Single-frequency continuous-wave optical parametric oscillators as new tools for high resolution molecular spectroscopy

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We present a new generation of continuous-wave optical parametric oscillators (OPOs) that have been developed specifically for applications in high resolution molecular spectroscopy. As a first demonstration of their capabilities, we have successfully employed them to perform Doppler-free Spectroscopy of a ro-vibrational transition in the Methane molecule at 3.39 ?m.

Continuous-wave OPOs offer a unique combination of features that make them attractive tools for molecular spectroscopy. After considerable development efforts in the last few years, such OPOs can now provide substantial amounts (10 – 250 mW) of coherent radiation over a very wide range of infrared wavelengths (0.8 – 4.0 ?m), featuring extremely narrow line-widths (< 50 kHz), continuous tunability (several GHz), and good long-term stability (< 100 MHz frequency drift per hour, < 5% power drift per hour) [1,2].

Until recently, one of the remaining problems has been to prevent gaps in the spectral coverage and thereby to guarantee access to every molecular transition of interest. In order to solve this problem, which is generally caused by imperfections and irregularities in the properties of the nonlinear crystal at the center of the OPO, we have sacrificed some of the compactness of our previous OPO designs in favor of an extended cavity setup with a specially designed *intracavity etalon*. The setup is shown in Fig. 1. Using this configuration, we have succeeded in obtaining the desired, well defined tuning behavior at output powers of more than 20 mW.

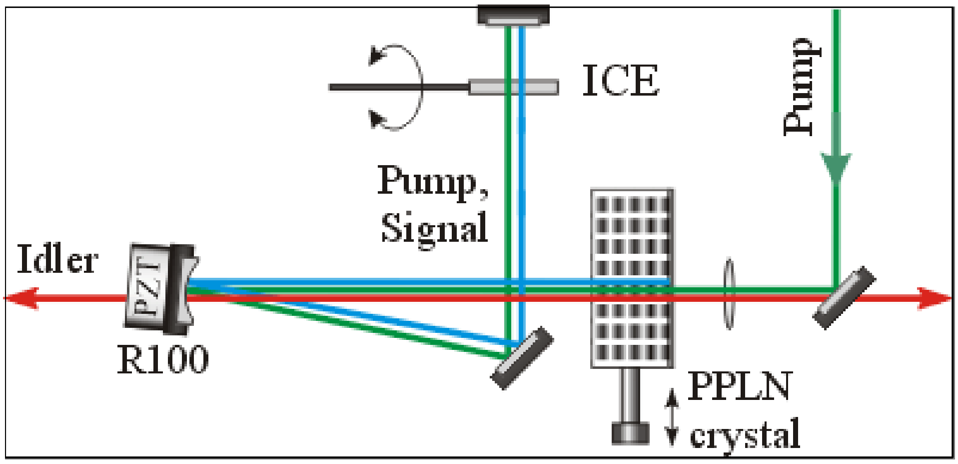

Fig. 1: The OPO is based on a periodically-poled $LiNbO_3$ crystal, pumped by a monolithic Nd-YAG laser. The pump- and the signal-wave are both resonated in the same, folded resonator, the length of which is locked to the pump laser. One mirror is directly coated onto the nonlinear crystal. The intracavity etalon should only affect the signal wave and is therefore anti-reflection coated for the pump-wave. The performance of this coating is crucial.

To demonstrate the capabilities of the new OPO design, we have performed Doppler-free spectroscopic measurements on methane. The methane molecule is an interesting candidate for a molecular frequency standard since, due to its symmetry, it exhibits low sensitivity to external field influences and low natural transition linewidth. Traditionally, the $F_2^{(2)}$ component of the P7 vibration-rotational transition (3,39 ? m) has been used as a high-stability reference line in He/Ne optical frequency standards. The use of OPOs would allow one to use transitions from low-lying vibration-rotational levels as new references which, at cryogenic temperatures, would increase the spectroscopic signal by 2-3 orders of magnitude and lead to higher performance frequency standards.

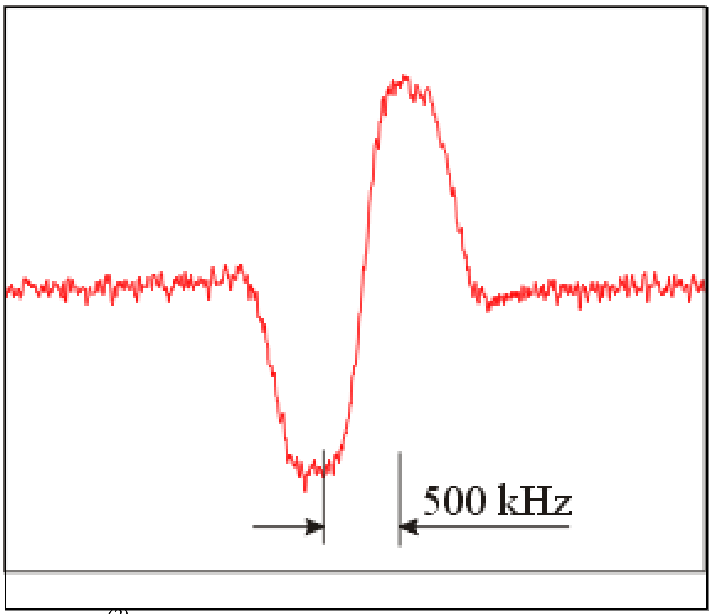

Fig. 2: Doppler-free $F_2^{(2)}$ component of the P7 ro-vibrational line of Methane at 3.39 µm.

To perform the spectroscopic measurement, the OPO output wavelength (idler) was tuned to 3.39 µm. The light was split and sent through a 2-meter long spectroscopy cell as two counter-propagating beams (pump and probe). The cell was filled with methane at a pressure of about 10 mTorr. Using the Pound-Drever-Hall frequency modulation method (1.24 MHz modulation frequency) and scanning the OPO frequency over a 5 MHz wide range, we obtained the Doppler-free dispersive signal shown in Fig. 2. The observed FWHM linewidth of 500 kHz can mostly be attributed to pressure broadening (300 kHz) and medium-term OPO frequency jitter (200-400 kHz).

In the future, we intend to improve the short term stability of the OPO by using a more rigid mechanical design and higher bandwidth lock-electronics. After these modifications we expect to replicate the linewidth (a few kHz) of our pump laser for the OPO output radiation. This would then open up the route to the spectroscopy of much narrower Doppler-free spectral lines, for example in ensembles of ultra cold molecules.

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[3] D. R. Weise, U. Strößner, A. Peters, J. Mlynek, S. Schiller, A. Arie, G. Rosenman; Opt. Comm. **184**, 329 (2000)