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Continuous Vernier filtering of an optical frequency comb for broadband cavity-enhanced molecular spectroscopy

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ABSTRACT

We have recently introduced the Vernier-based Direct Frequency Comb Cavity-Enhanced Spectroscopy technique which allows us to record broadband spectra at high sensitivity and GHz resolution (Rutkowski and Morville, 2014) [1]. We discuss here the effect of Vernier filtering on the observed lineshapes in the $3\nu+\delta$ band of water vapor and the entire A-band of oxygen around 800 nm in ambient air. We derive expressions for the absorption profiles resulting from the continuous Vernier filtering method, testing them on spectra covering more than 2000 cm^{-1} around 12,500 cm^{-1}. With 31,300 independent spectral elements acquired at the second time scale, an absorption baseline noise of 2×10^{-8} cm^{-1} is obtained, providing a figure of merit of 1.1×10^{-10} $cm^{-1}/\sqrt{Hz}$ per spectral element with a cavity finesse of 3000 and a cavity round-trip length around 3.3 m.

1. Introduction

Optical frequency combs (OFC), generated by femtosecond mode-locked lasers, are recognized to be very powerful devices for spectroscopy. They offer large optical bandwidth ($\Delta\nu$) discretized at the comb teeth frequencies. They are defined by the repetition rate of the femtosecond oscillator f_{rep} and by a carrier-envelope offset frequency f_{ceo} translating the entire comb of a constant value comprised between 0 and f_{rep}, each comb tooth frequency being indexed by an integer n and defined as $\nu_n^{las}=n\cdot f_{rep}+f_{ceo}$. Several approaches [2–8] have been designed to combine OFCs with the extended path lengths associated with high finesse cavities to attain high sensitivity in molecular absorption spectra. Some resolve the comb mode structure (with 1 GHz mode locked laser) [2,3], some are fast (around or below the ms timescale of acquisition) [4,5], some are particularly sensitive (baseline noise around 10^{-4}) [6], but none truly exploit the full bandwidth of the OFCs, restricting the accessible spectral range to roughly a few hundred wavenumbers, around ten percent of the entire range of a typical Titanium:Sapphire (Ti:Sa) mode-locked laser.

Recently, we have developed the Vernier continuous filtering technique, combining an OFC with a high finesse cavity to achieve the measurement of the full OFC spectrum with high sensitivity, GHz resolution and sub-second acquisition time [1]. The Vernier filtering deliberately mismatches in a controlled fashion the laser repetition rate and the cavity free spectral range FSR_c (as are the two scales of a Vernier caliper). The spectral cavity output then exhibits a new comb whose mode-spacing is sufficiently large to be dispersed with a low resolution optical grating. This Vernier filtering scheme was adopted as a strategy to calibrate telescope spectra, creating the so-called astro-comb [9,10]. Gohle et al. [2] first applied this approach to laboratory molecular spectroscopy, resolving the 1 GHz mode structure of a Ti:Sa mode-locked laser over 4 THz (130 cm^{-1}) in 10 ms, but at the price of a poor sensitivity (absorption baseline noise of a few 10^{-6} cm^{-1} with an effective optical path-length around 300 m). In 2014, the same philosophy was extended to a 250 MHz Erbium

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doped fiber mode-locked laser [8] without the comb mode resolution (a resolution of 1.1 GHz was obtained). A spectral coverage of $160\,\mathrm{cm}^{-1}$ and a sensitivity of $8\times10^{-8}\,\mathrm{cm}^{-1}$ (with an effective optical path-length of 11 km) was demonstrated on a time scale of 1 s. Still, only a small part of the OFC was exploited and sensitivity performances were again limited by the strong frequency-to-amplitude noise conversion resulting from the unstabilized cavity with respect to the stabilized OFC. In this work, we present our approach [1] characterized by the degree of mismatch between FSR_c and f_{rep}. In astro-comb or previous works in molecular spectroscopy, the mismatch is large enough to filter out the comb teeth adjacent to the tooth transmitted by the cavity. Each tooth of the new comb created at the cavity output thus corresponds to a single tooth of the original optical comb. The cavity length is slightly swept to extract spectroscopic information from the sample placed inside it, allowing to transmit the comb teeth successively through the cavity. The cavity transmission is thus modulated by the matching condition. Conversely, our scheme involves sufficiently small mismatches to simultaneously (partially) transmit several adjacent teeth. The matching condition is always fulfilled and the cavity length sweep induces a continuous transmission. This approach is conceptually similar to the so-called 'precision sweep of f_{rep} with cavity filtration' discussed by Thorpe et al. in [11] where f_{rep} is swept instead of the cavity length. In this paper, they provide a temporal cavity transmission revealing absorption lines of C_2H_2 without quantitative determination of the frequency and absorption scales. Here we show that, with an appropriate cavity locking scheme, this continuous Vernier filtering enables to probe the entire range of a free-running 100 MHz repetition rate Ti:Sa mode-locked laser, at GHz resolution and with a high sensitivity. With this approach implemented with an open-air cavity, an absorption baseline noise of $2\times10^{-8}\,\mathrm{cm}^{-1}$ is demonstrated with an effective optical path-length of 1.5 km and a spectral coverage larger than $2000\,\mathrm{cm}^{-1}$ with 31,300 independent spectral elements acquired in 1 s. We derive a full analytical model to describe the cavity transmission in presence of intracavity absorbing species for this continuous Vernier filtering regime. We show that both the real and imaginary part of the resonant molecular response need to be taken into account to explain the marked effect of the sign of the comb-cavity mismatch on the measured line profiles. Using the full model and a non-linear fitting algorithm, we present the results of the $3\nu+\delta$ water vapor band adjustment in ambient air over more than $100\,\mathrm{cm}^{-1}$ including more than 350 lines. We also assess the sensitivity capabilities of this approach from spectra of the first hot band of the $b^1\Sigma_g^+ \leftarrow X^3\Sigma_g^-$ doubly forbidden band of oxygen in ambient air.

2. The continuous Vernier filtering formalism

2.1. Identification of the perfect match

The Vernier approach relies on a controlled mismatch between FSR_c and f_{rep} from the reference position corresponding to the perfect match (PM). In the frequency domain, the PM occurs when both the scale and the origin of the OFC are tuned to match those of the cavity grid. In our system, FSR_c (controlled by the cavity length), and f_{ceo} are tuned to match f_{rep} and the cavity offset frequency f_0 respectively. A large fraction of the OFC modes are transmitted simultaneously through the cavity. It can be expressed, when an ideal cavity with no dispersion is considered, as $\nu_n^{las}=\nu_m^{cav}$ (for all $n=m$) where the integer m is the order of the longitudinal fundamental cavity mode. This reference position is easily identified by applying a small cavity-length modulation, for instance with one of the cavity mirrors mounted on a piezo-transducer (PZT). The global effect of this length-detuning is to dilate/compress the cavity grid, but at first order (if all intracavity dispersion sources such as mirror coatings or gas pressure are neglected) it shifts all the laser teeth out of resonance simultaneously. Using a detector with a bandpass lower than the laser repetition rate, the optical power signal measured at the cavity output when a small dither is applied around the PM resembles the signal obtained with a single frequency continuous-wave laser: a resonance curve corresponding to the Airy function (a Lorentzian line shape at high finesses) if the scanning speed guarantees the adiabatic response of the cavity, and the emergence of an increasingly pronounced ringing when the scanning speed is increased [12]. For a cavity length (round-trip) detuning of $\pm\lambda_n^{las}$, the longitudinal cavity mode of order $m=n\pm1$ reaches the laser mode n and secondary peaks appear in transmission. Due to the slight FSR_c variation, all the laser teeth can no longer enter in resonance simultaneously, and only a part of the OFC is transmitted at any time through the cavity: if the equality $\nu_n^{las}=\nu_m^{cav}(m=n\pm1)$ is satisfied for one comb tooth, the following tooth is slightly shifted with the corresponding cavity resonance, and hence partially transmitted through the Lorentzian line shape of the resonance. The secondary transmission peak is thus weaker in intensity and broader than the transmitted peak at the PM position. This situation is schematically depicted on Fig. 1. The pattern is symmetrical around the PM position. If f_{ceo} is not initially matched to the cavity offset, this symmetry is broken, resulting in a shifted and broadened main feature, less intense than the PM peak, and in un-symmetrical neighboring peaks.

Once this PM position is identified, a significant cavity length variation ΔL, which can be expressed as a large integer number k of λ_{las}, is applied. The chosen value of k depends on the cavity finesse. In the frequency domain, this new length creates a Moiré pattern between the laser comb and the grid of cavity resonances where some comb teeth are periodically coupled with a periodicity given by $1/\Delta L$, as we will show later. The comb-cavity beating creates at the cavity output periodic and controllable Vernier coincidences defining the Vernier comb.

2.2. The Vernier comb and its empty-cavity response function

The Vernier comb and its response function can be derived with the cavity transfer function $H(\nu)$ expressed