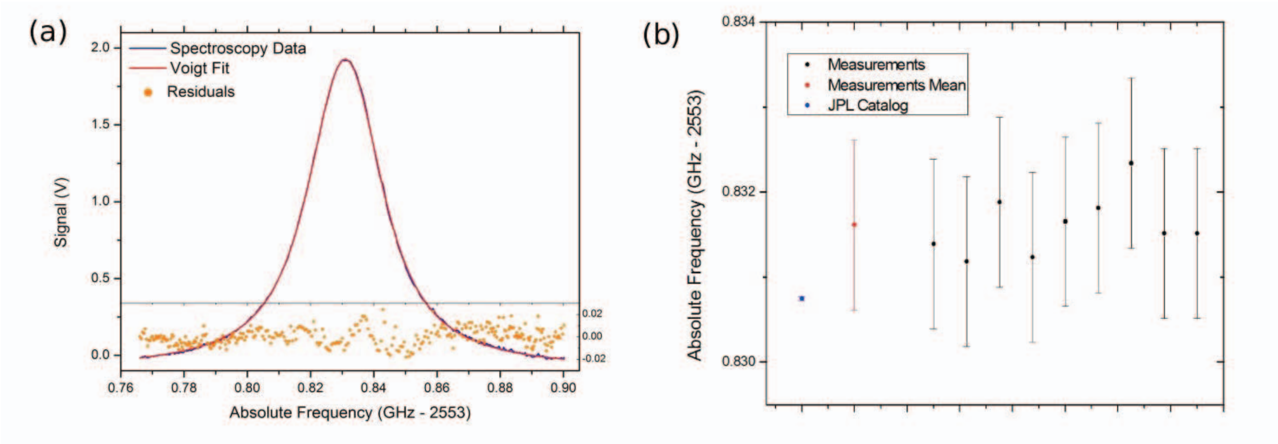

Fig. 1: (a) Absorption profile of the investigated molecular transition, and (b) comparison between our measurements and the calculated value from the JPL molecular catalogue.

References

[1] M.S. Vitiello, L. Consolino, *et al.* Quantum-limited frequency fluctuations in a terahertz, laser. *Nat. Photon.* **6** (2012).
[2] S. Barbieri, *et al.* Phase-locking of a 2.7-THz quantum cascade laser to a mode-locked erbium-doped fibre laser. *Nat. Photon.* **4** (2010).
[3] M. Ravaro, *et al.* Phase-locking of a 2.5 THz quantum cascade laser to a frequency comb using a GaAs photomixer. *Opt. Lett.* **36** (2011).
[4] L. Consolino, *et al.* Phase-locking to a free-space terahertz comb for metrological-grade terahertz lasers. *Nat. Commun.* 3:1040 doi:10.1038/ncomms2048 (2012).
[5] L.-H. Xu, *et al*, Torsion–rotation global analysis of the first three torsional states (vt = 0, 1, 2) and terahertz database for methanol, *J. Mol. Spectrosc.* **251** (2008).