

Femtosecond pulse-shape modulation at kilohertz rates

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We demonstrate a new scanning femtosecond pulse-shaping technique that allows pulse shapes to be modulated at kilohertz rates. This technique is particularly useful for lock-in measurements in which the signal is synchronized with the alternating pulse shapes. The pulse-shape lock-in technique is demonstrated in resonant coherent anti-Stokes Raman scattering, where it is shown to significantly improve the ratio of the resonant signal to both the nonresonant background and to noise. © 2004 Optical Society of America

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Femtosecond Fourier-domain pulse shaping^{1,2} has had a significant effect as an experimental tool in the past decade. This technique, capable of accurate control over femtosecond optical waveforms, has facilitated many advances in spectroscopy, nonlinear fiber optics, and high-field physics. Many of these applications require dynamic pulse-shape control. Several methods for applying dynamic Fourier-domain phase and (or) amplitude masks have been reported, such as liquid-crystal (LC) spatial light modulators (SLMs),² deformable mirrors,³ and acousto-optic modulators.^{4–6} Probably the most widespread approach is a one-dimensional array of LC elements used as a Fourier plane mask.² These SLMs provide great flexibility for pulse-shape control, allowing virtually continuous phase, amplitude, or polarization⁷ adjustment at every pixel, but usually suffer from a relatively low updatability rate of a few tens of milliseconds. A deformable mirror shaper is usually suitable for providing only smoothly varying phase variations because of the relatively small number of actuators and the upper limit of curvature that can be induced on the membrane mirror. On the other hand, in acousto-optic modulators, the acoustic grating wave, which fills the modulator's aperture, is not fixed. In spite of the fact that of every single femtosecond pulse the acoustic wave appears frozen on the time scale of propagation of that pulse, the pattern changes noticeably during the time between pulses or a typical mode-locked oscillator (~ 100 MHz). This practically limits the usage of the acousto-optic shapers to lower-repetition-rate systems such as femtosecond amplifiers.

It is often desirable to switch pulse shapes within a predetermined set of waveforms, yet at higher repetition rates than are practically achievable with a LC SLM. One such possible situation is switching quickly between two pulse shapes, one maximizing and one minimizing a particular effect. Specifically, this ability would be important for coherently controlled nonlinear spectroscopy and microscopy,⁸ in which pulse shapes that maximize and minimize a resonant coherent anti-Stokes Raman scattering (CARS) signal could be found, yet all other background and noise processes are insensitive to the exact pulse shape. In this situation one would prefer to detect only the difference signal when applying these two shapes, and this is

best done with lock-in detection while oscillating the waveform between these shapes. LC-based shapers are too slow to allow such a mode of detection; moreover, a LC SLM cannot change abruptly between two shapes—undesirable intermediate shapes may appear as one form is turned off and another is turned on.

In this Letter we demonstrate a scanning-mirror-based femtosecond pulse-shaping technique that uses a fixed two-dimensional reflective filter mask. In this shaper one dimension of the mask is used to encode the spectral pulse phase information and the other is the scanning dimension that allows one to choose a specific waveform. We demonstrate kilohertz-rate pulse-shape modulation for background-free CARS detection. We believe that this technique can be particularly useful for nonlinear imaging applications.

A schematic representation of our scanning shaper is shown in Fig. 1. The laser beam reflects from the scanner mirror (SM) and is imaged by the 4- f telescope (L1, L2) onto diffraction grating G1 (1200 mm^{-1}). We used a galvanometer scanner (Cambridge Technology Model 6220). This closed-loop scanner has analog addressing and feedback with accurate reading of the instantaneous angular position of the scanning mirror. This allows independent control of both scanning amplitude and frequency. A small torque

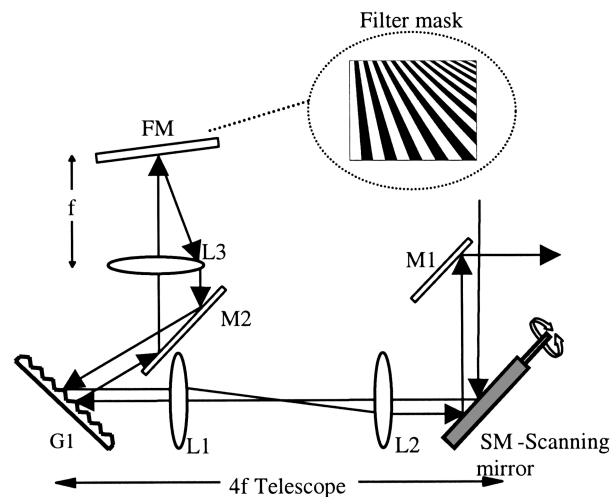


Fig. 1. Schematic representation of the scanning shaper. Inset, lithographic mask for the two-dimensional reflective phase filter.

$\sim 13 \text{ mm} \times 9 \text{ mm}$ mirror was used to ensure maximum positioning speed. This telescopic imaging system enabled us to rotate the small torque mirror instead of the relatively bulky grating, thereby allowing significantly higher updating rates. The diffracted beam reflects at a nearly Littrow angle on mirror M2, which directs it through the Fourier transform lens (L3) onto the Fourier plane filter mask (FM). The FM is tilted slightly so that the reflected beam is intercepted after the scanner by M1. This tilt is equivalent to a linear spectral phase or to a distortionless time shift of the output signal. In fact, a fast-scanning delay line was previously built with a similar system, but with the Fourier mask replaced by a simple scanning mirror.^{9,10} Note that in passing through the shaper the beam reflects from the scanner mirror surface twice on the scanning axis, so that its output direction is independent of the angular position of the scanner mirror. All lenses used in the setup are 100-mm achromatic doublets. By changing the angular position of the scanning mirror, we control the height of the horizontally diffracted spectral components of the pulse on the FM, thereby addressing different shapes on the two-dimensional mask. Note that the number of different spectral functions that can be realized by such a concept is given by the total vertical span covered by the scanner divided by the effective spot size in the Fourier plane, corresponding in our case to approximately several hundred.

We demonstrate this concept with a phase-only filter, realized as a relief-patterned reflecting mask. The binary mask was fabricated by a standard photolithographic process on a substrate of GaAs, etched to $\sim 200 \text{ nm}$ and coated with gold to produce π phase modulation for the reflected $\sim 800\text{-nm}$ light. The mask was patterned to provide periodic rectangular phase modulation of the spectral components of the ultrashort pulse. The period of the modulation varies linearly in the vertical direction across the mask, generating the fan-shaped pattern shown in the inset in Fig. 1. By changing the angular position of the scanning mirror, we can practically control continuously the modulation frequency of the spectral phase. Below the patterned area there is a flat surface left where we can place the scanning mirror to produce unshaped, transform-limited pulses.

As a first test we used the shaper in a second-harmonic generation (SHG) experiment. We addressed the scanner with a harmonic wave of 2.5 kHz , scanning around the interface between the patterned mask and the flat region of the mirror. The pulse from the shaper was directed into a SHG crystal. The actual scanning mirror angle, as well as the measured SHG signal, is shown in Fig. 2. The second-harmonic signal has an almost rectangular waveform, oscillating between a maximum signal (when the pulse is transform limited) to one third of this value when the shape is modulated. It is interesting to note that the SHG signal remains nearly constant in spite of the changing period of the spectral modulation. This can be easily understood: The periodic spectral phase modulation splits the transform-limited pulse into a train of pulses. The changing periodicity affects the

repetition rate of these pulses but not their relative heights. The SHG depends only on the instantaneous intensity, and therefore the integrated SHG signal is virtually constant regardless of the spacing between the pulses in the train. Indeed, calculation shows that a square-wave phase-modulated spectrum should reduce the total SHG by a factor of 3. This simple experiment demonstrates the ability of the scanning shaper to achieve shape-modulation rates greater than kilohertz.

In the second experiment we demonstrate the potential of lock-in measurements made possible by pulse-shape modulation in a nonlinear spectroscopy experiment. It was shown recently that CARS spectroscopy can be performed by a single ultrashort pulse, provided that it is spectrally broader than the relevant Raman levels.⁸ Selective Raman excitation is induced by periodic modulation of the spectrum, preferably phase modulation; the maximum resonant CARS signal is obtained when the Raman frequency is on integer numbers of periods, whereas the signal is minimal when it is on nearly half-integer numbers of periods, of the spectral phase function.¹¹ Both signals ride over a large nonresonant background, which, as with SHG in the previous experiment, is not sensitive to the modulation frequency. To obtain the contribution of the resonant CARS signal alone, we must sense the difference signal between two modulation patterns and eliminate all background signals. This is best done by alternating between the two patterns at a rate that is fast enough to eliminate other noise sources and by using lock-in detection.

In the experiment the pulse from the shaper was focused into a spectroscopic cell containing iodomethane (CH_3I), with a resonant Raman level at 523 cm^{-1} . The resulting CARS, at wavelengths just below the excitation spectrum, was filtered out by short-pass filters and detected by a photomultiplier. First, with a stationary mirror, we scan the filter mask along the vertical direction, thereby changing the spectral phase

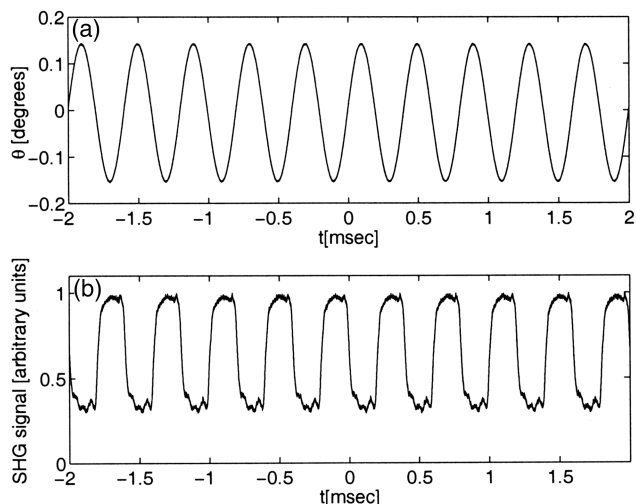


Fig. 2. SHG measurement with modulated pulse shapes at a rate of 2.5 kHz . (a) SHG signal. The mirror oscillates the spectrum between flat and modulated regions on the phase mask. (b) Scanning mirror angular position.

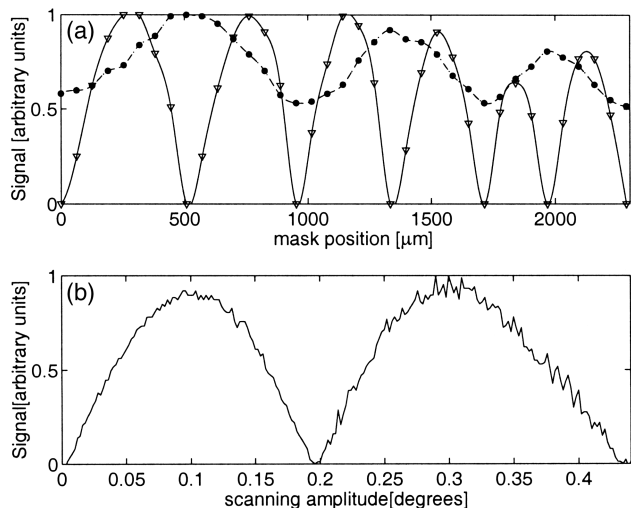


Fig. 3. Experimental CARS measurements of iodomethane (a single resonance at 523 cm^{-1}). (a) Circular data points, CARS signal as a function of mask position, when scanning mirror is not moving; triangular data points, CARS signal with lock-in on the pulse shape. Small angle (0.08°) amplitude scanning mirror modulation at 1000 Hz). (b) Lock-in CARS as a function of the scanning amplitude.

modulation frequency. The detected signal is shown by the filled circles in Fig. 3(a); it shows the total CARS signal, which is a combination of the resonant signal, which varies with the modulation frequency, and the nonresonant signal, which is independent of it. The spectroscopic information is obviously contained in only the resonant component. For most practical applications the nonresonant contribution is an undesirable background.¹²

Next we modulated the scanning mirror with a small-amplitude rectangular wave, moving it between two particular positions on the mask. Then we scanned the mask in the vertical direction as in the previous case. The signal from the photomultiplier was measured with a lock-in amplifier synchronized with the reference coming from the scanner drive signal. The resulting signal is shown in Fig. 3(a). The maxima are achieved when the mask is positioned in such a way that the scanner jumps between two pulse shapes—one maximizing the resonant signal and the other minimizing it—whereas minima are achieved when the scanner switches symmetrically between pulse shapes, providing equal signal contribution. This latter case corresponds to practically no signal in the lock-in measurement as can be seen

in Fig. 3(a). Another experimental manifestation of the same principle can be seen in Fig. 3(b). Here we increase the amplitude of the scanning, starting at some location inside the mask. As expected we observe the periodic behavior while the scanner goes through successive multiple orders of the molecular Raman resonance. Both measurements demonstrate the power of the lock-in technique to practically eliminate the background and increase the contrast of the measurement.

In summary, we have presented a practical femtosecond scanning pulse shaper that allows switching between pulse shapes at kilohertz rates. We note that with faster resonant optical scanners and by use of multistripped masks the rate can be increased by at least 1 additional order of magnitude. Lock-in on the pulse shape has been proposed and demonstrated, dramatically improving the signal-to-noise/background ratio. We expect the scanning shaper to be particularly useful in nonlinear imaging applications, in which a resonant signal should be discriminated from the nonresonant background.

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