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A pulsed optical parametric oscillator, based on periodically poled lithium niobate (PPLN), for high-resolution spectroscopy

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Abstract. We report a pulsed optical parametric oscillator (OPO), incorporating periodically poled lithium niobate (PPLN) and injection-seeded by a tunable diode laser (TDL). This is combined with an optical parametric amplifier (OPA) based on bulk lithium niobate (LNB) to form a modular, high-performance spectroscopic system. The instrument employs a novel intensity-dip control scheme to lock the length of the OPO ring cavity to the continuous-wave seed radiation from a 1.55-µm single-mode TDL. The coherent infrared signal and idler outputs of this TDL-seeded PPLN OPO/LNB OPA system are continuously tunable in the 1.55-µm and 3.4-µm regions, where their utility for high-resolution absorption and coherent Raman spectroscopic measurements are demonstrated by investigations of methane, in the gas phase and in a supersonic free jet. The output radiation has excellent beam quality and its optical bandwidth is shown to be 130 ± 10 MHz ($0.0043 \pm 0.0003\,cm^{-1}$), approaching the limit imposed by the Fourier transform of the pulse duration.

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Optical parametric oscillator (OPO) and amplifier (OPA) devices are widely recognised as versatile coherent tunable spectroscopic sources [1]. In a recent communication [2], we reported spectroscopic implementation of a pulsed OPO based on bulk lithium niobate (LNB). The tunability and optical bandwidth of the output radiation were controlled by injection seeding with a single-mode tunable diode laser (TDL), with a slightly misaligned passive ring cavity providing a low effective finesse for continuous tuning purposes, as in our previous work on OPOs based on β-barium borate (BBO) [3–6].

We now report a further advance that takes advantage of quasi-phase-matched (QPM) operation in periodically poled lithium niobate (PPLN) [7]. QPM devices offer higher nonlinear-optical coefficients and smaller size than birefringently phase-matched materials. As illustrated in Fig. 1, our new modular high-performance spectroscopic system

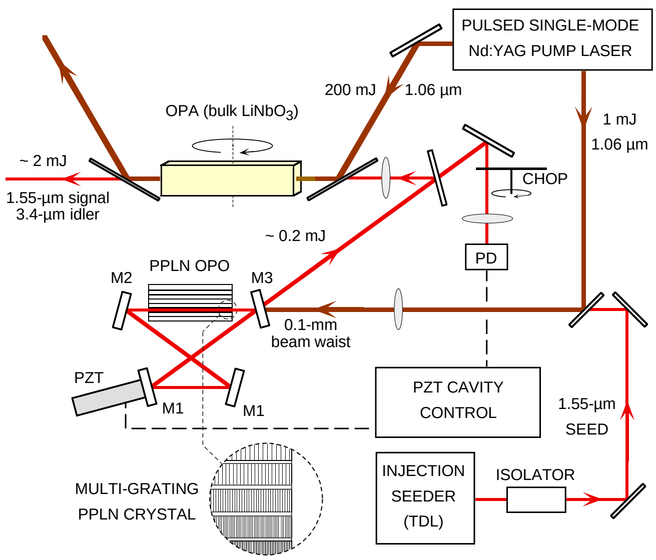

Fig. 1. Modular schematic of a narrowband, tunable PPLN OPO/LNB OPA spectroscopic system, pumped at 1.064 µm by a pulsed, single-mode Nd:YAG laser. The actively controlled PPLN OPO ring cavity is injection-seeded at its (resonant) signal wavelength of ∼ 1.55 µm by a 5-mW cw single-mode external-cavity diode laser (TDL). The output of the multiple-grating PPLN crystal (see inset) can be coarsely tuned by vertical positioning and/or temperature variation. The OPO output is amplified in an OPA stage based on bulk $LiNbO_3$. The corresponding idler output is at ∼ 3.4 µm. See text for further details

comprises a TDL-seeded PPLN OPO with an actively controlled cavity, *plus* a bulk-LNB OPA stage for some applications. The system is pumped (by a pulsed Nd:YAG laser) at 1.064 µm, injection-seeded (by a cw TDL) at ∼ 1.55 µm, and generates coherent narrowband infrared (IR) signal and idler outputs that are continuously tunable in the vicinities of 1.55 µm and 3.4 µm. These output wavelength regions coincide respectively with prominent first-overtone and fundamental absorption bands associated with bond-stretching (e.g., C−H) vibrations of hydride molecules (e.g., hydrocarbons). It is also possible to record coherent Raman spectra (e.g., of methane, CH_4 [2]) at the Raman-active difference frequency between the 1.064-µm pump and ∼ 1.55-µm sig-

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nal radiation. Our spectroscopic experiments with output from this pulsed PPLN OPO demonstrate a remarkably narrow optical bandwidth of 135 MHz (0.0045 cm^{-1}). This offers fresh prospects for high-resolution, time-resolved laser spectroscopy, particularly in molecular-beam environments or in gases at low pressure and temperature.

1 Instrumental development

The modular design of our PPLN OPO/LNB OPA spectroscopic system is depicted in Fig. 1. The PPLN OPO cavity is in the form of a bow-tie ring, constructed from tiny optical mounts; its optical path length of ∼ 12 cm corresponds to a free spectral range (FSR) of ∼ 2.5 GHz (∼ 0.08 cm^{-1}). It comprises four reflectors, two (M1) flat and two (M2 and M3) with 100-mm radius of curvature, and is resonant at the signal wavelength of ∼ 1.55 μm, with a 50% transmitting output coupler (M3). Round-trip losses at the idler and pump wavelengths (∼ 3.4 μm and 1.064 μm, respectively) are > 99.95%.

The PPLN crystal (supplied by Crystal Technologies Inc) has dimensions of 19 mm × 11.0 mm × 0.5 mm (length × breadth × thickness) and sits on a resistively heated fixed-temperature mount (settable within the range 20–80 °C). It is of multiple-grating form [8] with eight different grating periods ranging from 28.5 to 29.9 μm in steps of 0.2 μm, each grating stripe having a breadth of ∼ 1 mm. The ends of the PPLN crystal are uncoated, but their surfaces are wedge-polished at ∼ 2° relative to the long axis of the gratings, to reduce unwanted reflections in the cavity. The vertical position of the PPLN crystal (on its fixed-temperature mount) is finely adjustable by a precision translation stage, to enable selection of different grating periods for coarse QPM tuning purposes. Further tunability is attainable by varying the temperature setting of the PPLN crystal. This combination of the eight PPLN grating periods and temperature variation over ∼ 50 °C provides uninterrupted QPM tuning ranges of 1.45–1.55 μm for the signal and 4.0–3.4 μm for the idler.

For high-power applications, the 1.55-μm signal-wave output from the TDL-seeded PPLN OPO is coupled into an OPA pumped at 1.064 μm. This incorporates a LNB crystal (11 × 11 × 50 mm, cut at 47° for type-I phase matching in the vertical plane, broadband anti-reflection coated) as in our previously reported LNB OPO [2].

Pump radiation at 1.064 μm is generated in ∼ 10-ns pulses by a Q-switched, single-mode Nd:YAG laser (Spectra-Physics GCR-250, with model 6350 injection seeder) in a high-quality beam (> 85% fit-to-gaussian, 9 mm diameter, ∼ 45 MHz optical bandwidth), of which ∼ 200 mJ per pulse is typically used to pump the LNB OPA. Only a small portion of the pump beam (∼ 1 mJ per pulse) is needed to drive the PPLN OPO; this is focused by a 30-cm lens and coupled through mirror M3 into the cavity (along with collinear 1.55-μm seed radiation), forming a beam waist of ∼ 100 μm within the PPLN crystal. This corresponds to operation at 3–4 times the (unseeded) oscillation threshold for the PPLN OPO. The LNB OPA pump beam is vertically polarised, but all other input and output radiation is horizontally polarised.

The injection-seeding source is a single-mode external-cavity TDL (New Focus 6262 with model 6200 controller) operating in the range 1.50–1.59 μm and continuously tunable over an interval of ∼ 2 cm^{-1} (60 GHz) in fine scan control mode and over the entire range (∼ 350 cm^{-1}) under coarse scan control. The TDL delivers ∼ 5 mW of narrowband cw seed radiation to the PPLN OPO at its signal wavelength, via an optical isolator.

Active control of the length of the PPLN OPO ring cavity is necessary to avoid hopping between adjacent longitudinal modes as the narrowband seed wavelength is continuously scanned. This is because the fine beam waists of pump, signal and idler radiation in the PPLN crystal constrain the OPO to operate on a single transverse mode (nominally TEM_{00}) that is unable to resonate at certain seed wavelengths if the cavity length is held fixed. (This contrasts with the situation in TDL-seeded OPOs based on bulk nonlinear-optical media such as LNB [2] or BBO [3–5], where convenient continuous narrowband tunability can be achieved passively by using a slightly misaligned OPO cavity with a low effective finesse that enables slightly non-collinear phase-matching and operation on a variety of transverse modes.) The length of the PPLN OPO ring cavity is actively controlled by mounting one of the flat mirrors (M1) on a piezoelectric transducer (PZT) and locking the cavity length to an intensity dip arising from interference between seed beams reflected from mirror M3 and circulated in the OPO cavity. The combination of reflected and circulated cw seed beams is monitored by a germanium photodiode, which is protected from potentially damaging pulsed signal radiation by a mechanical shutter synchronised to the firing cycle of the pump laser. Further details of this novel cavity-control scheme will be reported elsewhere [9]. It is similar in some respects to previously reported cavity-locking schemes for narrowband pulsed OPOs [10–13], but is intrinsically simpler and/or does not require dithering of OPO cavity length.

Another module of the instrument (not shown in Fig. 1) is a sum-frequency generation (SFG) stage [2], which up-converts a portion of the IR signal beam to ∼ 630 nm by mixing it in a KTP nonlinear-optical crystal ($\theta = 51°$, $\varphi = 0°$, type-I phase-matched) with residual 1.064-μm pump radiation to enable diagnostics of optical beam quality and spectral purity by a fixed talon as well as wavelength calibration by a pulsed wavemeter (Burleigh WA-4500).

2 Performance characteristics of the PPLN OPO

The 1.55-μm signal-wave output from the TDL-seeded PPLN OPO has a typical pulse energy of ∼ 0.2 mJ per 7-ns pulse. Collimation by a single silica lens (30-cm focal length) delivers a remarkably high output beam quality, with low divergence and a quasi-Gaussian spatial profile (whether the OPO is seeded or not) that does not vary appreciably as the output wavelength is scanned. This aspect of performance is superior to that of TDL-seeded OPOs with passive cavities [2–5], where transverse mode effects prevail, as is verified by the coherent-Raman spectroscopic measurements reported below.

The continuous single-mode tunability of signal output radiation from the PPLN OPO is demonstrated in Fig. 2. Trace (a) shows the relative frequency (GHz) of the up-converted signal output at ∼ 630 nm, as measured by the SFG/wavemeter diagnostic module, plotted against the scan control voltage of the TDL used for injection seeding; the

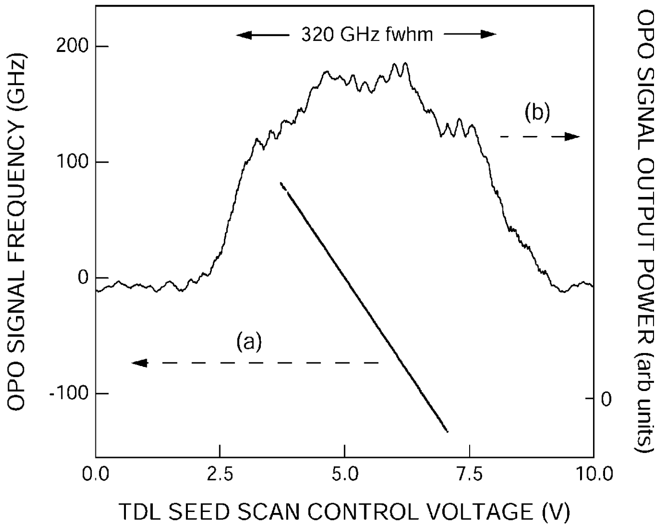

Fig. 2. (a) A plot of relative OPO signal frequency (GHz) of the tunable narrowband PPLN OPO system (measured by a SFG/wavemeter diagnostic module) against the scan control voltage of the TDL used for injection seeding. (b) Corresponding PPLN OPO signal output power as the TDL seed wavelength ($\sim 1.55\,\mu$m) is scanned across the 320-GHz QPM tuning range of a single 29.9-μm grating on the PPLN crystal at 35 °C

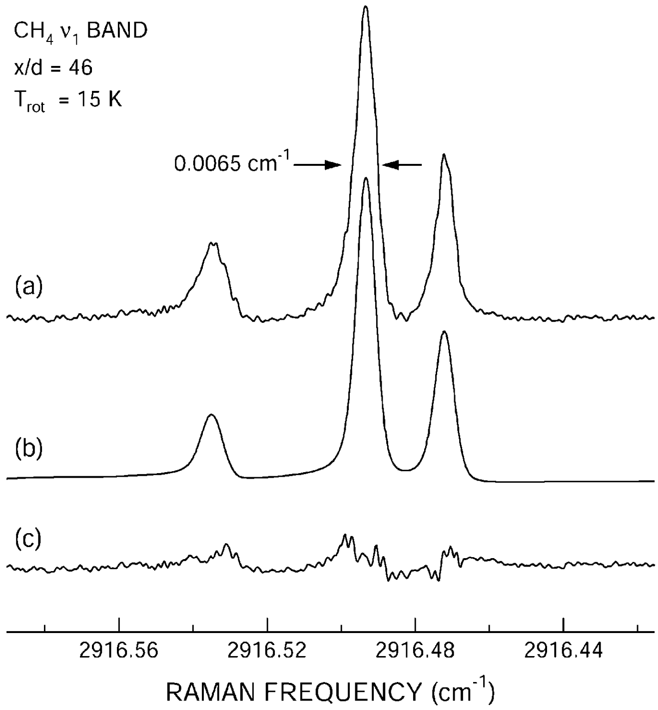

Fig. 3. CARS spectra of the Q(0), Q(1) and Q(2) lines in the 2916.5-cm^{-1} ν_1 Raman band of CH_4 in a supersonic free jet. The measured OPO CARS spectrum (a) is recorded with 1.064-μm Raman pump radiation (from the Nd:YAG laser) and tunable 1.54-μm Stokes radiation (signal from the TDL-seeded PPLN OPO). As in [2], the intensity distribution of the simulated CARS spectrum (b) arises from the three nuclear-spin modifications of the CH_4 molecule. The results are consistent with a rotational temperature of 15 K and with a fwhm optical bandwidth of ~ 135 MHz (0.0045 cm^{-1}) for the PPLN OPO signal radiation. Trace (c) shows the difference between traces (a) and (b). See text for further details

measurement range is limited by the acceptance bandwidth of the (fixed-orientation) KTP crystal used in the SFG process. Figure 2(b) plots the corresponding PPLN OPO output intensity as the TDL seed wavelength is scanned across the QPM tuning range of a single 29.9-μm grating on the PPLN crystal, with its temperature held at 35 °C. The half-maximum width of this tuning range is 320 GHz (10.7 cm^{-1}).

The spectral purity of the continuously scanned signal radiation from the PPLN OPO has been evaluated by means of a Fabry–Pérot étalon with free spectral range of 10 GHz and finesse of 1100 at $\sim 1.55\,\mu$m. After careful isolation from mechanical vibrations, the continuously scanning OPO is measured to have an output optical bandwidth of 125 ± 5 MHz, approaching the 55-MHz limit imposed by the Fourier transform of the pulse duration.

3 Coherent Raman spectroscopy with tunable OPO signal radiation

A more critical test of the signal radiation at $\sim 1.55\,\mu$m from the PPLN OPO is to use it as the Stokes beam for coherent anti-Stokes Raman spectroscopy (CARS) at Raman excitation frequencies of $\sim 3000\,\mathrm{cm}^{-1}$, with residual 1.064-μm Nd:YAG laser light as the Raman pump. This has been demonstrated by making OPO CARS measurements of the ν_1 band of CH_4 in the gas phase (as in [2], but with marginally better resolution) and in a pulsed supersonic jet (see Fig. 3). Arrangements for Doppler-free OPO CARS jet experiments are as described previously [2]. The OPO CARS spectrum in Fig. 3(a) was recorded with optical pulse energies of 10 mJ at 1.064 μm and 0.2 mJ at $\sim 1.55\,\mu$m and is effectively free of Doppler, collisional, saturation or Stark broadening. It is well simulated in Fig. 3(b), in a manner consistent with established CARS lineshape theory and spectroscopic data for CH_4 as in [2], by Voigt profiles in which a Lorentzian form (60 MHz fwhm) is convoluted with a Gaussian distribution function of 150 MHz fwhm, consistent with Raman pump (Nd:YAG laser) and Stokes (OPO) fwhm optical bandwidths of 45 MHz and 135 ± 5 MHz, respectively. The above ±5-MHz tolerance covers the extent to which the fitted rotational temperature (15 K) might be accompanied by some residual Doppler broadening (60 MHz fwhm at most).

4 Infrared spectroscopy with tunable OPO/OPA idler radiation

The combined pulse energy of signal and idler output from the PPLN OPO/LNB OPA system is typically ~ 2 mJ, with beam quality at least as good as that of the 1.064-μm Nd:YAG laser. The performance of this OPO/OPA system has been verified by an IR spectroscopic experiment that is sensitive to the stability, optical bandwidth, and continuous tunability of the pulsed idler output at $\sim 3.4\,\mu$m.

Figure 4(a) shows high-resolution photoacoustic absorption spectra of CH_4 gas ($T = 300$ K, $P = 10$ Torr) around the P(8), P(9) and P(10) clusters in the ν_3 (C−H stretching) fundamental band [14]. These are recorded in continuous scans of the idler wavelength, without correcting for any shot-to-shot variations of IR pulse energy. The corresponding simulated spectra Fig. 4(b) are generated by using the HITRAN code [15] to compute line positions and relative intensities, then convoluting them with an appropriate Voigt

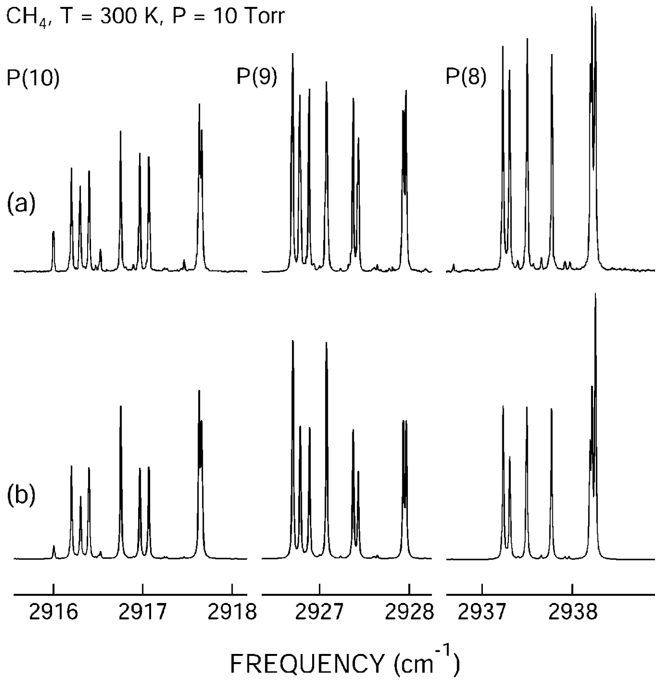

Fig. 4a,b. Measured (**a**) and simulated (**b**) IR absorption spectra of CH_4 gas ($T = 300$ K, $P = 10$ Torr) around the P(8), P(9) and P(10) clusters in the ν_3 band at $\sim 3.4\,\mu$m [14]. The Doppler-broadened spectra (**a**) have a fwhm spectroscopic linewidth of ~ 500 MHz ($\sim 0.017\,\text{cm}^{-1}$); they are photoacoustically detected by continuous scans of idler output from the TDL-seeded PPLN OPO/LNB OPA system, without normalising to IR pulse energy variations. The simulation (**b**) is based on the HITRAN code [15], as explained in the text

profile to reproduce the observed lineshapes (~ 500 MHz fwhm). After allowing for Doppler and pressure broadening [14, 15], it is deduced that the spectral profile of the idler radiation from the PPLN OPO/LNB OPA system comprises a Gaussian component of ~ 130 MHz fwhm and an additional Lorentzian component of ~ 300 MHz. The former is consistent with our measurements of the PPLN OPO alone; the latter is attributable to mechanical instability of the cavity, subsequently reduced when Fig. 3(a) was recorded, and/or possible nonlinear-optical broadening effects [10–12] in the LNB OPA stage, which was operated at ~ 3 times above threshold.

5 Concluding remarks

Injection seeding by an independent tunable laser is a well established way to control the wavelengths and monochromaticity of output from a pulsed OPO for spectroscopic purposes [1]. The results reported here depend on the QPM advantages (low threshold and high conversion efficiency, facilitating high spatial-mode purity) of PPLN [7, 8], combined with the power-amplification capability of bulk $LiNbO_3$ [2]. Optical bandwidths below 300 MHz ($0.01\,\text{cm}^{-1}$) have previously been demonstrated in a few pulsed tunable injection-seeded OPOs, including other TDL-seeded systems [2, 11, 16–18] and several OPO systems seeded by more elaborate colour-centre [10], Ti:sapphire [12] and cw dye [13] lasers. However, apart from our own earlier work on passive-cavity OPOs [2–5], instances of high-resolution spectra actually recorded by tuning injection-seeded pulsed OPO output radiation, either continuously [11, 13] or in fine wavelength steps [18], have been rare.

An outstanding feature of the actively controlled PPLN OPO system reported here is the very narrow optical bandwidth of its output radiation, which we infer from actual spectroscopic measurements in Sections 2 and 3 to be 130 ± 10 MHz ($0.0043 \pm 0.0003\,\text{cm}^{-1}$). Moreover, the locking circuit enables the output wavelengths to faithfully track the TDL seed wavelength, in continuous 300-GHz spectroscopic scans that extend over 2000 times the optical bandwidth of the OPO radiation. Other notable features of our system are its excellent output beam quality and its relatively compact, modular design. Such a TDL-seeded PPLN OPO spectroscopic system could, for some applications, be competitive with advanced grating-controlled OPO systems [19] that are commercially available. Finally, we note a recent report [20] of a single-mode PPLN OPO operating at fixed output wavelengths by means of an intracavity étalon.

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