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# Tunable second harmonic generation by phase-modulated ultrashort laser pulses

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**Abstract** We report on the generation of tunable light around 400 nm by frequency-doubling ultrashort laser pulses whose spectral phase is modulated by a sum of sinusoidal functions. The linewidth of the ultraviolet band produced is narrower than 1 nm, in contrast to the 12 nm linewidth of the non-modulated incident spectrum. The influence of pixellation of the liquid crystal spatial light modulator on the efficiency of the phase-modulated second harmonic generation is discussed.

## 1 Introduction

The outcome of nonlinear light–matter interaction can be controlled by shaping specific electric fields to produce either constructive or destructive interference among intrapulse spectral components during the interaction. This coherent control of light–matter interaction opens the possibility for applications such as selective two-photon microscopy [1], control of the outcome of both sharp and broad resonance systems [2, 3], and so on. The creation of a specific pulse shape can be accomplished by manipulating the phase and/or the amplitude of the spectral components of a femtosecond laser pulse using zero-dispersion pulse shapers. The most common pulse shaper uses a liquid crystal-based spatial light modulator (SLM) [4, 5], but other techniques for generating shaped ultrashort laser pulses, such as acousto-optic modulation (AOM) [6], acousto-optic programmable dispersion filter (AOPDF) [7], micro-electro-mechanical system (MEMS) [8], and deformable mirror [9] were also demonstrated.

The effect of spectral phase modulation on the second harmonic (SH) generation has been studied by a number of groups [10–14]. In particular, Hacker et al. [10] investigated the SH generation of phase-modulated pulses in BBO crystals with the purpose of producing shaped pulses in the ultraviolet (UV) spectral region for direct electronic excitation of molecular or atomic systems. Their motivation relies on the fact that one-photon excitations of quantum systems have selection rules different from those of two-photon absorption [2], in particular, different angular momentum transfer. In their study, frequency doubled sinusoidally phase-modulated fundamental pulses were used and in this case, the electric field of the SH can be approximated by a zero-order Bessel function whenever the modulation period is shorter than the pulse linewidth. The same sort of modulation was employed to demonstrate selective two-photon excitation of fluorescent probes for microscopy [1]. Later, an approach based on a binary phase shaping was demonstrated to spectrally narrow the multiphoton excitation, as required for selectivity in two-photon microscopy [11]. The proposed scheme improved the contrast ratio by a factor of 6 when compared to the sinusoidal phase alone, and the use of an evolutionary learning algorithm even improved the solution by a further factor of 2.5. Another interesting approach used for high-resolution, high-contrast, nonlinear optical spectroscopy, also based on phase-modulated ultrashort pulses, uses low-autocorrelation binary sequences that give rise to Galois fields [15]. This method allows achieving selective nonlinear optical excitation while strongly suppressing the background.

In this study, we show that it is possible to generate tunable, narrowband light around 400 nm by frequency-doubling ultrashort laser pulses whose spectral phase is

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modulated by a sum of sinusoidal functions with different frequencies. The idea behind this modulation scheme is that additive phase modulation results in a product of exponential functions, each one giving rises to a zero-order Bessel function. With periods and phases properly chosen, the product of these functions is nearly zero, except in a narrow band. This band can be continuously tuned across the whole spectrum, which is an important feature for selectivity in two-photon microscopy and one-photon spectroscopy in the UV region. A positive aspect of the phase-modulation approach presented here is that it relies in an analytical function and does not require any complex evolutionary learning algorithm to produce fairly good results.

## 2 Tunable second harmonic generation

The analytical description of second harmonic generation (SHG) of sinusoidal phase-modulated ultrashort laser pulses in a thin nonlinear crystal is presented in Ref. [10]. For a Gaussian incident pulse $E_1$, of frequency $\omega_1$ relative to the center frequency, carrying a spectral phase modulation of the type $\Phi \sin(\Delta t \omega_1 + \psi)$ the generated SH field $E_2$ of frequency $\omega_2$ is given by:

$$E_2(\omega_2) \sim \exp\left[-\frac{1}{2}\left(\frac{\omega_2}{\Delta\omega_1}\right)\right]^2 \times J_0\left[2\Phi \sin\left(\frac{1}{2}\Delta t \omega_2 + \psi\right)\right], \quad (1)$$

where $\psi$ is an arbitrary constant phase, $\Delta t$ is the spectral frequency, $\Phi$ is the modulation depth, and $\Delta\omega_1$ is the spectral width of the fundamental beam. This expression is strictly true for $\Delta t \Delta\omega_1$ sufficiently large, as it is usually the case. $2\Phi$ can be conveniently set to 2.4 rad to accomplish perfect modulation, which corresponds to the first zero of the Bessel function $J_0$.

In what follows, we assume a phase modulation of the type $\Phi \sum \sin[n_j(\Delta\omega_1 + \psi)]$, where $j$ is the summation index and the number $n_j$ can be arbitrarily chosen. By using the same mathematical procedure of Ref. [10], one can show that the generated SH field is written as

$$E_2(\omega_2) \sim \exp\left[-\frac{1}{2}\left(\frac{\omega_2}{\Delta\omega_1}\right)\right]^2 \times \prod J_0\left[2\phi \sin\left\{n_j\left(\frac{1}{2}\Delta t \omega_2 + \psi\right)\right\}\right]. \quad (2)$$

Each Bessel function presents a spectral modulation with a spectral frequency $n_j \Delta t$, as shown in Fig. 1a for the case where $n_j$ are integers. Therefore, the product of Bessel functions with different frequencies produces a narrow, well-defined maximum whose width is determined by the shortest period, as depicted in Fig. 1b. In this case, we have a situation that mimics a narrow-band filter made of a stack of tilted birefringent plates placed inside a laser resonator [16]. The idea of the method is that the maximum of a Bessel function with a given period coincides with the zero of another one, except when the argument of the sinusoidal function is multiple of $\pi$. Obviously, the use of Bessel functions is just an approximation. If we consider the Gaussian profile of the fundamental laser pulse and the applied spectral phase as a sum of sinus functions, we can numerically calculate the SH spectrum, as shown in Fig. 1c. A noticeable difference is that the sidebands are smaller in the case where the approximation with Bessel functions is considered. By changing the phase $\psi$, the output can be easily tuned across the broadband non-modulated SH profile, as required for one-photon spectroscopy and two-photon microscopy. Although this

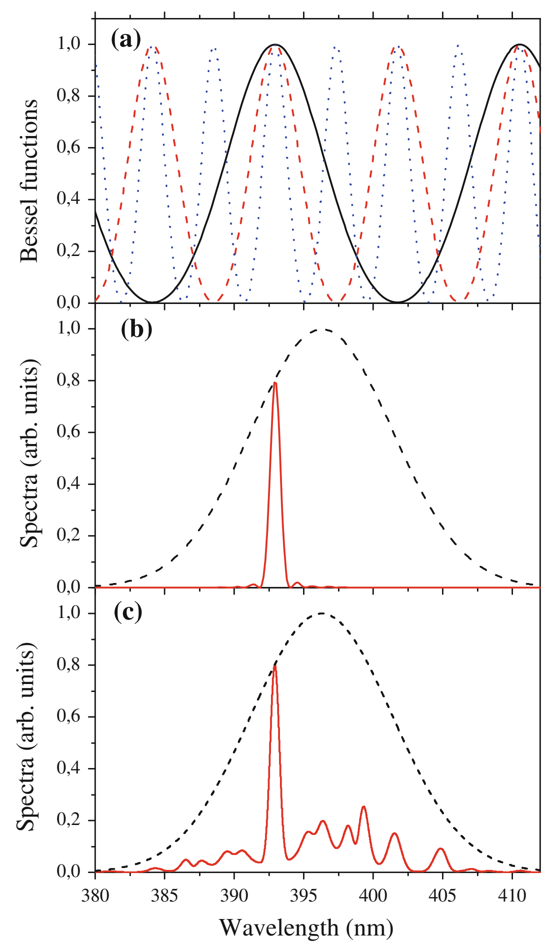


**Fig. 1** **a** Plot of $E_2(\omega_2)$ given in Eq. (1) for $n_j = 2$ (*solid line*), $n_j = 4$ (*dashed line*), and $n_j = 8$ (*dotted line*). $\psi$ is 0.31 rad and $\Delta t$ is set to 18 fs. **b** Spectra of the non-modulated second harmonic (*dashed line*) and modulated with a product containing $n_j = 1, 2, 4, 8$, and 16 in Eq. (2) (*solid line*), and **c** numerical simulation of the SH spectrum considering a Gaussian profile for the fundamental laser pulse and the applied spectral phase as a sum of sinus functions with the same $n_j$ as in (**b**)

choice for a set of $n_j$ theoretically produces the desired narrowing, the pixellation of the liquid crystal (LC) modulator leads to a significant limitation because the higher frequencies sinusoidal phases are transformed into staircase-shaped functions by the SLM. In this case, the Bessel function is not a good approximation and the result is that the product of functions is smaller than 1 at the maximum and sidebands start to show up. To circumvent this problem, we used a set of smaller $n_j$ that are not integers, as presented later.

## 3 Experimental details

The experimental arrangement used to manipulate the spectral phase of the fundamental beam is depicted in Fig. 2. Ultrashort laser pulses were delivered by a Ti:sapphire oscillator system (KM Labs, Ti:sapphire kit pumped by a 5 W Nd:$YVO_4$ laser operating at 532 nm). The central wavelength was set at 790 nm, the bandwidth was about 35 nm (FWHM), the pulse duration was 27 fs (FWHM, corresponding to a transform-limited (TL) pulse) at a pulse-energy level of 5 nJ and a repetition rate of 80 MHz. The pulses are sent to a phase-only pulse shaper, where the LC-based SLM is placed at the Fourier plane of a folded 4*f* zero-dispersion compressor consisting of a 600-grooves/mm grating and a spherical mirror of focal length 30 cm. By folding the optical path with a plane mirror, we save one diffraction grating and one spherical mirror, besides duplicating the phase change owing to the fact that the beam passes twice through the LC modulator. Careful positioning of the grating guarantees the TL condition, as confirmed by frequency-resolved optical gating (FROG) measurements carried out with a home-made apparatus. After passing through the SLM, the beam was focused into the crystal with a 10-cm focal-length lens. Coherent control of SHG using SLM has been investigated in a thin 300-μm-thick KDP crystal cut for type I phase matching. The SH generated was analyzed by means of a 0.3-nm-resolution UV portable spectrometer, although we also used a 60-cm monochromator, with a resolution better than 0.1 nm, to make sure that the resolution of the spectrometer did not affect the results.

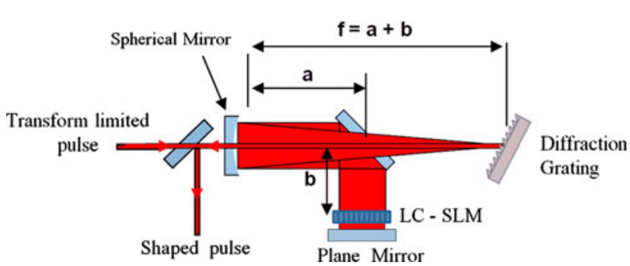


**Fig. 2** Layout of the folded SLM used to control the spectral phase of the fundamental beam

## 4 Results and discussion

Spectra of the non-modulated SH (dashed line) and modulated with products containing $n_j = 1$, 2, 4, 8, and 16 (solid lines) are shown in Fig. 3. Several values of $\psi$ were used to demonstrate the tunability. As we advanced earlier, the spectra obtained are narrow but the amplitudes are half of what should be expected from the product of Bessel functions shown in Fig. 1b. Moreover, there are a few small sidebands around the main peaks, which could produce some background noise in selective two-photon excitation microscopy.

As already mentioned, the spectral modulation should be a smooth function but the finite size of the individual modulator elements produces a staircase approximation that limits the phase control process and hence the efficiency of the phase-modulated SH. In other words, the Bessel solution of Eq. (1) is not a good approximation. To confirm this fact, we applied a single, variable frequency sinusoidal function and obtained the modulated signal as shown in the inset of Fig. 4, together with the non-modulated SH. By normalizing the modulated SH to the non-modulated one, we obtained spectral fringes similar to those of Fig. 1a, from which we can define the contrast as $\eta = (I_{\max} - I_{\min})/(I_{\max} + I_{\min})$, where $I_{\max}$ ($I_{\min}$) is the maximum (minimum) of the normalized fringe pattern. The result depicted in Fig. 4 shows that $\eta$ decreases for higher spectral frequencies. A similar behavior was

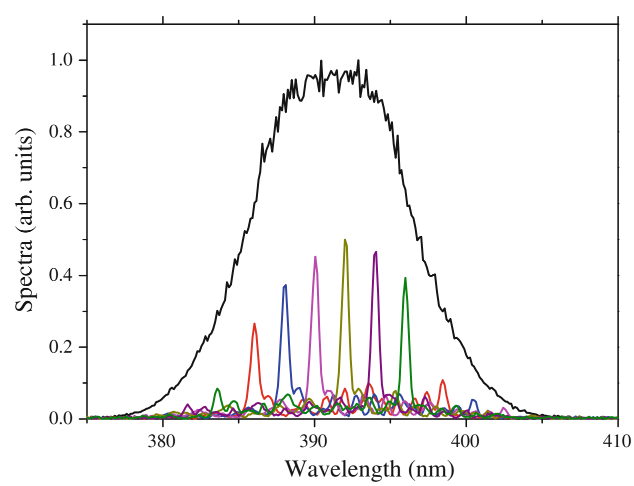


**Fig. 3** Spectra of the non-modulated second harmonic (*upper curve*) and modulated with a product containing $n_j = 1$, 2, 4, 8, and 16 (*lower curves*) for phases 1.26, 0.76, 0.26, −0.26, −0.76, and −1.26 rad, from *left* to *right*

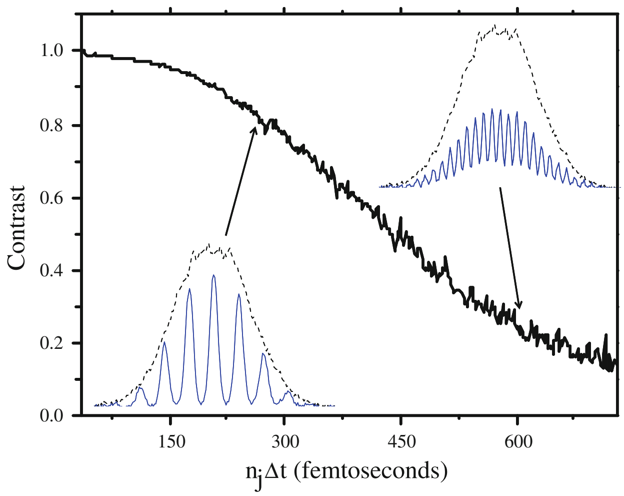


**Fig. 4** Contrast of the fringes obtained with a single sinusoidal modulation for different spectral frequencies. The *insets* show two examples of SH obtained with the single sinusoidal phase modulation together with the non-modulated phase

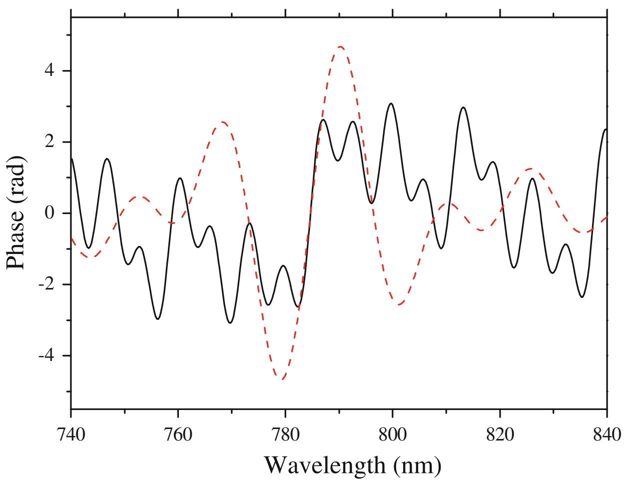


**Fig. 5** Phase modulation for $n_j = 1, 2, 4, 8$, and 16 (*solid line*) and $n_j = 3, 3.75, 4.5, 5.25$, and 6 (*dashed line*). $\psi$ is 0.31 rad and $\Delta t$ is set to 18 fs

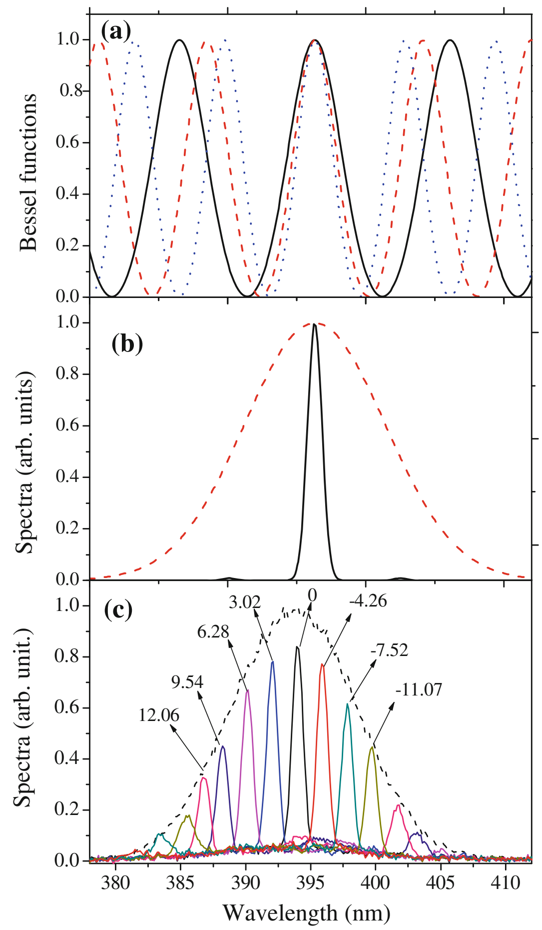


**Fig. 6** **a** Plot of $E_2(\omega_2)$ given in Eq. (1) for $n_j = 3$ (*solid line*), $n_j = 3.75$ (*dashed line*), and $n_j = 4.5$ (*dotted line*). **b** Spectra of the non-modulated second harmonic (*dashed line*) and modulated with a product containing $n_j = 3, 3.75, 4.5, 5.25$, and 6 (*solid line*) for $\psi = 0$ rad and $\Delta t = 18$ fs. **c** Spectra of the non-modulated second harmonic (*dashed line*) and modulated with a product containing $n_j = 3, 3.75, 4.5, 5.25$, and 6 (*solid lines*) for different phases given in rads by the *arrows*

observed in Ref. [10], although that effect was attributed to the spectrometer resolution, which is not our case.

To mitigate this sampling limitation one must use a phase modulation that varies sufficiently slow to be adequately sampled by the fixed modulator elements. This can be accomplished by using $n_j$'s that are closely spaced, without the requirement of being integers. As Fig. 5 shows, such choice of parameters produces a phase modulation that varies slower than the case where the $n_j$'s are integers and multiples. Plots equivalent to those of Fig. 1 are shown in Fig. 6a and b for a combination of $n_j$'s equal to 3, 3.75, 4.5, 5.25, and 6. Here, one has the product of five Bessel functions, which is 1 just at the origin, i.e., when the arguments of all sinusoidal functions are 0 (this happens at the center of the band for $\psi = 0$). When the arguments departs from 0, each Bessel function starts to decrease and their product can become a number much smaller than 1 if the number of Bessel function is high enough. Therefore, the linewidth is related mostly to the number of Bessel functions, in contrast to the case where $n_j$'s are integers and multiples, where the width is determined mainly by the shortest period of the sinusoidal function.

Figure 6c shows the spectra of the non-modulated SH (dashed line) and modulated with products containing $n_j = 3, 3.75, 4.5, 5.25$, and 6 (solid lines). The result shows that the amplitude of the modulated SH is closer the non-modulated case, although not 1 because the contrast of each Bessel function's fringe is slightly less than 1. Moreover, the linewidth is somewhat broader, but still acceptable for the purpose of one-photon spectroscopy and two-photon microscopy.

## 5 Conclusions

The phase-modulation approach based in a sum of sinusoidal functions produces the narrowing and tunability of the UV line necessary for selective two-photon microscopy. In addition, it does not require any complex evolutionary learning algorithm for its implementation. Two kinds of modulations can be employed that where $n_j$'s are integers and multiples produce narrower lines, but the amplitude is just half of the optimum non-modulated SH, while the one where the $n_j$'s are closely spaced, but not integers, give a more intense SH at the expense of the line broadening.

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