Multiphoton intrapulse interference 6; binary phase shaping

Matthew Comstock, Vadim V. Lozovoy, Igor Pastirk, and Marcos Dantus
Department of Chemistry and Department of Physics and Astronomy, Michigan State University, East Lansing, MI 48824

dantus@msu.edu

Abstract: We demonstrate a new approach to laser control using binary phase shaping. We apply this method to the problem of spectrally narrowing multiphoton excitation using shaped laser pulses as required for selectivity in two-photon microscopy. The symmetry of the problem is analyzed from first principles and a rational solution is proposed. Successful experimental implementation and simulations are presented using 10 fs ultrashort pulses. The proposed solution is a factor of 6 better than the sinusoidal phase used previously by our group. An evolutionary learning algorithm was used to efficiently improve the solution by a further factor of 2.5 because of the greatly reduced search space afforded by binary phase shaping.

OCIS codes: (320.0320) Ultrafast Optics; (320.5540) Pulse Shaping; (180.2520) Fluorescence Microscopy

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1. Introduction

Amplitude and phase shaped laser pulses could be used to control chemical reactions, and other physical phenomena. Because calculation of the required field would be prohibitively time consuming the use of evolutionary learning algorithms (ELA) guided by experimental results in a closed feedback loop was proposed [1]. Experimental implementation of this method and successes in control of chemical reactivity have been reported and reviewed [2-4].

In the low intensity regime, laser control is dominated by interference of different nonlinear optical pathways connecting the initial and final states [5]. The challenge is finding the proper phase for each frequency within the pulse to achieve constructive interference at the desired pathway and destructive interference elsewhere. Here we consider two-photon excitation of fluorescent probes, as used in two-photon microscopy, as the target for optimization. In particular we envision two chromophores with different two-photon absorption spectra and our goal is to achieve selective excitation by 'focusing' the energy available at a specific region of the two-photon spectrum, as shown in Fig. 1.

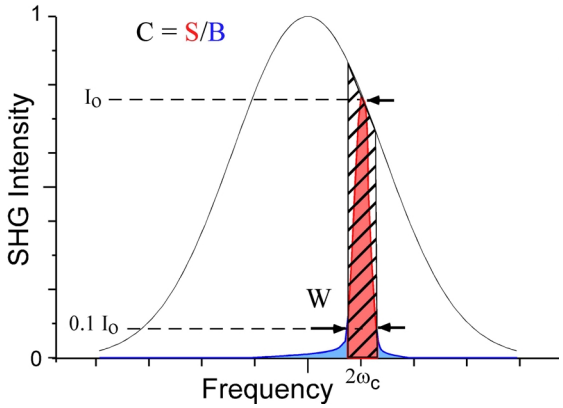

Fig. 1. Cartoon representation of the problem. The broad bandwidth second harmonic spectrum from transform-limited pulses is represented by a Gaussian (thin line). The objective is to introduce phase modulation to cause the two-photon spectrum to be intense only inside the window defined by frequency $2\omega_c$ and width W, and to minimize the background B outside the window. The contrast ratio C is defined as the integrated intensity inside W divided by the integrated intensity of light outside the window.

Experiments from our group have already shown that selective two-photon excitation is possible when one is able to tune the narrowed non-linear power spectrum to optimize excitation of one chromophore versus another [6,7]. Here, however, we realize that the phase between different frequencies should take only two values 0 or π to maximize or minimize a given pathway.

In order to control two-photon excitation, one needs to control the non-linear power spectrum of the laser $E^{(2)}(\omega)$ [8,9], which can be measured by obtaining the second harmonic spectrum generated using a thin second harmonic generation (SHG) crystal. The experimental and theoretical application of this new paradigm, which we call binary pulse shaping (BPS), is presented herein. The effect of spectral phase modulation on SHG has been studied by a number of groups and can be divided into broad [10-12] and narrow [13,14] phase matching bandwidth.

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Conceptually, the problem depends on the intrapulse interference between pairs of frequencies. If we only consider two phases, zero and π, we can analyze the problem in terms of symmetry. To maximize the SHG intensity at a frequency $2\omega_C$ the spectral phase needs to be symmetric or antisymmetric about ω_C, such that the frequencies interfere constructively. To minimize the background intensity at all other frequencies, the spectral phase must be asymmetric with respect to all other frequencies away from ω_C, so that destructive interference is maximized. Here we propose the use of prime numbers to help us generate the quasi-random phase changes required. The mask to be used to modulate the pulses is shown in Fig. 2, and is designed for a 128-pixel SLM.

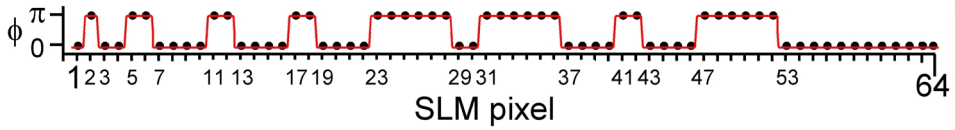

Fig. 2. Phase mask proposed based on the symmetry requirements of the problem, using the quasi randomness of prime numbers. This mask is reflected about pixel 64, and is designed to obtain a narrow second harmonic signal at the center of the spectrum.

2. Experimental section

The experiments are carried out using a titanium-sapphire oscillator laser system capable of generating pulses as short as 10 fs after a double pass prism compressor and a pulse shaper [6]. The spectral phase of the pulse is tailored using a computer-controlled pulse shaper of a design similar to those described by Weiner [15], and described elsewhere [6,7]. For the experiments shown here, the pulses were centered near 800 nm. The spectral phase of the laser pulses was corrected using the multi-photon intrapulse interference phase-scan (MIIPS) method, which compensates phase distortions to obtain transform-limited (TL) pulses [6,7]. The binary phase was introduced as an addition to the compensation phase.

The shaped laser pulses, with an energy of ~ 0.5 nJ per pulse and 87 MHz repetition rate, were focused mildly, to a spot size of ~ 100 microns in diameter, on a 20 micron thin βBBO SHG crystal. The frequency doubled light was collected with an optical fiber and dispersed on a compact spectrometer.

3. Results

First, we considered spectral amplitude restriction to solve the problem. We narrowed the spectrum of the laser using a slit as shown in Fig. 3(a), the SHG intensity obtained after amplitude restriction is shown in Fig. 3(b). For a spectral width of 10% of the available bandwidth, amplitude restriction produces 100 times less second harmonic intensity than transform limited pulses. Although the contrast ratio for this case is favorable, the low intensity obtained at the desired wavelength makes this method experimentally unpractical.

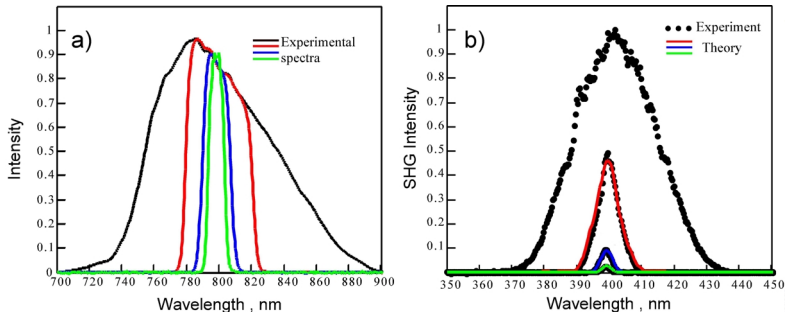

Fig. 3. Effect of spectral amplitude restriction on SHG. (a) Experimental spectrum of the laser before (black) and after filtering with windows of width 40 (red), 20 (blue), and 10 (green) nm. (b) Experimental (points) and simulation (continuous lines) for the second harmonic spectrum of TL and spectrally filtered pulses as indicated in panel (a).

#3579 - $15.00 US Received 22 December 2003; revised 1 March 2004; accepted 10 March 2004

The phase mask shown in Fig. 2 was programmed on the SLM and resulted in dramatic narrowing of the SHG spectrum as shown in Fig. 4. The contrast ratio as defined in Fig. 1 for this mask is 2.5 when the SHG peak is centered. By shifting the position of the mask on the SLM, hence tuning the center of symmetry, we can tune the SHG spectrum as shown in the animation included in Fig. 4. Sinusoidal phase modulation [6-9,11], can not produce contrast ratios greater than 0.5 in theory, and as the window is tuned away from the central frequency the contrast ratio drops below 0.1.

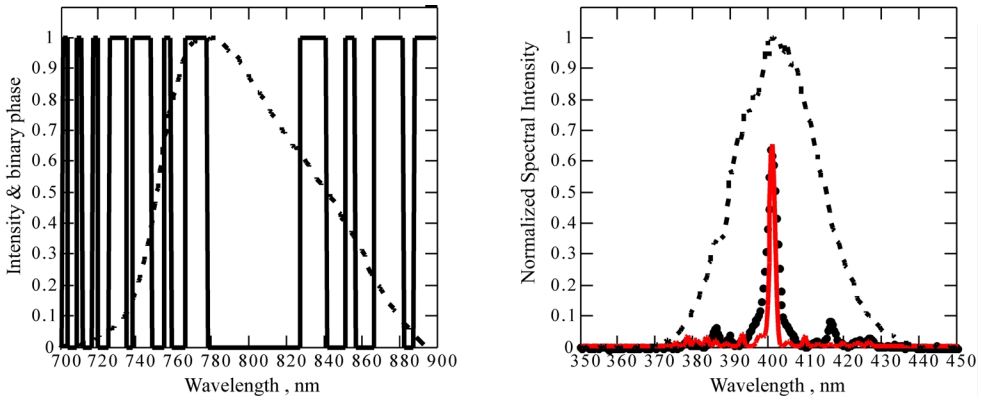

Fig. 4. Experimental results with binary phase shaping. (a) The spectrum of the laser (dashed lines) and the binary phase mask (0 or π) are shown as a function of wavelength. (b) The second harmonic spectrum of the TL pulses (dashed line) and second harmonic spectrum of the shaped pulses according to panel (a). The movie (428 kB) shows how translation of the binary phase mask across the spectrum tunes the frequency where the second harmonic is focused.

The red line in Fig. 4 corresponds to theoretical calculations. The absolute value of the spectral amplitude of the electric field was calculated from the experimental power spectrum of the fundamental pulse $I(\omega)$ using $|E(\omega)| = I(\omega)^{0.5}$. To simulate the experimental results we used a double Fourier transform method. The electric field in the time domain $E(t)$ was calculated as the Fourier image of the complex spectral amplitude in the frequency domain, with the formula

$$E(t) = \int |E(\omega)| \exp[i\phi(\omega)] \exp(-i\omega t) \mathrm{d}\omega, \quad (1)$$

where the spectral phase $\phi(\omega)$ is the function that is introduced by the SLM. The power spectrum of the SHG was calculated using

$$I_{\mathrm{SHG}}(\omega) = \left| \int E(t)^2 \exp(i\omega t) \mathrm{d}t \right|^2. \quad (2)$$

The SHG amplitude was normalized using the maximum of the SHG intensity calculated for TL pulses, $\phi(\omega) \equiv 0$.

The agreement between experiment and theory in Fig. 4 is satisfactory given that there are no adjustable parameters. Differences between experiment and theory are due to the resolution of the pulse shaper at the Fourier plane and errors in the applied phase mask due to cross-talk between adjacent pixels. These resolution and calibration issues cause the experimental SHG spectrum to be slightly broader than expected, though the contrast ratio remains 2.1, comparable to the theoretical contrast ratio of 2.7.

We wanted to determine if a better solution existed. For this we used an ELA. We assumed a Gaussian electric field corresponding to a 10 fs pulse centered at 800 nm. The SHG intensity was calculated according to

$$I_{\mathrm{SHG}}(2\omega_c) = \left| \int E(\omega_c - \omega) E(\omega_c + \omega) \mathrm{d}\omega \right|^2 \quad (3)$$

normalized to the maximum SHG amplitude for TL pulses.

#3579 - $15.00 US Received 22 December 2003; revised 1 March 2004; accepted 10 March 2004

BPS simplifies the calculations, especially if we assume the amplitude of the electric field to be a constant, that is, we set the spectral power equal to 1 in the allowed spectral region. Each spectral component of the electric field, linearly dispersed in the frequency domain, can be represented as a binary value (± 1) determined by $b_k = \exp(i\phi_k)$, for $\phi_k = 0$ or π, respectively. The intensity of the SHG signal measured at frequency $2\omega_k$ can be calculated with the formula

$$S_k=|\Sigma_j b_{k-j} b_{k+j}|^2 \qquad (4)$$

where the integral in Eq. (3) is now replaced by a discrete sum. The problem of spectral selectivity can now be formulated as finding a vector b_k such that $S_k = 1$ for $\omega_k = \omega_C$ and S_k is minimized at all other frequencies.

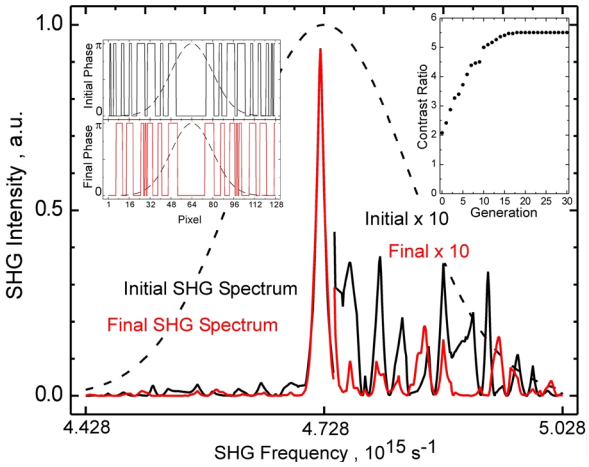

Fig. 5. Comparison of a prime number inspired phase mask (black) with a mask that was optimized using a computer based learning algorithm(red). The insets on the left depict the phase masks for each case. The inset on the right shows the improvement in the contrast ratio, as defined in Fig. 1, as the learning algorithm finds the best solution starting from the prime number phase mask.

In Fig. 5, we compare theoretical results from the phase described in Fig. 2 (black) and the result of the ELA (red), which was initiated with the proposed phase. The ELA used 100 individuals and one parent through 30 generations using single double and triple symmetric bit flips. We find that the result from the learning algorithm gives ~2.5 greater contrast. Experimentally contrast ratios greater than 5 are difficult to obtain when the spectrum is not linearly dispersed over the SLM pixels. Using an SLM with more pixels [16], and grouping those pixels such that each group covers the same frequency range, would improve the implementation and result in greater contrast, an observation based on our calculations.

4. Discussion

The goal of narrowing the SHG spectrum was motivated by the possibility of selective two-photon microscopy. To achieve this goal two conditions had to be satisfied, maximizing the nonlinear power spectrum at $2\omega_c$, and minimizing it elsewhere. Maximization has been investigated previously by Silberberg and coworkers, who identified the condition $\phi(\omega_c-\omega)=-\phi(\omega_c+\omega)$ for continuous functions [17,18]. Zheng and Weiner explored the narrow phase matching bandwidth SHG output at a single frequency, a problem that is analogous to two-photon excitation of an atom, using binary encoded pulses as used in communications [13,14]. Zheng and Weiner found that limiting the phase to values of zero and π led to the condition $\phi(\omega_c-\omega)=\phi(\omega_c+\omega)$. Under both shaping conditions the maximum signal approached that obtained with TL pulses.

#3579 - $15.00 US Received 22 December 2003; revised 1 March 2004; accepted 10 March 2004

The problem of spectral narrowing by pulse shaping under broad phase matching SHG conditions, or selective excitation of large organic dyes and chromaphores requires minimization of the nonlinear power spectrum away from $2\omega_c$. Upon study of the problem, as expressed in Eq 4, we realized that there is an analogy with convolution. In this case, some binary functions have the property of giving a sharp 'delta function' signal in one location and very low background elsewhere.

In principle, the solutions found in this study are members of the set of solutions obtained by arbitrary phase and amplitude pulse shaping. For a pulse shaper with *N* pixels, one can generate $(P*A)^N$ shaped pulses, where *P* and *A* are the number of different phases and amplitudes a pixel can take. If we assume 100 pixels, each taking 10 different amplitude values and 100 different phase values, the number of different pulses is of order of magnitude 10^{300}. This number is extremely large, therefore, while in principle, the solution exists to achieve the desired photonic transformation or excitation, finding it is very time consuming except for a convex problem with a clear maximum.

Experimentalists have reduced the search space of an ELA by reducing the number of active pixels by grouping, the type of phase functions that are allowed, modulating phase and not amplitude, and introducing phase functions with a reduced numbers of parameters [19-21]. We tried these approaches with limited success.

For one pulse experiments, the periodic nature of electromagnetic waves results in a great deal of redundancy in pulse shaping because nonlinear optical processes do not depend on the absolute phase or a linear variation of the spectral phase. This equivalence can be expressed by $\phi(\omega) \Leftrightarrow \phi(\omega) + a + b\omega$, where *a* and *b* are constants. Programming an ELA that acts on the second derivative of the phase can filter out this redundancy. The actual phase is obtained by integrating setting $a=b=0$. We used such an ELA to optimize smooth phase functions for spectral narrowing, but could not obtain a contrast ratio greater than unity.

The advantage of BPS is that it eliminates most redundancies. For BPS and 128 active pixels the search space is reduced to 2^{128}. If the problem has two-fold symmetry, for example two-photon excitation, then the search space is reduced to 10^{19}, a number that is at least 281 orders of magnitude smaller than would be considered for arbitrary phase and amplitude pulse shaping as discussed above. An ELA can quickly converge in this small search space without resorting to grouping pixels.

BPS may have significant technological advantages. A retardation equivalent to π is easy and fast to obtain and calibrate. Permanently etched masks can be made in advance and used for specific applications such as selective two-photon microscopy. Scanning the mask's position can yield two-photon excitation spectra.

In summary, we have demonstrated the use of BPS for selective two-photon excitation. The contrast ratio using BPS was 6 times greater than that obtained using sinusoidal phase masks. The amplitude of the signal is 100 times greater than would have been obtained by amplitude modulation. BPS makes it simple to analyze a laser control problem and to propose rational solutions, as demonstrated here with a phase mask inspired by the quasi-random gaps between prime numbers. An ELA can be used to improve on the proposed solution efficiently because of the greatly reduced search space.

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