Ultra-broadband mid-wave-IR upconversion detection

AJANTA BARH,* CHRISTIAN PEDERSEN, AND PETER TIDEMAND-LICHTENBERG

DTU Fotonik, Technical University of Denmark, DK-4000 Roskilde, Denmark
*Corresponding author: ajanta@fotonik.dtu.dk

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In this Letter, we demonstrate efficient room temperature detection of ultra-broadband mid-wave-infrared (MWIR) light with an almost flat response over more than 1200 nm, exploiting an efficient nonlinear upconversion technique. Black-body radiation from a hot soldering iron rod is used as the IR test source. Placing a 20 mm long periodically poled lithium niobate crystal in a compact intra-cavity setup (>20 W CW pump at 1064 nm), MWIR wavelengths ranging from 3.6 to 4.85 μm are upconverted to near-infrared (NIR) wavelengths (820–870 nm). The NIR light is detected using a standard low-noise silicon-based camera/grating spectrometer. The proposed technique allows high conversion efficiency over a wider bandwidth without any need for a shorter crystal length. Different analytical predictions and numerical simulations are performed a priori to support the experimental demonstrations.

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Recently, the detection and spectroscopy of a wide range of trace gases (e.g., CO₂, NOₓ, SO₂, CH₄, etc.) and compound molecules (H-O, As-H, C-H, HCHO, CCl₄) have become of prime importance owing to their inevitable impact on industry, environment, health, food, security, military, and so forth [1,2]. Those aforementioned specimens undergo strong molecular vibrational transition in the so-called mid-wave-infrared (MWIR) spectral regime (3–5 μm), which is abbreviated as the functional group regime. By carefully targeting such a spectral band, the salient features of those molecules can be probed very effectively. Apart from that, due to the coincidence with a clear atmospheric window, the 3–5 μm spectral range is particularly important for gas sensing and military applications [1,3]. One vital challenge is to detect the MWIR light, which cannot be seen by the naked eyes or by efficient Si-based detectors. Moreover, low noise detection is more challenging owing to the thermal background radiation and lack of specialized optics. Thus, unfortunately, unlike UV-vis-NIR, MWIR spectroscopy still lags behind in terms of speed, noise level, room temperature operation, user friendliness, and cost [4].

Most MWIR detectors are based on micro-bolometers or low bandgap material (e.g., InSb or HgCdTe) based detectors [4]. However, generally, the signal-to-noise level is quite low when compared to traditional NIR silicon detectors. MWIR spectroscopy is mostly based on Fourier transform infrared (FTIR) spectrometers, requiring high optical precision and mechanical stability, altogether resulting in high cost and complexity of the system. In 2012, Dam et al. re-introduced an alternative way to detect MWIR light by translating the MWIR signal to NIR wavelengths through nonlinear (NL) frequency upconversion in a χ⁽²⁾ material followed by simple, traditional cost-effective NIR Si-CCD/camera detection [5]. To date, this approach out-performs many other MWIR detection schemes in terms of conversion efficiency, noise level, mechanical tolerances, and ambient temperature operation. This approach has been exploited extensively both in imaging and for spectroscopy [1,2,6–8].

Though several problems have been overcome, the acceptance bandwidth of the upconversion process is still a limiting factor when applied for spectroscopy (tens to hundreds of nanometers at MWIR wavelengths). Various approaches have been tried to overcome this limitation. (1) Temperature tuning of the NL crystal [9] is a slow process so as to avoid any damage of coating on crystal facets and, hence, limits the acquisition speed. Moreover, it also brings instability in the system, particularly for an intra-cavity setup. (2) Using a chirped poled crystal improves the bandwidth at the cost of efficiency [10]. Furthermore, its design and fabrication is a complex task and requires extra care, i.e., apodization for smoothing of the efficiency curve [11]. (3) Sweeping the pump wavelength [12] is limited by the upconversion bandwidth and tuning range of the available source. (4) A multi-channel wavelength configuration [2] covers a several μm wide MIR spectrum, but adds complexity to the system.

In this Letter, the ultimate goal is to upconvert a broadband MWIR source followed by easy detection provided by a standard NIR spectrometer. We employ the well-known quasi-phase matching (QPM) technique, using a χ⁽²⁾ periodically poled NL crystal, which allows access to the largest nonlinearity of the material and design flexibility. In this design, we exploit the inflection point of collinear QPM, together with the turn-around point of non-collinear QPM to maximize the upconversion bandwidth. First, the collinear interaction among participating waves is studied for different NL crystals under...
various pumping conditions. Consequently, a periodically poled lithium niobate (PPLN) has been chosen as an NL crystal, combined with a 1064 nm pump laser. This choice is considered for the rest of the analysis. Subsequently, non-collinear QPM is exploited in the PPLN by placing it inside a high-power continuous wave (CW) intra-cavity setup. The high mixing power enhances the efficiency of the upconversion process. Finally, ultra-broadband upconversion, covering ~3.6–4.85 μm at MWIR wavelengths, is demonstrated at room temperature using a simple and compact design.

Frequency upconversion, or sum frequency generation in a χ(2) material, involves one pump photon of frequency ωp and one lower energy (MWIR) photon of frequency ωMWIR which are annihilated to generate one upconverted photon of frequency ωup satisfying both energy and momentum conservation (ωp + ωMWIR = ωup and Δk = kωp + kMWIR − kωp), where Δk is a measure of phase mismatch of the interacting waves. Δk should ideally be zero for maximal conversion efficiency, known as the phase-matching (PM) condition. Among various PM configurations, QPM in a periodically poled NL material with periodicity, known as the phase-matching (PM) condition. Among various PM configurations, QPM in a periodically poled NL crystal is chosen for the broadband upconversion, owing to its design flexibility and high effective nonlinearity [13]. In Fig. 1(a), a schematic diagram of a poled crystal with uniform poling periodicity (A) is shown with a pump and MWIR as input and the upconverted light as output. The design flexibility of such a structure enables us to investigate the effect on bandwidth for different NL materials, λ and pump wavelengths (λp).

First, we consider the collinear interaction of the participating waves (pump, MWIR, and upconverted light), propagating along the crystal length. The QPM condition for five different input pump wavelengths (λp = 914, 946, 1064, 1342, 1550 nm) in the case of two popular χ(2)-crystals, PP-KTP (KTiOPO4, transmission range: up to ~3.5 μm) and MgO-PPLN (MgO doped LiNbO3, transmission range: up to ~5 μm) are examined. The Sellmeier equations of [14,15] are used in the numerical calculation. The variation of a QPM period (A) covering the infrared range of 1–5 μm is plotted in Figs. 1(b) and 1(c) for PP-KTP and PPLN, respectively. In all cases, the phase-match curve has an inflection point around a certain phase-matched wavelength, which shifts for different mixing wavelengths λp and choice of NL material. This hints that, if we choose the Λ close to the inflection point, a wider wavelength band can be phase matched directly, using collinear interaction. In this simple way, efficient broadband (few 100’s of nm) upconversion of a specific infrared band can be achieved by only selecting the proper NL material, Λ and λp. Such a design criteria has previously been explored for a pulsed optical parametric generator in PPKTP [16] and PPLT [17].

In this Letter, we target to maximize the upconversion bandwidth in the 3–5 μm spectral range and, hence, as a first choice, we consider an MgO-PPLN crystal (Δωeff ~ 14 pm/V) with Λ = 23 μm and λp = 1064 nm. This choice corresponds to the solid blue circular spot in Fig. 1(c). The upconversion efficiency is calculated, assuming plane wave interaction, including the pump depletion. The efficiency is plotted in Fig. 2(a) for a 20 nm long MgO-PPLN, a pump power of 20 W, and a beam diameter of 300 μm. As predicted, a bandwidth of more than 200 nm is achieved, centered at the inflection point. The collinear QPM bandwidth can be further improved by decreasing the crystal length (L), however, reducing the maximum efficiency, which decreases with the square of L. This dependence is depicted in Fig. 2(b). Thus, depending on the specific application, different crystal lengths can be chosen.

In order to widen the bandwidth even further, we introduce non-collinear interaction of the participating waves [see Fig. 3(a)]. Since all the waves are extraordinary polarized (refractive index depends on the angle of propagation relative to crystal c-axis), their birefringence properties [13] also need to be incorporated in the numerical calculation. We assume that the pump is propagating along the PPLN length (z-axis), whereas the input MWIR wave and the corresponding upconverted wave make an angle θMWIR and θωp with the pump beam, respectively, inside the crystal. The specific angles are dictated by the QPM condition [see lower diagram of Fig. 3(a)], i.e., where both transverse and longitudinal phase matching are satisfied. The dependence of phase-matched wavelengths (λMWIR) on the incident angles (internal, θ) of the MWIR wave at room temperature is plotted in Fig. 3(b). The figure reveals that the QPM curve has a turnaround point at an internal angle θMWIR = 3.2°, which corresponds to λMWIR ~4.2 μm. This means that two different sets of λMWIR can be non-collinearly phase matched for the same input angle and, hence, a wider spectral band can be upconverted if launching input MWIR light over a small annular cone. According to Fig. 3(b), it can be seen that, by focusing the MWIR light inside the MgO-PPLN with a cone of half-angle of

Fig. 1. (a) Schematic diagram of a periodically poled ferroelectric material with periodicity, Λ, and the direction of the input (pump, MWIR) and output (upconverted) beams for collinear interaction. Variation of QPM period for a different combination of the pump and IR wavelengths in the case of (b) PP-KTP and (c) PPLN crystal; the blue circular spot indicates the inflection point for a 1064 nm pump.

Fig. 2. (a) Upconversion efficiency of collinear QPM in a 20 mm long MgO-PPLN. The FWHM bandwidth is ~230 nm around 4.2 μm. (b) Variation of the upconversion bandwidth (red dotted) and maximum efficiency (blue solid) for different crystal lengths.
3.2°, an ultra-wide spectral range extending from 3.6 to 4.85 μm can be upconverted directly; hence, a bandwidth more than 5 times wider (>1200 nm) can be achieved with this design compared to the collinear case.

To test our theoretical predictions and numerical calculations, we conduct an experiment where a high finesse, CW intra-cavity Nd:YVO₄ laser beam at 1064 nm (TEM₀₀) is used as a mixing source. The laser source is inside a closed box and pumped by an 880 nm laser diode delivering a maximum of 3 W of pump power [see Fig. 4(a)]. A hot soldering iron rod, emitting broadband black-body radiation, serves as the MWIR source, whereas a market available, cost-effective hot soldering iron rod serves as a MWIR source. (b) Upconverted image of the tip of a hot soldering rod heated to 430°C. The output power level is ~μW/nm. The spectral content of the image is captured by a fiber-coupled highly sensitive NIR spectrometer (Ocean Optics QE65000) at room temperature, and the spectrum is recorded with 100 ms integration time [see Fig. 4(c)]. For easier identification, a MWIR scale corresponding to the upconverted wavelength (NIR) is also included. As seen, we have successfully upconverted an ultra-wide MWIR light (3.6–4.85 μm) inline, which provides excellent agreement with our predicted numerical analysis of non-collinear upconversion [See Fig. 3(b)]. The resolution of the spectrum is approx. 1.9 nm at the NIR wavelengths limited by the size of entrance slit (25 μm) and grating element (300 LPM) of the spectrometer. This corresponds to a spectral resolution of approximately 45 nm at 4.2 μm. We may mention that a multi-mode fiber with a high NA (0.22) and large core size (300 μm) is used to efficiently collect the upconverted light. However, a significant amount of power is lost as this fiber couples to the much narrower entrance slit (25 μm). Therefore, it is important to balance between sensitivity and resolution of the detected spectra. Nevertheless, this all-optical setup enables fast detection of a broadband MWIR light. The gradual decrease in intensity toward longer wavelengths is primarily caused by the increasing absorption loss in the PPLN at longer wavelengths. Additionally, the angular dependent coupling to the fiber gives reduced signal intensity around the turnaround point.

Depending on the specific applications, the bandwidth and center wavelength of the upconversion detector can be tuned by varying different parameters. As discussed previously, tuning the pump wavelength (λ₀), a correct choice of NL material, and the length of crystal and its poling periodicity alter the bandwidth and center of the upconverted spectrum. Fine tuning can be obtained by temperature (T) tuning of the PPLN. This was investigated by heating up the crystal using a standard oven (Covesion Ltd.), keeping all other experimental conditions the same. The measured spectrum for four different T (40°C, 80°C, 100°C, 110°C) of the PPLN is shown in Fig. 5(a). It is observed that the spectrum gradually becomes narrower as the T increases, while keeping the central wavelength more or less constant. We also numerically calculate the phase-matching curve for those four temperatures, as shown in Fig. 5(b). Corresponding colors are used to plot graphs for the same T in both figures. Furthermore, we indicate the two extreme values of phase-matching wavelengths (for θₘ₉ = 0) using circular spots of
The intensity of the spectra is much higher for a wavelength with the hydrocarbon-based molecular emission (green curve). In Fig. 6(a), the spectrum of the hot candle flame is plotted, as detected by the upconversion module. The solid line acts as a temperature signature, as indicated in Fig. 5(a).

Another interesting point to be noted here is that the experiment captures the MWIR spectrum of atmospheric CO₂ (present between the IR source and PPLN), seen as a small absorption dip centered at 852 nm [±4.3 μm as indicated in Fig. 4(c)]. To explore this further, we examine the signature of CO₂ in our lab environment. We consider the flame of a candle as the IR source, which creates hot CO₂. By monitoring the flame, we have been able to detect the emission spectra of hot CO₂. In Fig. 6(a), the spectrum of the hot candle flame is shown, with wide spectral emission, along with the hydrocarbon-based molecular emission (green curve).

The intensity of the spectra is much higher for a wavelength ≥4.2 μm owing to the emission of different C-based molecules (CO, CO₂, etc.). The red curve is more important as the hot CO₂ (as well as H₂O) is forming just above the flame with high concentration (complete combustion). Thus, this region mainly works as an emission zone of CO₂ and contains its spectral signature. Note that the emission from CO₂ is re-absorbed along the path toward the upconversion detector (path length ~20 cm). Hence, the red curve in Fig. 6(a) depicts the combined emission and absorption spectrum of the CO₂ gas. This measurement convincingly follows the numerically calculated spectrum [Fig. 6(b)], where the IR emission from a hot CO₂ gas is transmitted through a 20 cm atmospheric path length. The spectrum is calculated using the HITRAN2012 database in SpectralCalc using 35 nm of spectral resolution.

In conclusion, we have presented an efficient and ultra-fast method to detect ultra-broadband MWIR light using a combination of a frequency upconversion module and a standard NIR spectrometer. We have theoretically analyzed collinear QPM conditions for frequency mixing in different crystals and identified an inflection point that maximizes the MWIR bandwidth up to 100’s of nm. Thereafter, we have explored non-collinear QPM and its turnaround point to further widen the detection bandwidth to ≥1.2 μm. Applying these techniques, we have experimentally demonstrated an ultra-wide frequency upconversion covering the 3.6–4.85 μm MWIR range using a compact intra-cavity setup and a 20 mm long MgO-PPLN crystal. We also experimentally checked that smaller crystal lengths do not widen the overall upconversion bandwidth further, as expected. A radiative heat source (hot soldering iron rod) was employed as broadband MWIR light source, and the upconverted light was detected in situ using a standard Si-camera/spectrometer. The targeted spectral band is very important for environmental studies, as it coincides with one of the atmospheric transmission windows and contains a molecular fingerprint of several trace gases, such as CO₂, CO, N₂O, and O₃. As an example, we measured the combined emission-re-absorption spectrum of CO₂ in the atmosphere by monitoring a candle flame. We believe that this approach will make a significant contribution to MWIR molecular spectroscopy and imaging.

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