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High harmonic generation in Solids driven by high energy fiber laser source

Djamila Boukhaoui^{*a}, Said Idlahcen^b, Jonathan Houard^a, Ivan Blum^a, Thomas Godin^b, Foued Amrani^c, Frédéric Gérôme^c, Fetah Benabid^{c,d}, David Gauthier^{e,f}, Willem Boutu^{e,f}, Hamed Merdji^j, Ammar Hideur^b and Angela Vella^a

^aUniv Rouen Normandie, INSA Rouen Normandie, CNRS, GPM UMR 6634, F-76000 Rouen, France. ^bUniv Rouen Normandie, INSA Rouen Normandie, CNRS, CORIA UMR 6614, F-76000 Rouen, France. ^cGLOphotonics, 123 avenue Albert Thomas, 87060 Limoges Cedex, France. ^dGPPMM Group, XLIM Research Institute, CNRS UMR 7252, University of Limoges, 87060 Limoges, France. ^eUniversité Paris-Saclay, CEA, LIDYL, 91191 Gif sur Yvette, France, ^fCY Cergy Paris Université, CEA, LIDYL, 91191 Gif sur Yvette, France. ^jLaboratoire d'Optique Appliquée, CNRS, Ecole Polytechnique, ENSTA Paris, Institut Polytechnique de Paris, Palaiseau, France

ABSTRACT

We present findings on high harmonic generation (HHG) in solids utilizing a high-energy fiber laser system operating at 1550 nm. The driving laser source comprises an erbium-doped fiber chirped pulse amplifier (EDFA) combined with a post-compression stage employing a hollow-core photonic crystal fiber (HC-PCF) filled with noble gases. Nonlinear self-compression in the HC-PCF enables the generation of ultrashort pulses with a duration of 50 fs and energy of 0.91 μ J at a repetition rate of 660 kHz. In a first step, harmonics up to H7 were observed when focusing the laser into small bandgap materials such as Zinc Oxide (ZnO). Subsequently, the system was enhanced to measure high harmonics in the extreme ultraviolet (XUV) range, with harmonics up to H25 observed using a large bandgap material, magnesium oxide (MgO). To the best of our knowledge, this represents the first solid-state HHG source driven by a high-energy few-cycle fiber laser in the telecom region.

Keywords: High harmonic generation, pulse self-compression, fiber laser, hollow core fibers

1. INTRODUCTION

High Harmonic Generation (HHG) refers to a nonlinear optical process where intense laser light interacts with atoms, molecules, plasma or solids. In this phenomenon, the target system emits light at frequencies equal to integer multiples of the frequency of the driving laser [1,2,3]. HHG is a promising research area from both fundamental and practical points of view and have enabled the generation of coherent extreme-ultraviolet radiation that has pioneered numerous applications. Solid-state HHG has gained attention for its potential to control and customize radiation properties. In solids, the mechanism involves inter- and intraband electronic dynamics, resulting in more efficient harmonics compared to gas. The coupling between electrons and holes allows excited electrons to recombine in neighboring sites, strengthening the HHG spectrum [3]. Additionally, it offers more degrees of freedom to act on the HHG emission by engineering the crystal structure, chemically or mechanically [4-7]. HHG strongly depends on the wavelength driving laser : while in gases the high harmonic cut-off depends on the square of the driving wavelength strength, it depends linearly on the laser wavelength in solids [8,9]. The use of long wavelength pulses (Mid IR) extends the cut-off [8,9]and increases damage threshold of bulk crystals Different driver laser sources have been used in the Mid IR region, mostly high power optical parametric chirped pulse amplifiers (OPCPA) [3,10-12] and optical parametric amplifiers (OPA) [13-17]. These systems offer high energy levels but they present very complex configurations.

The use of ultrafast fiber lasers in the mid-IR is a very promising route for the development of ultra-compact HHG platforms. This has been confirmed by several demonstrations of HHG driven by few-cycle mid-IR fiber lasers [7,18]. However, these sources based on soliton self-frequency shift in all-solid fibers are limited to few-10 nJ [7,18]. The best solution to obtain high energy levels in a fibered configuration involves the use of post-compression in hollow core (HC)

photonic crystal fibers (PCF) filled with rare gases. Such fibers provided the best self-compression qualities due to their high nonlinearity versus ionization rate ratio and do not exhibit any delayed Raman response compared to traditional pulse compression methods, as they use an atomic gas. Moreover, the dispersion of the waveguide is an additional degree of freedom for pulse propagation tailoring. Working in the anomalous dispersion regime indeed offers the possibility to generate Fourier limited pulses directly from the fiber, due to the interplay between self-phase-modulation-induced spectral broadening and simultaneous temporal compression due to the anomalous waveguide dispersion [19].

2. EXPERIMENTAL SETUP AND RESULTS

The schematic diagram of our HHG laser system is depicted in Fig. 1. The laser source is based on nonlinear pulse compression in a HC-PCF filled with gas and pumped by an erbium-doped fiber chirped-pulse amplifier system delivering sub-picosecond pulses with 600 fs duration and up to 3 μ J energy at 660 kHz repetition rate. The elliptically polarized beam is injected into the post compression stage based on a 2 m long hybrid Kagome HC-PCF (33 μ m diameter, $\beta_2 \approx -6300$ fs²/m @ 1550 nm) [20] filled with argon. We use a three lenses scheme to adapt the beam size and optimize the injection efficiency. The collimated beam at the output of the HC-PCF passes through a half wave plate ($\lambda/2$) and a thin film polarizer (TFP) that is used in combination with the waveplates before the HC-PCF to temporally purify the output beam from any residual light propagating in high-order modes and thus to enhance the output pulse contrast.

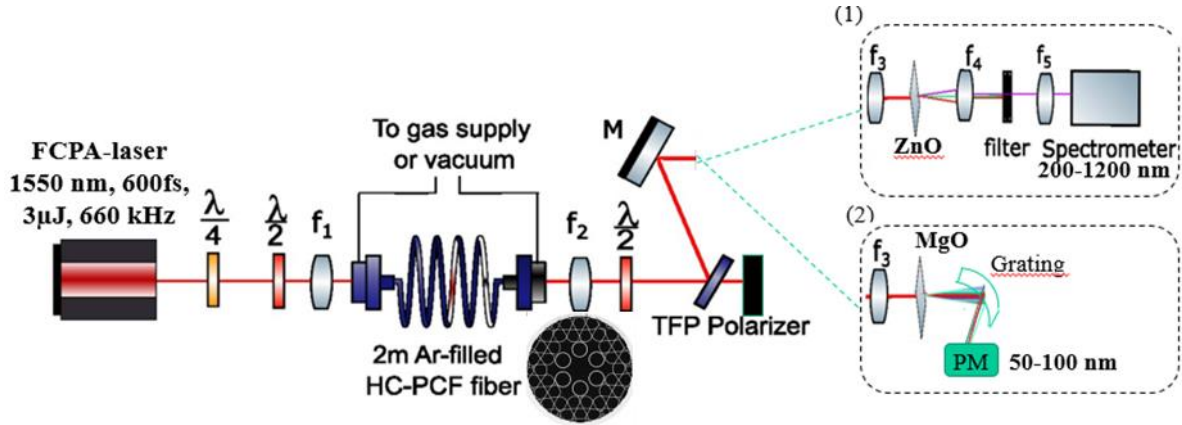


Figure 1. Experimental setup for the pulse self compression in the hollow core fiber and high harmonic generation in solids: (1) measurement in air and (2) measurement in vacuum. The FCPA laser consists in an erbium-doped fiber chirped-pulse amplifier system (CALMAR company, FLCPA-01C), f_1 is formed by a telescope ($f = 50$ mm, $f = 125$ mm) and lens of $f = 75$ mm to inject the beam into the hybrid Kagome hollow core photonic crystal fiber "HC-PCF (GLOphotonics company) filled with argon gas. $f_2 = 79$ mm, TFP: thin film polarizer, $f_3 = 35$ mm, $f_4 = 25$ mm, $f_5 = 50$ mm, different filters are used to select a specific harmonic order generated in the ZnO sample crystal in (1). In (2) the emitted radiation generated in a MgO crystal is recorded using an XUV spectrometer, which consists of a diffraction grating and a photomultiplier (PM).

In our setup, pulse self-compression is reached for a gas pressure of 13 bar and an input energy of 2.5 μ J with 600 fs pulses. At the HC-PCF output, we achieved strongly SPM-broadened spectra spanning from 1412 nm to 1675 nm as shown in Fig. 2.a. The corresponding autocorrelation trace shown in Fig. 2.b reveals a FWHM pulse duration of 50 fs, assuming a Gaussian shape. The output energy after polarization filtering is 0.92 μ J.

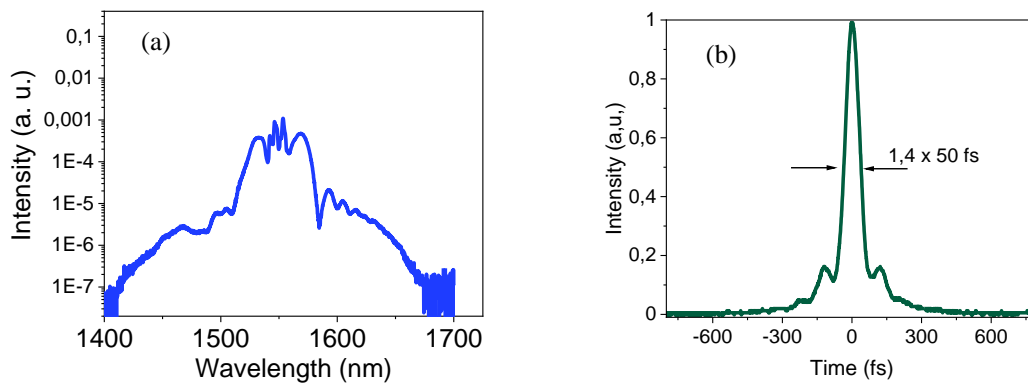


Figure 2. Experimental results for the output laser characteristics: (a) optical spectrum and (b) autocorrelation trace

To explore HHG, the post-compressed pulses were focused onto a 500 μm thick ZnO sample (spot size of 20 μm). The intensity at focus was estimated to 5.8 TW/cm^2 . HHG spectra measured using an Ocean Optics (Maya 2000 pro, 200 nm-1200 nm) spectrometer reveal the generation of perturbative and non-perturbative harmonics extending well above the band gap ($\Delta E=3.3$ eV) and luminescence generated at the excitonic band edge centered at 385 nm as shown in figure 3-a. In this experiment, we were limited to the measurement of H7 (seventh harmonic) due to our detection system.

The experimental setup is then updated in order to measure higher harmonics in the XUV range. The experiment, sketched in Fig. 1-(2), is carried out in vacuum. The laser beam is focused at normal incidence onto a 200 μm thick MgO bulk sample. The harmonic signal is optimized by moving the sample on a motorized translation stage along the optical axis. The crystal-orientation dependence of the HHG signal is investigated by rotating the crystal on a motorized rotation stage around the laser-propagation axis.

The interaction of the intense laser pulse with the MgO (band gap of 7.8 eV) sample results in the emission of coherent high-order harmonics in the XUV range, which co-propagate with the driving laser. The fundamental beam and high order harmonics are spectrally resolved by a home-made spectrometer composed of a reflective concave diffraction grating (McPherson 234/302, 2400 grooves/mm, Pt coated) and a photomultiplier. The range of detected harmonics spans from order 17 (13.6 eV) to 25 (20 eV).

Figure 3-b shows the first result of the measured HHG spectra from MgO at the laser intensity $I=3$ TW/cm^2 . As can be seen, nonperturbative harmonic up to H25 that well exceed the material band gap are generated. In the future, further investigation is required to fully characterize the harmonic yield.

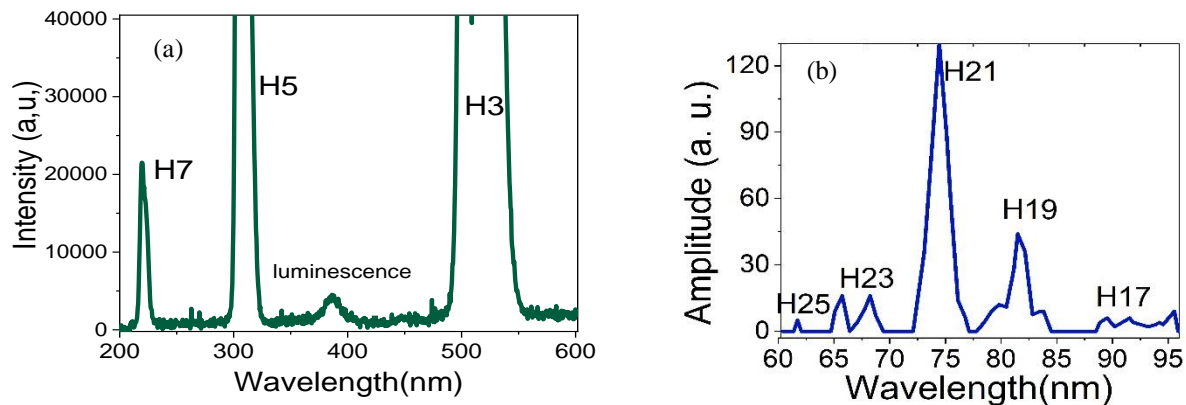


Figure 3 : Harmonics spectra emitted by (a) ZnO in the visible-UV range and (b) MgO in the XUV range.

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