

Ultrabroadband tunable continuous-wave difference-frequency generation in periodically poled lithium niobate waveguides

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We report, for what is the first time to our knowledge, widely tunable mid-IR light generation over a range of greater than 1000 nm in the 4 μm region by employing a single quasi-phase-matched periodically poled niobate waveguide at room temperature. The waveguide we used was fabricated by annealed proton exchange based on periodically poled lithium niobate. A peak conversion efficiency of 10%/W and a linewidth as small as 37 MHz were achieved. The developed mid-IR light generator may find wide applications in trace gas detection of multiple atmospheric species and high-resolution spectroscopy. © 2007 Optical Society of America

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Difference-frequency generation (DFG) has been demonstrated in quasi-phase matched (QPM) LiNbO₃ and AlGaAs waveguides in recent years [1,2]. It has impressive advantages such as a wide wavelength conversion bandwidth and sharp linewidth. Additionally, a laser-based sensor operating in the mid-IR spectral region is favored in the application of trace gas detection, since that region contains a large number of strong fundamental absorption bands of most atmospheric trace gases; applications include air-quality control in large buildings, hospitals, and aircraft, monitoring of emissions in mining and drilling, and combustion diagnostics [3].

In the mid-IR region, difference-frequency generation based on a QPM periodically poled lithium niobate waveguide (PPLN WG) has been shown to be capable of delivering sufficient output power, tunability, and spectral purity for trace gas detection at the parts per billion (parts in 10⁹) level [4]. Unlike in the bulk configuration, confinement of light in the waveguide has no trade-off issue of beam size and interaction length. As a result, a long interaction length and thus high conversion efficiency can be achieved in a PPLN WG [5]. Another feature of the waveguide configuration, which at first looks like a drawback but in many circumstances is beneficial to spectroscopy, is that it can support multiple guide modes at the pump wavelength λ_p and the signal wavelength λ_s while supporting only one guide mode at the idler wavelength λ_i when the proper waveguide design and fabrication conditions are used. Since the effective refractive index n is different for each guide mode, there are multiple sets of wavelengths that satisfy both the energy-conservation equation, $1/\lambda_p = 1/\lambda_s + 1/\lambda_i$, and the momentum conservation equation, $n_e(\lambda_p, T)/\lambda_p - n_e(\lambda_s, T)/\lambda_s - n_e(\lambda_i, T)/\lambda_i - 1/\Lambda_{\text{QPM}} = 0$ in a single waveguide with a given QPM period Λ_{QPM} [6]. As a result, a relatively broad tuning range can be obtained with a single waveguide.

Due to the excellent properties described above, QPM waveguide devices have been studied extensively. The Ti diffusion and annealed proton exchange technologies are widely used in forming waveguides on the surface of Z-cut lithium niobate [7]. In this Letter we present a ultrabroadband cw difference-frequency radiation based on PPLN waveguides. Device performance, including tunable bandwidth, conversion efficiency, and linewidth, is investigated.

The wavelength converter used in our study was fabricated on the +Z face of a PPLN substrate [8]. A periodical domain-inverted structure, which was used to satisfy the QPM condition, was obtained by the electrostatic technique, in which a static electric field is applied across the crystal. The period of the domain-inverted structure is 22.7 μm . The waveguide has a length of 35 mm and consists of a 10 μm wide waveguide formed by the annealed proton exchange method.

Figure 1 shows the experimental setup used in DFG. The pump laser was a cw Ti:sapphire ring laser (Coherent, CR899-29) that was tunable from 770 to 870 nm with an output power of several hundreds of milliwatts. The signal laser was a diode-

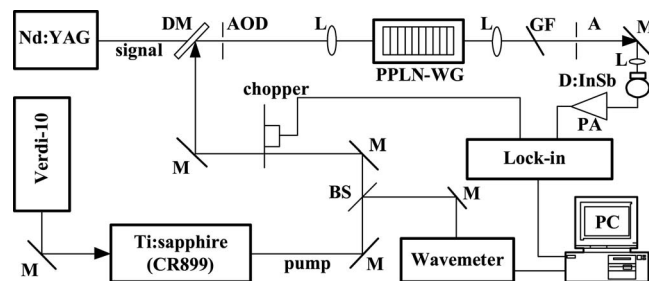


Fig. 1. Schematic diagram of the experimental setup used in DFG generation. M, mirror; DM, dichroic mirror; BS, beam splitter; L, lens; GF, germanium filter; PA, preamplifier; D, detector; A, aperture.

laser-pumped monolithic Nd:YAG laser (InnoLight, model Mephister1000) with 1 W output at 1064 nm. The Ti:sapphire laser with a linewidth of 500 kHz was pumped by a YAG laser (pumped by a diode laser, Coherent Verdi-10, $\lambda=532$ nm). The laser beams were combined at a dichroic beam splitter (high transmission near 1064 nm) and then launched into an end-polished QPM PPLN WG. A mechanical chopper was placed in front of the Ti:sapphire laser to chop the pump laser, and its chopping frequency can be varied from 0 to 4 kHz. The two laser beams, after being combined by a dichroic mirror, were then focused by an $f=20$ mm lens onto a PPLN WG. The generated mid-IR DFG light was collimated by a 20 mm focal length CaF_2 lens. In the waveguide, the combined beams were in transverse magnetic (TM) mode, which dominates DFG conversion efficiency according to the conversion $e+e \rightarrow e$ [9]. To obtain high-frequency conversion efficiency, the pump and signal laser beams have to be spatially overlapped within the PPLN WG crystal. Two apertures were placed along the laser beam to make the two beam spots as equal as possible.

Unconverted residual pump and signal beams were blocked by a Germanium low-pass filter that can absorb UV, visible, and IR up to $2 \mu\text{m}$ and is transparent up to $14 \mu\text{m}$. The idler radiation was focused on a liquid-nitrogen-cooled photovoltaic photodiode InSb detector (Hamamatsu Photonics Type:5968) with an active area of 3.14 mm^2 . During the experiment, the waveguide was mounted in a temperature-controlled oven, and its temperature was controlled by a thermoelectric temperature controller with a temperature stability better than $\pm 0.1^\circ\text{C}$. The output current signal, after being amplified by a low-noise (10 kHz low-pass filter) preamplifier, was sent to a lock-in amplifier for demodulation. Output of the preamplifier was acquired by a 12-bit analog-digital converter data-acquisition card and recorded by an autoscan personal computer.

Figure 2 shows the generated idler light power as a function of wavelength. During the experiment, the waveguide was mounted in the temperature oven at a constant temperature (24.5°C) and the signal laser was fixed at 1064 nm while the pump laser was

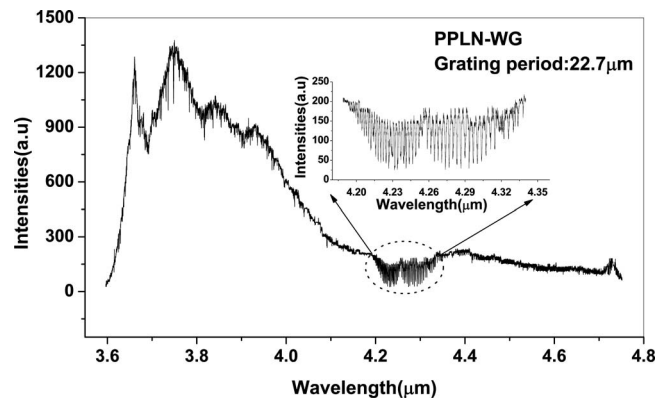


Fig. 2. Wavelength tuning curve for one waveguide with grating period of $22.7 \mu\text{m}$ at room temperature. The maximum conversion efficiency is about 10%/W. The inset shows the spectrum of the ν_3 band absorption of CO_2 .

swept from 820 to 870 nm. It is worth noting from the figure that, first, tunable mid-IR light is generated over a wavelength range greater than 1000 nm. To the best of our knowledge, this is the widest convertible wavelength range ever reported. The physical origins of such a broad range of wavelength conversion should be caused mainly by the multimode feature of the waveguide for the pump and signal wavelength as mentioned above [6]; at the same time, the waveguide with slightly varying nonuniformity and poling pattern might be another reason for the broadband region. We estimate that our waveguide supports six groups of modes that satisfy both energy conservation and momentum conservation. We were uncertain about the spatial modes that interacted to produce the multiple peaks in Fig 2. The spectrum contains multiple peaks because of the multiple waveguide modes at the pump wavelengths. The ambiguity of the multiple-peak feature in Fig 2 arises because the scan widths we obtained are quite large for the fundamental mode and because of the nonuniformity or poling pattern of the waveguide. Second, the generated mid-IR light is suitable for trace gas detection that includes a large number of strong absorption lines in the wavelength region, such as CO_2 , CH_4 , N_2O , CH_2O , and NO . As an example, the absorption spectrum of the ν_3 band (entire P , Q , R branch of the fundamental stretch-vibration band) of CO_2 in ambient air (absorption length is 30 cm) is shown in the inset of Fig 2.

We also notice in Fig. 2 that the idler power decreases with an increase of the idler wavelength. This may be caused by (i) low conversion efficiency at the long wavelength according to the simulation of idler power [10], (ii) a decrease in the pump power (shown in Fig 3), and (iii) absorption of lithium niobate itself (the transmission of the lithium niobate becomes lower for a wavelength of $4 \mu\text{m}$, especially above $4.5 \mu\text{m}$).

The maximum idler power measured with the detector at optimum phase matching was about $47 \mu\text{W}$, with an input power of 16 mW for the signal and 30 mW for the pump laser. Therefore a maximum conversion efficiency of 10%/W was achieved. According to Ref. [10], the theoretically predicted conversion efficiency is 109%; our experimental conversion efficiency was relatively low in comparison with the optimized one. The difference between the calculated and the experimental conversion efficiencies

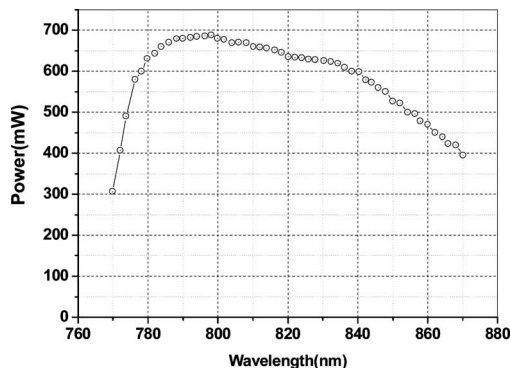


Fig. 3. Measured pump laser (Ti:sapphire) tuning curve.

might be caused by such factors as propagation loss and spatial overlap between the waveguide modes of the waves involved in the nonlinear interaction. We believe that the conversion efficiency could be further increased by using a pump and signal laser fiber combiner, optimizing the waveguide design and fabrication conditions, and minimizing the propagation loss.

The DFG linewidth is of great importance in that it determines the selectivity of the trace gas detection system. Selectivity is determined mainly by the linewidth and frequency stability, including the transient center frequency drift of pump and signal lasers. Thus, the system linewidth measurement of DFG is one of the key issues in the experiments. For this particular purpose, an additional cell with a single optical path geometry (optical path length 30 cm) was employed. Figure 4 shows the absorption spectrum of pure CO₂ at a reduced cell pressure of 0.3 Torr at room temperature. The data were averaged 1500 times during the scan. The background shape is the fringes of the Germanium etalon (thickness 1 mm).

To measure the linewidth of the mid-IR light system, the absorption line of ¹³CO₂(01111 ← 01101, P(17)) at 4.3767 μm was chosen, as shown in Fig 5. Deconvolving the measured linewidth of 132 MHz with the theoretical Doppler linewidth of 126.8 MHz results in a DFG linewidth of 37 MHz, which is larger than the estimated value of several megahertz. Generally speaking, the DFG linewidth is determined by the linewidth and the central wavelength fluctuation of the pump–signal laser, and the instantaneous linewidth of the pump–signal lasers we used were shown to be less than 1 MHz. The linewidth broadening of DFG source in our experiment may be caused by a transient power shift and central frequency shift of DFG light sources during data acquisition. For trace gas detection of small molecular species, nevertheless, the obtained linewidth is narrow enough to investigate Voigt or Lorentzian line shapes whose widths are several hundreds of megahertz or more. Therefore the obtained DFG system with its linewidth of 37 MHz is good enough to achieve the sufficient spectra selectivity required in

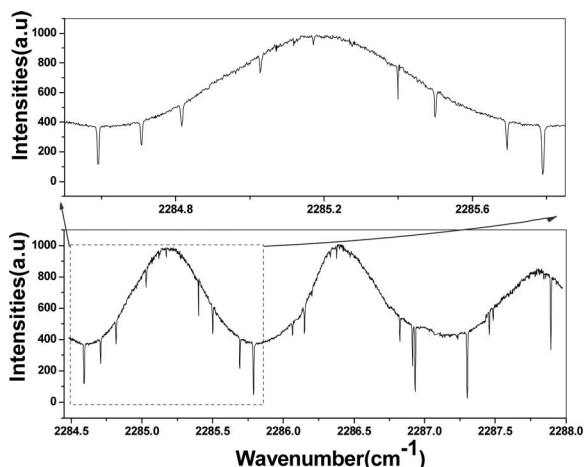


Fig. 4. High-resolution direct absorption spectrum of pure CO₂ at 0.3 Torr with 30 cm optical length; the enlarged shows part of the spectrum.

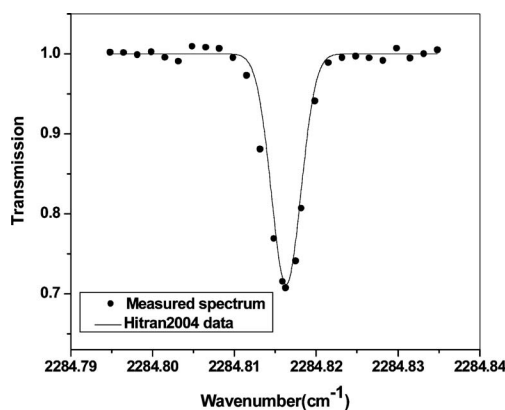


Fig. 5. Doppler-broadened absorption spectra of ¹³CO₂ by DFG frequency scan over 0.04 cm⁻¹. Pure ¹³CO₂ filled a 30 cm long cell with a pressure of 0.3 Torr.

trace gas detection.

In conclusion, we have generated mid-IR radiation from 3.6 to 4.7 μm in a single PPLN waveguide at room temperature. The mid-IR laser source was observed to be quite stable during the scanning time. The mid-IR output from a 35 mm long waveguide demonstrates excellent characteristics of the PPLN waveguide structure. The obtained results imply the possibility of achieving a compact, widely tunable, and highly efficient mid-IR source required in trace gas detection.

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