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Rapid communication

A new high-resolution femtosecond pulse shaper

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Abstract. A novel liquid crystal display (LCD) with 640 stripes was successfully implemented and investigated for femtosecond pulse shaping. As compared to previously used devices, the large active area allows for operation in high-power laser systems. The increased number of pixels greatly enlarges the manifold of accessible pulse modulations, making the device an ideal tool for coherent control experiments and optical information processing.

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Optical waveform synthesis of femtosecond laser pulses originally introduced by Heritage and coworkers [1] has become enormously important in many fields of research, from simple pulse compression to optical communication and control of quantum phenomena. It has been demonstrated that a pulse shaping device is able to automatically compress phase-modulated femtosecond pulses to their bandwidth-limited temporal width [2–5] or to generate user-defined pulse shapes [6]. It has also been shown that two-dimensional devices offer the possibility to control the temporal and spatial characteristics of short laser pulses at the same time [7]. Besides these more technical challenges, tailored femtosecond pulses have also been employed to efficiently generate a specific high harmonic via phase-matching in a gas jet [8]. They have been used to coherently control two-photon transitions [9, 10] and Rydberg wavepackets [11] in atomic systems. They have also been employed to control molecular [12, 13] and solid-state quantum systems [14]. In addition, pulse shaping may become a versatile tool in communication applications due to its ability to realize encoding and decoding devices [15].

Pulse tailoring allows one to simply delay a femtosecond laser pulse or, more importantly, to generate almost arbitrary intensity profiles starting from a bandwidth-limited femtosecond laser output. Almost all experimental implementations rely on imposing phase and/or amplitude modulation on the spatially dispersed frequency spectrum of the laser pulse. In order to realize a user-specified waveform mainly three experimental approaches are pursued, namely with liquid crystal displays (LCD) [1, 16], acousto-optic modulators [17], and deformable mirrors [18]. An excellent review of femtosecond pulse shaping using LCDs is given in [19]. Recent studies explore the possibilities to generate tailored femtosecond pulses in wavelength regimes where conventional pulse-shaping devices are opaque by using specially designed sum-frequency [20] or difference-frequency [21] generation schemes.

The fact that LCDs consist of discrete stripes parallel to each other limits the possible modulations. A number of recent applications have revealed that 128 pixels, as used by most standard devices, are not sufficient. Here, we report on a novel LCD that has 640 stripes and a 12-bit resolution. Combined with an all-reflective zero dispersion compressor, it overcomes two major problems of standard devices. First, due to the large active area it is suitable for shaping high-power laser pulses. Second, for the first time, it allows waveforms to be generated that were not accessible before using LCDs, but are required from optimal control theory in order to control quantum systems. In this paper, we will first discuss the experimental setup, then we will demonstrate the capabilities and finally explore the limits of the device.

1 Experimental setup

The main components of the experimental apparatus are shown in Fig. 1. A Ti:sapphire laser system operated at a repetition rate of 1 kHz and a center wavelength of 804 nm serves as the source for the ultrashort pulses. The pulse energy is 1 mJ, the spectral full width at half maximum (FWHM) 24 nm, and the temporal FWHM 50 fs (assuming a Gaussian intensity profile). The pulses traverse a zero-dispersion compressor and are subsequently characterized by a multi-shot FROG [22] based on second harmonic generation (SHG). The SHG spectra are recorded by a CCD array with a 16-bit reso-

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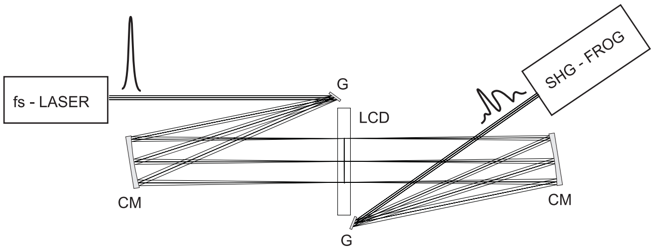

Fig. 1. Sketch of the fs-pulse-shaping apparatus. The spectral phase of a laser pulse is manipulated in a 4-f arrangement with the novel LCD in the Fourier plane; the pulses are subsequently analyzed by a SHG FROG. G: gold-coated gratings (1800 lines/mm); CM: cylindrical mirror, $f = 300$ mm; LCD: SLM-S 640/12

lution. The design of the reflective zero-dispersion compressor was optimized (using ZEMAX 9.0) for minimum phase-front distortion of the output pulses.

The novel LCD (SLM-S 640/12) has 640 individual stripes (pixels), the area of each single stripe is 97 μm ×10 mm and the gap between two stripes is 3 μm. At a wavelength of 800 nm the thickness of the liquid crystal cell allows a maximum phase shift of 6π (zero voltage). The average alignment time of the liquid crystals in the operating range between 0.25π and 2.25π is about 25 ms. The liquid crystals are aligned such that spectral components polarized perpendicular to the stripes may be retarded. Therefore, the LCD is perfectly matched to the high reflection efficiency for p-polarized light of the gratings in the visible and NIR spectral region. The two outer surfaces of the LCD have an anti-reflection coating and the LCD has a transmission of 95% in the wavelength range from 450 to 1500 nm. The damage threshold was measured to be 300 GW/cm^2 (at 800 nm, 45 fs). At higher energies the structured polyimide layer of the LCD is damaged. The phase retardance at three different wavelengths of the LCD was carefully measured and is shown in Fig. 2.

The two cylindrical mirrors have a focal length of 300 mm. Two 1800 lines/mm gold-coated gratings offer the possibility of shaping a spectral range of 96 nm at 810 nm with an average spectral resolution of 0.15 nm/pixel. Even

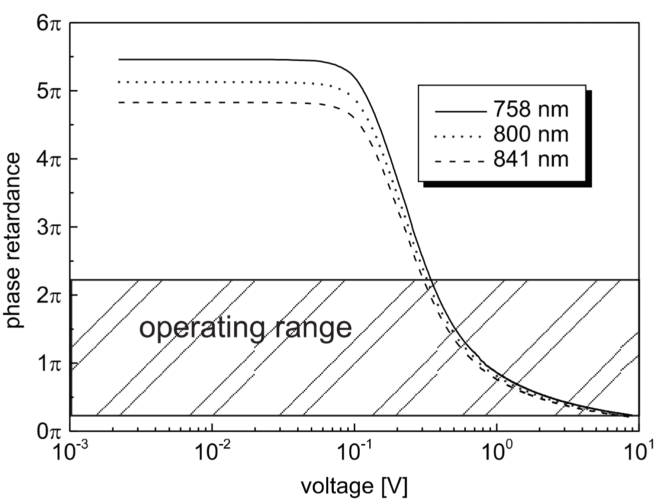

Fig. 2. Phase retardance of the LCD as a function of the applied voltage for three different wavelengths

for a pulse energy of 1 mJ, the intensity impinging on the LCD mask in this pulse-shaping setup is only about 5 MW/cm^2 and, therefore, well below the damage threshold. The pulse width impinging on one pixel was approximated by a Gaussian pulse having a spectral FWHM of 0.15 nm. The relatively low intensity per pixel is a direct consequence of the large area which is covered by the dispersed spectrum. The overall transmission through the pulse shaper was measured to 60% and is mainly determined by the reflectivity of the gratings. Since no polarization filters are used the device is operated as a phase-only shaper.

2 Results and discussion

Since we use phase-only linear spectral filtering, the electric field of the emerging pulse is

$$E_{\text{out}}(\omega) = E_{\text{in}}(\omega)\exp[-\mathrm{i}\Phi(\omega)]\,, \tag{1}$$

where the phase $\Phi(\omega)$ has a constant value between 0 and 2π in the range $[\omega_j, \omega_j + \Delta\omega]$ and $j \in [1 \dots 640]$. The frequency range $\Delta\omega$ that is covered by one pixel is related to the spectral resolution of the gratings, the focal length of the focusing optics, and the beam diameter. The discrete nature of the phase modulation is inherent to the LCD mask and limits the accessible phase modulations where the limit is set by Nyquist's theorem [23]. In other words the steepest slope of any applied phase function must not exceed π across one pixel ($\Delta\omega$). For exactly this reason it is desirable to have as many pixels on the LCD as possible. On the other hand, the possibility to wrap the phase increases the dynamical range [24], since it allows one to apply phase patterns modulo 2π. Technical restrictions have limited the number of pixels to 128 to the present day. With the new LCD described here, it is possible to access phase modulations with a highly increased variability. In order to test the novel mask and to demonstrate the enhanced performance, we performed a number of pulse-shaping experiments. In each experiment a specific type of modulation up to the corresponding Nyquist limit was applied to the mask in order to define the maximum working range of the device.

First a FROG trace of the unshaped laser pulse was recorded. A reconstruction of the electric field shows that the pulse has no second-order phase but some residual third-order phase. Applying a linearly increasing or decreasing phase allows one to shift the original pulse in time. For a spectral resolution of $\Delta\lambda = 0.15$ nm/pixel, the Nyquist limit for a purely linear phase is $\Phi_1 = \pm 7187$ fs.

$$\Phi_1 = \frac{\lambda_0^2}{2c\Delta\lambda}\,, \tag{2}$$

where c is the speed of light and λ_0 the center wavelength of the laser pulse. As expected, for even larger linear phase modulations, the original pulse structure is heavily distorted, indicating that the Nyquist theorem is violated. Nevertheless, the accessible delay range is about 14 ps with a resolution of 7 fs. This makes the pulse shaper a valuable delay generator