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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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11 **Abstract:** We show that ultra-short pulses can be generated in the mid-infrared when a gas filled
12 hollow-core fiber is pumped by a fundamental pulse and its second harmonic. The generation
13 process originates from cascaded nonlinear phenomenon starting from a spectral broadening of the
14 two pulses followed by an induced phase-matched four wave-mixing lying in the mid-infrared
15 combined with a dispersive wave. By selecting this mid-infrared band with a spectral filter, we
16 demonstrate the generation of ultra-short 60 fs pulses at 3-4 μm band and a pulse duration of 20 fs
17 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,
 46 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 μm with a duration of 20 fs.

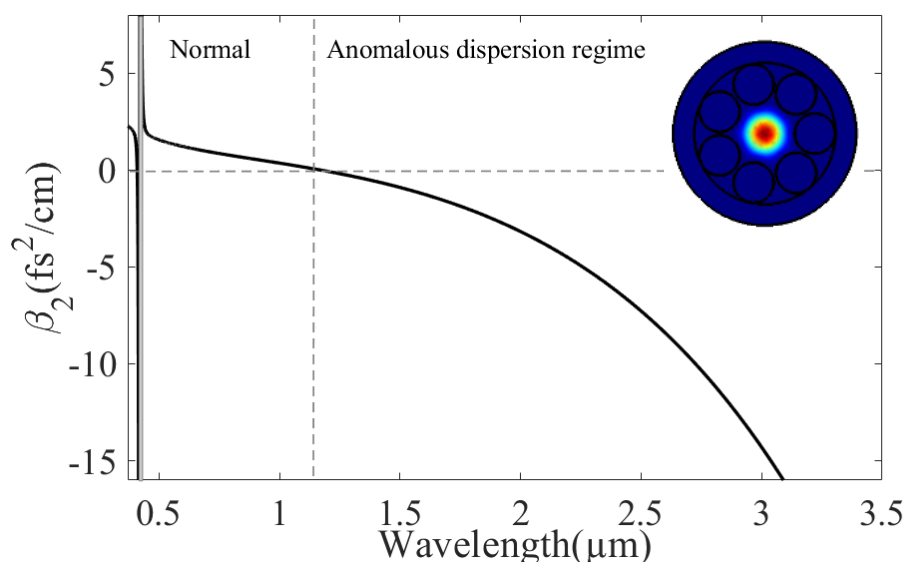
50 2. Anti-resonant fibers and numerical methods

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 LP_{01} 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 $D=150\ \mu\text{m}$ 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 $102\ \mu\text{m}$ to ensure a robust single-mode propagation. The wall thickness t equals to
 60 $0.2\ \mu\text{m}$ and the gap between tube g equals to $7.3\ \mu\text{m}$. The LP_{01} mode at $1030\ \text{nm}$ 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
 67 function of the wavelength for an HC-PCF filled with argon at a pressure of 5 bar. This pressure is
 68 chosen to obtain a zero-dispersion wavelength (ZDW) in the near infrared at $1170\ \text{nm}$. The effective
 69 area $A_{\text{eff}} \sim 992\ \mu\text{m}^2$ is also calculated from the model [22] and is assumed constant for the spectral
 70 range.



71
 72 **Figure 1.** Group velocity dispersion as a function of the wavelength for the HC-PCF filled with argon at a
 73 pressure of 5 bar. The grey area represents the resonant band. The inset represents a schematic view of the
 74 fiber with the LP_{01} mode at $1030\ \text{nm}$.

75

76 2.2. Numerical method for the nonlinear pulse propagation

77 The general principle for MIR generation is to pump the HC-PCF with two different ultra-short
 78 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

$$84 \quad \frac{\partial A}{\partial z} - \sum_{n \geq 2} \frac{i^{n+1}}{n!} \beta_n \frac{\partial^n A}{\partial T^n} = i\gamma(1 + i\tau_{shock} \frac{\partial}{\partial T}) |A|^2 A \quad (1)$$

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

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

88 ω_1 (ω_2) and A_1 (A_2) 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
 93 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 n_2 the argon

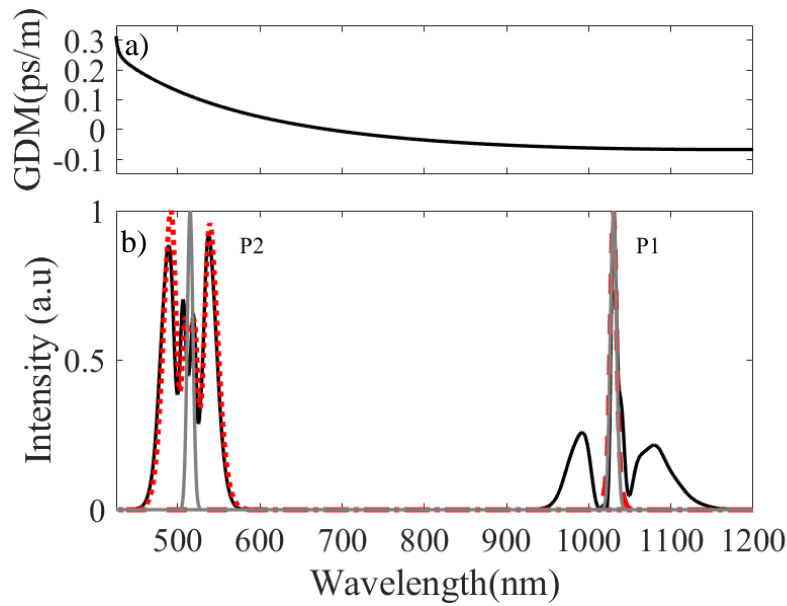
96 nonlinear index equals to $4.86 \times 10^{-23} \text{ m}^2/\text{W}$ and c , the speed of light.

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 μJ for P1 and 20 μJ 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 P2 is mostly due to self-phase modulation (SPM) since P1 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 P1 and the phase modulation on P1 is induced by P2. This cross
 112 phase modulation (XPM) allows to generate a broad-bandwidth centered at 1030 nm even for a
 113 weaker pulse P1 [25].

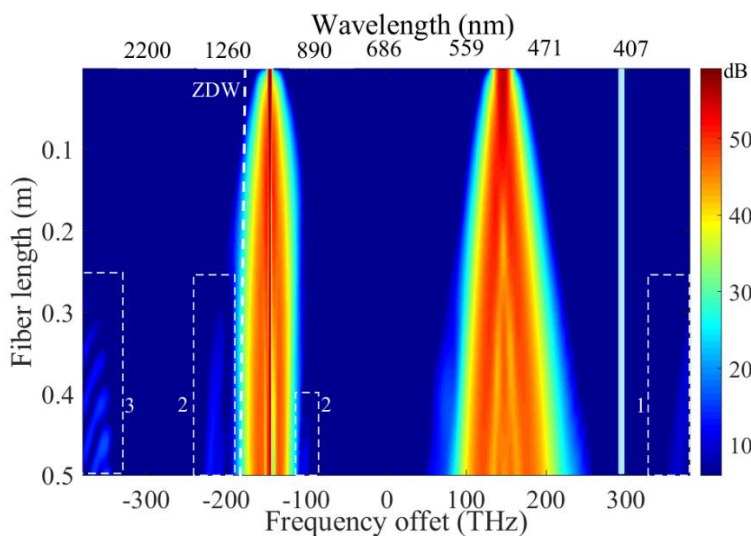


114

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.
 117 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 P₂ 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 $2\cdot\omega_2-\omega_1$.



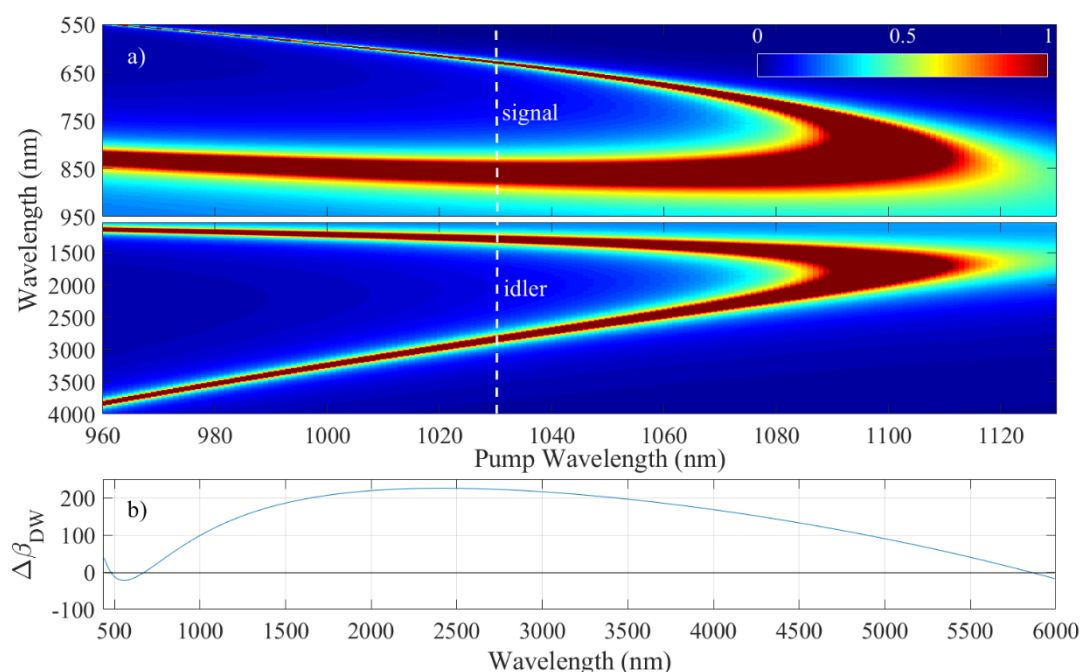
128

129 **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
 131 HC-PCF.

132 The other FWMs relies on a degenerated phase-matched process involving one strong pump pulse
 133 (i.e P₁) and a weak wave, at the angular frequency ω_s, which is originated from the broadening of the
 134 second pulse P₂. One idler band is generated in the MIR at the angular frequency ω_i, and then it will
 135 be selected with band-pass filters. In order to reach an ultrashort pulse, a broad-band FWM is
 136 required and we take benefit from a pump pulse (P₁) with a broad spectral bandwidth [26,27]. The
 137 phase-matching condition inside the HC-PCF, κ, is defined by the sum of a linear contribution that
 138 depends on the properties of the fiber, the gas and a nonlinear term :

$$139 \quad \kappa = \left\{ 2\beta_p(\omega_1) - \beta_s(\omega_s) - \beta_i(\omega_i) \right\} + 2\gamma P_{P_1} \quad (3)$$

140 With β_{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/κ, 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_{p1} is kept constant (W) although the instantaneous power variation modifies the
 144 phase matching condition.



145 **Figure 4.** (a) Coherence length (1/κ) as a function of the pump P₁ wavelength for a constant peak power of 63
 146 MW (from Eq. 3). (b) Phase mismatch of the dispersive wave (Δβ_{DW}) as a function of the wavelength for the
 147 pump P₂ (from Eq. 4).
 148

149 The coherence length is maximum when the pump wavelength lies in the normal dispersion regime
 150 (β₂>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 P₂
 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 μm. This structure is probably due to the modulated spectrum of P₁ that
 158 directly influences the shape of the band [26].

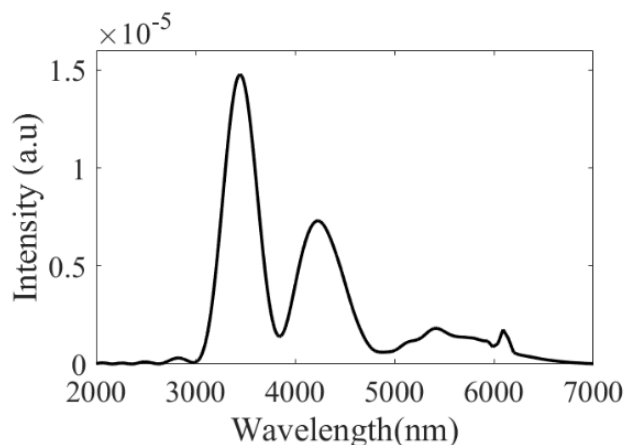


Figure 5. MIR spectrum at the HC-PCF output.

159

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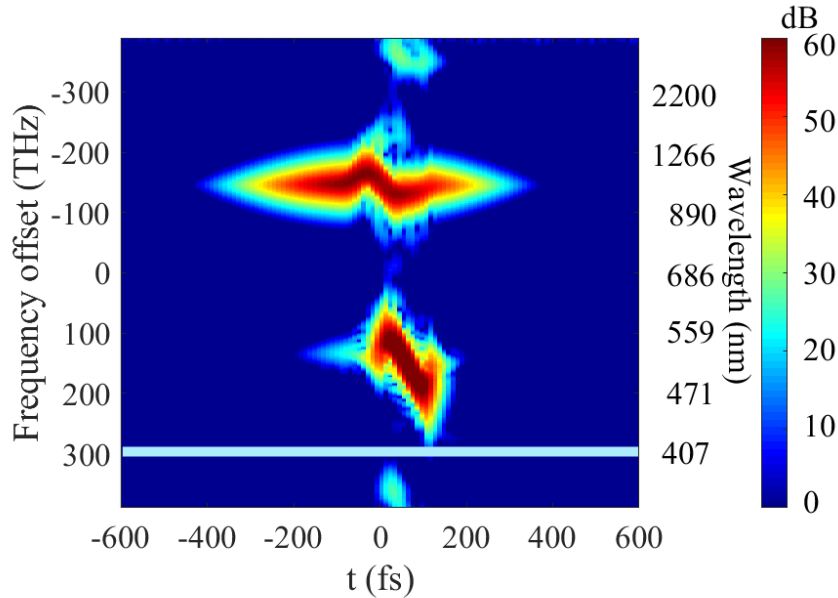
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162 In addition to this wide spectral band, we can also note that a weaker peak arises at ~5.5 μm defined
 163 as a dispersive wave (DW) seeded by spectral tail of the FWM band. This separate sideband radiation
 164 lies in the anomalous dispersion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

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

167 with P_{p2} is peak power of P₂. This phase-mismatch is plotted in Figure 4.b. The perfect phase-matching
 168 is obtained when Δβ_{DW} becomes zero at 5.8 μm in good agreement with the observed weak MIR band
 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 P₁ 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].

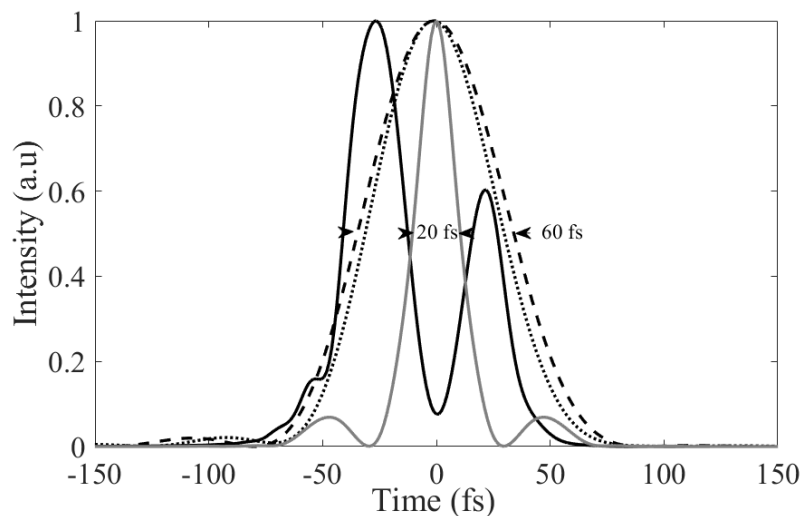


184

185 **Figure 6.** Spectrogram at the output HC-PCF. The light-blue line represents the absorption band of the HC-
186 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 μm , the temporal shape
190 exhibits two peaks separated by 48 fs (black solid-line in Figure 5). This structured temporal profile is
191 due 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 compensation (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

199 **Figure 7.** Temporal profiles of the MIR band (black solid-line) and filtered spectra at 3.4 μm or 4.2
200 μm (black dashed or dotted line). The grey solid-line corresponds to the pulse profile without the
201 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

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

215

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