Wavelength tuning of femtosecond pulses generated in nonlinear crystals by using diffractive lenses

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We demonstrate that diffractive lenses (DLs) can be used as a simple method to tune the central wavelength of femtosecond pulses generated from second-order nonlinear optical processes in birefringent crystals. The wavelength tunability is achieved by changing the relative distance between the nonlinear crystal and the DL, which acts in a focusing configuration. Besides the many practical applications of the so-generated pulses, the proposed method might be extended to other wavelength ranges by demonstrated similar effects on other nonlinear processes, such as high-order harmonic generation. © 2010 Optical Society of America

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The generation of nonlinear effects with high efficiency and flexibility has a clear implication for many applications, such as high-resolution optical microscopy [1], petawatt-class laser development via optical parametric amplification [2], or industrial pattern recognition [3]. For ultrafast optics, diffractive elements play a significant role in tailoring nonlinear processes, because they allow us to modify the spatial and temporal shape of the pulse. For example, diffractive gratings recorded in a birefringent crystal increase the efficiency of second-order nonlinear processes [4]. Moreover, Fresnel diffraction through circular apertures enhances supercontinuum generation by exploiting the lensing effect [5].

One of the most common diffractive optical elements is a diffractive lens (DL). This optical element focuses light by diffraction. The well-known Fresnel zone plate can be regarded as a poor diffraction efficiency DL. In experiments with nanosecond pulses, the Fresnel zone plate has demonstrated its usefulness to enhance the efficiency of third-harmonic (TH) generation up to a factor of 16 [6]. However, to the best of our knowledge, no attention has been paid to the basic problem of the generation of second-order nonlinear processes with DLs and femtosecond pulses. In this case, the large spectral bandwidth of the pulsed light and the strong dependence of diffraction on the wavelength cause new observable facts.

In this Letter, we experimentally study nonlinear frequency-conversion processes [7] of ultrashort pulses focused by a kinoform DL in nonlinear crystals. A kinoform DL is a phase DL that provides the highest diffraction efficiency [8]. Wavelength tuning of both second-harmonic (SH) generation and sum-frequency generation is demonstrated. The proposed method provides a novel technique to tune the central wavelength of the generated pulses with potential applications to other nonlinear processes.

Within the paraxial approximation, the generation of nonlinear effects by focusing a cw beam with refractive or DLs of the same NA is expected to show similar behavior. However, for a femtosecond pulse, strong differences appear owing to the large spectral bandwidth of pulsed radiation. In this case, the intensity distribution in the focal region of a refractive lens is different from that of a DL. Chromatic aberrations are the cornerstone of the above behavior. In particular, for a refractive lens, chromatic aberrations are caused by material dispersion [9], whereas, for a DL, they are due to the diffraction phenomenon [10]. The focal length of the DL, f, is related to the wavelength, λ, by the expression $\lambda f = \lambda_o f_o$, where the subscript o denotes the reference wavelength. For a DL made up of a low-dispersion material, the axial extension of the focal region is given by $\Delta f = f_o \Delta\lambda/\lambda_o$, where $\Delta\lambda$ is the bandwidth of the light source. This is several orders of magnitude larger than the focal region achieved with a singlet having a refractive index n, $\Delta f = -f_o \Delta\lambda (dn/d\lambda)_o/n - 1$. For an achromatic doublet, the focal region in the axial direction is the lowest due to the lack of chromatic aberrations. Note that the different spectral and spatial geometry of the focal volume will essentially affect the processes of frequency conversion induced with each lens.

The whole experimental setup is depicted in Fig. 1. The laser system was a Ti:sapphire regenerative amplifier (Spitfire, Spectra-Physics) that operates at a 1 kHz repetition rate, delivering pulses of $\tau_F = 120$ fs FWHM, and a central wavelength of 795 nm. The $1/e^2$ diameter of the beam is about 9 mm. A sample of the beam is taken for the SH generation experiment (dotted box) and is focused by a kinoform DL (Institute of Automatics and Electrometry, Russia) with a focal length of 150 mm, for the design wavelength of 565 nm. In our experiment, the focal distance for the wavelength of 795 nm corresponds to 106.6 mm. The DL was placed in a computer-
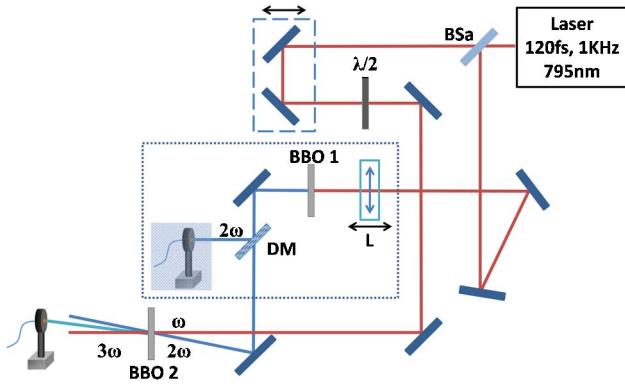
Fig. 1. (Color online) Experimental setup: BSa, beam sampler (10/90); $\lambda/2$, half-wave plate; BBO, nonlinear crystal; and L, diffractive/refractive lens mounted on a motorized stage.

controlled motorized stage that moves the lens along the propagation axis. Near the focal region, an uncoated Type-I β-BaB$_2$O$_4$ (BBO) crystal (10 mm × 10 mm × 0.5 mm, $\theta = 29.1°$, $\varphi = 0°$), denoted by BBO1 in Fig. 1, was aligned at quasi-normal incidence. Initially the BBO1 crystal was located at a distance $z = 104$ mm from the DL, and then this distance was increased with the motorized stage by steps of 75 μm. We captured the spectrum of the SH pulses, after reflecting at the dichroic mirror (DM), with a spectrometer (HR4000, Ocean Optics). The trace of the SH spectrum versus crystal–lens distance is shown in Fig. 2(a). For comparison, we also performed the above experiment replacing the DL by an achromatic doublet with a focal length of 100 mm (Linos G063144525), in order to get approximately the same NA. Results are shown in Fig. 2(b).

From Fig. 2, one can see that using the DL, the central wavelength of the SH pulse is tuned. Whenever the axial position of the BBO crystal coalesces with the focal position of the DL for a certain wavelength λ, then the central wavelength of the generated SH signal turns out to be
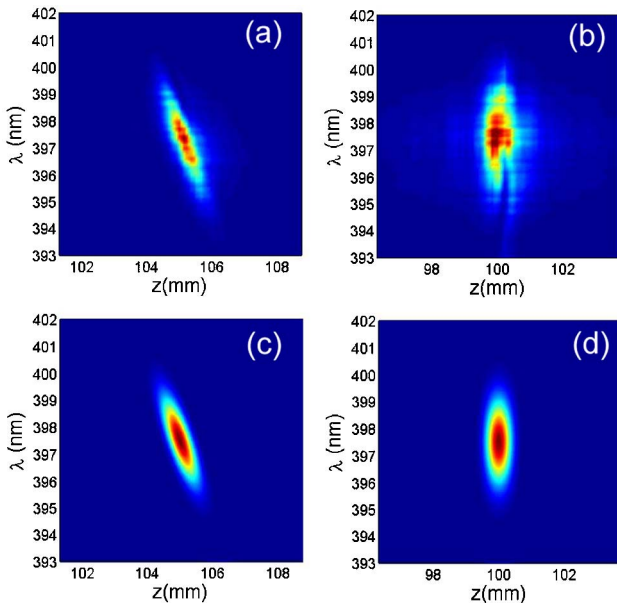

Fig. 2. (Color online) Measured SH spectrum versus the lens–crystal (BBO1) distance around the focal length of (a) a kinoform DL and (b) an achromatic doublet. (c) and (d) correspond to numerical simulations of the traces.

$\lambda/2$. Of course, the tunability range depends on both the spectrum of the incident pulse and the phase-matching spectral acceptance of the crystal. In our experiment, the crystal thickness is short enough to avoid limitation due to phase matching.

To simulate the above results, we use a very simple model based on the thin nonlinear crystal approximation with undepleted pump. We consider that a plane wave illuminates an ideal refractive or DL with 9 mm of aperture. The spectrum of the laser was modeled as a Gaussian spectrum. The power spectrum of the generated SH for each distance crystal–lens distance z can be roughly estimated by following the well-known expression $I(\lambda/2, z) \propto I^2(\lambda, z)[\sin(\Delta k_\lambda \cdot L/2)/(\Delta k_\lambda \cdot L/2)]^2$ [7], where $I(\lambda, z)$ is the power spectrum of the fundamental beam in each z position, L is the crystal length, and Δk_λ is the phase mismatch corresponding to the BBO crystal at normal incidence. In the simulation, we can consider the normal incidence approximation because the confocal parameter is greater than the crystal length. Results of these calculations are depicted in Figs. 2(c) and 2(d), showing very good agreement with experiments. It should be mentioned that a more accurate formulation can be used to simulate the experimental results; however, this simple approximation is enough to show the dominant effects.

To avoid damage of the crystal, the fundamental pulse energy was set in all cases to 44 nJ. SH conversion efficiency was measured for the two lenses at the crystal-lens distance that maximized the output energy. We obtained 25% and 10% of total efficiency with the refractive and the DL, respectively. The confocal parameter of the refractive lens ($b = 250$ μm) would require a larger crystal thickness to optimize the maximum efficiency condition for focused beams, $L = 2.84 \cdot b$ [7], but with such a choice, the phase-matching spectral efficiency would limit the converted spectrum. For the SH with a DL, the reduction in efficiency is mainly caused by two factors. The first factor is the spatial distribution of the spectrum that avoids the conversion of all the wavelengths at the same position of the crystal. The second factor is the limited diffraction efficiency of our DL for the central wavelength of the laser (about 69%). This efficiency can be improved with an optimal design of the DL for the desired wavelength. Then the efficiency of the SH generation with the DL would be close to the one obtained with the refractive lens.

From Fig. 2 we can also realize that the DL acts as a bandpass optical filter, reducing the SH spectral width. In particular, the spectral width of the SH pulses is 1.5 nm for the pulses generated with the DL and 2.4 nm for those generated with the achromat. This wavelength clipping reduces the well-known angular dispersion of the SH generated with focusing optics. Because of this fact, we did not introduce any correcting system. Moreover, an enlargement of the pulse duration is expected for such bandwidth reduction. Assuming a transform-limited pulse, the temporal duration of a SH pulse with 1.5 nm spectral width is 145 fs. The temporal characterization of the pulse was carried out by the cross correlation of the SH with the fundamental pulse. The setup is shown in Fig. 1. For these measurements, the DM was removed and the crystal–DL distance was fixed at the position of
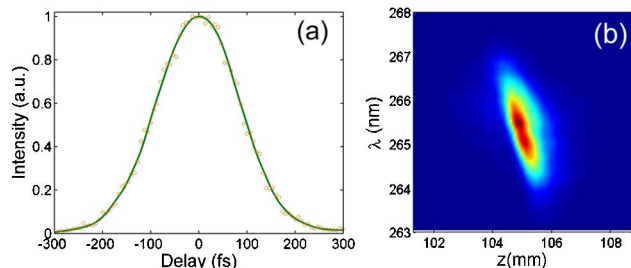
Fig. 3. (Color online) Experimental measurements: (a) cross-correlation trace of the SH generated with the kinoform DL and (b) TH spectrum versus the lens–crystal (BBO1) distance around the focal length of the kinoform DL.

maximum SH signal. The fundamental and the SH were mixed in a 100 μm Type I BBO crystal (BBO2 in the figure, 6 mm × 6 mm × 0.1 mm, $\theta = 44.3°$, $\varphi = 90°$), producing a TH signal whose power was measured in terms of the relative delay. The width of the recorded trace [depicted in Fig. 3(a)] was 200 fs. Such temporal width suggests [11] a duration of the SH pulses of ~150 fs (assuming an enlargement of the fundamental to 130 fs at BBO2), which is in fairly good agreement with the transform-limited pulse duration of the measured SH spectra.

Finally, we demonstrated the wavelength tuning of the TH signal by using the experimental setup for the cross-correlation measurements. The SH pulses were generated by focusing the beam in BBO1 and then mixed in BBO2 with the fundamental unfocused beam. To reduce the angle between the two beams, the last mirror was replaced by a DM. Next, the delay line length was fixed to maximize the TH signal and again the tunability was obtained by changing the distance between the DL and BBO1. The trace of the TH spectrum versus the DL–BBO1 distance is shown in Fig. 3(b). The TH signal can be tuned, thanks to the wavelength selection induced in the SH generation by means of the DL.

In conclusion, we have experimentally demonstrated a compact and robust technique using a DL that is able to achieve wavelength tunability in femtosecond pulses generated from second-order nonlinear optical processes by simply changing the DL–crystal length. An alternative technique to produce a similar tunability could be achieved by changing the orientation of a thick nonlinear crystal where the nonlinear process is taking place. However this choice is not suitable for ultrashort pulses because the shifting of the phase-matching curve in relation to the pulse spectrum will lead to the generation of a modulated spectrum in the SH pulse and therefore a degraded temporal shape. In addition to the direct application of the generated tunable pulses to experiments in spectroscopy, the proposed technique can be extended to other processes like optical parametric amplification or even to high-order harmonic generation.

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