DOPPLER-FREE SPECTROSCOPY OF CH_4 USING A CW OPTICAL PARAMETRIC OSCILLATOR

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We present the latest results on using continuous-wave optical parametric oscillators (cw-OPOs) for high-resolution Doppler-free saturation spectroscopy of methane and point out the resulting potential for future optical frequency standards. We also propose a new scheme for combining cw-OPOs with the recently developed femtosecond frequency combs to provide a versatile bridge between optical frequency standards in the visible, near-infrared and mid-infrared spectral ranges.

Stabilized He-Ne lasers, based on saturated absorption/dispersion resonances of the P7 $F_2^{(2)}$ rovibrational transition of the CH_4 molecule at 3.39 µm, are proven optical frequency standards with repeatability and accuracy at the level of 10^{-13}-10^{-14} [1]. The implementation of appropriate tunable laser sources, however, would allow one to use much stronger transitions from low-lying rovibrational levels as new references and thus lead to higher performance frequency standards [2]. Cw-OPOs are one of the most promising laser sources for this purpose [3]. Until recently, one of the remaining problems has been to prevent gaps in the spectral coverage of OPO and thereby to guarantee simple access to every molecular transition of interest.

To overcome this problem, we have developed a new singly resonant OPO that employs a specially designed intracavity etalon to provide a well-defined tuning behavior. The OPO is based on periodically poled lithium niobate, resonantly pumped by a monolithic Nd-YAG laser at 1064 nm (for more details see contribution of Peters *et al* in this volume [3] and in [4]). It features a narrow instantaneous linewidth of 12 kHz (Fig. 1a) and output power of more than 20 mW. We also succeeded in phase locking of the OPO idler frequency to a He-Ne stabilized laser (Fig. 1b) as well as to another, similar OPO.

To demonstrate the application potential of the new OPO, we performed Doppler-free saturation spectroscopy of methane at 3.39 µm (Fig. 2, [4]). The idler beam was split and sent as two counter-propagating probe (2 mW) and pump (10 mW) beams (Ø ~ 8 mm) through a spectroscopy cell filled with methane at a pressure of ~ 1 mTorr. Using the Pound-Drever-Hall frequency modulation technique (1.24 MHz modulation using an EOM), phase locking the OPO frequency to the He-Ne/CH_4 laser and tuning it over a 2-MHz wide range, we obtained the Doppler-free dispersive signal of the P7 $F_2^{(2)}$ line of methane shown in Fig. 2. The observed linewidth of 150 kHz can be attributed mainly to saturation broadening.

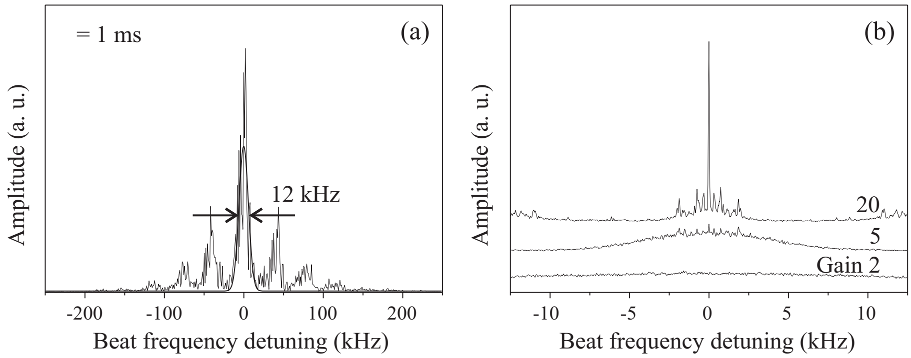

Figure 1. **(a)** Linewidth of the OPO idler output from a beat frequency measurement against a stabilized He-Ne/CH_4 laser. **(b)** Same beat signal when the OPO is phase locked to the He-Ne/CH_4 laser using three different relative gains of the PLL (bandwidth ~ 15 kHz).

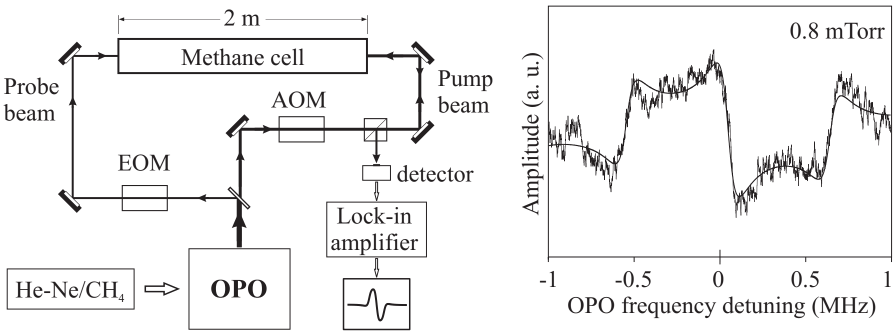

Figure 2. Setup for Doppler-free saturation spectroscopy of methane (left) and Doppler-free resonance of the P7 $F_2^{(2)}$ line with 150 kHz FWHM (right).

Given these capabilities, it should be possible to further improve the OPO frequency stability and use it to implement optical frequency standards based on the most suitable molecular transitions in the mid-infrared.

Another new perspective in optical metrology is opened up by employing OPOs in combination with the recently developed femtosecond frequency combs [5,6]. If the two sources are phase locked with respect to each other, the OPO may serve as a bridge that transfers the stability of frequency standards operating in the near- and mid-IR (e.g. He-Ne/CH_4 laser) to the visible spectral range and vice versa.

The method we propose utilizes the fact that the OPO emission spectrum includes not only the signal (S) and idler (I) waves (pump frequency P = S+I), but also a variety of linear combinations of these two frequencies generated by non-linear (generally not phase-matched) processes. Some of these lines (P+S, P+I, 2S) are located within the emission range of a femtosecond Ti:Sapphire laser. Thus, one can implement a four step locking scheme as illustrated in Figure 3:

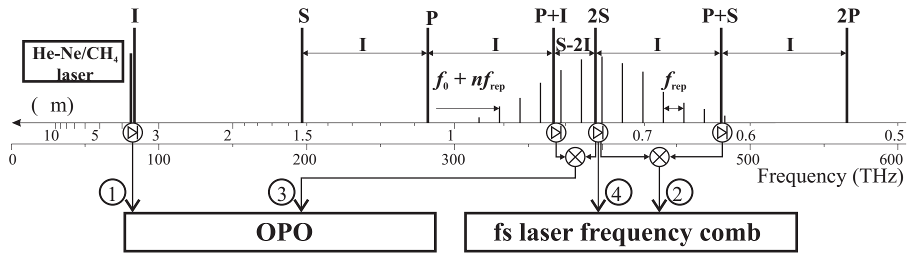

Figure 3. Proposed scheme of phase locking of the OPO output frequencies and a femtosecond laser frequency comb to a He-Ne/CH_4 frequency standard.

1. Phase-lock the OPO idler frequency I to a He-Ne/CH_4 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 Idler frequency I already stabilized in step 1).
3. Stabilize the separation between the lines 2S and P+I (which is equal to S-2I) using the comb. As a result, the OPO signal frequency S and pump frequency P will also be stable. Thus, the stability of the He-Ne/CH_4 standard is now transferred to the entire OPO output.
4. Stabilize the comb offset f_0 by phase-locking the comb relative to any OPO output line (e.g. 2S).

Using this scheme, all OPO output lines and the whole femtosecond frequency comb can be phase-locked to the He-Ne/CH_4 standard. The stability of the latter is thus transferred to the visible range, providing an optical frequency standard with better short- and medium-term stability [1] and lower cost than femtosecond frequency combs referenced to microwave standards (e.g. a cesium atomic clock). Note that for this scheme is generally *not* necessary to broaden the output spectrum of a Ti:Sapphire laser (assuming a pulse length of 30 fs).

Another possibility would be to lock the OPO to a femtosecond frequency comb referenced to a cesium fountain clock or another optical frequency standard and use it to provide stable emission in the IR: 1.48 - 1.93 μm (Signal) and 2.35 - 3.75 μm (Idler). Also, one could use the combination of OPO and femtosecond frequency comb to compare frequency standards of different physical nature and thus carry out tests of fundamental physics.

References

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