

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

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Abstract: We present a new continuous-wave optical parametric oscillator design specifically developed for high-resolution molecular spectroscopy. As a first application we have performed a Doppler-free measurement of a rovibrational transition in methane at 3.39 μm .

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OCIS codes: (190.4970) Parametrical oscillators and amplifiers

A new generation of continuous-wave optical parametric oscillators (OPOs) has appeared in the last few years as a coherent source that possesses an unique combination of advantages. After considerable development efforts such OPOs can now provide powerful (10 – 250 mW) coherent radiation over a very wide range of infrared wavelengths (0.8 – 4.0 μm), featuring extremely narrow line-width (< 50 kHz), continuous tunability (several GHz), and good long-term stability (< 100 MHz frequency drift per hour, < 5% power drift per hour) [1-3]. A particularly valuable feature of the cw OPOs is that their tuning range covers the mid-infrared region around 3 μm where fundamental vibrational modes of molecular bonds containing hydrogen occur.

Until recently, one of the remaining problems has been to prevent gaps in the spectral coverage of OPO 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 OPO's nonlinear crystal and optical coatings, 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 (Fig. 1). Using this configuration, we have succeeded in obtaining the desired, well defined tuning behavior of output radiation with linewidth of about 100 kHz and output power of more than 20 mW.

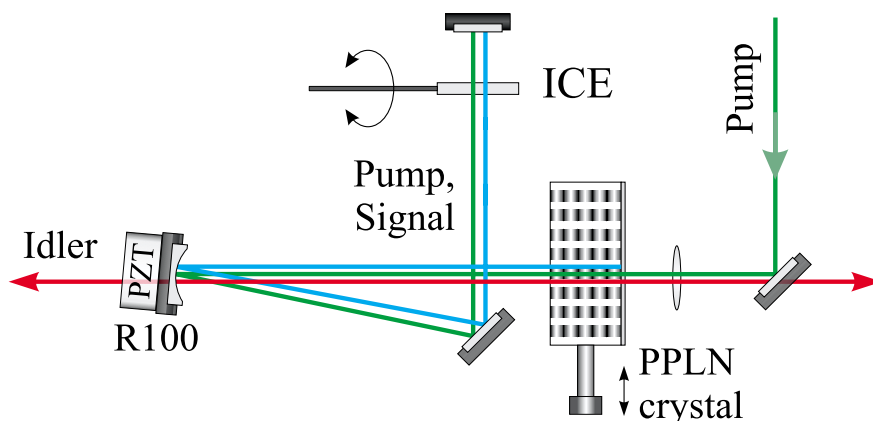


Fig. 1. The OPO is based on a periodically-poled LiNbO_3 crystal, pumped by a monolithic Nd-YAG laser. The pump and the signal wave are both resonated in the same, folded resonator, the length of which is locked to the pump laser. One mirror is directly coated onto the nonlinear crystal. The intracavity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

To demonstrate the capabilities of the new OPO design, we have performed Doppler-free spectroscopy on methane. The OPO output wavelength (idler) was tuned to 3.39 μm . The light was split and sent as two counter-propagating beams through a 2-meter long spectroscopy cell filled with methane at a pressure of about 10 mTorr. Using the Pound-Drever-Hall frequency modulation technique (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 linewidth of 500 kHz can mostly be attributed to pressure broadening and medium-term OPO frequency jitter.

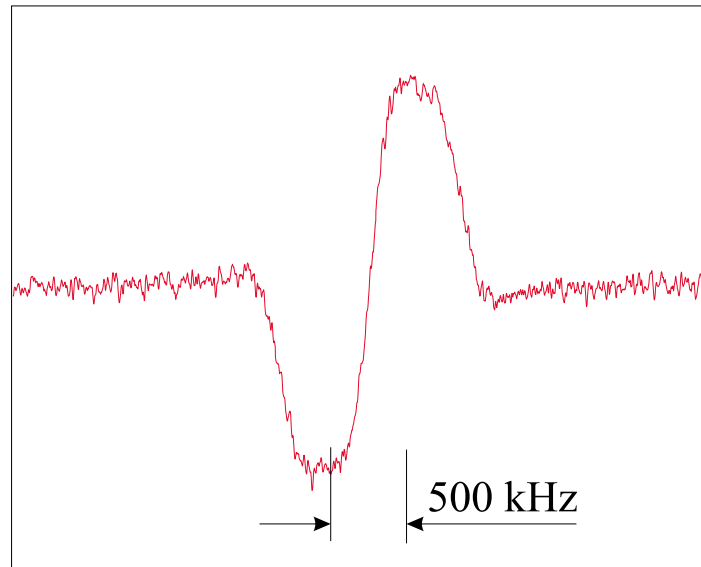


Fig.2. Doppler-free resonance of the $F_2^{(2)}$ component of the P7 rovibrational line of methane (3.39 μm).

Traditionally, the $F_2^{(2)}$ component of the P7 rovibrational transition of methane molecule (3.39 μm) 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.

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