CONTINUOUS-WAVE OPTICAL PARAMETRIC OSCILLATORS AS NEW TOOLS FOR HIGH RESOLUTION SPECTROSCOPY

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We review the development of continuous-wave optical parametric oscillators (OPOs) at the University of Konstanz as coherent light sources for high resolution spectroscopy. We describe various implementations of such devices covering a wide range of wavelengths (550 – 4000 nm), discuss their performance and present first applications to Doppler-free spectroscopy of methane at 3.39 µm and molecular iodine at 580 nm.

1 Introduction

The increased availability of novel nonlinear materials crystals, high performance optical coatings and ultra-stable solid-state pump lasers has recently led to a new generation of continuous-wave optical parametric oscillators (cw-OPOs). After considerable development efforts undertaken in several laboratories around the world such OPOs can now provide powerful (10 – 800 mW) coherent radiation over a very wide range of near- and mid-infrared wavelengths (660 – 4000 nm). They feature extremely narrow line-widths (< 50 kHz), continuous tunability (up to tens of GHz), and good long-term stability (< 30 MHz frequency drift per hour, < 5% power drift per hour)[1-7]. By subsequent frequency doubling the emission range can be extended further into the visible (550 – 760 nm) with output powers up to 160 mW [14]. This unique combination of properties makes OPOs attractive and promising tools for applications in high resolution spectroscopy. In particular, they could remedy the general lack of widely tunable high power laser sources in the mid-infrared and offer a more convenient alternative to dye lasers in the visible spectral region.

Several Doppler-limited spectroscopic applications of single frequency cw-OPOs were demonstrated since 1998, emphasizing the potential outlined above [4,6-10]. Doppler-free spectroscopy, however, remained a unresolved challenge until recently demonstrated by our group [2]. The main difficulty has been a certain degree of unpredictability in the tuning behavior of the OPO which led to gaps in the spectral coverage and thus often precluded access to the atomic and molecular transition of interest. In the following we present two markedly different OPO systems that both address those issues.

2 Singly-resonant OPO pumped at 1064 nm

To generate output wavelengths in the near-infrared and mid-infrared, we employ a singly resonant OPO in which the pump wave at 1064 nm is resonantly enhanced in the same cavity as the signal wave (PR-SRO). To maintain the resonance condition with respect to the pump laser the cavity length is actively controlled. Besides leading to a lower oscillation threshold, this scheme offers the advantage of transferring the excellent spectral properties of a stable pump laser to both output waves (signal + idler).

As nonlinear material we use a periodically poled lithium niobate (PPLN) crystal with 33 different grating periods. At crystal temperatures between 145 and 170°C they provide quasi-phasematching for signal waves in the wavelength range 1.48 - 1.93 µm, corresponding to 2.35 - 3.75 µm for the idler. For coarse tuning of the output wavelength the crystal can be translated to select one of the gratings, while finer tuning is achieved by varying the crystal temperature and tuning the pump laser frequency.

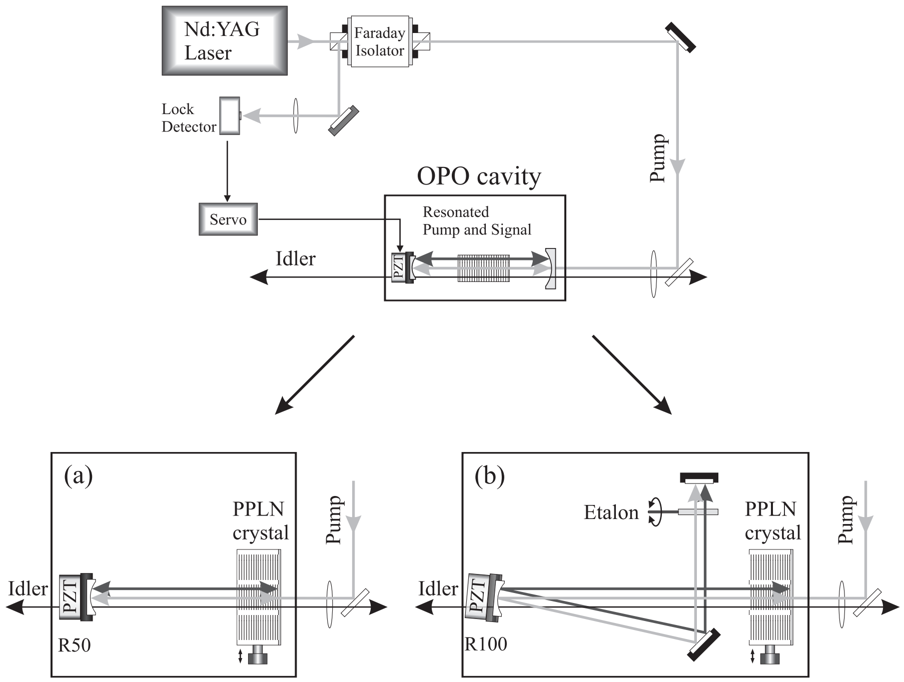

Figure 1. Two variations of a PR-SRO pumped at 1064 nm.

Figure 1 shows two variations of the OPO setup. In the original semi-monolithic version of the cavity (a) is formed by a single external mirror and a second mirror coated directly onto one face of the nonlinear crystal. While this system generally shows excellent performance at fixed output wavelengths [4], it exhibits unpredictable tuning behavior caused by irregularities in the nonlinear crystal and etalon effects due to residual reflectivity of AR-coatings.

To overcome these problems, we changed the setup to an extended cavity design (b) with a specially designed intracavity etalon (ICE) for additional mode selection. Unlike the etalons commonly used in single-frequency lasers and also a variety of singly-resonant OPOs [5-7], this etalon is located inside a cavity with two resonated waves, of which it should only affect one (signal) and not the other (pump). It is therefore coated to provide a low reflectivity of 0.75% per surface for pump compared to ~ 15% for the signal.

The influence of the etalon on the OPO performance is shown in Figure 2a. Whereas without etalon no reliable tuning over more than a few hundred MHz could be achieved, the introduction of the etalon provides the desired well-defined tuning behavior and access to any wavelength of interest. Continuous tuning over 1.8 GHz for the signal and 1 GHz for the idler waves is possible by synchronously sweeping the pump frequency and tilting the etalon using a galvanometer scanner. The idler output power generally exceeds 20 mW and the instantaneous linewidth is of the order of 10 kHz (Fig. 2b). Typical threshold powers are ~ 300 mW with, and ~ 200 mW without etalon.

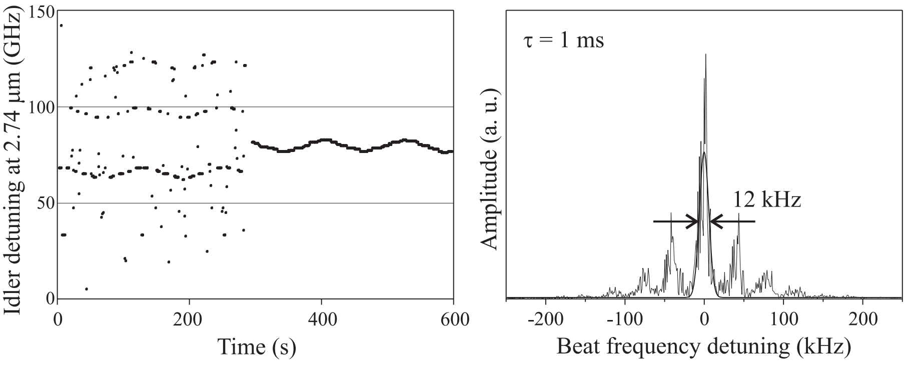

Figure 2. (a) Tuning behavior of the idler output of the PR-SRO of Fig. 1 without (left part) and with (right part) etalon. Tuning is performed by varying the pump frequency. (b) Instantaneous linewidth of the PR-SRO obtained from a beat measurement against a methane stabilized He-Ne laser.

3 Doppler-free spectroscopy of methane at 3.39 µm

To demonstrate the capabilities of the new PR-SRO design, we have performed Doppler-free spectroscopy on methane. For this purpose, the idler output of the OPO was split and sent as two counter-propagating beams (8 mm diameter) through a 2-meter long spectroscopy cell filled with methane at a pressure of 0.1 – 50 mTorr. Using the Pound-Drever-Hall frequency modulation technique (1.24 MHz modulation frequency) we obtained the Doppler-free dispersive signals shown in Figure 3.

For the measurement at 6 mTorr (Fig. 3a) the OPO idler frequency was frequency-locked (bandwidth < 100 Hz) to a methane stabilized He-Ne laser in order to suppress slow frequency drifts. The observed linewidth of 500 kHz can be attributed to a combination of pressure broadening (200 kHz) and medium-term OPO frequency jitter (200 – 400 kHz). For the measurement at 0.8 mTorr (Fig. 3b) the OPO idler frequency was phase-locked (bandwidth ~ 15 kHz) to the same He-Ne laser. The observed lineshape agrees with a theoretical model for a linewidth of 150 kHz (mostly attributable to saturation broadening).

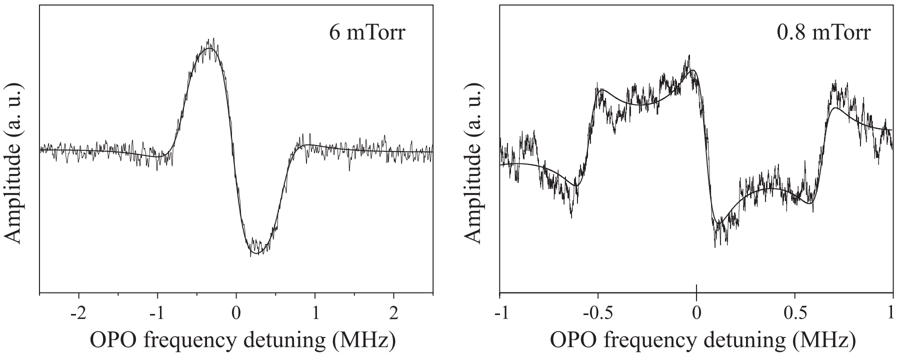

Figure 3. Doppler-free resonance of methane at 3.39 µm for two different pressures.

4 Singly resonant OPO pumped at 532 nm

While infrared pumped cw-OPOs such as the one described above are now relatively mature and are starting to become commercially available, the development of cw-OPOs with emission covering the visible spectrum is still a major challenge. After initial developments toward this goal [12-13], we have now implemented an OPO system that emits single-frequency radiation from the green to the mid-infrared spectral range, 550 – 2830 nm [14]. This is, to our knowledge, the largest tuning range of any single-frequency source reported so far. The system also exhibits excellent spectral properties such as ultra-narrow linewidth, high absolute frequency stability and a large continuous frequency tuning range.

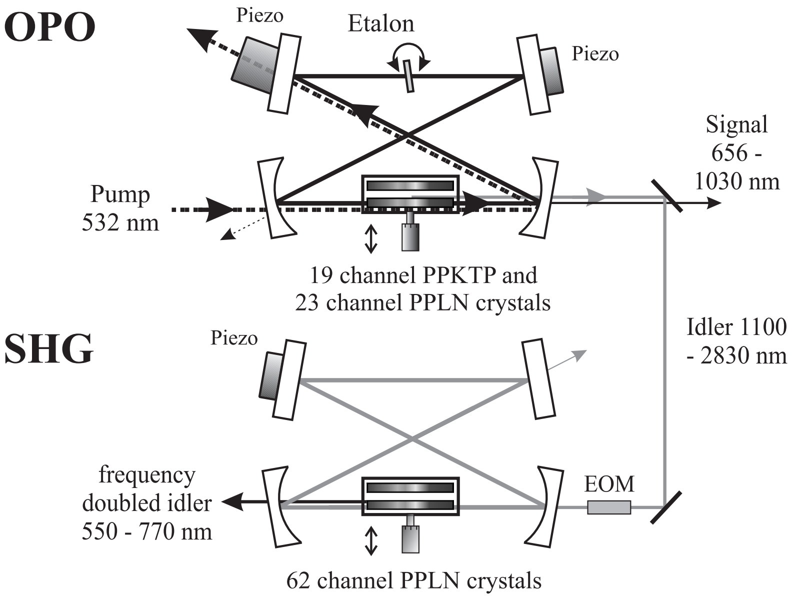

Figure 4. Setup of 532 nm pumped SRO and subsequent frequency doubler.

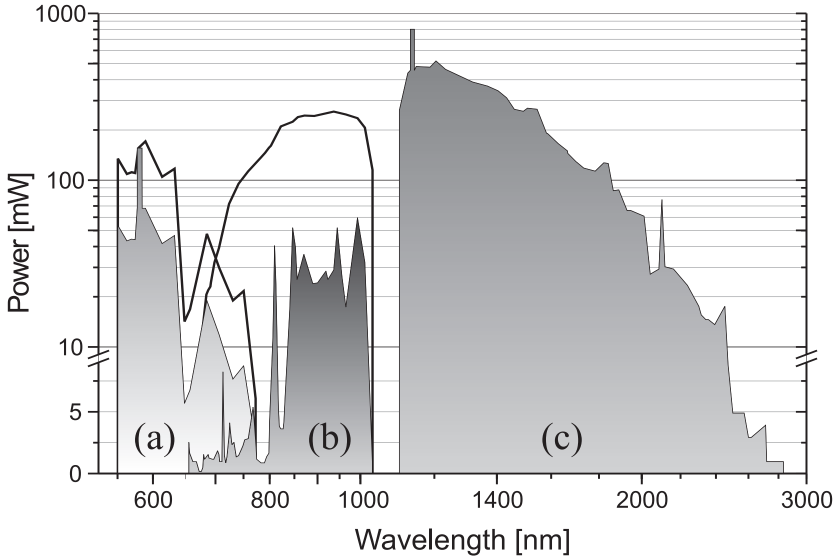

Figure 5. Output power versus wavelength for the 532 nm pumped SRO system. (a) Frequency doubled idler, (b) signal, (c) idler. The solid lines indicate the estimated performance with optimized in-coupling and out-coupling mirrors.

Figure 4 shows a schematic of the system consisting of a singly-resonant OPO (SRO) and a resonant frequency doubler for the idler wave. As nonlinear material for the SRO we use PPKTP as well as PPLN multi-grating crystals with a total of 19 and 23 different poling periods. The resonator is configured as a bow-tie shaped ring cavity for the signal and is pumped in a single pass (i.e., the pump wave is not resonated) by a commercial 10 W single-frequency solid-state laser at 532 nm (Coherent Verdi V10). The resonator was designed to tolerate strong thermal lenses with a focal length down to 4 mm. An optional intra-cavity etalon is used to prevent hops between different longitudinal resonator modes for the signal. The idler output of the OPO is frequency doubled in an external resonator using 2 PPLN crystals with a total of 62 different poling periods.

Figure 5 summarizes the emission range and output powers. The total emission range is covered by the idler (1096 – 2830 nm, up to 800 mW), the signal (656 – 1035 nm, up to 60 mW), and the frequency doubled idler (550 – 770 nm, up to 70 mW). Furthermore, up to 1.25 mW of blue light are generated by non-phasematched frequency doubling of the signal in the OPO crystal. Pump powers ranged from 0.8 to 3.3 W. Threshold powers varied from 290 mW at 830 nm to more than 2000 mW at both ends of the emission range. Signal and doubled idler powers are both expected to increase to a level well above 100 mW for optimized in-coupling and out-coupling mirrors (solid lines in Fig. 5) [14,15]. The validity of this conjecture was demonstrated by generating 160 mW of yellow light at 580 nm.

The free running OPO shows a short term linewidth of 20 kHz over 50 µs (Fig. 6b) with 5 MHz jitter over 1 s and a long term frequency drift of less than 50 MHz/h. This information was obtained from a beat measurement against a 946 nm Nd:YAG laser (Innolight Mephisto). For the linewidth measurement shown in figure 6 the OPO was frequency locked to the Nd:YAG laser with a low bandwidth.

The signal and idler can be tuned continuously (without modehops) by changing the cavity length using a piezo actuator and synchronously tilting the etalon, either controlled by a feedforward circuit or a feedback loop. Thus, maximum tuning range of 38 GHz was achieved with PPKTP as nonlinear material (16 GHz for PPLN) and an 0.5 mm etalon, limited by the walk-off losses induced by the etalon tilt.

5 Doppler-free spectroscopy of molecular iodine at 580 nm

To demonstrate the capabilities of the 532 pumped SRO system (including frequency doubler) as a versatile new light source in the visible spectral region, we have performed Doppler-free saturation spectroscopy of molecular Iodine at 580 nm [16]. For this experiment two counter-propagating beams of 1 mm diameter were overlapped inside a 100 mm long cell containing iodine at 24° C and a corresponding vapor pressure of ~ 400 mTorr. The light powers in the pump- and probe-beam were 50 and 5 mW, respectively. Figure 6a shows the resolved hyperfine structure of the molecular transition recorded by the Pound-Drever-Hall frequency modulation technique (12.7 MHz modulation frequency). Note that no pre-stabilization of the OPO to an external reference cavity was necessary to obtain this spectrum and that the OPO linewidth (Fig. 6b) is much narrower than the observed (pressure broadened to ~ 5 MHz) or even natural (~ 200 kHz) linewidth of the molecular transition.

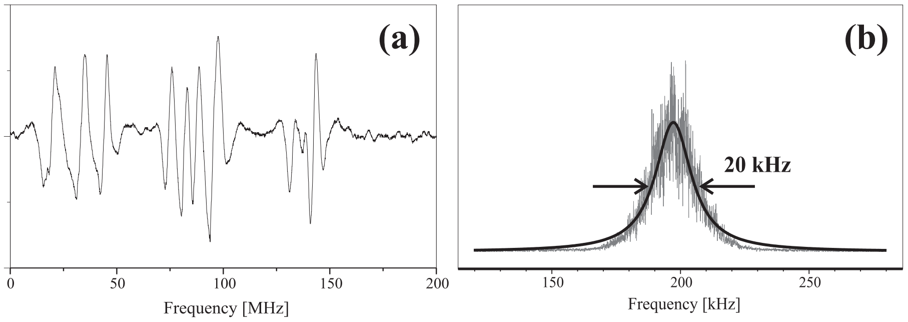

Figure 6. (a) Doppler-free resonances of molecular iodine at 579,6605 nm (line 1617) obtained by scanning the frequency doubled 532 nm pumped SRO. (b) OPO linewidth from a beat frequency measurement against a 946 nm Nd:YAG laser.

6 Summary and outlook

In conclusion, we have presented two different cw-OPO systems suitable for high resolution spectroscopy and demonstrated their capabilities by performing Doppler-free saturation spectroscopy in the mid-infrared and visible spectral region.

We have shown that reliable continuous tuning can be achieved either with a tunable or a non-tunable pump laser, by choosing the appropriate OPO type. Furthermore, the required threshold powers of typically a few hundred mW are easily obtainable using commercially available pump lasers. The already excellent spectral properties of the OPO output, narrow linewidth and small absolute frequency drift, could be enhanced even further by stabilizing the pump frequencies, improving the mechanical stability of the OPO resonator, or adding an external reference cavity (as is done in cw-Ti:Sapphire and dye lasers). For applications in optical metrology a combination with the recently developed femtosecond optical comb generators seems especially promising [17].

Future development should allow further expansion of the spectral coverage towards shorter wavelengths and a substantial increase in output power, possibly exceeding 1 W. For the latter, an continued effort in utilizing and testing new periodically poled materials as they become available is of special importance. Other improvements might include continuous tuning over several nm and a fully monolithic design for enhanced reliability and ease of use.

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