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Compensation of second-order dispersion in femtosecond pulses after filamentation using volume holographic transmission gratings recorded in dichromated gelatin

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Abstract We have designed and developed a pulse compressor with volume transmission holographic gratings to be implemented in post-compression experiments based on filamentation in gases. Pulse compression down to 13 fs has been demonstrated. The gratings have been recorded in commercial PFG-04 dichromated gelatin emulsions with a recording wavelength of 532 nm, attaining sufficient index modulation to achieve high efficiency when they are illuminated by an 800-nm laser.

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1 Introduction

During the mid 1990s, post-compression techniques were adapted to the mJ regime by means of the hollow-fiber system [1]. This technique opened the path toward very short laser pulses. Some years later, the filamentation process was also applied to obtain few-cycle laser pulses [2]. In both cases, nonlinearities in the propagation medium are used to broaden the laser spectrum. In the case of filamentation, for example, when a femtosecond laser beam (propagating in air or other gases) is focused by a lens and the laser power exceeds the critical power, a balance between the Kerr effect, which leads to beam focusing, and the defocusing effect due to the plasma generated during the collapse of the beam forms a filament of light [3–5]. In this process, the spectral bandwidth of the laser pulses is increased due to all the nonlinear effects present during the propagation. Once a broad spectrum has been obtained, it is possible to post-compress the pulses after compensating their spectral phase. Typically, this compensation is achieved by means of chirped mirrors or prism compressors. In the present work, we study the possibility of compensating the GDD (group delay dispersion) after the filamentation stage by using volume transmission holographic gratings (VTHGs).

Other kinds of transmission gratings can be used to construct compressors, especially to be implemented in chirped pulse amplification (CPA) configurations [6]. For example, relief gratings recorded in fused silica with electron beams or ion etching exhibit high performance and are commercially available. We have chosen VTHGs because they can be recorded in a conventional holography laboratory and also because the gratings can be tailored with the suitable spatial frequency and size for each application at low cost.

In previous works [7, 8], VTHGs have been used to design compressors to be implemented in CPA configurations, in which a large compression factor is required, and consequently gratings with high groove density. These gratings can introduce a significant amount of angular dispersion with high efficiency, but the drawback is the narrowing of the bandwidth of the spectrum transmitted when the spatial period decreases, which limits their use to pulses of tens of femtoseconds.

However, in post-compression experiments, where large compression factors are not required, the use of VHTGs allows the design of compact and versatile compressors. In this case, it is necessary to study how the chromatic selectivity of the volume gratings might affect the compression when they are used with pulses with broad spectrum.

Some of the materials that have been used successfully to record volume gratings are dichromated gelatin (DCG) emulsions, owing to their optical characteristics, such as a high dynamic range of refractive-index modulation [9, 10], a high diffraction efficiency (with values close to 100%), low scattering, low absorption, and high resolution [11]. Since DCGs' plates have a high damage threshold, they can be used with high-energy pulses [8]. Currently, there are commercial DCG plates available, manufactured by the Slavich Company and commercialized as PFG-04 [12]. The values of index modulation that can be reached in this emulsion are sufficient to record volume transmission gratings with high efficiency when the wavelength of reconstruction is 800 nm. Accordingly, they can be used with a femtosecond Ti:Sa laser source [13].

Here we have designed and developed compressors with two VTHGs recorded on Slavich PFG-04 plates. With these commercial emulsions, it is possible to record multiple replicas of tailored gratings at low cost. These compressors were designed to work in post-compression experiments involving two femtosecond laser systems: a CPA laser from Spectra Physics (Spitfire Ti:Sa laser, 10 Hz, 10-nm bandwidth centered at 795 nm) and a CPA laser from Amplitude Technologies (10 Hz, 55-nm bandwidth centered at 805 nm).

The structure of this paper is as follows. In Sect. 2 we report some tests of the volume grating compressor with pulses before the post-compression step in order to study the bandwidth limitation of the gratings and how to improve it. In Sect. 3 we focus on the post-compression setup and the broadened spectra, designing some specific new volume holograms able to work under those conditions and presenting the compression results obtained. Finally, we end in Sect. 4 with the conclusions.

2 Design and characterization of the pulse compressor

The compressors developed here were used to compress pulses whose spectra were around 800 nm, such that the gratings had to be highly efficient at this wavelength. In a previous work [13], we characterized the Slavich PFG-04 dichromated gelatin emulsions, recording gratings with a laser source emitting in the green spectral region ($\lambda = 532$ nm), where they have enough sensitivity without sensitizing dyes. The dynamic range of the emulsion allowed us to record highly efficient gratings when they were illuminated at a wavelength different from the one used in the recording, for instance at 800 nm. The emulsion is coated on a glass substrate 2.5-mm thick and it is necessary to seal the grating after the processing step with a glass plate (we have used one of 1.6-mm thickness).

In order to assess the usefulness of VTHGs in ultra-high-power pulse compressors, the damage threshold of the emulsion under femtosecond pulse exposure must be characterized. The measurement of this threshold is very difficult provided that it depends strongly on irradiation conditions such as the pulse duration or the beam size, and no standard procedure exists so far. As a first approach, we have determined the ablation threshold of the emulsion when exposed to 120-fs pulses (795-nm central wavelength) focused with a $f = 100$ mm lens (spot diameter at the focus was 11.3 μm). We followed the technique described in [14] and the values obtained were 0.74 J/cm^2 for single-pulse ablation and 0.34 J/cm^2 for 100-pulse ablation. This decrease in the ablation threshold with increasing number of pulses is related to incubation effects at the sample [15]. Although no damage was evident at the surroundings of the ablation craters, it is expected to appear when using large-area beams due to the presence of surface defects that will also result in the decrease of the ablation threshold [16]. Due to the similarities found in the ablation threshold and optical behavior between this material and some photoresists, it is reasonable to assume similar damage thresholds for large-beam irradiation. In particular, for a 120-fs pulsed beam with radius 3.5 mm (similar to one of those used in the compression experiments presented here), no damage was observed up to a pulse energy of 5 mJ (0.013 J/cm^2 peak fluence) [17]. All the tests of the VTHG compressor were done with fluences under this value to ensure safe operation.

To develop the VTHG compressor, two diffraction gratings were recorded by interference of two beams forming an angle of 22° in a symmetric configuration (unslanted gratings). The grating frequency was 730 lines/mm. Figure 1 shows the zero-order efficiency curve (s polarization) vs. the reconstruction angle when the reconstruction wavelength was 800 nm. In that figure, the reflection losses have been taken into account. The minima obtained at 17° correspond to the Bragg angle for $\lambda = 800$ nm. The relative efficiency is around 95% (note that minima do not reach the zero value) and the absolute efficiency is around 73%. The Fresnel losses on both sides of each grating are 8%. We have measured absorption and scattering losses in glass of 12%. From these data and the maximum value of the zero-order efficiency (Fig. 1), absorption and scattering losses of around 2% in the emulsion layer are estimated.

The grating compressor employs a similar scheme to that introduced by Treacy with reflection gratings [18]. The scheme of the pulse compressor composed of two identical volume transmission holographic gratings is shown in Fig. 2a.

The output pulse duration, τ_2, and the transform-limited pulse duration, τ_0, are related by the chirp coefficient, b, introduced by the compressor, as can be seen in (1), where $b_{\min}$ is the chirp coefficient when $\tau_2 = \tau_0$ [19].

$$\tau_2 = \tau_0 \sqrt{1 + \frac{(b - b_{\min})^2}{\tau_0^4}}. \tag{1}$$

The coefficient b (please note that $b = 2 \times \mathrm{GDD}$, GDD being the group delay dispersion) that characterizes the compressor is given by the following expression [19]:

$$b = \frac{\Psi''}{2\pi^2} = -\frac{2\lambda_0 d}{\pi c^2 \cos\theta_0} \frac{\lambda_0^2}{\Lambda^2 - (\lambda_0/2)^2}, \tag{2}$$

where λ_0 is the central wavelength; d is the distance between the gratings; θ_0 is the angle of the reconstructed wave at λ_0, and Λ is the period of the gratings. Figure 2b shows a photograph of the compressor. Both gratings must be oriented to work at the Bragg angle to achieve maximum efficiency at 800 nm. The second grating and the mirrors are mounted on a translation stage, such that the distance d can be varied from 3 to 53 mm. The value of the chirp coefficient b in this configuration varies from -6500 to -114000 fs^2. Due to the glass used to seal both gratings, it is not possible to join them completely to obtain $b = 0$. Additionally, the glass substrate and the sealing glass of each grating (4 mm) introduce a positive chirp. The dependence of the chirp coefficient in a dispersive material, b_g, with wavelength λ is given by the following equation [19]:

$$b_g = \frac{\lambda_0^3}{\pi c_0^2} \frac{d^2 n(\lambda)}{d\lambda^2} z, \tag{3}$$

where z is the propagation length of the pulse inside the dispersive material. Using the Sellmeier equation and coefficients [20], the chirp coefficient of glass at the output of the compressor can be estimated as 1500 fs^2. The compressor must compensate this chirp by increasing the distance d.

Even though the relative efficiency of each grating was higher than 90%, the overall efficiency of the compressor

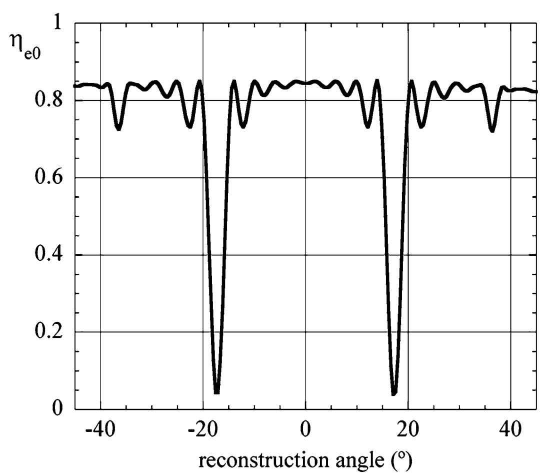

Fig. 1 Zero-order efficiency curve vs. reconstruction angle of a grating recorded with 532 nm and illuminated with an 800-nm laser source. Reflection losses are taken into account

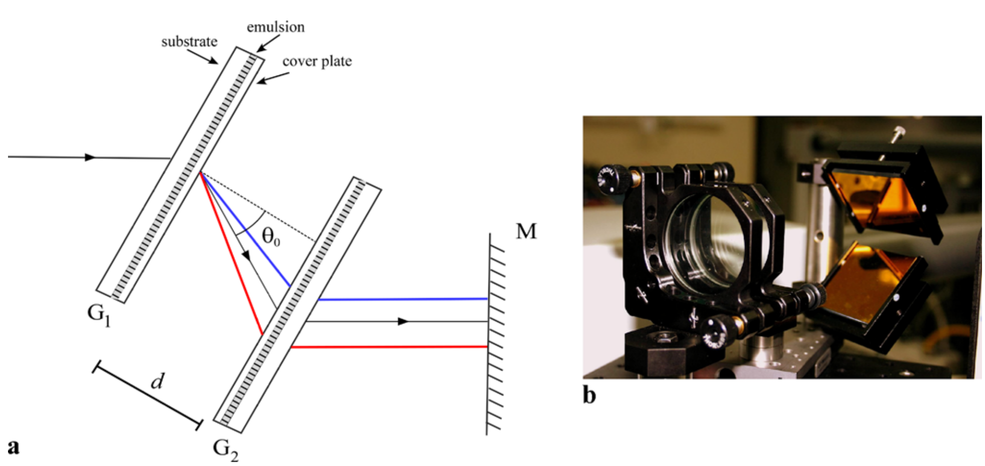

Fig. 2 (**a**) Scheme of the compressor: G_1 and G_2 are the volume holographic transmission gratings; M is a periscope, (**b**) photograph of the compressor setup

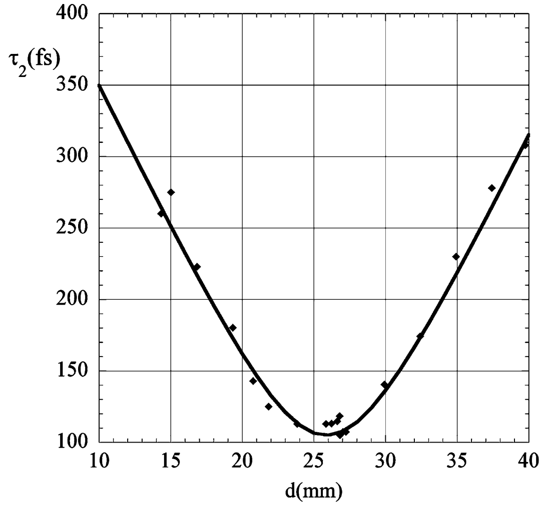

Fig. 3 Temporal width of pulses at the compressor output as a function of the distance between gratings for an input pulse duration of 580 fs. *Squares*: experimental results measured with a single-shot autocorrelator, *solid line*: theoretical curve taking into account the dispersion due to glass

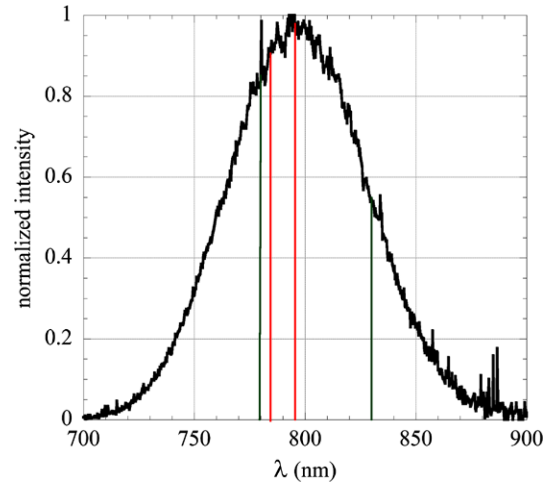

Fig. 4 Intensity at the output of the compressor vs. wavelength curve (gratings of 730 lines/mm) when the incident light is a white light source. The highlighted region of the curve corresponds to the 10-nm spectral bandwidth of the Spectra Physics laser pulses used in the characterization of the compressor (*red*) and the 55-nm spectral bandwidth of the Amplitude Technologies laser pulses (*green*)

was around 30%, because of the additional losses (reflection, absorption, and scattering) previously commented on. This value could be made higher (more than 50%) with an antireflection coating on both sides of each grating and minimizing the losses on the glass by choosing a sealing glass with low absorption and scattering at 800 nm. Since we were working with commercial emulsions, there was no possibility of changing the glass support.

In order to test our compressor, we used a 100-fs, 795-nm Ti:Sa laser (Spectra Physics Spitfire), modifying the grating distance of the CPA compressor to obtain 580-fs chirped pulses. We measured the temporal width of these pulses at the output of our VTHG compressor as a function of the grating distance by means of a single-shot autocorrelator, assuming a Gaussian pulse profile. The results are shown in Fig. 3, together with the theoretical curve, taking into account the dispersion due to the glass. The minimum autocorrelation full width at half maximum (FWHM) obtained was 150 ± 10 fs, which corresponds to an almost Fourier-transform-limited pulse of around 106 fs (assuming a Gaussian pulse shape).

One disadvantage of using volume holographic gratings is the chromatic selectivity. The maximum efficiency of each grating is obtained for a given wavelength (in our case, 800 nm), but it declines as the wavelength varies. In Fig. 4 we show the curve of the spectral transmittance at the output of the compressor vs. the incident wavelength when the incident light is a white light source. It is possible to note the chromatic selectivity due to the volume gratings, which can distort the output pulse shape and limit the minimum pulse duration that can be obtained with the compressor. As shown in Fig. 4, the FWHM measured with a spectrometer was 70 nm. The chromatic selectivity did not affect the pulses in the case of those used to test the compressor, since the efficiency of the gratings remained constant in the 10-nm spectral bandwidth of the pulses.

In order to test whether chromatic selectivity might alter the shape or the pulse duration in the case of larger-bandwidth pulses, we repeated the previous measurements but this time with 38-fs pulses (delivered by a CPA laser from Amplitude Technologies) of 55-nm bandwidth centered at 805 nm. Therefore, we pre-chirped the pulses positively by means of the CPA compressor and used the volume holographic grating compressor to achieve full compression. Figure 5 shows the pulse spectra and the corresponding phases of the chirp-compensated original pulse and the output of our compressor when compensating a pre-chirped pulse. Figure 6 shows the intensity in the time domain of the original pulse and the output of our compressor compensating the pre-chirped pulse, measured with SPIDER (Spectral Phase Interferometry for Direct Electric-field Reconstruction) [21]. As can be seen, the FWHM was not significantly distorted by the chromatic selectivity of the volume gratings, obtaining a 42-fs pulse, although the VTHG compressor introduces a higher-order dispersion which alters slightly the pulse shape and increases the satellite pulse structure.

Although the compression of the 38-fs pulses was successful, the compressor bandwidth will hinder the compression of shorter pulses. For this purpose, it was necessary to address the spectral selectivity of the volume holograms, as we will comment on in the following section.

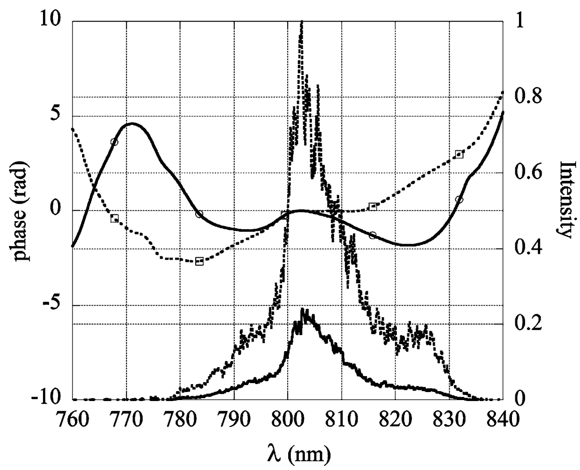

Fig. 5 Power spectra and spectral phases of the original 55-nm broadband pulses (*dashed lines*) and the pulses compressed by the VTHG compressor after being positively chirped (*solid lines*)

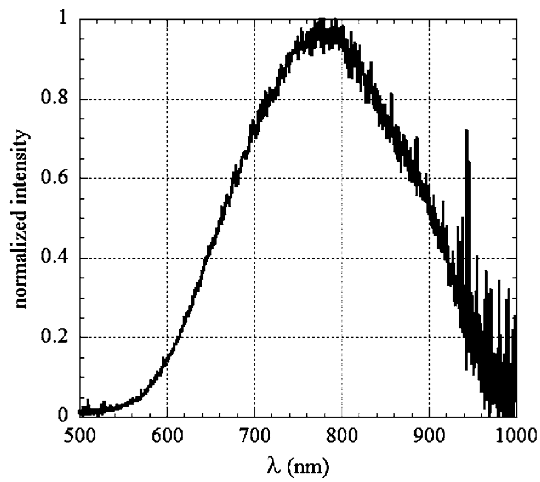

Fig. 7 Intensity at the output of the compressor vs. wavelength curve (gratings of 330 lines/mm) when the incident light is a white light source

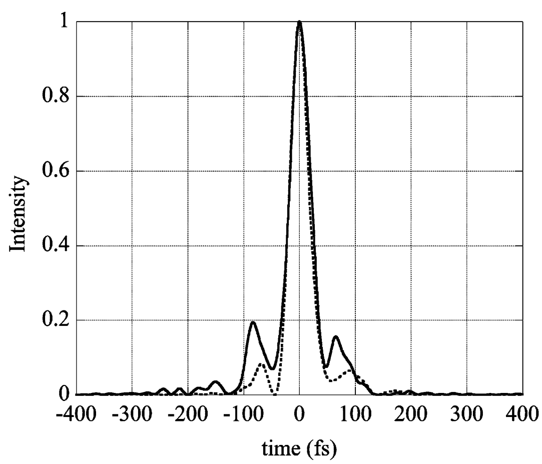

Fig. 6 Intensity in the time domain: original pulses (*dashed line*) and pulses compressed by the VTHG compressor after being positively chirped (*solid line*), measured with SPIDER

3 Adaptation of the compressor for broadband spectra. Application to pulses coming from the filamentation process

We have previously reported that the compressor can be used to compensate the GDD of high-intensity laser pulses down to 38 fs. In this part of our work, the compressor was implemented for a specific application: to achieve shorter pulses by post-compression based on filamentation in gases. The role of the VTHG compressor consists of compensating the GDD of the pulse after the filamentation process.

The spectral bandwidth of pulses obtained after filamentation in air or in other gases is usually broad, due to self-phase modulation (SPM), and therefore allows the possibility of a further compression of the pulses. Thus, the chromatic selectivity of the gratings can affect the shape or duration of the compressed pulse and it was necessary to reduce it in order to obtain a VTHG compressor suitable for our purposes. As is well known [22], the chromatic selectivity of a volume phase hologram depends on the thickness of the emulsion and the grating period. Since the thickness of the PFG-04 emulsion is provided by the manufacturer, the grating spatial frequency must be decreased in order to decrease the chromatic selectivity. As the thickness of the emulsion is 30 μm, gratings down to 330 lines/mm can be recorded, keeping the volume regime. This value marks a limit, since lower spatial frequencies lead to a non-volume condition and hence the efficiency of the gratings decreases.

Additionally, reducing the spatial frequency of the gratings leads to a smaller compression factor, and therefore the distance between gratings must be increased in order to obtain the same GDD correction. Fortunately, in post-compression setups the amount of second-order dispersion to be compensated is not too large (within the range of some hundreds or a few thousands of fs^2), so the distances between the gratings do not have to be long (a few centimeters). A new compressor was constructed using these 330 lines/mm gratings, permitting the higher spectral bandwidth of the VTHG setup but remaining within the volume hologram condition. In this case, the FWHM spectral transmission at the compressor output would be around 225 nm, as shown in Fig. 7. The filament is generated by focusing the 55-nm bandwidth laser beam with a lens (focal length 1.5 m) in a tube containing nitrogen at 2.5 bar. Chirped pulses with broadband spectra were obtained at the output of the filamentation stage. Those spectra presented typically a major component, ranging from 650 to 850 nm, and a lower but broad pedestal covering the visible range, with values as low as 350 nm being achieved.

Figure 8 shows the spectrum of the pulses before and after their passage through the compressor. The spectrum at

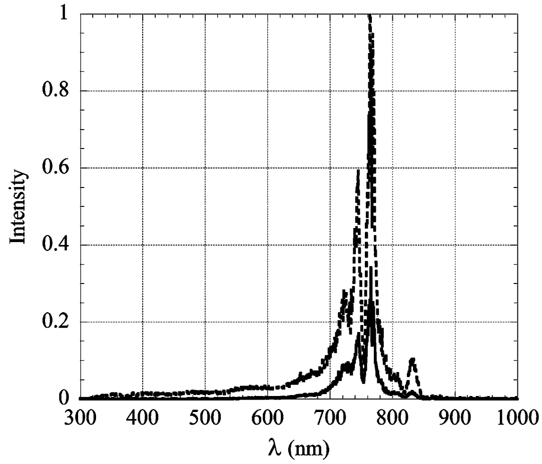

Fig. 8 Spectrum of the pulses generated by filamentation in nitrogen: at the input (*dashed line*) and output (*solid line*) of the compressor

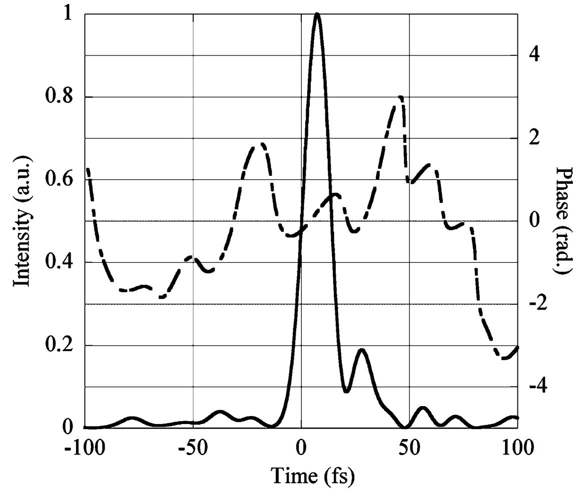

Fig. 9 SPIDER measurements of the temporal profile of the pulses obtained by filamentation at the output of the compressor (*solid line*) and its temporal phase (*dashed line*)

the output of the compressor shows that the chromatic selectivity of the gratings affected the shape of the input spectrum, presenting important losses below 700 nm. On the other hand, the major component of the spectrum (from 650 to 850 nm) remained almost unchanged. This new spectrum at the compressor output corresponded to a Fourier-transform-limited pulse with a duration of 7.8 fs.

In order to measure the pulse duration and retrieve the pulse shape at the output of the compressor, we used a SPIDER device. Since our pulse reconstruction system allowed us to retrieve pulses down to 10 fs (FWHM) and a spectral range between 700 and 900 nm, we were not able to reconstruct the pulses with such a spectrum as the one presented in Fig. 8. In order to know the effect of the VTHG compressor on the spectral phase over the 700–900 nm range, we have worked in conditions where the broadened spectrum was within the mentioned range. We adapted the filamentation compression conditions in order to obtain output pulses within this duration range. Thus, 12.75-fs pulses were measured, while the Fourier-transform limit was around 12 fs.

In Fig. 9 we show the intensity profile (solid line) and the temporal phase of the compressed pulse (dashed line). It can be deduced from the spectral phase variations that some third- and higher-order dispersion remained uncompensated at the output. This is probably due to the grating compressor characteristics, for example, the angle of the gratings, and the nonlinear process during the filamentation, that usually introduce third- and higher-order dispersion creating satellite pulse structures [2, 23].

In summary, the VTHG compressor can be applied to post-compression setups, attaining pulses around 12 fs.

4 Conclusions

We have developed a volume holographic grating compressor for the compensation of the GDD for second-order dispersion in experiments of post-compression based on filamentation in gases, obtaining pulses of 12.75 fs, starting out from pulses of 38 fs. The spectral transmission suggests that this kind of compressor may operate under sub-10-fs conditions. The compressor was composed of two volume holographic transmission gratings recorded in commercial dichromated gelatin emulsions (Slavich PFG-04). These emulsions allowed us to record gratings with an appropriate index modulation value to operate with high efficiency at 800 nm. The damage threshold of the plates enables their use with ultra-intense laser pulses. In this case, it is very useful to record the gratings with a suitable spatial frequency in order to minimize the chromatic selectivity typical of volume gratings.

The use of commercial dichromated gelatin emulsions provides an economical way to record volume gratings with sizes up to 60 mm. It is possible to record them with a spatial frequency and a maximum efficiency wavelength suitable for each experiment. Although the overall efficiency of the compressor was 30%, it could be increased to more than 50% with an antireflection coating on both sides of each plate and by choosing a sealing glass with low absorption at 800 nm.

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