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OPTICAL FREQUENCY COMBS FOR ATOMIC CLOCKS AND CONTINENTAL FREQUENCY DISSEMINATION

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Nearly 25 years ago, Optical Frequency Combs (OFCs) have revolutionized practically from one day to the next the metrology of optical frequencies. Before that, only a few labs in the world could connect the optical (~10¹⁵ Hz) and the microwave (~10¹⁰ Hz) domains, at the price of the

complex operation of a chain of multiple non-linear frequency conversions. In contrast, OFCs provided a tabletop instrument, compact, reliable and operable by a single scientist. Unmistakeable sign: only a few years had passed before pioneering inventors of OFCs Theodor Hänsch and John Hall were awarded the 2005 Nobel prize in Physics, together with Roy Glauber.



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he principle of operation relies on a femtosecond pulsed laser, featuring a spectrum of typically ~100 000 phase-locked modes contributing simultaneously. The field can therefore be defined as:

$$E_{\rm comb}(t) = \sum_{n} E_n e^{i\phi_n(t)}$$

The average value of each phase $\phi_n(t)$ can be described by $2\pi (nf_{rep})$ $(+ f_0)t + \phi_n(0)$, where f_{rep} is the repetition rate, f_0 the offset frequency, *n* an integer number and $\phi_n(0)$ a mode-dependent offset. The Fourier transform of this field shows that a comb is strictly equivalent to a ruler in the frequency space: while f_0 is the offset of the first tooth of the ruler, f_{rep} is the difference between adjacent teeth. Very importantly, the coherence is preserved throughout the whole spectrum, which makes it a unique tool to connect reliably very different spectral domains. Beatnotes between an oscillator at v_{osc} and the comb feature a frequency at $f_{osc} = v_{osc} - n_{osc} f_{rep} - f_0$, which leads to a complete determination of v_{osc} provided parameters f_{osc} , n_{osc} , f_{rep} and f_0 are measured accurately.

The advent of OFCs renewed considerably the interest of the Time and Frequency community in the metrology of optical atomic transitions. Even though the definition of the SI second had been based on the hyperfine transition of Cs since 1967, researchers early realized that the use of atomic transitions in the optical domain would dwarf radically most of possible systematic shifts affecting clock frequencies. Nevertheless, if single ion clocks had been under development since the early 80's, the very cumbersome connection to the SI second was considerably hindering the perspective of a new definition. OFCs changed completely this perspective, and it is about at the same time, in 2003, that a researcher from the University of Tokyo, Hidetoshi Katori, proposed a scheme allowing the trapping and high resolution spectroscopy of many

neutral atoms simultaneously. The race between optical clocks based either on single ions or neutral atoms had started and is still ongoing today: with published uncertainties down to 1×10⁻¹⁸ or below, instruments operated notably with Sr, Yb, Hg, Yb⁺, Al⁺ or Sr⁺ have outperformed even state-of-theart Cs clocks by more than 2 orders of magnitude. OFCs are used to compare optical clocks either to the SI second or directly one with another: the span of the comb spectrum is so large that it allows comparing clocks that are hundreds of nanometers apart. In the perspective of the roadmap for a new definition the second, aiming at 2026, possible contenders to new primary and secondary representations of the SI second are thus compared frequently and reliably in order to progressively refine, year after year, the knowledge of their frequency ratios.

Progressively, the technology supporting OFCs improved and industrial devices became available. If the original mode-locked sources were Titanium-Sapphire lasers, they often missed the reliability necessary to measurements bound to last days, if not months. The alternative offered by fiber lasers in the near infrared region soon released this constraint: erbium-doped fiber laser based OFCs notably offer a mode-lock so reliable that a year of continuous operation is

Figure 1. Architecture of atomic clocks, oscillators and frequency combs at SYRTE. The three operational OFCs act as a bridge between microwave and optical oscillators, probing respectively the SYRTE microwave fountains and optical lattice clocks.

not an issue. More recently, research on compact combs based on micro-resonators or semiconductor systems has raised a strong interest in the community. Despite their low power and high repetition rate, making notably the measurement of f_0 challenging, the potential of devices that could be easily installed in a large range of academic or industrial applications is very appealing. Generally, the technical progress opened the door to contributions to a large variety of scientific and technical fields, way beyond frequency metrology, as detailed in reference [1]. Striking applications were for example published in molecular spectroscopy (dualcomb spectroscopy, analysis of gas sample to track pollution, analysis of human breath), astronomy (calibration of spectrograph), or distance measurements (laser ranging).

At SYRTE (Systèmes de Référence Temps-Espace), the French National Metrology Institute for Time and Frequency, OFCs are used on the one hand for accurate and high resolution frequency metrology and on the other hand for the transfer of spectral purity from one frequency domain to another. In the following sections, we describe how this is applied to atomic clocks comparisons, to the construction of very low noise sources in the optical and microwave domains and to the dissemination of frequency standards on national and even international distances.

ACCURATE AND HIGH RESOLUTION ATOMIC CLOCK COMPARISONS

In the last 30 years, the SYRTE teams have developed and improved an ensemble of seven atomic



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clocks strongly contributing to the accuracy of international timescales UTC (Coordinated Universal Time) and TAI (Temps Atomique International), and to the SI second monthly calculated by the BIPM (Bureau International des Poids et Mesures). This clock ensemble includes four microwave fountains, three based on cesium and one on rubidium, all featuring an accuracy of a few 10⁻¹⁶ and regularly contributing to the steering of TAI. In addition, we are developing three optical lattice clocks operated with strontium or mercury that reach an uncertainty of a few 10⁻¹⁷ and already provided calibrations reports to the BIPM. This is one of the most complete sets of clocks existing in the world, and in order to compare frequently all these instruments, we have developed an ensemble of three operational OFCs. The goal of this redundancy is twofold: ensure the accuracy of the measurements, and evaluate the stability of the comparisons (figure 1)

Our architecture relies on two Erbium doped fiber laser based OFCs (Menlo Systems, FC1500) featuring a spectrum in the near infrared, spanning from 1 μ m to 2 μ m after broadening. This allows a straightforward connection to the telecommunication bands around 1.5 μ m, but does not permit to address directly all optical clock transitions (typically between 200 and 800 nm). Modules doubling the frequency of the combs in specific spectral windows are

therefore necessary, for instance to generate teeth at 698 nm and 813 nm to connect respectively the strontium clock and lattice lasers (figures 2 and 3). This is complemented by a former generation titanium-sapphire laser based OFC, with a spectrum after broadening (500 nm to 1.1 µm) enabling the direct comparison of all SYRTE optical clocks. We chose to phase-lock all our frequency combs to a cw ultrastable laser (frequency instability below 10⁻¹⁵ between 0.1 and 1000 s timescales) in order to reach the so-called narrow linewidth regime: this ultrastability is transferred to each tooth of the comb, which leads to beatnotes with external oscillators that can be filtered in a narrow band. After mixing out f_0 from all the beatnotes f_{osc} , we effectively generate a virtual comb at $f_0 = 0$ after isolating the RF single tone at v_{osc} - $n_{osc}f_{rep}$, thus rendering every following measurement insensitive to f_0 . From this point, phase-locking a single beatnote between the comb and an ultrastable laser by applying feedback to the optical length of the femtosecond laser cavity, so as to keep f_{osc} - $n_{osc} f_{rep}$ constant, is sufficient to 'freeze' entirely f_{rep} and therefore the position of the comb in frequency domain. Comparing the frequency of the resulting f_{rep} signal (in the RF or microwave domain and easily obtained by photodetection of the pulse train emitted by the femtosecond laser) with signals referenced to microwave frequency standards •••

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(atomic fountain clocks in particular) and to the SI second produces a high-precision measurement of the absolute frequency of optical clocks. Furthermore, by appropriate arithmetic combination of the various signals involved, it is even possible to extract directly the frequency ratio between two optical clocks without involvement of microwave standards and even independent of the residual noise of the OFC itself! [2]

REFIMEVE FIBER NETWORK AND DISSEMINATION OF AN ACCURATE 1542 NM REFERENCE

The research infrastructure REFIMEVE¹ is a network of optical fibers dedicated to the ultra-low noise dissemination of an ultrastable carrier at 1542 nm throughout the French territory. It provides sustainably and reliably (uptime > 85%) this infrared reference to many academic institutions and other research infrastructures, state agencies and industrials. The source of the disseminated signal is a SYRTE 1542 nm ultrastable laser, which defines the stability and the accuracy received by the end users. Both operational Erbium combs are phase locked to this laser, which in turns allows us to lock its frequency to a hydrogen maser on long (>100 s) timescales, while benefiting from the stability of a high finesse Fabry-Perot cavity on shorter timescales. The active compensation of the propagation noise in the network yields a signal at the disposal of the users featuring a stability below 2×10^{-15} at any timescale, and an accuracy of 1×10^{-14} on the fly, that can be pushed down to $< 1 \times 10^{-15}$ on demand.

International connections to similar networks have enabled comparisons on a continental scale between SYRTE and PTB (Germany, since 2015), NPL (UK, since 2016), INRIM (Italy, since 2020) and soon CERN (Switzerland). All together, this is an ensemble of ~12 optical clocks that are interfaced with this European 1542 nm reference by OFCs. These instruments are frequently compared one to the



Figure 3. Operational OFC 'Er2' at SYRTE. The comb is equipped with optical setups to form ultrastable optical beatnotes with various lasers. At the forefront, an electronic setup is being set up to generate operational low-noise microwave to replace the cryogenic sapphire oscillator in the long run.

others, in order to ascertain the reproducibility of frequency ratios between the various contenders to a new definition of the second. The long term monitoring of these ratios also contributes to tests of fundamental Physics, such as the search for a possible drift of fundamental constants. Finally, in the last years, the field of Earth Sciences has expressed a growing interest in optical clocks: as their frequency is sensitive to the local gravitational potential, this allows them to contribute to the refined mapping of the geopotential and to an improved determination of the geoid (equipotential approximating the mean sea level). It is in this spirit that SYRTE started recently the development of a transportable ytterbium optical lattice clock, that will be moved in the future along the REFIMEVE¹ network and remotely compared to the stationary European clocks. This new device will be equipped with a transportable OFC in order to measure the frequency of the clock versus the 1542 nm carrier, thus opening the possibility to exploit the ~60 outputs of the network over the metropolitan territory.

¹ Network under supervision of Université Sorbonne Paris Nord, Observatoire de Paris-PSL and CNRS

TRANSFER OF SPECTRAL PURITY

Beyond their measurement capacity, OFCs can also be utilized for novel applications where the excellent spectral purity of a state-of-the-art optical oscillator (cw laser) serves as a reference to create an ultra-pure radiation in an entirely different part of the electromagnetic spectrum, where high purity sources are difficult or even impossible to realize by any other technique. The idea is simply that when appropriately phase-locked to a high spectral purity source (in the optical or near-infrared domain), each tooth of the comb reproduces this spectral purity, and can be used to generate and/or phase-lock other sources in different spectral domain. Provided the phase comparison/ phase-locking processes are set up with extreme care and expertise to be sufficiently low noise, the final radiation will reproduce the spectral purity of the initial state-of-the-art reference, but transferred to a different spectral domain.

At SYRTE, we have first demonstrated the use of this technique to transfer the spectral purity from an ultra-stable laser in the near-infrared domain (1542 nm) to an other laser in the same domain but at a significantly different wavelength (1062 nm) [3]. We showed how, with extreme precautions, this could be realized with a minute added noise corresponding to a few 10⁻¹⁸ at a 1s timescale, well below the residual frequency fluctuations of any existing ultra-stable laser. In this case, the idea is simply to directly phaselock the 1062 nm laser onto an existing tooth of the comb close to it in frequency. Significant care must, however, be taken to minimize or cancel the noise resulting from the propagation of the laser radiations in fibered or free-space optics where even minute fluctuations arising - for example - from temperature changes or air flow currents may degrade the final performance.

In collaboration with our colleagues from LPL (Laboratoire de Physique des Lasers, CNRS/Université Sorbonne Paris Nord), we have also demonstrated a transfer of spectral purity from a 1542 nm wavelength laser to a ~10 µm wavelength radiation emitted by a Quantum Cascade Laser (QCL) [4]. In this case, an extra level of difficulty arose from the physical separations between SYRTE (hosting the 1542 nm laser) and LPL (where the QCL, the OFC, the equipment to phase lock it and characterize its spectral purity, was residing). This difficulty was solved by the use of the REFIMEVE network which connects the two laboratories. In this case, the 10 μ m wavelength spectral range is not directly accessible by existing OFCs, and we utilized non-linear optics (difference frequency generation) to generate a comparison signal between the near-infrared OFC and the 10 μm QCL. This technique demonstrated not only the most spectrally pure QCL-laser emission ever produced, but also provided absolute and SI-traceable referencing of its frequency.

Last, in collaboration with our industrial partners from MenloSystems GmBH and Discovery Semiconductor Inc., we have demonstrated how realizing the transfer of spectral purity from a near-infrared laser (1542 nm) to the microwave domain (12 GHz) was able to produce the lowest phase-noise microwave signal ever demonstrated by any existing technique [5]. This is of large interest - in particular - for radar applications where the quest for lowphase noise carrier is one of the sources of resolution improvement. In this case, the microwave signal is simply produced by photo-detecting the train of pulses emitted by the OFC. Mathematically

speaking, if the spectral purity transfer is perfect, since the emitted microwave signal is phase-coherent with the comb and the near-infrared reference laser, its phase fluctuations are imposed by those of the reference laser at 1542 nm, but with a very large division factor (of the order of 17000... the ratio between the optical and the microwave frequency). In order to be as close as possible to this mathematical ideal, an ultra-low-noise OFC was developed for this purpose by MenloSystems, and a very high linearity photo-diode was developed by Discovery Semiconductor. This is paramount for extreme performance, in particular because amplitude-fluctuations will generate phase fluctuations in the photo-detection process (even though we use special "magic point" conditions in the vicinity of which this effect is strongly reduced), and because high optical power needs to be used for photodetection in order to minimize the effect of thermal and quantum noise. We demonstrated an absolute phase noise of -173 dBc/Hz at 10 kHz and -106 dBc/Hz at 1 Hz from a 12 GHz carrier.

CONCLUSION

We have presented how optical frequency combs, both utilized as measurement tools and as mean for transferring spectral purity between various spectral domains, can achieve an extremely high level of metrological performance and be put to use in the most demanding high-precision measurements apparatus. Extreme care must however be taken in order to achieve the best result, but as commercially available systems are constantly improving, such performances will progressively become available for non specialists in turn-key systems.

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