

Continuous-wave optical parametric oscillator enable high resolution molecular spectroscopy in the infrared “stretch” region

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Molecular spectroscopy is applied to trace gas detection, combustion research, and quantum technology. Vibrational energy transitions are excited by photons in the mid-infrared, and vibrations that correspond to stretches of the molecule typically lie in the 2500 – 3700 cm-1 (2.7 – 4.0 µm) range. Due to the lack of convenient, narrow linewidth, high power sources that are widely tunable over this frequency range, TOPTICA Photonics has developed a continuous wave optical parametric oscillator. This laser source has the widest tuning range (1.45 – 4.00 µm) in the mid-infrared with the narrow linewidth needed for gas-phase molecular spectroscopy. Mature near-infrared techniques can be applied to the OPO pump and signal to perform sensitive advanced spectroscopic techniques with the idler in the mid-infrared. Here, we present absorption spectroscopy in acetylene and methane. We show linewidth measurements and significant linewidth narrowing, along with sideband modulation transferring from the seed source to the idler.

Keywords: Molecular spectroscopy, optical parametric oscillator, CH stretch, OH stretch, NH stretch, trace gas detection, combustion spectroscopy, ultracold molecules, cavity ring-down spectroscopy.



Figure 1. TOPTICA’s continuous-wave optical parametric oscillator. Pictured are the OPO head with pump, signal, and idler outputs, and the digital laser controller DLC pro. System also includes a seed laser and high-power fiber amplifier.

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Introduction

Molecular spectroscopy is an important technique for detecting molecules, for refining our understanding of them, and for better harnessing molecules for quantum technologies. The unique spectra of molecules can be used to detect the presence of that molecule. Greenhouse gas generation can be detected by spectroscopic signatures, allowing the sources to be mitigated. Molecules characteristic of explosives can be monitored in sensitive areas like airports, molecular indicators of disease can be detected in human breath, and more efficient fuels can be designed by monitoring the generation of intermediate molecules during combustion processes.

After decades of effort the vibrational energies of many molecules are well known, but fundamental research is still ongoing to understand the interactions between electronic and vibrational levels. Spectroscopy is used to determine the spectral line positions and line shapes of ultracold molecules in vacuum, allowing precision measurements of fundamental properties of our universe. A number of techniques have been demonstrated, with cavity ringdown or NICE-OHMS methods [1] allowing for highest detection sensitivities. Furthermore, vibrational states and their manipulation are a promising toolbox for applications in quantum technologies and make cold molecules a promising platform for quantum sensors and quantum computation.

Naturally, a laser source is required at the wavelengths of interest in order to perform precision spectroscopy of a target molecule. Tunable diode lasers can be used to probe energy levels (typically electronic) from the ultraviolet to the near infrared (5000cm^{-1} and higher, or up to 2 µm). Quantum cascade lasers can scan over vibrational levels between 800 - 2700 cm^{-1} (4 - 11 µm), each laser with a tuning range of approximate 100 – 200 cm^{-1}. But there remained a large gap in the availability of widely tunable laser sources with direct emission between 2500 – 4500 cm^{-1} (2.2 – 4.0 µm). Vibrations in this frequency range correspond to molecular stretches, where atoms within the molecule oscillate towards and away from each other as though on a spring. The frequencies of different stretch transitions are given in Table 1.

Absorption (cm^{-1})	Stretching Group	Compound Class
3700-3584	O-H	alcohol
3550-3200	O-H	alcohol
3500	N-H	primary amine
3400-3300	N-H	aliphatic primary amine
3350-3310	N-H	secondary amine
3300-2500	O-H	carboxylic acid
3200-2700	O-H	alcohol
3000-2800	N-H	amine salt
3333-3267	C-H	alkyne
3100-3000	C-H	alkene
3000-2840	C-H	alkane
2830-2695	C-H	aldehyde
2600-2550	S-H	thiol
3700-3584	O-H	alcohol
3550-3200	O-H	alcohol

Table 1. Frequencies corresponding to different molecular stretching transitions.

DLC TOPO

When laser sources with direct emission at a given wavelength are non-existent or show poor performance, nonlinear frequency conversion is used to fill the spectral gap. TOPTICA has decades of experience building high-efficiency, continuous-wave, frequency converted systems. These systems rely on high–power, narrow-linewidth near-infrared (NIR) fundamental lasers and resonant build-up cavities to efficiently perform second harmonic generation (SHG), conversion of two input photons into one output photon with twice the energy of the input photons. TOPTICA's SHG pro systems are known for their stability, ease-of-use, and hands-free operation, and spanning the 190 – 780 nm spectral range [2].

To generate mid-infrared radiation, TOPTICA has leveraged this experience, and uses the same nonlinear optics technology in a modified configuration. TOPTICA's continuous-wave optical parametric oscillator (OPO), the "DLC TOPO" is shown in Figs. 1,2. The TOPO is pumped at 1.06 µm (Fig. 2b) by a distributed feedback diode laser (TOPTICA's DFB pro) seeding an ytterbium fiber amplifier. In the optical parametric generation process, one pump photon is converted to two lower energy photons, the signal and the idler (Fig. 2c). The signal is the output photon with higher energy, the idler with lower. A cavity, which in the case of TOPO is resonant for the signal wave, is used here to improve efficiency and to narrow the linewidth. When a signal photon is generated, it is trapped in the cavity and passes through the nonlinear medium to stimulate conversion of additional signal (and thus idler) photons. The

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amplified signal then is amplified even further upon multiple passes through the crystal for high efficiency conversion from the pump, much like photons passing multiple times through a gain medium in a traditional laser oscillator. In this way, > 60% of the pump power can be converted to the signal and idler.

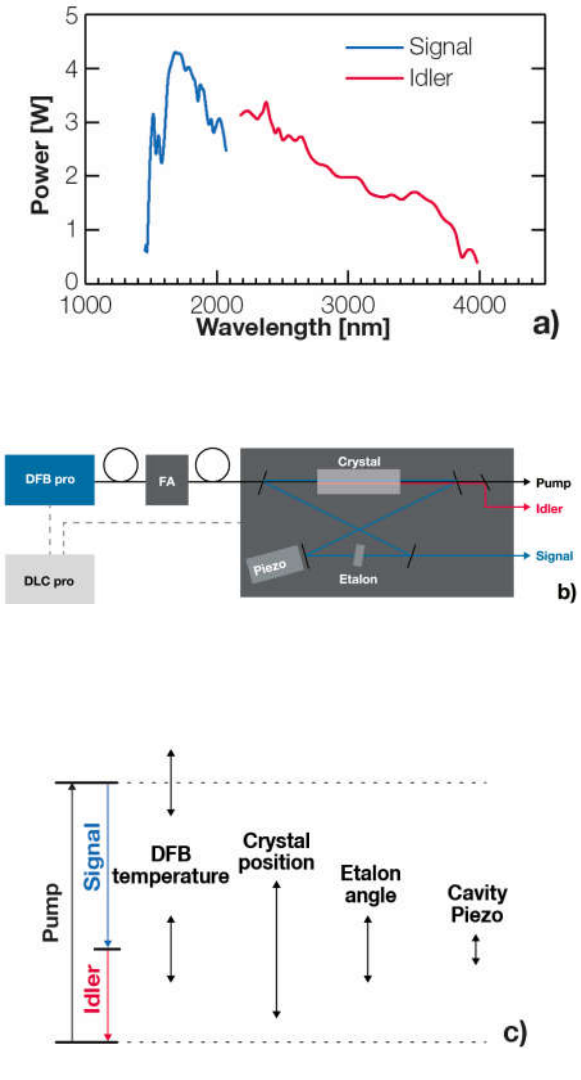


Figure 2. Diagram of TOPTICA's DLC TOPO.
(a) Characteristic tuning curve, (b) system diagram, and (c) diagram of optical parametric generation process, including influence of tuning mechanisms.

The TOPO signal is tunable between 1.45 – 2.07 µm and the idler is tunable between 2.20 – 4.00 µm. A typical curve of power as a function of output wavelength is shown in Fig. 2a. There are four major tuning mechanisms, and each can be adjusted using the system controller DLC pro, or software for remote hands-free tuning, without manual alignment or optics exchanges over the full tuning range:

- Coarse tuning is performed by motorized adjustment of the nonlinear crystal position, which changes phase-matching conditions. This tunes the signal and idler in linear steps of approximately 1 THz across the entire tuning range.
- The cavity includes an etalon to ensure oscillation on a single cavity mode. This etalon sits on a motorized rotation mount, and it can be tilted to tune the signal and idler. Combined adjustments of the crystal position and etalon tilt improve tuning resolution to < 30 GHz.
- A piezoelectric transducer on one of the cavity mirrors can be scanned for continuous tuning of the signal and idler.
- The seed laser can be tuned continuously, e.g. by tuning the DFB diode temperature, to continuously tune the idler while the signal frequency remains fixed. In this way the idler can be tuned by 30 – 300 GHz before phase-matching conditions force the signal to hop to a new cavity mode.

To reach a target wavelength, each of these actuators can be adjusted individually and iterated while the user monitors the output wavelength This is a deterministic tuning method, and all tuning actuators are controlled by system software. Such, TOPTICA has developed software for automatic tuning and spectroscopy with feedback from a wavelength meter. A lookup table of the coarse tuning actuators is generated first, and then the user can input start wavelength, stop wavelength, and step size for a spectroscopy scan. The system will refer to the wavelength meter as it tunes the system, stitching together individual mode hop-free regions to generate wide, high resolution continuous idler frequency scans.

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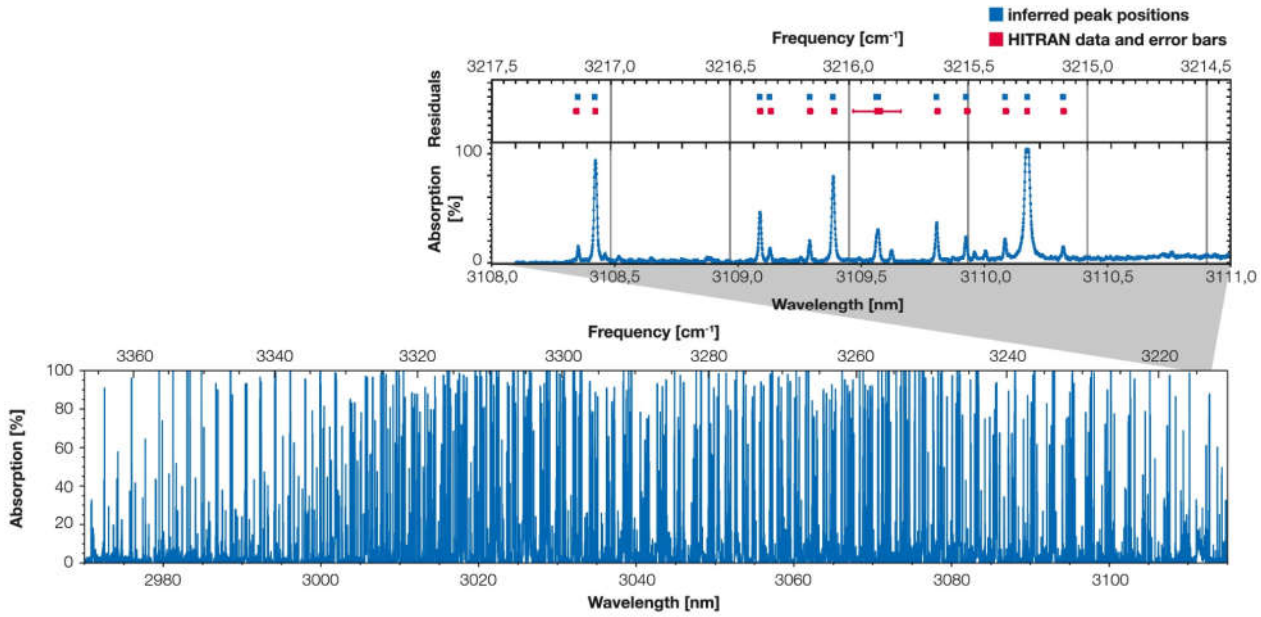


Figure 3. Absorption spectroscopy of C-H stretching in acetylene and methane.

DLC TOPO and Molecular Spectroscopy

To demonstrate use of the TOPO, a molecular spectrum was obtained by automatically tuning the system and passing the idler beam through a 20 cm gas cell filled with 10 Tor C_2H_2 and 10 Torr CH_4 at room temperature. Power transmitted through the cell was measured using a HgCdTe photodetector, and the resulting absorption spectrum is plotted in Fig. 3. The scan covers a frequency range of 3210 – 3365 cm^{-1} with a step size of $3x10^{-3}$ cm^{-1} given by the wavelength meter resolution[1]. The system has a mode hop-free tuning range of 1 – 10 cm^{-1}, so this scan demonstrates automatic stitching of > 10 mode hop-free regions.

Highly sensitive spectroscopy methods like cavity ring-down spectroscopy and frequency modulation spectroscopy may require a source with even narrower linewidth than the standard TOPO, or fast laser frequency modulation or sideband generation. The TOPO can take advantage of readily available TOPTICA toolbox of tunable narrow-linewidth seed sources and frequency locking solutions in the near infrared, and transfer the performance to the mid-infrared using the optical parametric process.

In order to demonstrate the linewidth and modulation capabilities of TOPO, we performed a beat frequency measurement[2] where

- we beat the pump and signal waves of one TOPO system with a TOPTICA DFC CORE + frequency comb,
- and we beat the idler waves of two independent TOPO systems.

While the standard DFB seed laser has a linewidth of < 2 MHz[3], the resonant parametric process yields a spectral purification similar to laser action in single frequency laser cavities, which can be pumped by an incoherent source. Such, the signal linewidth is on the order of 10s of kHz, see Fig. 4a.

Because the optical parametric process is entirely coherent, the idler linewidth is a convolution of pump and signal linewidths. Because of the signal being much narrower than the DFB seed laser, the seed frequency noise is essentially converted to the idler. Hence, the idler approximately matches the < 2 MHz DFB linewidth in the standard TOPO system. This is demonstrated in Fig. 4a.

[1] The scan is continuous, while the wavelength meter has a finite resolution.

[2] We note that in each beatnote we measure the convolution of frequency noise of both sources taking part in the experiment (DFC CORE + or TOPO). Such, any linewidth information obtained from the beatnote shall be considered an upper limit for the respective linewidth (or frequency noise contribution from the TOPO system).

[3] Yet another unit! For convenient conversion between cm^{-1}, µm, MHz, use TopticCalc: https://www.toptica.com/products/optical-isolators-photonicals/topticalc/

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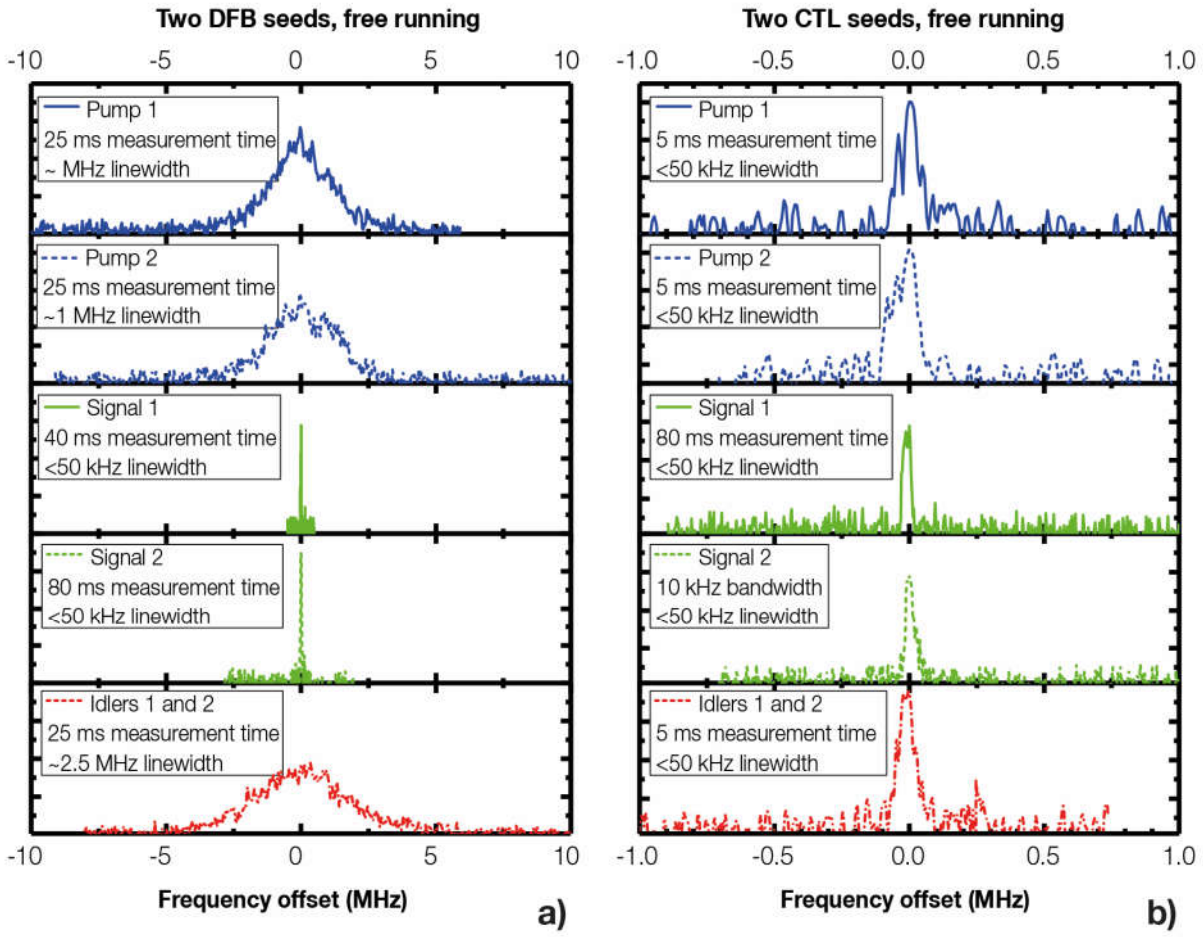


Figure 4. Linewidth measurements of TOPO pump, signal, and idler. To measure pump and signal linewidths, each output is beat against TOPTICA's DFC CORE + frequency comb, which has 20 kHz tooth linewidth. The idler linewidth is measured by beating the idlers of two TOPOs together. In this case, the measured beat linewidth is approximately a factor of $\sqrt{2}$ broader than the idler linewidth. (a) Pump and signal linewidths of two DFB-seeded TOPOs, and beat width of their idlers. (b) Pump and signal linewidths of two CTL-seeded TOPOs, and beat width of their idlers (notice the 10-fold reduction of the frequency scale with respect to the DFB seeds in (a)).

In addition to DFB diode laser systems, TOPTICA manufactures the CTL 1050, a near infrared external-cavity diode laser that has wide continuous tunability near 1.06 µm and narrower linewidth (<10 kHz) than the DFB laser (< 2 MHz). Using CTL 1050 as an alternative seed source for the TOPO, in Fig. 4b) we demonstrate a significant reduction of idler the linewidth, down into the 10- kHz range.

Similarly, a seed laser with frequency sidebands, when convoluted with the narrow linewidth signal, will yield an idler with frequency sidebands of the same spacing. This capability finds its application in Pound-Drever-Hall error signal generation and frequency-locking to mid-infrared resonant optical cavites as required for cavity ringdown or NICE-OHMS spectroscopy applications. For the demonstration of modulation of the seed radiation and its transfer to the mid-infrared idler output, sidebands were generated via direct diode current modulation on the seed laser of one of the two TOPOs in the experiment. This was performed with both DFB and CTL seed laser configurations. The resulting idler beatnotes are plotted in Fig. 5a) and 5b), and it is evident that the sidebands are present on the idlers.

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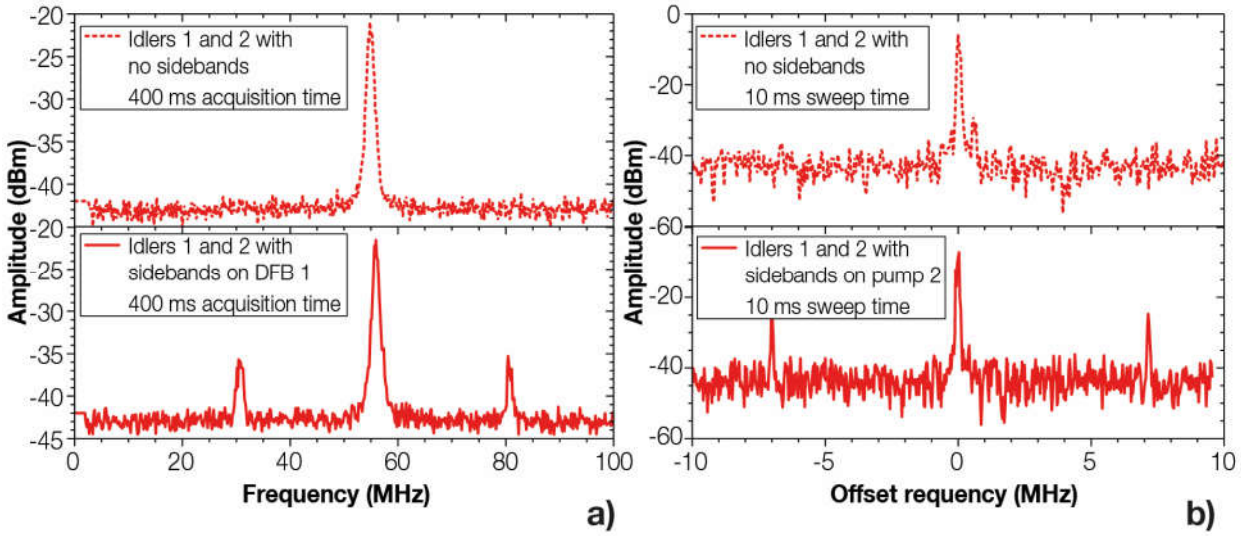


Figure 5: (a) DFB-seeded idler beat measurements with and without sidebands on the DFB seed of one TOPO. (b) CTL-seeded idler beat measurements with and without sidebands on the CTL seed of one TOPO.

TOPTICA's DLC TOPO has the widest spectral coverage amongst tunable, narrow linewidth CW laser sources in the mid-infrared. It fills the critical gap in wavelength coverage between near infrared diode lasers and mid- to far-infrared quantum cascade lasers, providing a source for spectroscopy in the molecular stretch region. It has automatic, hands-free tuning for easy integration in experiments. Sidebands can be generated for frequency modulation spectroscopy and for locking to cavities for ring down spectroscopy. TOPTICA can also provide locking solutions for stabilizing the idler to cavities, and combinations similar to the DFC CORE + and TOPO presented here have been demonstrated to provide even narrower linewidths. Ultimately, phase coherent links between distant ranges in the electromagnetic spectrum have been demonstrated [3], opening new pathways for precision metrology and quantum sensing. Reach out to TOPTICA's team to discuss how to integrate TOPO into your experiment!

References

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