A combination of continuous-wave optical parametric oscillator and femtosecond frequency comb for optical frequency metrology

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We combine a tunable continuous-wave optical parametric oscillator and a femtosecond Ti:Sapphire laser frequency comb to provide a phase-coherent bridge between the visible and mid-infrared spectral ranges. As a first demonstration of this new technique we perform a direct frequency comparison between an iodine stabilized Nd:YAG laser at 1064 nm and an infrared methane optical frequency standard at 3.39 µm.

Continuous-wave optical parametric oscillators (cw-OPOs) are one of the most promising infrared (IR) laser sources for high-resolution molecular spectroscopy. They offer wide tuning range and high output power levels as well as narrow linewidth and the possibility of phase locking to stable optical references. Combining them with recently developed optical frequency combs based on femtosecond lasers opens up new perspectives in optical metrology. In such a system the special properties of the OPO allow it to serve as a bidirectional coherent bridge linking the IR and visible spectral ranges (Fig. 1). For example, an infrared methane optical frequency standard can be used as a highly stable reference for an OPO and a visible frequency comb. Alternatively, an OPO phase locked to a visible frequency comb, which itself is referenced to a high performance microwave or optical clock, can provide stable emission with known absolute frequency in both the mid-IR and telecom spectral ranges. In this Letter we present the first implementation of this new technique.

Methods of femtosecond optical frequency metrology are based on the establishment of a phase-coherent relation between an optical frequency and that of a primary microwave clock or of an optical frequency standard. This is achieved using a comb of equidistant lines with frequencies \( f_n = n f_{rep} + f_0 \), where \( n \) is an integer, \( f_{rep} \), the repetition rate, and \( f_0 \), the carrier-envelope offset frequency of the femtosecond laser, with \( f_0 < f_{rep} \). Current optical frequency combs are predominantly based on mode-locked Ti:Sapphire and Er:fiber lasers. Their output is thus restricted to the visible and near-IR spectral ranges, while building an optical clockwork capable of addressing the mid-IR range has remained a challenge. Application of frequency combs for this purpose requires their combination with additional steps of sum- or difference-frequency generation (SFG and DFG).

SFG clockworks need for their realization additional transfer oscillators such as diode lasers or a commercial cw-OPO. Offset-free DFG combs, which are produced in the IR by mixing either the output of two synchronized visible frequency combs or different spectral components of a specially designed Ti:Sapphire laser, have very low output power (tens of µW total, with < 1 nW per comb mode). All these approaches phase-coherently connect IR frequencies and primary microwave clocks via \( f_{rep} \) but not the optical frequencies. They are either tailored to very specific goals or quite complex, and generally they require a number of non-linear crystals and supplementary lasers to make them useful for spectroscopic applications. Thus, a specific benefit of the new method presented here is that it consolidates all these subsystems into a single cw-OPO, providing direct phase-coherent link between two optical frequencies — the visible and the infrared.

The scheme itself is based on the fact that a singly resonant + pump resonant cw-OPO emits not only strong signal (S) and idler (I) waves, but also a set of weak (a few µW) components resulting from non-phase-matched mixing processes of the resonated pump (P=S+I) and signal waves. Some of these (2S, P+S, 2P) are located within the emission range of a Ti:Sapphire femtosecond laser comb. Forming suitable differences of the heterodyne beat frequencies with adjacent comb lines then allows mutual phase locking of OPO optical frequencies and the microwave frequencies \( f_{rep} \) and \( f_0 \) characterizing the femtosecond comb. This can be implemented following a variety of schemes, e.g. the basic four-step locking scheme as illustrated in Fig. 2.

1. Phase lock the OPO idler frequency I to a He–Ne/CH₄ standard.
2. Phase lock the comb spacing \( f_{rep} \) to the frequency difference between the lines P+S and 2S, which is equal to the already stabilized idler frequency I, \((P+S)-2S=I\).

![FIG. 1: OPO-comb tandem as a bidirectional coherent link between IR and visible spectral ranges.](https://example.com/fig1.png)
3. Measure and stabilize the difference between frequencies (P+S) and P, and therefore the signal frequency S using the comb, [(P+S)-P=S]. As a result, the stability of the methane standard is transferred to the signal and pump frequencies, and thus the entire OPO spectrum.

4. Measure and stabilize the comb offset frequency $f_0$ relative to one of the OPO lines, e.g. P.

The first two steps by themselves are already sufficient to implement a methane-based infrared optical molecular clock analogous to that reported by Foreman et al.\textsuperscript{19} The last two steps then phase lock the whole femtosecond frequency comb and all OPO output lines to the He–Ne/CH$_4$ standard.

As a first realization of this method we have performed a direct comparison between a Nd:YAG laser (1064 nm) stabilized on the $a_{10}$ line of the R(56) 32–0 iodine transition near 532 nm\textsuperscript{19} and a He–Ne laser stabilized on the $\sim$300 kHz wide $P(7)F_2^{(2)}$ line of the methane molecule at 3.39 $\mu$m. The latter is integrated in a transportable He–Ne/CH$_4$ frequency standard and serves us as a highly stable (over hundreds of seconds) IR reference laser.\textsuperscript{20} The methane standard and reference laser were both previously characterized during several absolute frequency comparisons.

The experimental arrangement of the frequency comparison is shown in Fig. 3. The cw-OPO was specifically developed for applications in high-resolution Doppler-free molecular spectroscopy and metrology.\textsuperscript{1,2} It is similar to a system described earlier although modified to be mechanically more stable and featuring improved cavity lock electronics. The setup is based on a periodically-poled lithium niobate (PPLN) crystal with multiple grating periods, pumped by a monolithic 1-W Nd:YAG laser. The pump and the signal waves are both resonated in the same, folded cavity, the length of which is locked to the pump laser using a piezoelectric transducer (PZT). A specially designed temperature-stabilized intracavity etalon allows controlled access to any desired wavelength in a wide OPO emission range: 1.5–1.9 $\mu$m (signal) and 2.4–3.7 $\mu$m (idler). Using this configuration, we obtain well-defined tuning behavior of the idler output radiation at power levels of >50 mW at 3.39 $\mu$m, exceptional long-term stability, and an instantaneous linewidth of about 10 kHz. The idler frequency can easily be phase locked to any optical reference using a phase-locked loop (PLL) with a bandwidth of $\sim$15 kHz and by applying the correction signal to the pump laser PZT.\textsuperscript{20}

Our frequency comb is based on a femtosecond Ti:Sapphire ring laser (GigaOptics, GigaJet-20) with a repetition rate $f_{\text{rep}}$ $\sim$750 MHz. A continuum between 0.5 and 1.1 $\mu$m with a total power of $\sim$300 mW is generated in a photonic crystal fiber and then split with a diffraction grating into three spectral parts, centered around the 1064 nm, 775 nm and 631 nm — corresponding to the OPO lines P, 2S and P+S. After additional filtering with Fabry-Perot etalons these beams are overlapped with the related OPO components and sent to avalanche photodiodes. Typical power levels here are 10, 0.3 and 1 $\mu$W for P, 2S and P+S, respectively. The three resulting OPO beat signals with adjacent comb lines typically have signal-to-noise ratios of 25-40 dB in a resolution bandwidth of 100 kHz. In a slight modification of the more general scheme presented above we measured the iodine frequency relative to the OPO pump laser.

During the comparisons only $f_{\text{rep}}$, was phase locked to the methane standard while the comb offset $f_0$ remained free-running, similarly to the approach of Telle et al.\textsuperscript{20} Frequency differences between the relevant filtered and amplified beats were processed using double-balanced mixers. Frequency generators and counters involved were referenced to a Rubidium microwave standard. The signal used for phase locking of the comb spacing to the stabilized idler frequency and the final signal corresponding to the iodine stabilized laser frequency were both refined using tracking oscillators. The iodine frequency signal was then recorded using a PC-based counter board.

Fig. 4 shows the resulting relative Allan deviation averaged over different measurement runs (counter gate times between 1 ms to 10 s). The result of the comparison is limited by the iodine stabilized laser performance, which is known from independent measurements relative to a cryogenic optical resonator (CORE).\textsuperscript{19} The accuracy limitation of the new comparison method itself is
We also succeeded in reversing the scheme and phase locking with respect to a cryogenic optical resonator (CORE) (open circles) and known performances of the methane standard\textsuperscript{12} (solid line) and the CORE\textsuperscript{22} (dashed line). Inset: typical beat signal of the I\textsubscript{2} vs CH\textsubscript{4} comparison, counted with a 1 s gate time.

FIG. 4: Result of the frequency comparison between the iodine and methane stabilized lasers. Also shown are independent measurements of a similar iodine system\textsuperscript{10} with respect to the CORE (open circles) and known performances of the methane standard\textsuperscript{12} and the CORE\textsuperscript{22} (solid line). Inset: typical beat signal of the I\textsubscript{2} vs CH\textsubscript{4} comparison, counted with a 1 s gate time.

expected to be much lower than $10^{-13}$. Continuous measurement time was restricted by the slow degradation of the photonic crystal fiber, which led to non-uniformities in the comb spectrum and insufficient power levels. The OPO showed very reliable operation and remained phase locked to the methane stabilized laser over several days. We also succeeded in reversing the scheme and phase locking $f_{\text{rep}}$ and the idler frequency to the iodine stabilized laser.

As the current comparisons do not yet challenge the accuracy performance of the system, upcoming measurements will use better optical frequency references (e.g. CH\textsubscript{4} vs CORE) to explore the actual limitations. The ease-of-use of the setup could be improved by using specially designed PPLN crystals in order to enhance the power of the non-phase-matched OPO components 2S and P+S. Another line of development is to use the opposite direction of stability transfer in order to provide stable emission for the purposes of high-resolution Doppler-free molecular spectroscopy in the infrared with output power levels up to several Watts using a modified design\textsuperscript{15}. Specifically, this should lead to applications in precision IR spectroscopy, such as metrology of the strong transitions of cold CH\textsubscript{4} molecules\textsuperscript{8} and the study of rotation-vibrational transitions in decelerated and trapped OH-radicals\textsuperscript{9}.

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