3.5-µm high-resolution gas sensing employing a LiNbO$_3$
QPM-DFG waveguide module

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Abstract Diode laser technology coupled with a wave-
length-conversion unit to produce mid-infrared narrow
bandwidth laser light applicable to trace-gas detection and
with the potential for high-resolution spectroscopy is de-
scribed. Quasi-phase-matched difference-frequency gener-
ation (QPM-DFG) in a compact and fibre-coupled periodi-
cally poled lithium niobate (PPLN) waveguide module mix-
ing 1063 and 1525-nm radiations has been adopted for gen-
erating 34 µW of 3.5-µm wavelength laser light. Optical de-
tection methods, including sensitive wavelength modula-
tion spectroscopy and a rapid wavelength chirp technique, have
been employed with a single-pass cell to investigate methane
and formaldehyde absorption profiles around 2855 cm$^{-1}$, as
proof of principle experiments for high sensitivity and res-
olution spectroscopy on atmospherically important mole-
cules.

PACS 07.07.Df · 33.20.Ea · 42.55.Px · 42.62.Cf ·
42.65.Wi · 42.65.Ky

1 Introduction

For quantitative analysis of trace levels of gases important
for environmental monitoring and medical diagnostics, it is
desirable to combine sensitive optical approaches with the
strong ro-vibrational molecular transitions associated with
fundamental vibrational modes, many of which exist within
the mid-infrared spectral window. The 3-µm region is par-
ticularly significant as fundamental C–H, N–H, and O–H
stretching modes give rise to strong absorptions, resulting
in spectroscopic fingerprints for a large variety of molecules
in the gas phase.

Over the last decade, developments in semiconductor
laser technology have been made in the attempt to pro-
duce high quality narrow bandwidth mid-IR sources suit-
able for optical sensors with the benefits of wide tunabil-
ity and room-temperature operation. Advances have been
made with quantum cascade (QC) and interband cascade
(IC) lasers [1, 2], with improvements being made continu-
ously in tunability, power (cw operation in the mW regime),
line width, spectral coverage, and the device’s thermal re-
quirements. QC lasers are now available in external-cavity
geometries resulting in broadband mode-hop-free operation,
although not yet in the 3-µm region [3]. IC lasers are still
largely development devices that need cryogenic cooling for
operating at these wavelengths [4].

A well-established alternative for generating mid-infrared
laser light lies in the combination of diode-based (solid-
state) laser light sources and difference-frequency gener-
ation (DFG), which results from the nonlinear optical re-
sponse to an intense optical field in appropriate dielec-
tric materials. As a result, the advantages characteristic
of diode lasers are translated into the mid-infrared region,
making nonlinear wavelength conversion strategies attrac-
tive for spectroscopic applications in the gas-sensing field
[5–12]. Periodically poled lithium niobate (PPLN) is a
domain-engineered crystal extensively adopted in the gen-
eration of 2–5-µm light via quasi-phase-matched differ-
cence-frequency generation (QPM-DFG) as a consequence of its
transparency in this wavelength range and its large non-linear response [13]. Recently, particularly high efficiencies in conversion have been reported employing PPLN engineered as a ridge waveguide coupled with commercially available diode laser pumps, generating 10–10⁴ µW of mid-infrared light, potentially sufficient for optical cavity based spectroscopy techniques [14–16].

In this work, various spectroscopic applications of a LiNbO₃ waveguide wavelength converter based on QPM-DFG emitting around 3.5 µm are presented as proof of principle studies for further sensitive measurements.

2 QPM-DFG theory

In the presence of an intense applied optical field consisting of two distinct components at frequencies ω₁ and ω₂, DFG is one of the interactions arising from the second-order nonlinear response of the irradiated dielectric medium [17]. This nonlinear process implies that the material converts the ω₁ photon in a process stimulated by the field at frequency ω₂, generating radiation at frequency ω₃ = ω₁ − ω₂. The efficiency of the conversion process is limited by chromatic dispersion; therefore, a phase-matching condition must be maintained over the crystal length so that the incident power is efficiently transferred into the generated radiation.

QPM is a well-established and commonly adopted phase-matching strategy for experimentally satisfying this coherence requirement [18]. This technique requires the nonlinear material to be periodically poled, and this periodic alternation in the sign of the second-order susceptibility enables compensation for the dispersion even in crystals displaying insufficient birefringence for perfect phase-matching conditions. Furthermore, since no restrictions on the combination of polarizations of the interacting radiation fields are imposed, the largest coefficient of the susceptibility tensor characteristic of the nonlinear medium (dᵢⱼ) is accessible, and as a result the highest conversion efficiency can be achieved.

To account for the periodic poling of the crystal, the mathematical analysis of QPM interactions includes a modification to the effective value of the nonlinear coefficient, attenuated by a 2/mπ factor dependent on the mth order of the Fourier component describing the spatial variation of dᵢⱼ. The wavevector mismatch (Δk) is also modified in accordance with the following equation:

\[
Δk = k₁ - k₂ - k₃ - \frac{2πm}{Λ},
\]

where \(k = \frac{2πn}{λ}\) is the wavevector describing each of the frequency components of the interacting radiation, \(n\) is the refractive index, and \(Λ\) is the poling period of the crystal [19].

In this work, mid-infrared laser light was efficiently produced via first-order (\(m = 1\)) QPM-DFG accessing the \(d₃₃\) coefficient of a periodically poled lithium niobate (PPLN) crystal constructed as an optical waveguide. This wavelength-conversion device was engineered to provide a high confinement of the interacting optical fields, efficiently guiding them along the periodically poled nonlinear material surrounded by a lower refractive index environment. The increased effective spatial overlap of the radiations enables higher conversion efficiencies to be achieved than those with a bulk crystal. For DFG in a bulk PPLN crystal using focused laser beams, there is a trade-off between highest intensity and longest interaction length of the interacting fields, limiting the conversion efficiency.

The expected mid-infrared power produced by the DFG module was calculated in the low-conversion limit, assuming the signal and pump fields incident on the waveguide to be undepleted by the nonlinear interaction. Under this experimental condition the idler (mid-IR) power is given by

\[
P_i = \frac{ηP_sP_p}{100},
\]

where \(η\) is the device’s overall conversion efficiency expressed in % W⁻¹ [20].

3 Experimental set-up

The apparatus used for generation of mid-IR wavelengths, and its spectroscopic applications, is illustrated schematically in Fig. 1. The radiations required in the nonlinear mixing process for generation of the 3.5-µm idler radiation were supplied by a distributed Bragg reflector (DBR) 1063-nm diode laser (Eagleyard Photonics) and a narrow line width fibre pigtailed 1525-nm distributed feedback (DFB) diode laser (NTT Electronics), employed as pump and signal, respectively. The pump beam was directed through a 48 dB optical isolator (Thorlabs) to avoid back reflection and its vertical polarization was restored by a half-wave plate. The signal beam was amplified using a telecoms variable-gain erbium doped fibre amplifier (EDFA) designed to provide a saturated output power of up to 26 dBm given an input power within the 0–14 dBm range (Nuphoton Technologies).

The pump and signal beams were combined with a polarization-maintaining three-port wavelength-division multiplexer (WDM) (OZ Optics) connected to the input pigtail fibre of the waveguide unit (980-nm PANDA fibre), feeding into the module 8.1 and 198 mW of the pump and signal radiations, respectively. This wavelength converter (NTT Electronics) was engineered by directly bonding a PPLN wafer onto a LiTaO₃ support, and dicing the layers to fabricate a ridge waveguide with 10.7 µm × 16.6 µm rectangular section. The input fibre and the waveguide were