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Review

Intense Few-Cycle Infrared Laser Pulses at the Advanced Laser Light Source

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To push the generation of isolated attosecond pulses towards shorter XUV wavelengths (0.3 to 1 keV), intense few-cycle driving pulses are required whose center frequency is significantly red shifted compared to established Ti-Sa technology. A simple route for generating these pulses is demonstrated using an optical parametric amplifier (OPA). Its infrared (IR) multi-cycle laser pulses with millijoule of energy are spectrally broadened by nonlinear propagation in a hollow-core fiber (HCF) filled with Argon. For the OPA Signal wavelength (λ = 1.4 micron), chirped mirrors were used for dispersion compensation with a resulting pulse duration of 13.1 fs. For the OPA Idler wavelength at 1.8 micron, a new compression scheme has been developed with which 11.2 fs laser pulses have been characterized (1 cycle = 6 fs). Here, pulse compression is achieved through the combined effects of self-steepening during nonlinear propagation in the HCF followed by anomalous dispersion during linear propagation in fused silica.

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I. INTRODUCTION

The discovery of X-rays in 1895 by Dr. William Roentgen has been an important step towards major scientific breakthroughs in the last century. One can hardly imagine the development of society without the progress made in medicine by providing physicians with unique tools for X-ray diagnostics and treatments. In scientific research, X-rays are used for spectroscopic and structural characterization of atoms, molecules, biological structures and materials. Today, researchers from a broad diversity of scientific horizons anticipate using ultrashort X-ray laser pulses to probe dynamical processes with combined high spatial and high temporal resolutions. This quest motivates the construction of X-ray free electron laser (XFEL) facilities providing femtosecond X-ray laser pulses of high brilliance [1].

In parallel to the development of those infrastructures for ultrafast X-ray science

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research, there are major ongoing activities to develop table-top approaches [2–5]. Among them, X-ray sources based on the process of high harmonic generation (HHG) [6–9] provide the ultimate temporal resolution with a world record of 67 attoseconds pulse duration [10]. With those pulses, researchers now possess a unique metrology to probe ultrafast electronic dynamics [11–15]. Currently, this technology has been developed in the spectral range of 20 to 100 eV, which limits the temporal resolution but also important applications where higher photon energies are required [16]. So far, isolated attosecond pulses have been generated using Titanium-Sapphire (Ti-Sa) laser technology which is centered at 800 nm wavelength. Based on the three-step model introduced by Corkum in 1993 [17], the maximum photon energy obtained from HHG is given by I_p+3.17Up with $Up \sim I\lambda^2_{\text{laser}}$, where I is the intensity and λ_{laser} the laser wavelength. From this simple formula, one may conclude that an ever increasing intensity would provide higher and higher photon energy. This is neglecting the fundamental coherent nature of HHG where the emission arises from the interference of the returning electron wavepacket with the bound atomic one. The latter must not be fully depleted by ionization, which will prevent emission, thus limiting the maximum intensity (I) that can be used. For helium, this intensity is in the range of 1×10^{15} W/cm^2, corresponding to a Up of 60 eV at 800 nm. Thus, with Ti-Sa, the maximum photon energy expected is in the range of 200 eV. Phase matched HHG from helium using the capillary approach has been extended up to 150 eV [18]. Therefore, to further increase the maximum photon energy from HHG, one has to increase λ_{laser}. This was first illustrated by Shan and Chang [19], followed by several groups [20–23]with now a maximum photon energy of 1.6 keV by using a driving laser field at 3.8 micron [24]. Despite this great progress, isolated attosecond pulses in the sub-keV spectral range have not yet been demonstrated. This requires the development of intense few-cycle infrared laser pulses.

In contrast to other research groups worldwide working on the development of few-cycle infrared laser sources [25–28], we have decided to develop this technology using spectral broadening of millijoule multi-cycle OPA laser pulses through propagation in a HCF filled with argon. Intense few-cycle pulses have been generated at both the Signal and Idler wavelengths [29–31]. The use of HCF to spectrally broaden pulses, followed by pulse compression at the output, was first introduced by Nisoli and co-workers for a Ti-Sa (800 nm) laser system [32]. In this proof of concept experiment, pulse compression was achieved using a prism pair. Nowadays, chirp mirrors allow the generation of 1.5 cycle pulses [33]. It is important to note that this development in the late 90s has been one of the key steps, together with carrier envelope phase stabilization [34, 35] and the attosecond streak-camera [36], to enable the generation and the characterization of attosecond pulses in 2001 [6].

II. METHODOLOGY

The long wavelength few-cycle laser sources (1.4 and 1.8 micron) have been integrated to the 800 nm, 100 Hz, 100 mJ, 35 fs, Titanium-Sapphire (Ti-Sa) laser beamline of the Advanced Laser Light Source (ALLS, located at INRS-ÉMT). Using an OPA based on parametric superfluorescence (He-TOPAS, Light Conversion), we convert 6 mJ from the

800nm beamline to the Signal and Idler wavelengths, shown as red and blue lines in Figure 1.

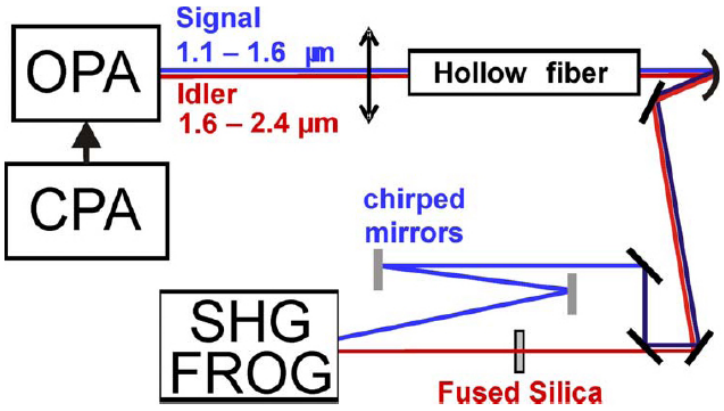

FIG. 1: Experimental layout for the generation of intense few cycle infrared laser pulses. Millijoule level OPA pulses are broadened in a hollow-core fiber and subsequently compressed by chirped mirrors in the Signal spectral range. Idler laser pulses are compressed using the anomalous dispersion provided by 3 mm of fused silica.

Typical pulse energy is 1.3 mJ and 0.9 mJ, for the Signal and Idler respectively. By using different wavelength separators either Signal or Idler laser pulses can be selected for the experiment as shown in Figure 1. In both cases energy fluctuations of the OPA are about 3% RMS, and the laser pulse duration is about 60 fs. The beam is coupled into the HCF setup using an f = 1 m focusing lens. The fiber diameter is 400 m with a length of 1 m. It is installed in a closed gas cell and supported on an aluminum V-groove. Typical pressure ranges from 1–2 bars are used and the fiber transmission efficiency is between 40% and 60% depending on the mode quality of the OPA. At the fiber output, the laser beam was collimated using an f = 1 m concave silver mirror. The confined propagation in the fiber provides conditions for nonlinear propagation where the light pulse itself modifies the medium properties through which it propagates. As a consequence the refractive index of the medium becomes intensity dependent and hence time dependent which causes a temporal variation of the instantaneous phase, known as self-phase modulation (SPM). Following the basic relations of Fourier transformation this temporal variation of the phase is associated with spectral broadening in the frequency domain. That means the HCF output spectrum is significantly broadened with respected to the input spectrum as can be seen in Figure 2.

After spectral broadening, the laser pulses have to be compressed in time, which is achieved in two ways; with chirped mirrors for the Signal [29] and using the anomalous dispersion of bulk material for the Idler [30, 37]. Those approaches are described in the next section. Replacing the OPA based on parametric superfluorescence by a white light seed version enabled to passively stabilized the carrier envelope phase of the Idler pulses [23,

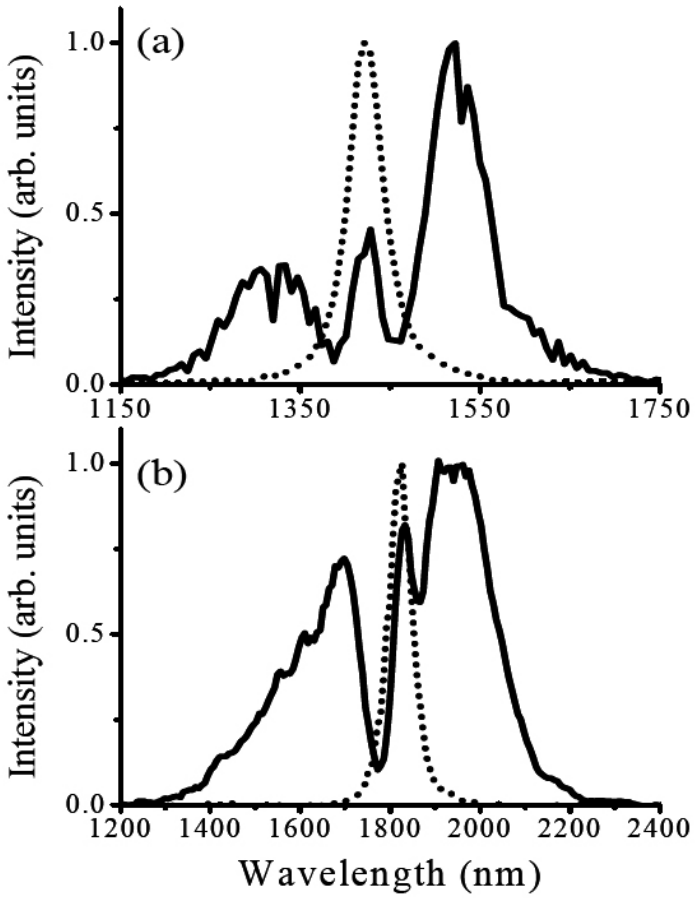

FIG. 2: OPA spectra for (a) Signal wavelength (1425 nm) and (b) Idler wavelength (1823 nm). Dotted curve: Spectra at the output of the OPA. Solid line: Spectra at the output of the hollowcore fiber.

31, 38].

Temporal characterization of the intensity profile of the few-cycle pulses has been achieved with a home built dispersion free second harmonic generation frequency resolved optical gating (SHG-FROG) [39]. To avoid dispersion we do not use a traditional beam splitter. Instead we divide the beam geometrically using a pair of 2 mm diameter pinholes that are separated by 4 mm. The resulting beams are delayed with respect to each other and focused in a non-collinear geometry with an f = 500 mm silver mirror. A very thin type I BBO crystal ($\Phi = 21°$) of 10 μm thickness guarantees ultra broadband phase matching over the whole spectral bandwidth of our pulses.

III. RESULTS

First we describe results achieved when coupling the OPA Signal pulses into the HCF and subsequent compression with chirped mirrors. Because the spectral phase introduced by SPM can be approximated by a quadratic function (positive group velocity dispersion (GVD)), it can be compensated by negative GVD as provided by chirped mirrors. However, those mirrors were not commercially available in the Signal wavelength range and first had to be designed in collaboration with Femtolasers (Vienna, Austria). Each mirror consists of 73 alternating layers of SiO_2 and Nb_2O_5 to provide sufficient reflectivity ($R > 99.5\%$) over a bandwidth ranging from 1000–1700 nm. This multi layer design provides a large GVD of -300 fs^2 (per roundtrip) and a third order dispersion (TOD) of -15 fs^3 at 1425 nm per single bounce. Fine tuning of dispersion was achieved by introducing small amounts of positive dispersive material like SF10 into the beam.

In Figure 3, we present the retrieved temporal intensity profile of the laser pulses. Those results have been obtained by combining one round trip on the chirped mirrors with ~ 1.5 mm of SF10. The fact that the obtained FWHM duration of 13.1 fs is only 1.07 times the Fourier limit (12.2 fs assuming flat spectral phase) proves the good compressibility of the broadband spectra at 1425 nm. At 1425 nm, 13.1 fs is less than 3 optical cycles, equivalent to about 7.5 fs at 800 nm. Moreover, we have calculated that 70% of the energy is concentrated in the central peak of the laser pulse temporal intensity profile. The direction of the time axis was determined by overcompensating the dispersion with one round trip on the chirped mirrors and added positive dispersion with SF10 windows.

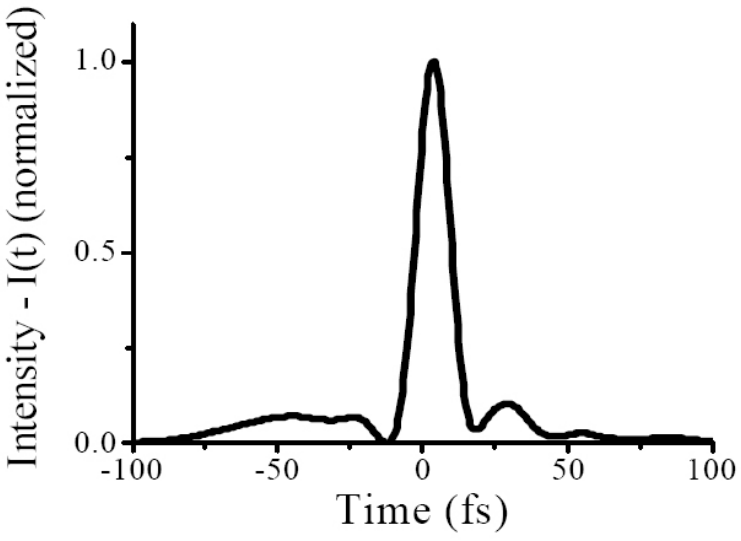

FIG. 3: Retrieved intensity profile for the sub three cycle Signal laser pulses. Full width half maximum is 13.1 fs corresponding to 2.7 optical cycles.

After discussing the established approach for pulse compression via spectral broaden-

ing in HCF followed by dispersion using chirped mirrors, we now describe a new compression scheme in the Idler spectral range which utilizes the anomalous dispersion of bulk material. The GVD and third order dispersion (TOD) curves for the glasses used in this setup are shown in Figure 4 from which one can deduce several things. First, bulk material like fused silica (FS) provides negative GVD predominantly in the Idler spectral range. Thus, it cannot be used to compress the Signal laser pulses to sub three cycle pulse duration. Second, CaF2 exhibits the lowest absolute GVD and TOD in the spectral range of interest and hence was chosen as material for transmissive optics like lenses and cell windows. Third, regardless of the sign of GVD all glasses provide only positive TOD throughout the entire spectral range.

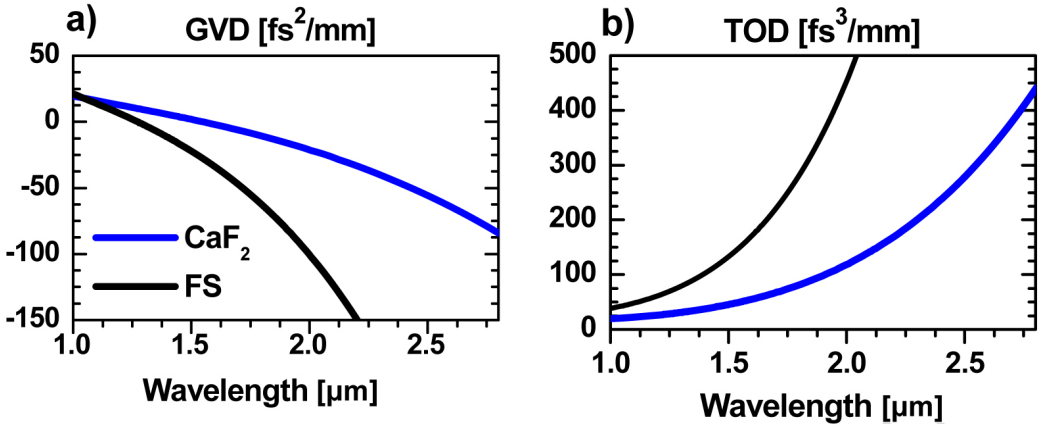

FIG. 4: GVD (a) and TOD (b) curves for the transmissive optics used in the setup. The negative GVD of FS in the anomalous dispersion regime can be utilized for sub-two cycle pulse compression if the positive TOD is compensated by self-steepening during nonlinear propagation in the fiber prior to linear propagation in the glass.

That means dispersion with bulk material will be limited by TOD which never becomes negative. Surprisingly, the compressed pulse of 11.5 fs shown in Figure 5 is very close to the transform limit of 10.1 fs. For the corresponding experimental spectrum of Fig. 2 one can calculate that the TOD of 3 mm FS and 1 mm CaF_2 (cell window at the HCF output) would broaden the pulse to about 14.9 fs which is much longer than the experimentally observed duration.

The explanation for this unexpected result, which cannot be provided by the bulk material alone, lies in the nonlinear propagation itself. SPM is not the only effect taking place in the fiber. Self-steepening is also present in which not only the gas medium is modified as function of laser pulse intensity but also the time dependent medium couples back to the temporal properties of the driving pulse causing an asymmetric power-spectral shape and phase. The later is the key ingredient for subsequent pulse compression with bulk material because the spectral phase asymmetry due to self-steepening opposes the

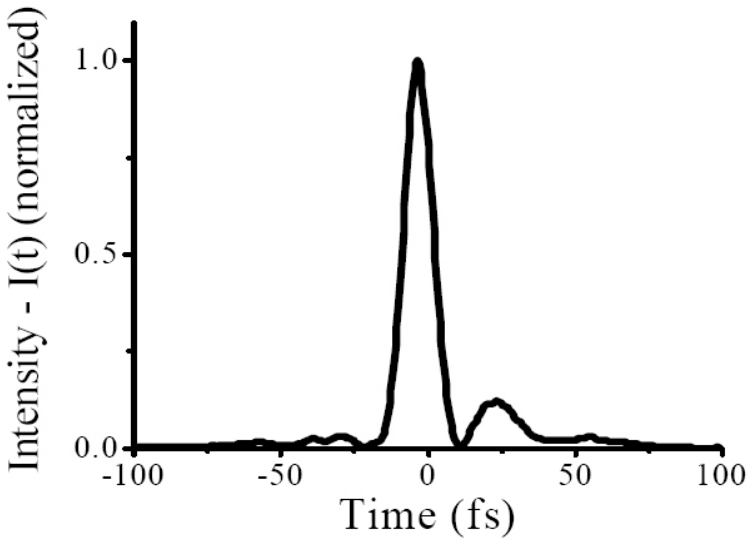

FIG. 5: Retrieved intensity profile for the sub two cycle Idler laser pulses. Full width half maximum is 11.2 fs corresponding to 1.9 optical cycle.

positive TOD component of the glass. This was confirmed through numerical simulations where experimental results were reproduced with a level of accuracy [30, 37].

IV. CONCLUSIONS

One of the motivations to develop intense few-cycle infrared laser pulses is to use the process of HHG to enable the generation of shorter XUV wavelengths (100 eV to 1 keV) laser pulses, with femtosecond to attosecond duration [16]. The establishment of such a table-top ultrafast X-ray source will open new scientific horizons by providing the ultimate temporal resolution to probe ultrafast processes in the condensed matter, enabling applications that are nowadays not possible. Among a large variety of possible applications, the water window spectral range (280 to 540 eV) is highly attractive for biological imaging [40], and the range of 700 to 900 eV is in high demand to study of ultrafast demagnetization of ferromagnetic materials [41]. In addition to those applications, intense few-cycle infrared laser pulses provide high cut-off for low ionization potential atoms and molecules thus are ideal to perform HHG spectroscopy [23, 42, 43], and to control of molecular electronic dynamics in intense laser fields [44].

To conclude, we are convinced that this large variety of applications for intense few-cycle infrared laser pulses makes compression using the HCF approach very attractive for the ultrafast community considering that the only requirement is combine Ti-Sa systems with a commercially available OPA.

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