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1 Article

2 Mid-infrared ultra-short pulse generation in a gas-

3 filled hollow-core fiber pumped by two pulses 4 [Invited].

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Abstract: We show that ultra-short pulses can be generated in the mid-infrared when a gas filled hollow-core fiber is pumped by a fundamental pulse and its second harmonic. The generation process originates from cascaded nonlinear phenomenon starting from a spectral broadening of the two pulses followed by an induced phase-matched four wave-mixing lying in the mid-infrared combined with a dispersive wave. By selecting this mid-infrared band with a spectral filter, we demonstrate the generation of ultra-short 60 fs pulses at 3-4 µm band and a pulse duration of 20 fs can be reached with additional phase compensator.

18 Keywords: Mid-infrared pulse generation, Four-wave mixing, Anti-resonant hollow-core fiber

19

20 1. Introduction

21 Since these recent years, an interest is growing for the mid-infrared (MIR) sources driven by 22 demanding applications as gas sensing [1], food inspection [2], life and molecular sciences [3] or the 23 creation of secondary sources [4]. An excellent method for the light generation in the MIR is to 24 combine some laser properties with nonlinear photonic devices to create down-converted 25 frequencies. For example, high power sources can be made from several efficient nonlinear crystals, 26 chosen from their optimal phase matching conditions and absorption bands to achieve an optical 27 parametric amplification effect or difference frequency generation. Other few alternative devices at 28 lower power are based on other transparent materials as in silicon-, lithium niobate- or chalcogenide-29 based waveguides [5-7] or specific nonlinear fibers with soft-glass materials [8].

30 In this manuscript, we propose to generate ultra-short pulses in the MIR by pumping a gas-filled 31 hollow-core photonic crystal fiber (HC-PCF) with two different pulses to create an efficient phase-32 matched process. The HC-PCFs bring significant advantages to handle and convert ultra-short 33 pulses. Particularly, they offer a high damage threshold which allows the propagation of high peak 34 power [9]. In addition, the frequency conversion from the pump to the MIR can be engineered with 35 the dispersion of the waveguide and the gas contribution, i.e the phase matching can be tuned in real 36 time by changing the gas pressure. Outstanding progresses have also been achieved to reduce the 37 loss of HC-PCFs in the MIR [10,11], for example, with a transmission loss of ~25-50 dB/km now being 38 obtained by minimizing the interaction between the core guided mode and the silica walls of the 39 cladding of an inhibited-coupling hollow-core fiber [10]. Recent anti-resonant HC-PCFs have also 40 gained interest [12] due to their properties including a wide tunability of the spectral transmission 41 bands from the Ultra-violet (UV) to MIR [13,14,15] with low loss, high damage threshold [16] and a 42 single mode propagation by choosing specific geometries [17]. Several impressive works on ultra-43 short pulse generation in the MIR with gas filled HC-PCFs have been reported, such as pulse

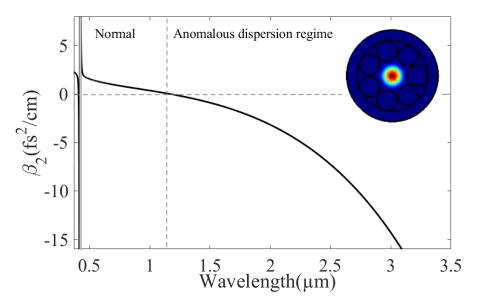
44 compression [18], supercontinuum and MIR dispersive wave generation [13,19,20] and adiabatic

- 45 down conversion from four wave mixing (FWM) processes in a tapered fiber [21]. In this later case,
- the HC-PCF is pumped by two picosecond pulses and seeded with a chirped signal to overlap with
- 47 the conversion spectral band. In our approach, only two ultra-short pump pulses are injected and
- 48 spectrally broadened in the HC-PCF. FWM are directly induced from the two broad-band pulses
- 49 capable to generate pulses at 3 and 4 μ m with a duration of 20 fs.

50 2. Anti-resonant fibers and numerical methods

51 2.1. Properties of the hollow-core photonic crystal fiber

- 52 The chosen HC-PCF geometry is a negative curvature fiber based on a single-ring structure 53 surrounding by the hollow-core. This type of fiber generally supports a large family of high-order 54 modes but in a specific condition (d=0.68D with d and D the tube and core diameters, respectively), 55 it can provide a single mode LP₀₁ operation [17]. A schematic view is shown in the inset of Figure 1 56 with our chosen parameters.
- 57 The HC-PCF structure is composed of a central hollow core with D=150 μm surrounded by seven
- 58 evenly and non-touching capillaries attached to the inner surface of a thick-walled capillary. The tube
- 59 diameter is set at 102 μ m to ensure a robust single-mode propagation. The wall thickness t equals to 60 0.2 μ m and the gap between tube g equals to 7.3 μ m. The LP₀₁ mode at 1030 nm supported by this
- 61 HC-PCF has also been simulated by the finite element method (insert in Figure 1) and it also confirms
- 62 the great advantage to propagate waves in the MIR with low loss since the overlap of the spatial
- 63 profile with the silica structure is weak even if the glass is highly absorbent in the MIR.
- 64 The group velocity dispersion, β_2 , is firstly analytically calculated for the fundamental mode from
- 65 empirical formulae [22] derived from a modification of the Marcatelli and Schmelzer's capillary
- 66 model [23] to take into account the anti-resonant reflection. Figure 1 shows the dispersion curve as a
- function of the wavelength for an HC-PCF filled with argon at a pressure of 5 bar. This pressure ischosen to obtain a zero-dispersion wavelength (ZDW) in the near infrared at 1170 nm. The effective
- 69 area A_{eff} ~992 µm² is also calculated from the model [22] and is assumed constant for the spectral
- 70 range.



71

Figure 1. Group velocity dispersion as a function of the wavelength for the HC-PCF filled with argon at a
 pressure of 5 bar. The grey area represents the resonant band. The inset represents a schematic view of the
 fiber with the LP₀₁ mode t 1030 nm.

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The general principle for MIR generation is to pump the HC-PCF with two different ultra-short pulses, P1 and P2, with spectra centered at 1030 nm and 515 nm corresponding to the emission of an

- 79 ultra-fast ytterbium doped fiber laser and a second harmonic generator. The two pulses lie in the
- 80 normal dispersion regime and the spectra broaden during the propagation in the fiber. Then, the MIR
- 81 light is generated from a phase matched FWM between several created spectral components. In order
- 82 to deeply understand the concept and the dynamics, we conducted numerical simulations by
- 83 integrating the generalized nonlinear Schrödinger equation
- 84 $\frac{\partial A}{\partial z} \sum_{n \ge 2} \frac{i^{n+1}}{n!} \beta_n \frac{\partial^n A}{\partial T^n} = i\gamma (1 + i\tau_{shock} \frac{\partial}{\partial T}) |A|^2 A$ (1)

85 describing the evolution along the fiber length z of the complex total electric field A(z,T).

86 The initial field corresponds to the total field composed of P1 and P2; i.e

87
$$A(z=0,T) = A_1 \exp(i(\omega_1 - \omega_c)T) + A_2 \exp(i(\omega_2 - \omega_c)T)$$
(2)

88 ω₁ (ω₂) and A₁ (A₂) are the angular frequency and the electric field amplitude of the pump P1 (P2),
89 respectively. In our simulation, we choose to center the frequency grid between the two pumps; at ω_c
90 corresponding to a wavelength of ~686 nm. Accordingly, we choose T in equation (1) as the time in

91 the frame moving at the group velocity β_{1c} at ω_c .

92 The series expansion coefficient of the dispersion term β is calculated in the frequency domain from

- the empirical model (section 2.1). The time derivative in the right-hand side is important to take into
- 94 account the shock term on a time scale of $\tau_{shock} = \frac{1}{\omega_c} \left[\frac{1}{n_{eff}(\omega)}\frac{\partial n_{eff}(\omega)}{\partial \omega}\right]_{\omega c}$ [24]. γ is the nonlinear term

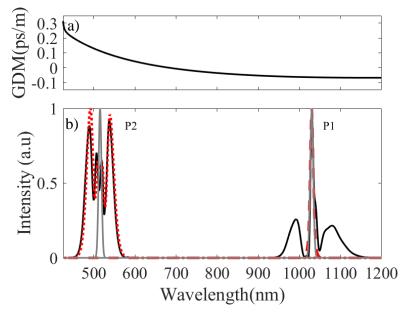
95 allowing to generate the new frequency components according to $\gamma = \frac{\omega_c \cdot n_2}{A_{eff} \cdot c}$ with n₂ the argon

- 96 nonlinear index equals to $4.86 \times 10^{-23} \text{ m}^2/\text{W}$ and c, the speed of light.
- 97

98 3. Continuum generation

99 3.1. Generated continuum from a dual-pump scheme

100 The light is generated in the MIR from a cascade of nonlinear processes in the argon filled HC-PCF 101 pumped by the two pump pulses. The initial pulses P₁ and P₂ have a duration of 180 fs and 50 fs at 102 Full-Width at Half Maximum (FWHM) at the Fourier transform limit. The corresponding spectral 103 bandwidth are $\Delta\lambda_1 \sim 9$ nm and $\Delta\lambda_2 \sim 8$ nm at FWHM. The group delay mismatch (GDM) between the 104 two pulses is ~176 fs/m (Figure 2.a) and we consider a total fiber length of 50 cm to ensure a temporal 105 overlap during the propagation. At the input fiber, the energies are 12 μ J for P₁ and 20 μ J for P₂ such 106 as the peak powers reach 63 MW and 390 MW. At first, each spectrum broadens along the fiber. 107 Figure 2.b shows the spectra for a 50 cm long fiber (black solid-line). For comparison, the spectra are 108 also displayed when only P1 (red dashed-line) or P2 (red dotted-line) is injected in the HC-PCF. 109 Clearly, the spectral broadening of P_2 is mostly due to self-phase modulation (SPM) since P_1 does not 110 affect significantly the spectral shape; i.e the spectra are similar with or without P1. Alternatively, 111 SPM has a negligible contribution on P_1 and the phase modulation on P_1 is induced by P_2 . This cross 112 phase modulation (XPM) allows to generate a broad-bandwidth centered at 1030 nm even for a 113 weaker pulse P₁[25].



114

115 Figure 2. (a) Group delay mismatch as a function of the wavelength. (b) Output spectrum for L=50 cm when P1

116 (red dashed-line), P2 (red dotted-line) or both pulses (black solid-line) are injected in the 50 cm long HC-PCF.

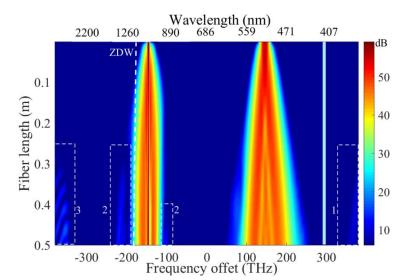
The input spectra are also displayed (grey solid-lines).



118 *3.2. Frequency generation mechanism and dynamics*

119 Figure 3 displays the total spectrum as a function of the fiber length with a logarithmic scale. During 120 the pulse propagation, the two pumps symmetrically undergo spectral broadenings due to the phase 121 modulations and a portion of the pump energy enters in the anomalous dispersion regime at z~10 122 cm. In addition, the spectral tail of the broadened pulse P_2 extends and overlaps with phase matched 123 frequencies in which the energy transfer process takes place. Other spectral components are 124 generated in the UV at ~374 nm (zone 1), in the near-IR (zone 2) and MIR (zone 3) due to the 125 interaction of the two main pulses with a large spectral bandwidth to achieve phase match processes 126 mostly from a FWM. For example, the radiation at (zone 1) results from the FWM process between

127 the pump (P₂) seeded with P₁ leading to an angular frequency of $2.\omega_2-\omega_1$.



128

Figure 3. Total spectrum as a function of the fiber length with a logarithmic scale. Zone 1, 2 and 3 correspond

130 to some generated bands in the UV, near IR and MIR. The light-blue line represents the absorption band of the

HC-PCF.

138 depends on the properties of the fiber, the gas and a nonlinear term :

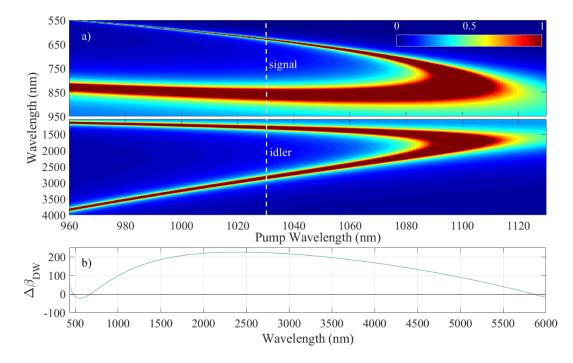
139
$$\kappa = \left\{ 2\beta_p(\omega_1) - \beta_s(\omega_s) - \beta_i(\omega_i) \right\} + 2\gamma P_{P1}$$
(3)

140 With $\beta_{p,s,i}$ the wave vector of each photon (pump, signal, idler) and P_{p1} the pump peak power. Figure

141 4 correspond to the analytically calculated coherence length, i.e $1/\kappa$, as the function of the pump 142

wavelength. This wavelength range corresponds to the bandwidth of the pulse P₁. For this estimation, 143 the peak power P_{P^1} is kept constant (W) although the instanstaneous power variation modifies the

144 phase matching condition.



145

146 **Figure 4**. (a) Coherence length $(1/\kappa)$ as a function of the pump P₁ wavelength for a constant peak power of 63

147 MW (from Eq. 3). (b) Phase mismatch of the dispersive wave ($\Delta\beta$ Dw) as a function of the wavelength for the pump P₂ (from Eq. 4).

148

149 The coherence length is maximum when the pump wavelength lies in the normal dispersion regime 150 $(\beta_2>0)$ and only a part of the P₁ spectrum plays a role in the FWM. Several bands arise around the 151 pump wavelengths. For example, two side-band pairs (signal/idler) are located at 0.85/1.2 µm and 152 0.62/2.8 µm for a pump wavelength sets at 1030 nm (vertical white lines in Figure 4.a). The locations 153 of these bands are in good agreement with those observed in the numerical simulations (zone 2 and 154 3, Figure 3). In addition, it is important to note that the visible band overlaps with the spectrum of P_2 155 that allows to generate the MIR band. The MIR spectrum obtained from the numerical simulations 156 (Figure 3) is displayed in Figure 5. It has a bandwidth (FWHM) of 1000 nm and exhibits several lobes 157 with maxima at 3.4 and 4.2 μ m. This structure is probably due to the modulated spectrum of P₁ that 158 directly influences the shape of the band [26].

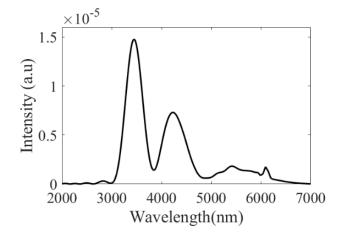


Figure 5. MIR spectrum at the HC-PCF output.

161

162 In addition to this wide spectral band, we can also note that a weaker peak arises at \sim 5.5 µm defined 163 as a dispersive wave (DW) seeded by spectral tail of the FWM band. This separate sideband radiation

as a dispersive wave (DW) seeded by spectral tail of the FWM band. This separate sideband radiation lies in the anomalous dispersion regime and is resonantly amplified by the pulse P₂ in the normal

165 dispersion regime [20,28], according to the phase matching condition of the DW given by [19] :

166
$$\Delta \beta_{DW} = \beta(\omega) - \left\{ \beta(\omega_2) + \beta_1(\omega_2) \left[\omega - \omega_2 \right] + \gamma P_{p2} \left[\frac{\omega}{\omega_2} \right] \right\}$$
(4)

167 with P_{P^2} is peak power of P_2 . This phase-mismatch is plotted in Figure 4.b. The perfect phase-matching 168 is obtained when $\Delta\beta_{DW}$ becomes zero at 5.8 µm in good agreement with the observed weak MIR band 160 (Figure 5)

169 (Figure 5).

170 As observed in Figure 3, the MIR band is only efficiently generated when the signal at ~620 nm is 171 created from the early-spectral broadening process of P2. Firstly, the P2 spectrum broadens during 172 propagation due to SPM and at ~30 cm (Figure 3), the spectral tail reaches the visible band of the 173 FWM and MIR waves are generated. In Figure 3, we also observed that the band at 1.2 µm arises at 174 z~30 cm once the signal at 850 nm arises from the spectral broadening of P₁. This MIR band evolves 175 during the propagation in the HC-PCF and their origins can be also discussed from the spectrogram 176 (Figure 6) where we show the time-frequency distribution of the total field at the fiber output. During 177 the propagation, the two pulses accumulated some temporal phase and therefore the instantaneous 178 frequencies is quasi-linearly spread in time. As the spectral broadening of P_1 is due to the XPM with 179 P₂, this chirp is only observed during the pulse overlap. Similarly, the additional bands (in the UV, 180 NIR and MIR) can be generated only once the two pumps overlap. Each pulse owns a defined spectral 181 bandwidth that evolves along the fiber (Figure 3). From Figure 6, we can also observe that the MIR 182 band has a spectro-temporal distribution. In fact, the pump and signal chirps are transferred to the

183 idler [27] creating the spectro-temporal distribution [26, 29].

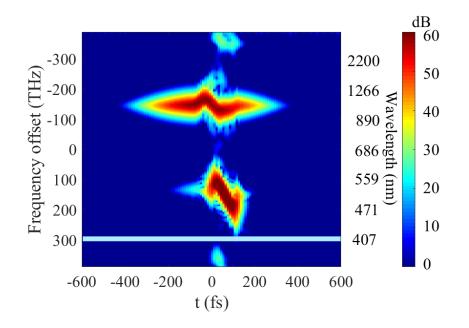
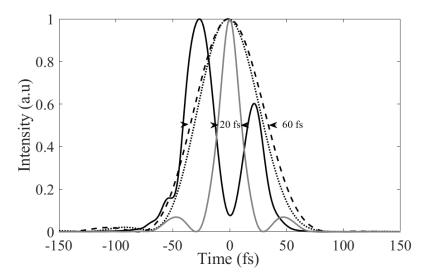


Figure 6. Spectrogram at the output HC-PCF. The light-blue line represents the absorption band of the HC PCF.

187 3.3 Pulse properties of the filtered mid-infrared continuum

188 At the output of the HC-PCF, the MIR band is selected with a flat-top bandpass filter and the pulse 189 shape is directly calculated. By selecting the MIR band between 3 and 5 μ m , the temporal shape 190 exhibits two peaks separated by 48 fs (blak solid-line in Figure 5). This structured temporal profile is 191 dued to the uncompensated phase acquired during the propagation and the nonlinear conversion in 192 the fiber. At the HC-PCF output, the accumulated phase can be compensated in order to reach an 193 ultra-short pulse duration of ~20 fs (grey solid-line). When each lobes are filtered, the temporal profile 194 has a bell-shape with a duration of ~60 fs at 3.4 µm or 4.2 µm without any phase conpensation (black 195 dashed and dotted-lines-Figure 3). An energy of few nJ is obtained at the fiber output. Therefore, this 196 scheme is highly desirable to develop a simple and robust pre-amplifier to seed subsequent ultra-fast

197 amplifier in the MIR.



198

Figure 7. Temporal profiles of the MIR band (black solid-line) and filtered spectra at 3.4 μm or 4.2
 μm (black dashed or dotted line). The grey solid-line corresponds to the pulse profile without the filter when the total phase is compensated.

202 5. Conclusion

203 We numerically demonstrated that ultra-short pulses at 3 and 4 µm can be generated through the 204 interaction of a fundamental pulse and its harmonics in a HC-PCF filled with argon. The generation 205 process originates from cascaded nonlinear phenomenon starting from spectral broadening of the 206 two pulses followed by an induced phase-matched four wave-mixing. The selected MIR band can 207 directly provide sub-60 fs pulses and a duration of ~20 fs is expected if a system of phase 208 compensation is used. This technique can be easily implemented since it relies on available laser 209 systems and state of the art anti-resonant fibers. The method is very promising to access the MIR 210 spectral ranges in the ultra-short pulse regime in a robust fiber system.

211

Author Contributions: C. F-D., O Z-M. and D.B. conceived, developed the model and performed the full
simulation; C.F-D and D.B analyzed the data and wrote the manuscript. D.B and PM supported this project. PM,
O. Z-M discussed the results, and reviewed the manuscript.

215

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218 Conflicts of Interest: "The authors declare no conflict of interest."

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