Full control of the spectral polarization of ultrashort pulses

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The spectral polarization of an ultrashort pulse is fully controlled by a novel pulse shaper design, including three liquid-crystal spatial light modulator arrays at three different orientations. The added degree of controllability permits generation of previously unattainable pulse shapes, with possible applications in multidimensional spectroscopy, coherent control, and ultrafast polarization gating. © 2006 Optical Society of America

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The ability to tailor the temporal properties of ultrashort pulses has recently had a significant effect on a variety of subjects, such as photochemistry[1] and high-harmonic generation.[2] The main tool in most of these experiments is the Fourier-domain pulse shaper.[3] In this apparatus the pulse is first separated into its frequency components by a grating and a lens. At the Fourier plane, each of these is manipulated separately, usually by a liquid crystal or an acousto-optic spatial light modulator (SLM). The pulse is then reconstructed by a second lens–grating combination. Until recently, pulse shapers manipulated only the spectral phase and amplitude of ultrashort pulses, resulting in a modified temporal profile of the pulse but maintaining its polarization properties. Brixner and Gerber,[4] and Brixner *et al.*[5] have recently introduced polarization pulse shaping, by which not only the phase but also the polarization state of each frequency component was manipulated separately. Their approach utilizes two liquid-crystal SLMs at the Fourier plane, and has proved beneficial in several applications,[6] including nonlinear spectroscopy,[7] control of angular momentum states,[8] and photochemistry.[9,10] The design of Brixner and Gerber, however, is unable to arbitrarily control the spectral polarization. In short, it enables us to introduce only polarization states that are obtainable by use of a wave plate oriented at 45° to the input polarization. Moreover, the output spectral polarization is distorted by elements with spectrally dependent transmission or reflection, such as diffraction gratings or dielectric mirrors. In particular, the effect of elements that introduce a spectrally varying phase difference between the two polarizations cannot be easily compensated for by the use of standard polarization compensators. Here we show how, by adding a third SLM at the appropriate orientation to the pulse shaper, both the phase and the polarization state of each frequency component can be controlled arbitrarily. This degree of controllability also enables us to precompensate for any distortion of the spectral polarization along the optical path. We demonstrate this by constructing a temporal polarization profile that was unattainable with previous pulse-shaping techniques.

Needless to say, simple pulse sequences where the various pulses differ in polarization have been used extensively in time-resolved studies.[11] These pulse sequences are typically generated by an interferometric setup. The main difficulty in many of these experiments is the need for active phase stabilization of all interferometer arms. More complicated temporally varying polarizations have also been realized. A rapidly rotating linear polarization (optical centrifuge) has been achieved by means of two counterpropagating circularly polarized beams.[12] Polarization gating of high-harmonic generation[13] has been realized using a combination of wave plates.[14,15] Nevertheless, the versatility and robustness of ultrafast pulse shaping permits generation of much more complicated, inherently phase-coherent waveforms.

As noted by Brixner and Gerber, full control of the temporal polarization of ultrashort pulses requires four independent degrees of freedom for each frequency component.[4,5] Our setup provides only three, maintaining the total energy content of the pulse. Both the spectral polarization and the overall phase of each frequency component are, however, fully controllable. This is done by a sequence of three liquid-crystal SLMs, the first two with their preferential axes at right angles to each other and at 45° relative to the input (linear) polarization, and the third with

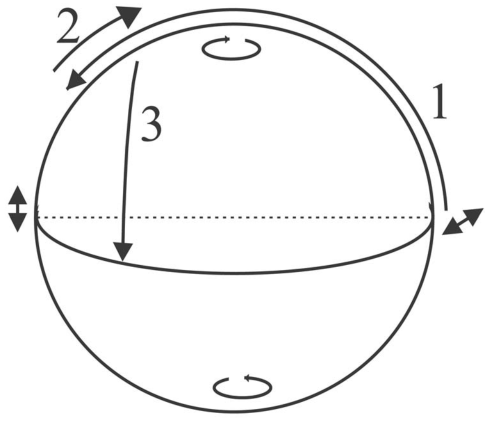

Fig. 1. Poincaré representation of the effect of the three LCD arrays on the spectral polarization. The first two arrays provide phase control and a polarization rotation along the circle connecting the poles and the input polarization state. The third array provides a rotation about the axis connecting the horizontal and the vertical linear polarizations (dashed line).

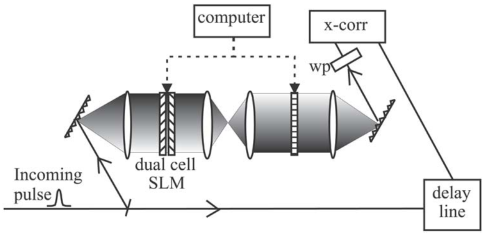

Fig. 2. Experimental setup of the full spectral polarization shaper. It consists of an 8*f* lens system with a dual cell SLM, whose preferential axes are oriented at ±45° to the input polarization at the first Fourier plane and a single SLM oriented parallel to the input polarization at the second Fourier plane. Pulses are passed through a wave plate and characterized by cross correlation with an unshaped pulse.

its preferential axis parallel to the input polarization. Considering the Poincaré sphere representation of the polarization transformations, as shown in Fig. 1, it is easy to see how an arbitrary output polarization is achieved. Starting from an initial linear polarization (along the horizontal axis), the first two SLMs can move it only along a circle passing through the poles (arrows 1 and 2 in Fig. 1). Since the two SLMs are at right angles, the motion induced by the second is exactly opposite to that induced by the first, making possible control of the overall path length on the Poincaré sphere, or the spectral phase. As can be seen, from the expression in Eq. (1), however, the relative retardance between the horizontal and the vertical axes is fixed at $\pm\pi/2$. The third SLM enables one to change this relative retardance, which in the Poincaré representation is motion perpendicular to the path induced by the first two SLMs (arrow 3).

By writing the matrix operations for the polarization rotations of all three SLMs, the necessary coordinate transformation can be calculated in the *x*-*y* basis to be

$$\vec{E}_{\text{out}}(\omega) = \frac{1}{2}\begin{bmatrix} R_x(\omega)^2 e^{i\phi_3}(e^{i\phi_1}+e^{i\phi_2}) & R_x(\omega)R_y(\omega)e^{i\phi_3}(e^{i\phi_1}-e^{i\phi_2}) \\ R_x(\omega)R_y(\omega)(e^{i\phi_1}-e^{i\phi_2}) & R_y(\omega)^2(e^{i\phi_1}+e^{i\phi_2}) \end{bmatrix}\vec{E}_{\text{in}}(\omega), \tag{1}$$

where R are the grating reflectivities and ϕ_1, ϕ_2, and ϕ_3 are the relative phase retardances induced by the three SLM arrays.

In building the experimental setup that enabled us to perform such manipulations we used only off-the-shelf components. Since a single SLM with three LCD arrays is unavailable, instead we built an 8*f* pulse shaper, in which the first Fourier plane is imaged onto a second by a 1× magnification telescope. A phase and amplitude pulse shaper (CRI SLM-256) is located in the first Fourier plane, and a phase-only pulse shaper (CRI SLM-128 phase) is located at the second Fourier plane. This experimental setup is shown schematically in Fig. 2. In the experiments we used 25 fs pulses at 810 nm from a home-built Ti:sapphire laser oscillator. These pulses were spectrally separated by 1200 line/mm gratings and collimated by $f=150$ mm achromats. The 1× telescope was built using $f=100$ mm achromats. The spectral resolution, as determined by the spot size on the SLM plane was 0.4 nm. Each SLM pixel covered a bandwidth of ~0.6 nm. Alignment of the two SLMs was performed by optimizing a 45° polarization rotation of a single pixel. To do so, we induced a $\lambda/4$ relative phase shift between the two arrays of the first SLM, resulting in a circular polarization, and modified it to a linear polarization by a second $\lambda/4$ shift in the second SLM. The total energy passing through a polarizer at 45°, as measured by a spectrum analyzer, was maximized by spatially overlapping the second SLM and the image of the first SLM. The overall efficiency of the pulse shaper was ~30%. The outgoing pulses pass through a quarter-wave plate and a half-wave plate and are characterized by cross correlation with an unshaped 25 fs pulse in a thin $LiIO_3$ crystal, resulting in a cross correlation of the horizontal polarization component only (due to the phase matching geometry).[16]

To exemplify the capabilities of this new pulse shaper we demonstrate controlled generation of a pulse that evolves along a predetermined path on the Poincaré sphere, evolving from a linear polarization along the *x* axis, through a circular one, to a linear polarization at 45° to the horizontal axis, and back to a linear polarization along the *x* axis. To do so, we chirp the shaped pulse to a duration of 3 ps by shifting the recollimating grating from the 8*f* position. An appropriate phase mask was chosen on all three SLMs to induce the required polarization evolution.

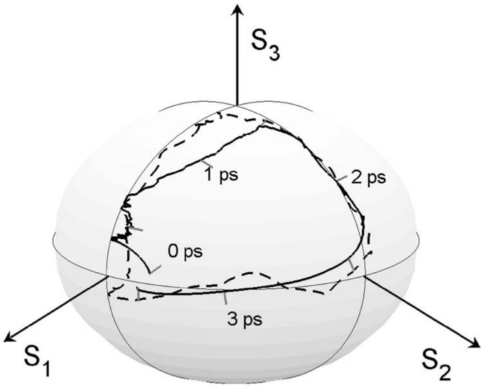

Fig. 3. Experimental (solid curve) and calculated (dashed curve) results for the evolution of the polarization state of the shaped pulse on the Poincaré sphere. Note that the measured polarization state is obtained up to the sign of S_2, so that $|S_2|$ is plotted. Some third-order dispersion that was present in the experiment is neglected in the calculation. Time tick marks are indicated as gray lines on the experimental graph every 0.5 ps.

The pulse was characterized by performing four cross-correlation measurements for vertical, horizontal, and left-, and right-circular polarizations using the wave plates, which determine the Stokes parameters S_1, S_2, and S_3 up to the sign of S_2. The experimental results are presented in Fig. 3, together with the expected results, as calculated from the laser parameters. As can be seen, the polarization begins as linear horizontal and then evolves to circular through elliptic polarizations, then again to linear polarization at 45° to the initial horizontal polarization. Finally it evolves on the Poincaré equator through different linear polarizations to the initial horizontal polarization. Two factors contribute to the small discrepancy between the two: a small third-order dispersion of the reference arm, and the fact that the quarter- and half-wave plates used are not achromatic.

While phase-controlled ultrafast waveforms have been used in a variety of applications in the past decade, polarization-shaped pulses have only recently begun to be used as an experimental tool.[7–10] The pulse-shaping apparatus that we present here significantly broadens the range of obtainable polarization-shaped waveforms, with obvious implications in the fields of adaptive quantum coherent control,[17] molecular alignment,[18,19] and coherently controlled advanced spectroscopy methods.[20,21] Moreover, it inherently overcomes some of the difficulties involved with the current polarization-shaping technique, particularly due to the ability to precompensate for distortion of the spectral polarization in the beam path. In fact, the presented setup can also be used to perform an opposite task, compression of a pulse with a distorted polarization profile, by simply inverting the direction of propagation of the beam through it. Unlike in Ref. 22, this setup is capable of correcting any distortion to the spectral polarization of the pulse. Finally, we note that such a device can be practically incorporated into a single SLM device including three LCD arrays, significantly simplifying the optical setup by eliminating the need for an imaging telescope.

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