Mid-infrared wavelength- and frequency-modulation spectroscopy with a pump-modulated singly-resonant optical parametric oscillator

I.D. Lindsay, P. Groß, C.J. Lee, B. Adhimoolam and K.-J. Boller

Laser Physics and Nonlinear Optics Group, Faculty of Science and Technology, University of Twente,
P.O. Box 217, 7500 AE Enschede, The Netherlands
i.d.lindsay@tnw.utwente.nl

Abstract: We describe the implementation of the wavelength- and frequency-modulation spectroscopy techniques using a singly-resonant optical parametric oscillator (OPO) pumped by a fiber-amplified diode laser. Frequency modulation of the diode laser was transferred to the OPO's mid-infrared idler output, avoiding the need for external modulation devices. This approach thus provides a means of implementing these important techniques with powerful, widely tunable, mid-infrared sources while retaining the simple, flexible modulation properties of diode lasers.

OCIS codes: (190.4970) Parametric oscillators and amplifiers; (140.3510) Lasers, fiber; (140.3600) Lasers, tunable; (300.6380) Spectroscopy, modulation.

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1. Introduction

Frequency modulation techniques are widely used in laser spectroscopy to achieve a high signal-to-noise ratio and, therefore, sensitive spectroscopic detection. These techniques exploit the fact that, on passage through a medium having frequency-dependent absorption, frequency-modulation of a laser results in a transmitted power variation at the modulation frequency and its harmonics. Phase-sensitive detection at frequencies beyond the range of technical noise sources is thus possible. In practice, these techniques are typically classified into two approaches [1,2]. Wavelength-modulation spectroscopy (WMS) conventionally describes the case where the modulation frequency is much less than the width of the spectral feature of interest and the modulation index is high. In practice this typically corresponds to modulation frequencies from a few kilohertz to a few megahertz. In the case usually termed frequency-modulation spectroscopy (FMS) the modulation frequency is comparable to, or greater than, the spectral width of the target feature and the modulation index is sufficiently low that only the first two sidebands of the modulated laser spectrum have significant amplitude. In this case, modulation frequencies are typically in excess of 100 MHz.

Both techniques have been most widely applied in diode-laser spectroscopy due to the ease with which diode lasers can be frequency modulated via their injection current. In contrast to external electro-optic modulators required by other laser types, current modulation of diode lasers can be achieved over broad bandwidths, extending to several gigahertz, with minimal RF power requirements and simple control of the modulation index over a wide range. This flexibility allows the same laser system to be easily reconfigured for different FM techniques [2]. Many variations on these techniques have been demonstrated with diode lasers including two-tone FMS [2], high modulation index WMS [3] and photo-acoustic WMS [4]. To access fundamental molecular vibrational bands in the mid-infrared (mid-IR) FM spectroscopic techniques have been demonstrated with lead salt diode lasers [1], and, more recently, quantum cascade lasers [5], which offer similar advantages to diode lasers in terms of ease of modulation. Mid-IR WMS with milliwatt-level powers has also been demonstrated by difference frequency generation (DFG) between amplified near-IR diode lasers, whose modulation then transfers to the mid-IR output [6].

Advances in nonlinear optical materials and pump lasers have made continuous-wave singly-resonant optical parametric oscillators (OPOs) attractive sources for mid-IR spectroscopy. These devices can produce watt-level output powers, far exceeding those of other mid-IR sources, while having tuning ranges of hundreds of wavenumbers [7]. Combining these attributes with the benefits of WMS and FMS techniques would, therefore, be highly attractive. Hybrid wavelength-amplitude modulation has been used with a pulsed OPO for systematic background cancellation in photoacoustic spectroscopy [8]. However, the intrinsically low modulation frequency (30Hz) would preclude many of the signal to noise advantages WMS usually offers. WMS and FMS in the conventional sense, as described above, appear never to have been demonstrated with an OPO source.

Recently, we have shown that pump-tuned singly-resonant OPOs represent particularly attractive spectroscopic sources [9]. In this case, the resonant signal wave remains fixed in frequency and tuning of the pump is transferred directly to the mid-IR idler output. This approach allows rapid tuning over hundreds of wavenumbers [10], wide-range continuous tuning [9], and mid-IR tuning with narrow linewidth [11]. In this paper, we extend this approach to include transfer to the idler of pump laser modulation and use this to demonstrate mid-IR WMS and FMS detection. Use of a diode laser-based pump source allows this to be achieved while retaining simplicity of modulation and avoiding the requirement for an external modulator in the mid-IR. We believe this to be the first reported demonstration of the WMS and FMS detection techniques with an OPO.

2. Experimental arrangement

The optical configuration used for both WMS and FMS investigations is shown schematically in Fig. 1. The pump source consisted of a commercial multi-section DBR diode laser seeding

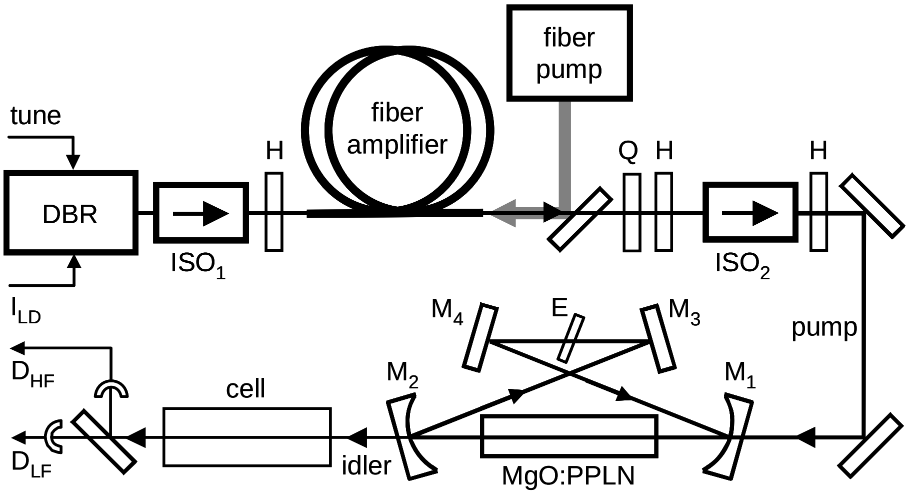

Fig. 1. Optical configuration for WMS and FMS experiments. DBR: multi-section DBR diode laser, ISO_1: 60dB isolator, ISO_2: 30dB isolator, Q and H: quarter- and half-wave plates, $M_{1\text{-}4}$: OPO cavity mirrors, E: intracavity etalon, tune: tuning input to DBR laser, I_{LD}: DBR laser injection current supply, D_{HF}: fast mid-IR detector output, D_{LF}: slow mid-IR detector output. Electrical connections relate to instrumental configuration shown in Fig. 2.

a fiber amplifier. The seed laser (Eagleyard Photonics) produced up to 100mW of output power at around 1063nm with a linewidth of 30MHz. Following beam shaping and 60dB isolation, up to 60mW of output was available to seed the fiber amplifier. This amplifier was based on a commercial ytterbium-doped, double-clad, large-mode-area fiber (Liekki YB1200-20/400DC) having a core of 20μm diameter with an NA of 0.07 surrounded by a 400μm diameter inner cladding with an NA of 0.46. The 4.5m length resulted in 95% pump absorption at 976nm. The fiber was pumped from the output end, with an estimated coupling efficiency of around 80% into the inner cladding, by a fiber-coupled, 30W, 976nm diode bar. Both end facets of the fiber were polished to an 8^o angle to avoid unwanted feedback. Single-mode output, with $M^2 < 1.1$, was achieved by coiling the fiber with a diameter of 7.5cm [12], and optimizing seed overlap with the fundamental mode. In contrast to our previously reported work [9], modulation-induced spectral broadening of the seed laser was not required for stable operation. The seed modulation parameters could, therefore, be chosen arbitrarily. Up to 12W of polarized output were produced after isolation for as little as 30mW of seed with over 99.5% of the output power concentrated at the seed wavelength.

The singly-resonant OPO, was based on a 50mm long crystal of 5% MgO-doped periodically-poled $LiNbO_3$ (MgO:PPLN), having grating periods ranging from 28.5 to 31.5μm. This was placed in a bow-tie ring resonator, similar to that described in [9], formed by two 50 mm radius mirrors (M_1, M_2) and two plane mirrors (M_3, M_4), resonant for the signal wave only. The temperature of the MgO:PPLN crystal mount was stabilized at around 50^oC. To ensure signal resonance on a single longitudinal mode of the cavity, a 200GHz FSR solid etalon with 22% reflectivity coatings was inserted between M_3 and M_4. In operation, the OPO exhibited a threshold of 2.5-3.5W, depending on wavelength, and up to 900mW of idler output was obtained for 8W pump power. At a temperature of 48^oC, operation was observed for four poling periods from 29.5μm to 31.5μm spanning an idler wavelength range of 3.15-3.8μm. Coarse (discontinuous) tuning of the DBR laser over its full 3nm tuning range, resulted in idler tuning of between 20nm and 30nm for each poling period. Full coverage between poling periods could be achieved by temperature tuning of the MgO:PPLN crystal.

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As previously reported [9], continuous tuning ranges in excess of 100GHz could be achieved on millisecond timescales with high linearity by appropriate control of the DBR laser.

The idler output of the OPO was passed through a 90cm long absorption cell with 3^o wedged CaF_2 windows and containing 0.77mbar of CH_4 buffered to 52.3mbar in pure nitrogen. Pressure broadened FWHM values for the principle CH_4 absorption lines in the spectral region investigated were estimated at around 150MHz from HITRAN data [13]. Doppler broadening was calculated to result in FWHM values of 300MHz and was thus the dominant broadening mechanism. The transmitted beam was split using a CaF_2 wedge with a 50% reflective aluminium coating on the front surface. Half the light was directed onto a high-speed photoconductive detector (Vigo System S.A, type R005-3 with high-speed amplifier) having a bandwidth >150MHz. This was used to monitor the modulated absorption signal. The remaining light was incident on a low frequency mid-IR photodiode (IBSG) having a bandwidth of approximately 0.7 MHz. This detector, with further low-pass filtering to block the modulation frequencies, was used to monitor the direct absorption signals.

Figure 2. shows the instrumental configuration for both WMS and FMS detection. In both cases a ramp signal applied to the tuning controller of the DBR diode laser provided linear tuning over the required spectral range. The transmission of the idler through the cell, was recorded by the slow photodiode (D_{LF}), as the DBR laser frequency was swept. In the case of WMS, shown in Fig. 2a, modulation was applied to the DBR laser via the injection current driver (Thorlabs type LDC205) which had a bandwidth of 150kHz. A lock-in amplifier (Stanford Research Systems SR530) was used to demodulate the signal from the fast detector, D_{HF}. The demodulated signal was recorded simultaneously with the direct absorption signal on a digital storage oscilloscope. The configuration for FMS (Fig. 2b.) was broadly similar except that modulation was applied to the DBR laser via a bias-tee unit (Minicircuits ZFBT-4R2GW) and demodulation was carried out by mixing the signal from D_{HF} with the local oscillator in a double-balanced mixer (Minicircuits ZFM-3) with the high-frequency components coupled to ground via a small capacitor.

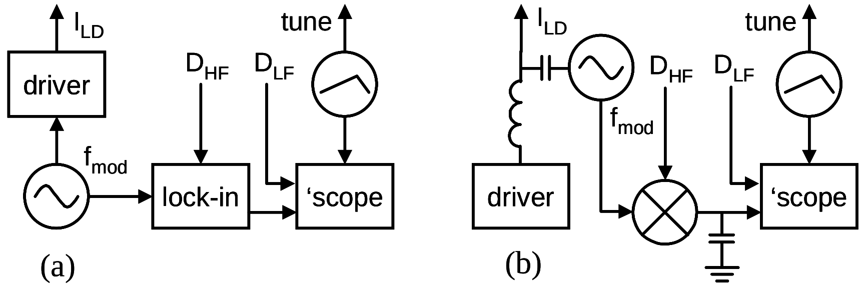

Fig. 2. Instrumental configuration used for (a) WMS and (b) FMS detection. I_{LD}: Injection current to DBR laser, tune: ramp signal to DBR tuning controller, D_{HF}: fast mid-IR detector signal, D_{LF}: slow mid-IR detector signal, driver: DBR laser injection current supply, ‘scope: digital storage oscilloscope.

3. WMS and FMS detection

To investigate application of the OPO system to the WMS and FMS detection techniques, the tuning parameters of the DBR laser were adjusted to repeatedly tune the OPO idler continuously over a 30GHz range containing a number of absorption features. Wavemeter measurements of the pump and signal allowed determination of the absolute idler frequency, enabling comparison of the positions of spectral features with standard data. For WMS detection, 50kHz modulation was applied to the DBR laser with an amplitude adjusted to give a frequency excursion of approximately ±150MHz, corresponding to the calculated Doppler half-width of the spectral features to be detected. The time constant of the lock-in amplifier

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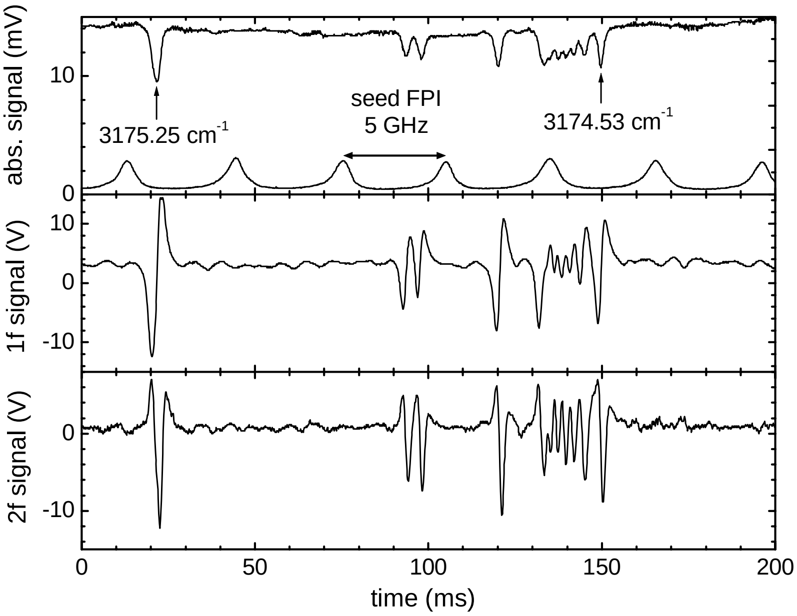

Fig. 3. WMS spectra demodulated at the 50kHz modulation frequency (1f) and its second harmonic (2f). Also shown is the simultaneously recorded direct absorption spectrum (upper plot, top trace) and DBR seed laser transmission through a reference Fabry-Perot interferometer (upper plot, lower trace). Line assignments indicated on the direct absorption spectrum were made from the HITRAN database.

was set to 1ms and the tuning rate of the DBR laser was set to 0.17 GHz/ms, above which noticeable distortion of the WMS signal occurred. Spectra demodulated at both the modulation frequency (1f) and its second harmonic (2f) were recorded. The results are shown in Fig. 3. It can be seen that the 1f and 2f spectra show, respectively, the first and second derivatives of the features observed in the direct absorption spectrum, confirming successful transfer of the DBR laser modulation to the OPO idler and resulting WMS detection.

To demonstrate FMS detection a modulation frequency of 153MHz was used, again chosen to correspond approximately to the Doppler half width of the observed absorption features. The precise frequency was arrived at by adjustment to achieve the correct phase relationship between the detected FM signal and the local oscillator at the mixer to yield the in-phase demodulated signal, corresponding to absorption, rather than the in-quadrature signal, corresponding to dispersion [14]. In practice, it was found that modulation frequencies up to 200MHz could be used, limited by the bandwidth of detector D_{HF}. The modulation amplitude was adjusted to give a DBR laser spectral distribution having two sidebands, each containing around 5% of the total power, as observed using a high-finesse Fabry-Perot interferometer. Under these conditions –10dBm was supplied by the RF signal generator, in total, to both modulate the DBR laser and act as a local oscillator for demodulation.

The results are shown in Fig. 4. Again, it can be seen that a signal corresponding to the first derivative of the direct absorption spectrum was acquired, confirming that modulation of the DBR laser was transferred to the OPO idler allowing successful FMS detection. The calculated cell transmission based on Doppler-broadened HITRAN data [13], is also shown in Fig. 4. The excellent correspondence of line positions with the experimental data confirms the high degree of linearity previously noted for OPO tuning by this approach [9]. It should also be noted that the FMS approach allowed a tuning rate approximately 20 times higher than that used for WMS, which was limited by the 1ms time constant of the lock-in amplifier.

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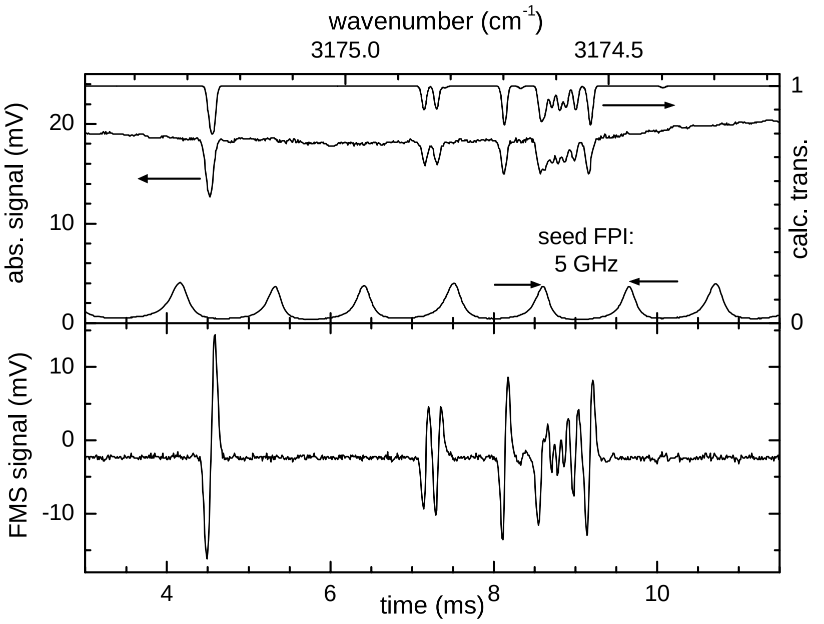

Fig. 4. FMS spectrum acquired with f_{mod}=153MHz. Also shown is the simultaneously recorded direct absorption spectrum (upper plot, center trace) and DBR seed laser transmission through a reference Fabry-Perot interferometer (upper plot, lower trace). The calculated cell transmission, based on HITRAN data, is shown in the upper plot against the frequency scale indicated on the top x-axis.

4. Summary and conclusions

The results shown in Figs. 3 and 4 clearly demonstrate that a singly resonant OPO pumped by a fiber-amplified diode laser can be used to implement WMS and FMS detection in the mid-IR simply by appropriate modulation of the near-IR diode laser. The approach described provides a means to implement these important spectroscopic techniques with powerful, widely tunable mid-IR sources. At the same time, modulation via the diode-based pump source, without further inputs to the OPO itself or the use of external modulators, retains all the advantages that make diode lasers attractive sources for implementing these spectroscopic techniques. We believe the results presented here to be the first demonstration of the WMS and FMS techniques with an OPO.

As the results here demonstrate, a major advantage of the modulation approach used is the simplicity with which the system can be reconfigured to address different modulation regimes. As a result, we believe this approach offers a simple route to implement, with high mid-IR powers, any of the wide range of variations on the WMS and FMS techniques previously demonstrated with diode lasers. Of these, two-tone FMS is of particular interest as it offers the means to carry out FMS at the gigahertz modulation frequencies compatible with pressure-broadened absorption features while avoiding the need for high-bandwidth detectors [2], which have highly limited availability in the mid-IR. The approach described would also be highly attractive for use in photo-acoustic variations of WMS [4], where the high mid-IR powers available from the OPO would provide significant improvements in sensitivity.

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