

# High-resolution Doppler-free molecular spectroscopy with a continuous-wave optical parametric oscillator

E. V. Kovalchuk, D. Dekorsy, A. I. Lvovsky, C. Braxmaier, J. Mlynek, and A. Peters

*Fachbereich Physik, Universität Konstanz, M696, 78457 Konstanz, Germany*

S. Schiller

*Institut für Experimentalphysik, Heinrich-Heine-Universität Düsseldorf, 40225 Düsseldorf, Germany*

Received February 13, 2001

We present a reliable, narrow-linewidth (100-kHz) continuous-wave optical parametric oscillator (OPO) suitable for high-resolution spectroscopy applications. The singly resonant OPO with a resonated pump is based on periodically poled lithium niobate crystal and features a specially designed intracavity etalon, which permits precise tuning to any desired wavelength in a wide range. We demonstrate Doppler-free spectroscopy of a rovibrational transition of methane at  $3.39 \mu\text{m}$ . © 2001 Optical Society of America

OCIS codes: 300.6390, 190.4970.

Considerable technological progress in the fabrication of periodically poled nonlinear crystals, durable optical coatings, and ultrastable solid-state pump lasers recently led to a new generation of continuous-wave (cw) optical parametric oscillators (OPOs) with single-frequency output covering a very wide range of the near-IR spectrum ( $0.8\text{--}4.0 \mu\text{m}$ ). These OPOs can now provide substantial amounts (10–250 mW) of coherent radiation with narrow-linewidth ( $<160\text{-kHz}$ ), continuous tunability (several gigahertz), and excellent long-term stability.<sup>1–7</sup> This unique combination of features makes them attractive and promising tools for molecular spectroscopy. Accordingly, several Doppler-limited spectroscopic applications of single-frequency cw OPOs have been demonstrated since 1998.<sup>5–10</sup> However, to our knowledge no experiments employing cw OPOs for Doppler-free spectroscopy have been reported. In this Letter we describe a new OPO design specially developed for high-resolution spectroscopy and present its application to observation of Doppler-free resonances in methane at  $3.39 \mu\text{m}$ .

Until recently, one of the remaining challenges in using cw OPOs for spectroscopic applications has been to prevent gaps in the spectral coverage and thereby to guarantee access to every atomic and molecular transition of interest. Such gaps were generally caused by irregularities in the nonlinear crystal and etalon effects that are due to residual reflectivity of antireflection coatings, resulting in spurious modulation of the wide OPO gain profile. Although OPOs showed excellent performance at local gain maxima, attempts to tune them to a desired wavelength led to unpredictable frequency hops between neighboring maxima. This behavior explains numerous past failures to apply cw OPOs for precision spectroscopy.

To avoid these problems, it is desirable that additional mode selection be provided. To this end, we have implemented a singly resonant OPO with a resonated pump in an extended cavity setting with a specially designed intracavity etalon (ICE). Unlike the etalons that are commonly used in single-frequency lasers and also in a variety of singly-

resonant OPOs,<sup>6,7,11</sup> this ICE is located inside a cavity with two resonated waves, of which it should affect only one (signal) and not the other (pump). It is therefore antireflection coated for the pump wave. The performance of this coating is crucial for the proper operation of the OPO.

The OPO arrangement is shown in Fig. 1. A monolithic diode-pumped Nd:YAG ring laser with an intrinsic linewidth of  $\sim 5 \text{ kHz}$  and 1-W single-frequency output power at  $1064 \text{ nm}$  is used as a pump source. We can tune the frequency of the laser by up to 40 GHz (9 GHz without mode hops) by changing the laser's crystal temperature. An optical isolator prevents backreflections from the OPO cavity. The OPO is based on a periodically poled lithium niobate (PPLN) crystal of 19-mm length and  $0.5 \text{ mm} \times 50 \text{ mm}$  aperture. The crystal contains 33 gratings with periods ranging from  $28.98$  to  $30.90 \mu\text{m}$ . At crystal temperatures from  $145$  to  $170 \text{ }^\circ\text{C}$ , the gratings provide quasi-phase matching for signal waves in the wavelength range  $1.48\text{--}1.93 \mu\text{m}$ , corresponding to  $2.35\text{--}3.75 \mu\text{m}$  for the idler. The crystal is temperature stabilized within  $\pm 2 \text{ mK}$ , and we can translate it to select one of the gratings.

The extended OPO cavity consists of three plane mirrors and one concave mirror. One mirror, M1, is

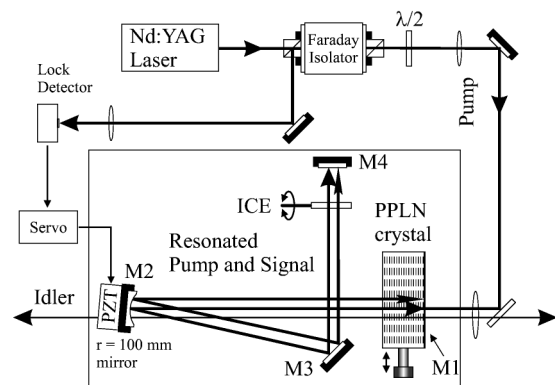


Fig. 1. Schematic of the OPO setup.  $\lambda/2$ , half-wave plate. See text for other abbreviations.

directly coated onto the plane entrance face of the nonlinear crystal and is highly reflecting for the pump (98%) and the signal (>99% for 1.35–1.65  $\mu\text{m}$ , with a maximum of 99.9% at 1.5  $\mu\text{m}$ ) but highly transmitting (>98%) for the idler (3–4  $\mu\text{m}$ ). The concave mirror, M2, of 100-mm curvature radius, and the plane end mirror, M4, have a reflectivity of better than 99.8% for the pump and signal and 1% for the idler. The plane mirror, M3, minimizes the angle of incidence at the concave mirror and therefore astigmatic aberrations of the resonator mode. At 45°, M3 reflects 99.8% of the pump and the signal and an average of 3% of the idler. The second plane face of the crystal is antireflection coated for all three wavelengths, with residual reflectivities of 0.15% for the pump, <1% (1.35–1.65  $\mu\text{m}$ , minimum 0.1% at 1.5  $\mu\text{m}$ ) for the signal, and <2% for the idler. The idler wave is thus not resonated and leaves the cavity through mirrors M1 and M2.

The concave mirror is located 43 mm from the crystal surface and is mounted on a piezoelectric transducer (PZT). The optical cavity length is 420 mm, corresponding to a free spectral range (FSR) of 360 MHz. One waist of the fundamental cavity mode, with radius  $w_0 = 35 \mu\text{m}$  (for the pump), is located at the entrance face of the crystal; the other, with  $w_0 = 440 \mu\text{m}$ , is located at the end mirror. The pump beam is focused into the PPLN crystal with an effective mode matching to the fundamental cavity mode 99%. The pump and signal waves both resonate in the same folded cavity, whose length is locked to the pump laser<sup>1,2</sup> by use of the Pound–Drever–Hall method.

The ICE is made from a 1-mm-thick undoped yttrium aluminum garnet plate (83-GHz FSR) that is coated to provide a reflectivity of 15% per surface for the signal. To minimize walk-off losses, the ICE is placed near the cavity end mirror, where the beam is almost collimated. The walk-off losses were negligible compared with the etalon effects caused by the residual 0.75% reflectivity of the ICE surfaces for the pump wave. The etalon effects, while affecting the threshold and tuning behavior of the OPO, did not prevent its stable operation. The influence of the ICE on the OPO performance is shown in Fig. 2. Although without the etalon no reliable tuning could be achieved, the introduction of the ICE provides well-defined tuning behavior and access to any desired wavelength.

The minimum threshold pump power of the OPO was 305 mW at a signal wavelength of 1.55  $\mu\text{m}$ . A finesse of 85 and a measured input mirror transmission of 1.8% for the pump wavelength yield round-trip cavity losses of 5.5%, of which 1% can be attributed to the etalon. At a pump input power of 808 mW the total idler output power (through M1 and M2) at 3.39  $\mu\text{m}$  was 58 mW, limited mainly by the nonoptimal transmission of M1 for the pump.<sup>12</sup>

By synchronously sweeping the pump frequency and tilting the ICE with a galvanometer scanner, we achieved continuous tuning over 1.8 GHz for the signal (corresponding to 5 FSRs of the cavity) and 1 GHz for the idler waves. Tuning was limited by the PZT servo amplifier and could be easily extended.

We observed mode-hop-free operation of the OPO over several hours, with a typical idler frequency drift of 0.5 MHz/min. To determine the spectral characteristics of the OPO, we performed a beat measurement between the idler wave and the narrow-linewidth (50-Hz) radiation of a transportable He–Ne/CH<sub>4</sub> stabilized laser<sup>13</sup> at 3.39  $\mu\text{m}$ . For an integration time of 500  $\mu\text{s}$  we found a linewidth of  $\sim 100$  kHz (Fig. 3). For integration times longer than 10 ms the output linewidth became dominated by frequency jitter of the pump laser.

To demonstrate the capabilities of the new OPO, we performed Doppler-free spectroscopy on the  $F_2^{(2)}$  component of the  $P7$  rovibrational transition of the methane molecule at 3.39  $\mu\text{m}$ . This line is traditionally used as a high-stability reference line in He–Ne optical frequency standards. The measurements were performed by use of the frequency-modulation saturation technique<sup>14</sup> in a 2-m-long methane cell (Fig. 4). The diameter of the collimated counterpropagating probe (2 mW) and saturating (10-mW) beams inside the cell is  $\sim 8$  mm, giving a transit-time broadening of approximately 30 kHz and a saturation parameter of  $\sim 0.1$  at a methane pressure of 6 mTorr. Using orthogonal polarizations for the pump and probe beams, together with the 80-MHz frequency shift provided by the acousto-optic modulator, helps very effectively to prevent perturbations of the OPO as a result of backreflections. To suppress slow frequency drifts of the OPO, we frequency locked the idler frequency

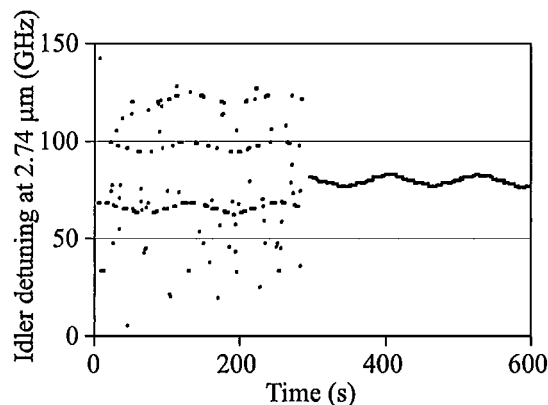


Fig. 2. Tuning behavior of the OPO idler output (left) without and (right) with ICE. We tuned the OPO by varying the pump frequency.

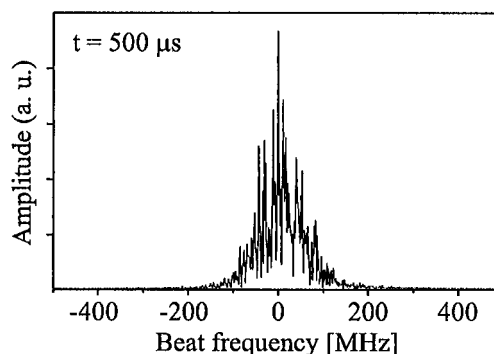


Fig. 3. OPO idler output linewidth.

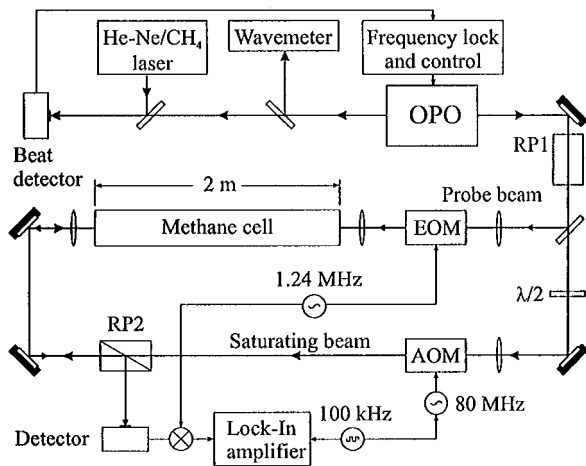


Fig. 4. Doppler-free spectroscopy setup. One OPO idler output is split into saturating and probe beams and sent through the methane cell. The probe beam is phase modulated by an electro-optic modulator (EOM) at 1.24 MHz (modulation index,  $\sim 1$ ). The saturating beam is chopped by the acousto-optic modulator (AOM) at 100 kHz. After the cell the probe beam is sent to a fast photodetector, whose output is analyzed by a phase-sensitive detection circuit, and a lock-in amplifier that extracts the spectroscopic signal. The second OPO idler output is used for frequency monitoring by a wavemeter and for frequency locking to a He-Ne/CH<sub>4</sub> laser. RP1, RP2, Rochon prism polarizers.

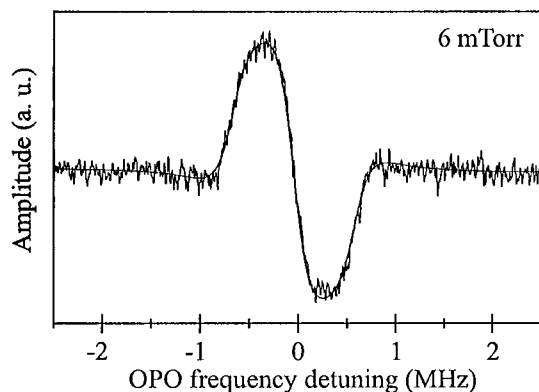


Fig. 5. Doppler-free resonance of methane at 3.39  $\mu\text{m}$ , obtained by scanning of the OPO idler frequency over 5 MHz in 50 s and use of a lock-in time constant of 30 ms. Curve, fit according to a theoretical model.<sup>14</sup>

(bandwidth,  $<100$  Hz) to the stabilized He-Ne laser with an offset of 40 MHz. We then scanned the idler frequency linearly over the saturated resonance by changing the offset frequency.

Doppler-free dispersion resonances were detected for methane pressures from 6 to 54 mTorr. At 6 mTorr the observed linewidth of 500 kHz (Fig. 5) can be attributed to a combination of pressure broadening (200 kHz) and medium-term OPO frequency jitter (200–400 kHz). At higher pressures the linewidth was dominated by pressure broadening.

In the future, we intend to reduce the linewidth of the OPO further by using a more rigid mechanical design and a higher-bandwidth cavity lock. After these modifications we expect the OPO output linewidth to

replicate that of a pump laser. Once such OPOs become available they should finally make up for the notorious lack of tunable narrow-linewidth light sources for high-resolution spectroscopy in the mid IR. One of the many possible applications of such a source would be in the development of a new generation of optical frequency standards based on strong molecular rovibrational transitions.<sup>13</sup>

In conclusion, we have demonstrated a new design for a pump-resonant singly resonant OPO with a tunable intracavity etalon, which permits access to any desired frequency in the wide OPO emission range. We have demonstrated the spectroscopic capability of the OPO by recording Doppler-free resonances in methane. This work opens up the path to a much wider range of applications for cw OPOs, especially in the fields of precision spectroscopy and metrology.

We are grateful to J. P. Meyn and R. Wallenstein for providing PPLN crystals and to Mikhail Gubin and his team at the P. N. Lebedev Institute (Moscow, Russia) for making the transportable He-Ne/CH<sub>4</sub> optical frequency standard available. We acknowledge financial support from the German Ministry of Education and Research, the German-Israeli Foundation, the Optik-Zentrum Konstanz, and the Gerhard-Hess Program of the German Science Foundation. A. I. Lvovsky is supported by the A. von Humboldt Foundation. Address all correspondence to evgeny.kovalchuk@uni-konstanz.de.

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