Precision and high sensitivity molecular spectroscopy with frequency comb based sources in the mid-IR

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Trace gas detection of molecular species plays a key role to access knowledge about physical, chemical, and biological processes. Its applications span from fundamental physics to applicative fields as diverse as environment, medicine, security or archaeology/cultural heritage. A privileged window for molecules detection is the mid and far-infrared (IR) part of the spectrum, also named “fingerprint region", where the strongest absorption lines, belonging to fundamental ro-vibrational bands, can be excited. Progress in physics and chemistry due to atomic and molecular laser spectroscopy around the visible and near-IR spectral range has benefit of well-assessed metrological-grade laser sources and spectroscopic techniques in these spectral regions. Recently, even in the mid and far-IR, the growing number of powerful, stable and tunable coherent cw sources, such as nonlinear-based sources [1-3] or quantum cascade lasers [4-7] are definitely improving access to most spectroscopic techniques. In particular, their combination with optical frequency comb synthesizers (OFC’s) has improved frequency stability to reach unprecedented resolution, precision and accuracy on molecular spectroscopic measurements. Moreover we could set an unprecedented limit in trace gas detection, accessing the part-per-quadrillion (ppq, 10^{-15}) concentration range, when implemented with appropriate spectroscopic technique [8,9].

In this conference, I present a review of our recent results in trace gas sensing of molecular species by using these new hybrid frequency comb based IR sources and their combination with a new spectroscopic approach, named Saturated-CAvity Ringdown (SCAR) spectroscopy [8]. In particular, two applications will be presented. First, the trace gas detection of a very elusive molecule as radiocarbon dioxide ($^{14}C^{16}O_2$) at concentrations well below its natural abundance (i.e 10^{-12}) [9]. This all-optical setup, orders of magnitude more compact and less expensive than accelerator mass spectrometry (AMS), is a complementary alternative to AMS for radiocarbon based applications, in particular dating of organic samples [10]. Second, a spectroscopic test for possible violation of the symmetrization postulate (SP), one of the basic postulates in quantum mechanics. The experiment searches with extremely high sensitivity for (very weak) molecular lines involving the forbidden states due to the required symmetry of the wave function under exchange of identical nuclei in molecules. The present result improves about four times the upper limit for possible violation for exchange of bosonic nuclei by using $^{12}C^{16}O_2$ molecule [11].

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