# Generation of CW mid-infrared radiation in the mW power range and tuneable over 400 nm

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11 Abstract: Miniaturization of mid-infrared (MIR) spectroscopy sources has progressed significantly during the past two decades, but a solution able to provide full integration, high 12 optical power and wide tuneability in the so-called atmospheric window (2.5 - 5  $\mu$ m) is still 13 missing. In this context, we investigated a broadband frequency-tuneable source relying on 14 difference frequency generation (DFG) in a periodically poled Lithium Niobate (PPLN) ridge 15 waveguide. By employing tuneable lasers for the pump and signal wavelengths emitting at 16 around 1 um and 1.55 um, respectively, we were able to fully cover the  $\approx 3$  - 3.5 um spectrum. 17 thus translating the technological maturity of data communication photonic sources to the MIR 18 wavelength band. Moreover, the use of a relatively large cross-section for the here-proposed 19 PPLN ridge waveguide compared to commonly employed thin-film Lithium Niobate (TFLN) 20 waveguides has allowed us to achieve low propagation and coupling losses together with high 21 damage threshold, thereby allowing us to reach mW-level power in the MIR wavelength band. 22

## 23 1. Introduction

Mid-infrared (MIR) optical sensors based on spectroscopy represent powerful tools in several 24 applications, such as environmental monitoring, industrial process control and petrochemical 25 industry [1,2]. The ability to accurately identify different chemical species requires the use 26 of laser sources with spectral lines in the kHz range, output powers of several mW, and fine 27 tuneability in frequency over an extended band of the order of several tens of nm. In particular, 28 the so-called atmospheric window that ranges from 2.5 to 5  $\mu$ m is of high interest as it contains 29 the spectral fingerprints of several pollutants, such as N<sub>2</sub>O, CO<sub>2</sub>, NO<sub>2</sub>, HCl, NH<sub>3</sub>, C<sub>2</sub>H<sub>6</sub> and 30 others. The availability of a widely tuneable and powerful MIR source suitable for integration 31 on a photonic integrated circuit (PIC) platform and for mass production is still missing and 32 would represent a revolutionary opportunity in this rapidly expanding application area. Important 33 technological advances for MIR spectroscopy have been the development of room-temperature 34 quantum cascade lasers (QCL) [3–5] and interband cascade lasers (ICL) [6,7], which have 35 extended the operating range of tuneable semiconductor lasers from the visible and near-infrared 36 (NIR) to the MIR. With these sources, a novel generation of sensors suitable for real-time in 37 situ detection of gases is now available. However, such tools, which rely on the assembly of 38 discrete optical components, still fail to meet some key requirements for many applications. The 39 40 first limitation is related to their limited tuneability, which usually limits their application to the detection of a single species for a given laser source. 41 In the present scenario, the exploitation of efficient nonlinear integrated photonic platforms could 42

43 represent an appealing solution for MIR light generation and amplification with a very compact

<sup>44</sup> footprint [8,9]. Indeed, the exploitation of nonlinear processes could allow to efficiently translate

the radiation emitted by mature and low-cost telecom optical sources, with excellent linewidth

<sup>46</sup> properties, modulation capabilities and wide tuneability, to the MIR spectral region.

Lithium Niobate (LN) is one of the most mature material platforms for the realization of 47 second-order nonlinearity-based integrated photonic devices [10, 11]. In recent years, a few 48 results have been reported on the generation of continuous wave (CW) and tuneable MIR 49 radiation exploiting LN-on-insulator (LNoI) platforms. Nonlinear waveguides usually rely on 50 ridge structures obtained by dry etching a very thin LN core layer (typically 300 - 900 nm 51 thick [12–14]) bonded on top of a silica cladding. Such geometry allows obtaining strong modal 52 confinement devices, which were not available in the past; this fuelled a significant research 53 activity based on the exploitation of the LNoI platform for nonlinear applications. The strong 54 modal confinement and flexibility in the waveguide geometry and cladding materials offered 55 by the LNoI platform guarantees new degrees of freedom in tailoring waveguide dispersion, 56 enabling both frequency generation over an ultra-wide bandwidth [15] and intermodal phase 57 matching for multi-wavelength generation in a single waveguide [16]. Despite its outstanding 58 efficiency and versatility, the LNoI platform suffers from some fundamental limitations. Indeed, 59 the typically strong modal mismatch between focused input beams and LNoI waveguide modes 60 at the integrated chip facet commonly results in a very poor butt-coupling efficiency of pump 61 and signal. Typical reported values are in the order of  $\sim 1 - 2\%$  [17], which makes this factor 62 relevant when considering the nonlinear performance of the system [18]. In principle, better 63 coupling efficiency can be obtained by the exploitation of grating or edge couplers with advanced 64 designs at the expense of an increased complexity in the waveguide fabrication and a limitation 65 in the operational bandwidth. Moreover, huge field intensities result from the extreme mode 66 confinement in the LNoI waveguides (mode effective area =  $0.2 - 1.5 \ \mu m^2$ ), which allows 67 achieving very high nonlinear efficiencies. However, absolute values of the MIR output power in 68 the order of a few tens of  $\mu W$  are typically reported, which are insufficient for most spectroscopy 69 applications. In this work, we demonstrate the generation of MIR radiation through difference 70 frequency generation (DFG) tuneable from 3 to 3.5 µm with output power in the mW range. This 71 result is obtained by exploiting a nonlinear LNoI platform based on a ridge waveguide with a 72 cross-section enabling optimal coupling efficiency and low propagation losses for the wavelengths 73 involved in the nonlinear process. This ridge structure consists of a periodically poled LN crystal 74 that guarantees phase-matched interactions over a band exceeding 400 nm. We believe that the 75 proposed device can represent a significant breakthrough towards the development of integrated 76 CW sources able to provide high-power and tuneable radiation in the MIR range for spectroscopy 77 applications. 78

## 79 2. Design and simulations

<sup>80</sup> The proposed device is based on the use of the DFG process exploiting the high second-order <sup>81</sup> nonlinear coefficient d<sub>33</sub> (25.2 pm/V) of LN. Considering a pump wave at a wavelength  $\lambda_p$ <sup>82</sup> and a signal at a wavelength  $\lambda_s$ , the wavelength  $\lambda_i$  of the generated radiation (idler) is obtained <sup>83</sup> according to the energy conservation principle:

$$\frac{1}{\lambda_i} = \frac{1}{\lambda_p} - \frac{1}{\lambda_s} \tag{1}$$

<sup>84</sup> By considering a pump wavelength  $\lambda_p$  ranging from 1 to 1.1 µm and a signal tuneable in the <sup>85</sup> optical communication C-band (1.53 - 1.565 µm), a MIR radiation in the range from 2.75 to <sup>86</sup> 3.91 µm could be generated. The efficiency of the nonlinear process is mainly dictated by the <sup>87</sup> phase matching (PM) condition that has to be fulfilled between the interacting fields. As already <sup>88</sup> extensively reported in the literature, such a condition can be satisfied by periodically poling the <sup>89</sup> ferroelectric domains of the LN waveguide. In this case, the phase-matching condition can be <sup>90</sup> written as [19]:

$$\Delta\beta = \beta_p - \beta_s - \beta_i - \frac{2\pi}{\Lambda} \approx 0, \text{ with } \beta_j = \frac{2\pi n_{eff,j}}{\lambda_i}, \tag{2}$$

where  $\Delta\beta$  is the phase-mismatch term,  $\beta_i$  is the propagation constant of the fundamental TM<sub>00</sub> 91 mode employed in this work at the pump (p), signal (s) and idler (i) wavelengths,  $n_{eff,i}$  is the 92 effective refractive index,  $\lambda$  is the wavelength and  $\Lambda$  is the poling period selected to achieve the 93 phase-matching. The designed ridge waveguides exhibited a cross-section of  $7 \times 9.6 \,\mu\text{m}^2$  and 94 was 3 cm-long. Since the inversion of the ferroelectric domains is induced by placing a mask on 95 top of the LN wafer, the orientation of the poling requires to couple the radiation as transverse 96 magnetic (TM). The waveguides are placed on a  $0.6 \,\mu\text{m}$  silicon dioxide (SiO<sub>2</sub>) layer, bonded 97 through a 0.2 µm gold (Au) layer to a silicon (Si) wafer (see Fig. 1a). The spatial distribution of 98 the fundamental TM<sub>00</sub> mode in the wavelength ranges of  $\lambda_p = 1 - 1.1 \mu m$ ,  $\lambda_s = 1.53 - 1.57 \mu m$ 99 and  $\lambda_i = 2.7 - 4 \mu m$ , their respective effective refractive indices and effective areas were obtained 100 through numerical simulations based on a commercial software (MODE Solutions from ANSYS 101 Inc.). The mode field diameters (MFDs) simulated at  $\lambda_p = 1 \ \mu m$  and  $\lambda_s = 1.55 \ \mu m$  were 6.248 102 µm and 6.303 µm, respectively, thus providing a very good mode matching with the fundamental 103 mode of the optical fiber used for edge coupling (MFD =  $6.6 \,\mu m @ 1 \,\mu m$ ). From the numerical 104 simulations performed at  $\lambda_i = 4 \mu m$ , it can be seen that the fundamental TM<sub>00</sub> mode (see Fig. 105 1b) is very well confined in the LN ridge waveguide with a MFD of 6.631 µm and with a limited 106 extent into the silica layer, thus limiting additional losses that might arise at MIR wavelengths 107 due to the  $SiO_2$  cladding material absorption. Addionally, it is worth noting that the mode size 108 is weakly dependent on wavelength, resulting in an optimal spatial overlap between the three 109 interacting wavelengths. 110



Fig. 1. (a) Cross-sectional view of the designed waveguide and dimensions; (b) Spatial distribution of the electric field of the fundamental  $TM_{00}$  mode numerically simulated at 4  $\mu$ m.

The poling period was determined through numerical simulations to achieve quasi-phase matching across the desired wavelength range, with the results reported in Fig. 2a. Fig 2b reports the phase-mismatch diagram that can be obtained considering a poling period equal to 25 µm. In this configuration, the phase-matching for the DFG nonlinear process can be preserved over a broad wavelength range from 2.7 to 3.5 µm by tuning the pump wavelength. By considering a LN waveguide with a different poling period, it is possible to shift the phase matching band to longer or shorter wavelengths.

The coupling efficiencies between a cleaved fiber (MFD =  $6.6 \,\mu\text{m} @ 1 \,\mu\text{m}$ , as in the experiments) and the fundamental and higher-order TM modes of the designed LN waveguide were numerically calculated. The overall coupling efficiency was found to be equal to 80%, with most of the power coupled to only two modes: 73.2% of the total power was coupled into the TM<sub>00</sub> mode and 6.2%was coupled into the TM<sub>02</sub> mode. The maximum coupling to all the other modes was found to be negligible.



Fig. 2. (a) Numerical simulations of the required poling period (in  $\mu$ m) to satisfy the phase-matching condition as a function of the pump and idler wavelengths considering a signal wavelengths in the range 1530 - 70 nm; (b) Normalized phase mismatch diagram numerically simulated for a poling period equal to 25  $\mu$ m.

### 124 3. Fabrication

The first stage of the fabrication was to periodically pole a 500 µm thick, 4 inches diameter 125 commercial z-cut congruent  $LiNbO_3$  wafer by a standard technique involving the application of 126 an intense electric field at room temperature [20] using a photo-resist pattern [Fig. 3 (a)]. Several 127 poling periods near 25 µm were inserted in the mask and realized to keep into account fabrication 128 variations. In a subsequent stage, a SiO<sub>2</sub> layer was deposited by ICPECVD (Inductively-Coupled 129 Plasma-Enhanced Chemical Vapour Deposition) onto one face of the poled wafer followed by 130 the sputtering of a 100 nm-thick gold layer [Fig. 3 (b)]. A high-flatness silicon wafer was also 131 coated with a 100 nm-thick gold layer. The metallized faces of both the PPLN and silicon wafers 132 were then placed into contact and pressed in a wafer bonding machine [Fig. 3 (c)]. This metal 133 diffusion bonding process was realized at room temperature which prevents mechanical stress 134 that could occur due to the dissimilar temperature coefficients of the two wafers. The bonding 135 procedure was completed by applying a strong pressure to the stack which yields more than 136 98% of the surface bonded, as observed by an ultrasound characterization technique. At the 137 end of the process, a 1 mm thick hybrid structure composed of a silicon substrate bonded to a 138 PPLN wafer is obtained. The structure was mechanically polished to obtain a 7 µm thick PPLN 139 layer [Fig. 3 (d)]. Note that this method was used earlier to produce adhered non-linear ridge 140 waveguides [21–23] where either epoxy glue or direct bonding was used to fix the PPLN wafers 141 onto LiNbO<sub>3</sub> or LiTaO<sub>3</sub> substrates. At this stage, the layers' thickness was examined to select the 142 most homogeneous areas on the wafer. In the next step, two parallel trenches were cut in the 143 PPLN layer by a precision dicing saw to form the lateral sides of the ridge waveguide [Fig. 3 (e)]. 144 The dicing parameters were set to minimize the roughness of the cut surfaces [24, 25]. Finally, 145 the hybrid wafer was diced to achieve polished input and output faces for the ridge waveguides. 146

### 147 4. Experimental setup

A schematic of the experimental setup used for the device characterization is reported in Fig. 4. It consists of a fiberized input side to couple the optical pump and signal into the PPLN waveguide and a free-space detection side to collect the output beams.

A custom-made CW tuneable laser source, able to generate Watt-level optical power in the wavelength band 1020 - 1065 nm, was employed as the optical pump. The laser consists of an oscillator with 6 mW maximum output power amplified through a co-propagating amplification



Fig. 3. Fabrication schematic: (a) periodically poling of a LiNbO<sub>3</sub> wafer; (b) deposition of silica and sputtering of gold buffer; (c) bonding of the PPLN wafer on a silicon wafer; (d) grinding and polishing of the PPLN side to reach the desired thickness; (e) dicing of the ridge waveguides and of the hybrid wafer.



Fig. 4. Experimental setup used for the nonlinear DFG experiments. The photo shows the edge-coupling between a cleaved optical fiber and a PPLN waveguide. PBS: polarization beam splitter, PM WDM: polarization-maintaining wavelength division multiplexer, PM coupler: polarization-maintaining coupler, LSA: laser spectrum analyser, MIR PD: MIR photodiode. The fiberized/free-space blue, green and orange lines indicate the paths of the pump, signal and idler beams, respectively.

stage. The oscillator's active medium is a Yb-doped polarization-maintaining (PM) optical 154 fiber pumped with a 300 mW laser diode at 976 nm. The emission wavelength is set with a 155 1200-lines/mm grating mirror. The output of the oscillator, from a fiber coupler, is sent through 156 a pump combiner to a double-clad Yb-doped PM amplifier pumped by a 10-W single-emitter 157 multimode laser diode at 915 nm. The amplified output is separated from the residual pump by 158 a dichroic mirror and an isolator is added to avoid back reflections into the oscillator. A  $\lambda/2$ 159 waveplate and a C-coated lens are used to couple the laser output radiation in a PM optical fiber, 160 which is then connected to a 99:1 PM fiber coupler. The power of the pump laser is monitored 161 through a power meter that detects the 1% output of the 99/1 PM fiber coupler. The signal beam 162 is generated by a CW laser (hp8168F) tuneable in the S-C bands, with a 100 kHz linewidth and 163 0.035 nm tuning accuracy. The signal is then amplified using an Erbium-doped fiber amplifier 164

(EDFA) with maximum output power of 1W. Since the laser and the EDFA are not PM devices, 165 a manual fiber polarization controller and a fiber polarization beam splitter (PBS) are used to 166 precisely control the polarization of the signal coupled into the PPLN waveguide. The PBS 167 limits the maximum signal power to around 500 mW. The polarization extinction is controlled by 168 monitoring the power on one output of the PBS using a power meter. The pump and signal are 169 combined together with a PM wavelength division multiplexer (WDM). The light is coupled into 170 the PPLN waveguides through edge-coupling using a cleaved PM fiber (fiber PM980-XP), whose 171 angle is controlled using a fiber rotator to excite the fundamental  $TM_{00}$  mode of the waveguide. 172 The chip is mounted on a metal support equipped with a Peltier cell which can be used to change 173 the temperature. 174

In the edge-coupling procedure, the spatial modes at the optical pump wavelength at the chip 175 output facet were imaged using a CCD camera (Thorlabs DC1545M-G1) to maximize the power 176 coupled into the  $TM_{00}$  mode of the waveguide. A free-space coupling scheme is set up to collect 177 the output radiation from the chip using a  $BaF_2$  lens with an anti-reflective coating between 2 - 5 178 um to initially collimate the output light. The different wavelength components of the collimated 179 output beam are split using a germanium (Ge) window with an anti-reflective coating between 180  $1.9 - 6 \,\mu\text{m}$  and a dichroic mirror. The pump and signal beams are measured using two power 181 meters (Ophir 3A-SH and Thorlabs PM16-122, respectively). The MIR beam component is 182 modulated using a chopper and it is then measured initially through a laser spectrum analyser 183 (BRISTOL 771-B MIR) to determine the wavelength of the generated idler and then using an 184 InAsSb photodiode (Thorlabs PDA07P2) to retrieve the MIR power at the corresponding idler 185 wavelengths. The modulated signal from the InAsSb photodiode is finally acquired through a 186 USB oscilloscope (Digilent Analog Discovery 2). 187

### 188 5. Waveguide characterization

A linear characterization of the waveguides was initially carried out by measuring the waveguide 189 propagation losses using the Fabry-Perot interferometric method, utilizing both fringe contrast 190 and Fourier analysis [26]. We measured propagation losses in the S- and C-bands of  $\approx 0.026$ 191 dB/cm. The coupling losses were also evaluated and we measured coupling efficiency of  $\approx 75\%$ 192 using a simple 90°-cleaved optical fiber, in good agreement with numerical simulations. These 193 results highlight the advantages of using large waveguides compared to commonly used thin-film 194 LN (TFLN) waveguides, both in terms of propagation and coupling losses. In particular, the high 195 coupling efficiency was achieved without the use of lensed optical fibers thanks to the similar 196 MFD of the PPLN waveguide and the single-mode fiber, thus providing even better long-term 197 power stability in the experiments thanks to the relaxed tolerance in terms of fiber misalignment. 198 Nonlinear experiments were then carried out on a 3 cm-long PPLN waveguide with a poling 199 period of 24.6  $\mu$ m, and side dimensions of 7 × 9.6  $\mu$ m<sup>2</sup>. 200

The overall conversion bandwidth was evaluated by varying the pump wavelength  $\lambda_p$  at 5 201 nm steps from 1020 to 1065 nm. The signal wavelength  $\lambda_s$  was fine-tuned across the EDFA 202 amplification spectrum for each  $\lambda_p$  value. The results of this characterization are reported 203 in Fig. 5a (note that values indicated in the figure are normalized and refer to absolute MIR 204 power values between 220 and 300  $\mu$ W that are dependent on the pump power availability at 205 the specific wavelength of operation). As expected from numerical simulations, we observed 206 that the maximum efficiency of the DFG process remains almost constant over a wide band in 207 the MIR, approximately 460 nm. By finely tuning the wavelengths of both the pump and signal, 208 it is possible to achieve the maximum efficiency for any desired MIR wavelength within this 209 bandwidth. According to numerical simulations, using laser sources for the pump and signal 210 with a broader tuneability, the range of the generated idler wavelengths could be further enlarged. 211 The recorded peak normalized efficiency is equal to 3.24 %/W/cm<sup>2</sup>, while the simulated one is 212 equal to 6.73 %/W/cm<sup>2</sup>. The efficiency is normalized by accounting for C-band losses and the 213

nonlinear effective length of the waveguide, as in Ref. [15]. The nonlinear effective length of the 214 waveguide accounts for the length of the waveguide over which the phase-matching condition can 215 be considered actually satisfied and an effective nonlinear process takes place. This was evaluated 216 by fixing the wavelength of the pump and varying the wavelength of the signal to retrieve the 217 signal-conversion bandwidth. By comparing the measured full width at half maximum (FWHM) 218 to the simulated one, an effective length of  $\approx 70\%$  of the total waveguide length was found. The 219 reason behind this could be variations in the size and poling period along the waveguide. The 220 difference between simulated and experimentally measured efficiencies could be likely attributed 221 to higher MIR losses compared to the C-band losses and also to the limited nonlinear effective 222 length of the waveguide. Through a comparison between experimental and simulated efficiencies, 223 it is also possible to determine the maximum value of the loss in the MIR being equal to 1.5 224 dB/cm, by setting a nonlinear effective length equal to 70% of the real waveguide length. 225



Fig. 5. (a) Normalized conversion efficiency as a function of the idler wavelength for different wavelengths of the pump source; (b) Idler output power as a function of the output pump power. The input signal power is fixed at 0.5 W; the wavelengths are set as:  $\lambda_p = 1040$  nm,  $\lambda_s = 1537.8$  nm and  $\lambda_i = 3212.76$  nm.

To evaluate the idler output power as a function of the pump power, we set the pump wavelength 226  $\lambda_p = 1040$  nm, the signal wavelength  $\lambda_s = 1537.8$  nm with an EDFA output power  $P_s = 0.5$  W 227 and we continuously varied the pump power, recording the idler output power at  $\lambda_i = 3212.76$ 228 nm. The results are reported in figure 5b. The output idler power grows linearly with the pump 229 power up to the mW level, which satisfies the typical power requirements for direct absorption 230 spectroscopy (DAS). DAS is one of the simplest methods for trace gas analysis and consists of 231 the detection of a residual MIR beam that has propagated through a gas sample. The typically 232 minimum required power for nearly shot-noise-limited MIR detection is of the order of 0.1 mW, 233 depending on the detector [27]. Due to the input pump power limit in this test, we did not reach 234 the saturation point of the conversion that we analytically estimated would be reached with a 235 pump power in excess of 20 W, nor the damage threshold of the waveguide. By considering a 236 similar waveguide structure [28], we estimate that the maximum power we could tolerate inside 237 the waveguide before reaching the damage threshold is 4 W (CW regime), occurring considerably 238 sooner than the pump depletion-induced saturation point. Therefore, we can assume that is 239 possible to generate even higher MIR optical power with the proposed device. 240

#### 241 6. Conclusion

In conclusion, we discussed the design, fabrication and characteritazion of a PPLN waveguide
 for broadband, mW-level, NIR to MIR wavelength conversion. The generated MIR light was

tuned by changing the wavelength of a  $1.02 - 1.065 \,\mu\text{m}$  pump and a telecom C-band signal.

<sup>245</sup> Compared to conventional TFLN waveguides on silica devices [15], the here-proposed waveguide

<sup>246</sup> configuration benefits from a larger cross-section to achieve better mode confinement and lower

<sup>247</sup> propagation and coupling losses, thus allowing higher optical power levels for the generated idler.

The demonstrated broadband fine-tuning in the  $\approx 3 - 3.5 \,\mu m$  wavelength range, achievable output

<sup>249</sup> power level and the capability of translating the mature telecom technology in the MIR can be

exploited for spectroscopy applications. Higher powers and broader conversion bands may be

demonstrated in the future by improving the input laser sources and changing the chip temperature.

Further progress in the realization of the poling mask may also enable the exploitation of the entire length of the waveguide for the DFG nonlinear process, increasing both efficiency and

entire length of the waveguide forachievable output power.

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