

Referencing Femtosecond Laser Frequency Combs to a He–Ne/CH₄ Optical Frequency Standard

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Abstract—The combination of a cw OPO and femtosecond laser frequency combs enables several different approaches for the realization of CH₄ optical molecular clocks. These are based on a fs Ti:Sapphire or an Er-fiber laser comb, whose repetition rate frequency is phase locked to the frequency of the He–Ne/CH₄ standard. Both systems can also be operated in reverse mode for precise optical synthesis and high-resolution spectroscopy in the mid-infrared and telecom spectral ranges.

I. INTRODUCTION

Methane stabilized He–Ne lasers have historically been recognized as one of the important secondary mid-infrared optical frequency standards (OFSs). Recently, several infrared molecular optical clocks based on a He–Ne/CH₄ system have been demonstrated, where the repetition rate frequency of a femtosecond Ti:Sapphire or an Er-fiber laser comb was phase locked to the frequency of a methane standard using a difference-frequency generation process [1], [2].

Here we consider different approaches for the realization of CH₄ clocks using a combination of a continuous-wave optical parametric oscillator (cw OPO) and a femtosecond laser comb. The first methane optical clock developed in our group is set up on a fs Ti:Sapphire comb referenced to the frequency of a He–Ne/CH₄ standard. This clock was involved in a direct frequency comparison with an iodine stabilized frequency doubled Nd:YAG laser at 532 nm, which was reported previously [3], [4].

II. IMPLEMENTATION OPTIONS FOR CH₄ OPTICAL CLOCKS

For more compactness and reliability of future CH₄ optical clocks, we intend to combine a cw OPO and a femtosecond Er-fiber laser comb with a new monolithic He–Ne/CH₄ standard.

A. New developments of He–Ne/CH₄ standards

By now, a new generation of methane reference oscillators is under development at the P. N. Lebedev Institute. A new design of the He–Ne laser cavity with an internal methane cell based on a low-expansion Sitall glass monoblock has led to compact and reliable optical references at 3.39 μm . The first prototypes of such lasers exhibit a frequency stability of $\sim 10^{-13}$ at 1 s, exceeding that of high performance commercial hydrogen masers up to 100 s. A number of technological solutions have been implemented in the next generation of monolithic He–Ne/CH₄ optical frequency standards, which should be available very soon [5]. It is expected that this will increase their short-term stability by at least one order

of magnitude and improve the long-term performance to the level of 10^{-15} at 1 day (see the contribution by M. Gubin in these Proceedings).

Thus, the new methane optical clocks can be regarded in many precise frequency and time measurements as an interesting alternative to the best commercial microwave clocks due to their extremely low phase noise and very good short- and mid-term stability.

B. cw OPOs for metrology

High-resolution Doppler-free spectroscopy and metrological applications of cw OPOs have been made possible by solving the long-standing problem of controlled access to any desired wavelength in the wide emission range of OPOs, including the ability to precisely tune the output frequency over the molecular and atomic transitions of interest. For this, a new design for the OPO cavity with an intracavity etalon was implemented, extending the concept of a cw tunable singly resonant OPO with resonated pump wave [6], [7], [8]. The developed OPO, operating at wavelengths from 1.5 μm to 1.9 μm and from 2.4 μm to 3.7 μm , demonstrates very good intrinsic stability and spectral properties, which were determined in direct beat frequency measurements with a methane infrared optical frequency standard. Thus, an idler radiation linewidth of ~ 12 kHz and mode-hop-free operation of the OPO over several days were observed. For the first time, it was shown that an OPO can be phase locked to a highly stable optical reference and thus much more precisely controlled and tuned [9].

C. Combining a cw OPO and a femtosecond laser comb

In the past, we have already reported on the combination of this OPO with a femtosecond Ti:Sapphire optical frequency comb to realize our concept for a phase coherent link between the visible and infrared spectral ranges [3], [4]. The idea is based on the fact that pump-enhanced singly resonant cw OPOs emit not only strong signal (S) and idler (I) waves, but also a variety of weak components in the visible spectral range corresponding to their linear combination. The OPO output spectrum extends over more than six octaves and partly overlaps with femtosecond Ti:Sapphire and Er-fiber laser comb spectra as shown in Fig. 1. When the OPO is pumped by a Nd:YAG laser at 1064 nm, some of these lines (2S, P+S, 2P) are located within the emission range of a Ti:Sapphire

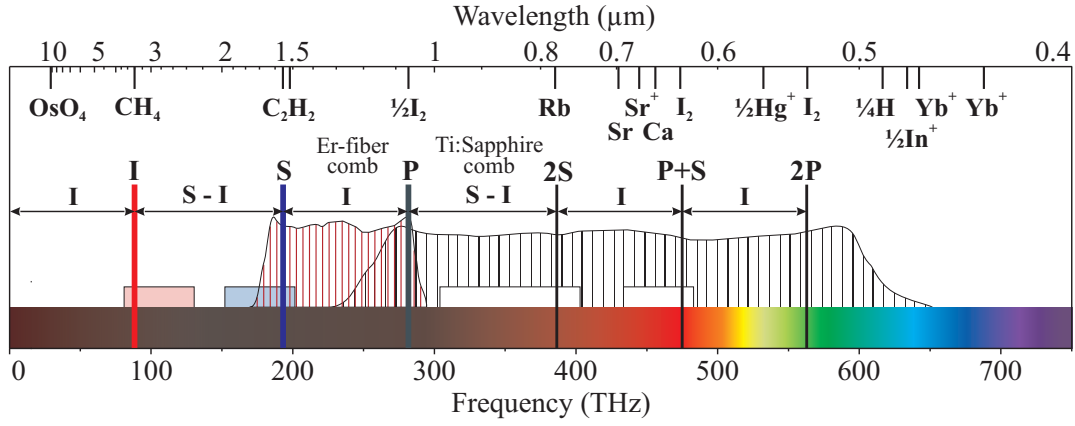


Fig. 1. Schematic of an OPO output spectrum, covered by a femtosecond Ti:Sapphire and an Er-fiber laser frequency comb. Boxes under the OPO lines mark their tuning ranges. Positions of some modern optical frequency references are also shown.

comb. The pump and signal frequencies are also covered by an Er-fiber comb spectrum.

The separation between some of the OPO output lines is exactly equal to the idler frequency, which can be phase locked to a methane optical standard at $3.39 \mu\text{m}$. The simultaneous measurements of the appropriate beat notes with the adjacent comb lines, followed by the processing of the frequency difference signals, provide then a phase coherent connection between the near-infrared optical frequency and the microwave comb repetition rate frequency as well as a direct link to all optical frequencies of the whole comb.

Thus, several opportunities are opened up by combining an OPO and a femtosecond laser comb (see Fig. 2):

- the repetition rate frequency of either fs Ti:Sapphire or Er-fiber laser comb can be referenced to the OPO idler frequency phase locked to a He-Ne/CH₄ standard, providing a microwave output signal of the methane optical clock;
- the OPO and femtosecond laser comb referenced to a mid-infrared methane frequency standard can be used for precise frequency measurements in the visible optical range;
- taking advantage of the new developments in visible

optical frequency standards, a highly-stable infrared radiation within the OPO signal and idler tuning ranges can be synthesized.

In these schemes a cw OPO referenced to the methane standard serves as a multi-frequency transfer oscillator, phase-coherently linking widely separated frequency ranges. Consequently, it becomes much easier to compare optical frequency standards of different nature.

III. CH₄ OPTICAL CLOCK BASED ON A FS ER-FIBER COMB

To gain the most benefit from the latest developments of very compact and efficient femtosecond Er-fiber laser combs and monolithic He-Ne/CH₄ optical frequency standards, we want to combine them with our cw OPO to build a small integrated methane optical clock. The principal schematics of this clock is quite simple as depicted in Fig. 3.

First, the idler frequency (I) has to be phase locked to a methane optical reference. Then, the beat notes of the pump (P) and signal (S) waves with the nearby Er-comb lines should be measured simultaneously. The corresponding beat frequencies can be expressed as $\delta_2 = P - (n_2 f_{\text{rep}} + f_0)$ and $\delta_1 = S - (n_1 f_{\text{rep}} + f_0)$, where f_{rep} is the repetition rate of

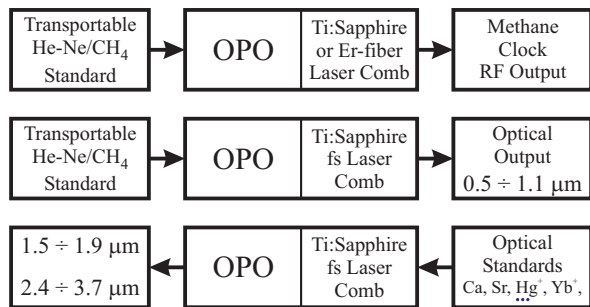


Fig. 2. Some possible schemes to phase coherently link mid-infrared, visible and microwave frequency ranges, using a combination of a cw OPO and femtosecond laser frequency combs.

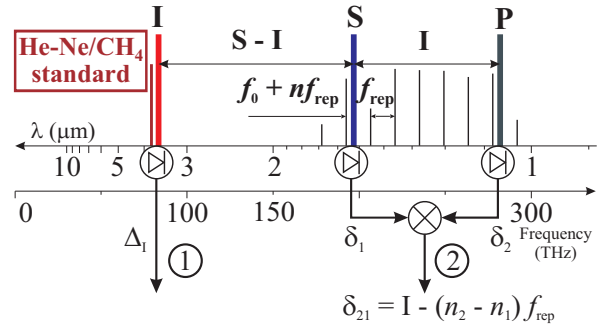


Fig. 3. Scheme of a CH₄ optical clock based on a fs Er-fiber laser comb referenced to a He-Ne/CH₄ optical frequency standard. See text for abbreviations.

the femtosecond Er-fiber laser (typical values for f_{rep} are 0.1–0.3 GHz) and $f_0 < f_{\text{rep}}$ is the offset frequency due to pulse-to-pulse carrier-envelope phase shift, and n_1 and $n_2 \sim 10^6$ are integers.

The beat note difference $\delta_{21} = \delta_2 - \delta_1 = (P - S) - (n_2 - n_1) f_{\text{rep}} = I - n_{21} f_{\text{rep}}$ is independent from the comb offset frequency and can be used to phase lock f_{rep} to the idler frequency (I). If we phase lock δ_{21} to a synthesized radio-frequency Δ_{12} , we obtain a simple relation between the comb repetition rate and the methane standard frequencies:

$$f_{\text{rep}} = \frac{(\nu_{\text{CH}_4} + \Delta_{\text{I}} - \Delta_{12})}{n_{12}},$$

where ν_{CH_4} is optical frequency of the methane standard, and Δ_{I} is a synthesized offset radio-frequency used for the idler phase lock loop.

The infrared optical frequency ν_{CH_4} of the methane standard is at least 10^6 times higher than the frequencies Δ_{I} and Δ_{12} , which are phase locked to a microwave reference. Thus, the resulting instability of f_{rep} is mainly determined by the performance of the methane optical reference. The comb repetition rate frequency phase locked to the He–Ne/CH₄ standard finally provides the microwave output of the CH₄ optical clock.

IV. CONCLUSION

In this paper we discussed our efforts on referencing femtosecond laser combs to a near-infrared methane frequency standard. We have developed a CH₄ optical clock based on a femtosecond Ti:Sapphire laser comb. Our intention is to implement a compact methane clock relying on the new generation of the monolithic He–Ne/CH₄ standards combined with Er-fiber laser comb and cw OPO.

The upcoming direct comparison of these methane clocks with respect to each other or similar optical clocks should reveal their performance. Both systems can also be operated in reverse mode for precise optical synthesis and high-resolution spectroscopy in the mid-infrared and telecom spectral ranges.

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