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Frequency doubling of phase-modulated, ultrashort laser pulses

M. HACKER
R. NETZ
M. ROTH
G. STOBRAWA
T. FEURER
R. SAUERBREY ✉

Institut für Optik und Quantenelektronik, Friedrich-Schiller-Universität, Max-Wien-Platz 1, 07743 Jena, Germany

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ABSTRACT We have investigated theoretically and experimentally the generation of shaped pulses at 400 nm by frequency-doubling of phase-modulated, ultrashort laser pulses. We present an analytical description of frequency-doubled pulses with a sinusoidal spectral phase modulation. It is shown that such a phase modulation can be transferred completely into a spectral amplitude modulation with a resolution determined only by the phase-modulating device. A criterion for achieving the maximum modulation contrast is given.

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1 Introduction

The techniques of generating programmable ultrashort laser pulses [1, 2] allow the creation of nearly any pulse shape. This opportunity has found its applications in dispersion control, in communication systems, including encryption and decryption of information [3], or and the generation of complex pulse structures for the coherent control of atomic [4, 5], molecular [6, 7] or solid-state systems [8]. The most common technique to achieve modulated fs pulses is the manipulation of the spectral phase [9] and/or amplitude [10] of the laser pulses using a zero-dispersion compressor setup with a liquid crystal [1], an acousto-optical modulator (AOM) [2] or a deformable mirror [11] acting in the Fourier plane. Because of the limited range of transparency of most modulators, these techniques are restricted to the visible and near-infrared (NIR) spectral regions. Recently reported experiments have extended pulse-shaping techniques to other wavelength regions [12, 13]. Although deformable mirrors in principle offer the possibility of directly accessing other spectral regions, the range of producible phase modulations is restricted to rather smooth functions with small amplitudes. These devices are therefore only preferred for the compensation of residual phases in laser systems. For direct electronic excitation of molecular or atomic systems, shaped pulses in the ultraviolet spectral region are essential. One-photon excitations offer new possibilities for transitions in quantum systems as a consequence of the selection rules, considering the different angular momentum transfer compared to two-photon absorption excitation schemes [5].

Because the phase modulation of ultrashort laser pulses in the NIR has become a state-of-the-art technique, including the availability of quite sophisticated phase shaping devices [14, 15], we investigate the second harmonic generation (SHG) of phase-modulated pulses as a way of generating shaped ultrashort pulses in the ultraviolet region. Several methods are available to extend the spectral acceptance of thick nonlinear crystals [16–19]. Therefore, we restricted our studies to SHG in thin crystals that accept the full input bandwidth. To the best of our knowledge, the only known technique for generating shaped SH pulses is the use of fixed, longitudinally nonuniform, quasi-phase-matching (QPM) gratings [20].

In this paper we first discuss the general theoretical relationship between phase-modulated fundamental pulses and the corresponding SH pulses. A theoretical examination of frequency-doubled sinusoidally phase-modulated fundamental pulses is specifically presented. This type of phase modulation is not only important for the generation of pulse trains, but also is central for coherent quantum control of atomic systems by two-photon transitions [5]. The non-resonant, two-photon absorption probability in an atomic medium driven by sinusoidally phase-modulated laser pulses can be derived from the description of the SH spectrum of the same frequency-doubled pulses. Finally, we discuss the experiments and compare them to the theory.

2 Second harmonic generation of phase-modulated pulses

For a theoretical investigation of SHG of ultrashort laser pulses in a nonlinear crystal, the coupled differential equations describing the three-wave mixing of the involved pulse spectra [21] have to be evaluated. Several program packages [22, 23] are available that solve these equations numerically.

Assuming a crystal thin enough to accept the whole incident fundamen-

✉ Fax: +49-3641/947-202, E-mail: sauerbrey@ioq.uni-jena.de

tal pulse spectrum, and imposing no appreciable phase modulation on the pulse, the acceptance function η may be approximated as $\eta \simeq 1$. In the case of a type-I SHG process and assuming a non-depleted fundamental field, the coupled differential equations reduce to

$$E_2(\omega_2) \propto \int E_1(\omega_1) E_1(\omega_2 - \omega_1) \mathrm{d}\omega_1 \,, \tag{1}$$

i.e.

$$E_2(t) \propto E_1^2(t) \tag{2}$$

for the generated SH field envelope E_2 of frequency ω_2. The quantities E_1 and ω_1 characterize the fundamental field envelope.

As long as (2) is valid, frequency mixing does not affect the temporal pulse shape, apart from the squaring of the temporal amplitudes. But because (1) is a convolution integral, a mixing of frequency components within the pulse spectrum occurs which may influence both the spectral phase and amplitudes. Whether the phase modulation and the spectral phase are conserved in the frequency-doubling process depends on the nature of the phase modulation applied to the input pulse and cannot be easily determined. For example, linear and quadratic spectral phase modulations applied to Gaussian pulses are transferred to the SH without having any influence on the shape of the generated spectrum. Therefore, the phase modulation still carries the information about the temporally shaped pulse, and only a spectral broadening by a factor of $\sqrt{2}$ occurs. In contrast, a variety of phase modulations exists which change the shape of the generated SH spectrum: examples are cubic and sinusoidal phase modulations. In this case, information about the temporal pulse shape is transferred from the spectral phase to the spectral amplitude. Consequently, the SH spectrum experiences an amplitude modulation, and the spectral phase modulation may even vanish.

In the following we will discuss the effects of this type of phase modulations with the help of an exemplary problem. For the input field we assume a Gaussian pulse carrying a sinusoidal phase modulation with a spectral modulation frequency Δt:

$$E_1(\omega_1) \sim \exp\left[-\left(\frac{\omega_1}{\Delta\omega_1}\right)^2\right] \times \exp\left[\mathrm{i}\Phi \cos(\Delta t \cdot \omega_1 + \psi)\right] \,, \tag{3}$$

where ω_1 is the frequency relative to the center frequency, $\Delta\omega_1$ the spectral width, Φ the modulation amplitude and ψ an arbitrary constant phase. This field corresponds to a pulse train with a temporal separation Δt between subsequent pulse maxima:

$$E_1(t) \sim \sum_{n=-\infty}^{\infty} J_n(\Phi) \times \exp\left[\mathrm{i}n\left(\frac{\pi}{2} - \psi\right) - \frac{1}{4}(n\Delta t + t)^2 \Delta\omega_1^2\right] . \tag{4}$$

By inserting the field $E_1(\omega_1)$ into (1), we obtain the following result:

$$E_2(\omega_2) \sim \exp\left[-\frac{1}{2}\left(\frac{\omega_2}{\Delta\omega_1}\right)^2\right] \times \sum_{n=-\infty}^{\infty} a_n \,, \tag{5}$$

where

$$a_n = J_n\left(2\Phi \cos\left(\frac{1}{2}\Delta t \cdot \omega_2 + \psi\right)\right) \times \exp\left[\frac{1}{2}\mathrm{i}n\pi - \frac{1}{8}(n\Delta t \Delta\omega_1)^2\right] . \tag{6}$$

Obviously, only terms containing Bessel functions of low orders dominate the result, since the terms a_n exponentially decrease with $|n|$. Therefore, if $\Delta t \cdot \Delta\omega_1$ is sufficiently large, (5) can be approximated by the term containing the zero-order Bessel function:

$$E_2(\omega_2) \sim \exp\left[-\frac{1}{2}\left(\frac{\omega_2}{\Delta\omega_1}\right)^2\right] \times J_0\left(2\Phi \cos\left(\frac{1}{2}\Delta t \cdot \omega_2 + \psi\right)\right) . \tag{7}$$

A qualitative criterion for the validity of this approximation is given by the following inequality:

$$\Delta t \cdot \Delta\omega_1 > \sqrt{-8 \ln p} \,, \tag{8}$$

where p defines the ratio $|a_1|/|a_0|$.

Clearly, the resulting SH spectrum contains no phase modulation anymore but is strongly amplitude modulated instead, with the same modulation frequency as the phase modulation applied to the input pulse. Note that for every modulation frequency it is possible to achieve perfect modulation contrast, i.e. to produce periodic dips in the SH spectrum by choosing the appropriate phase modulation amplitude Φ. This would not be possible by SHG of a perfectly amplitude-modulated fundamental spectrum. In this case the SH spectrum would be subject to a spectral smearing caused by the convolution. Therefore the modulation contrast would decrease with an increasing spectral modulation frequency. If only the central spectral component $S_2 = E_2(0)$ of the SH is considered, (7) reduces to $S_2 \sim J_0(2\Phi)$, which is equivalent to the result obtained for the transition probability for non-resonant two-photon absorption in an atomic medium [5].

3 Experimental setup

We used a pulse shaper and ultrashort pulse diagnostics to study experimentally the dependence of the SH spectrum on the modulation frequency Δt for different relative phases ψ and on the phase-modulation amplitude Φ. The schematic arrangement used for the experiments is depicted in Fig. 1. Ultrashort laser pulses were delivered by a Ti:sapphire oscillator–amplifier system. The center wavelength was 810 nm, the bandwidth about 25 nm, the pulse duration 50 fs at a pulse-energy level of 1 mJ and a repetition rate of 1 kHz.

First, the laser pulse was divided by a glass plate used as a beam-splitter into a main pulse, which was sent to a phase-only pulse-shaper, and an unmodified reference pulse, which was required for the subsequent diagnostics. A detailed description of the phase-only pulse-shaper is given in [15]. After the main pulse was modified by the pulse-shaper it was either directly characterized by a cross-correlator or sent to a 100- or 500-μm-thick BBO crystal to generate shaped SH pulses. The cross-correlator used for the characterization of the shaped fundamental pulses was based on SHG in a 100-μm-

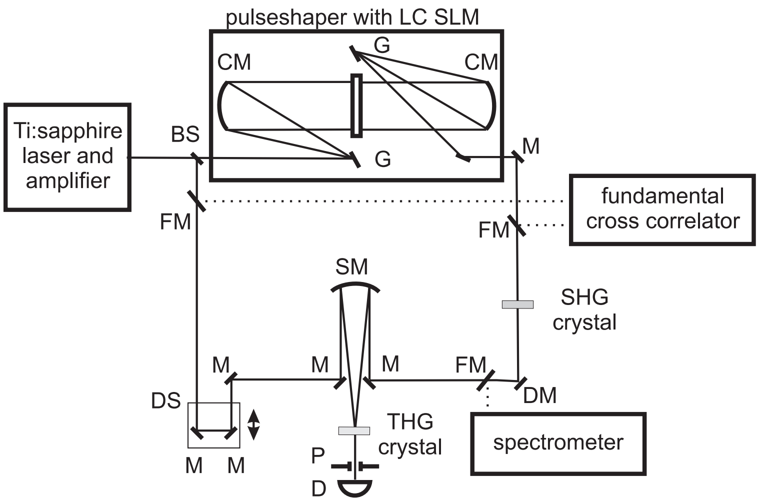

FIGURE 1 Experimental setup. BS: beam-splitter; CM: cylindrical mirror; D: detector; DM: dichroic mirror; DS: delay stage; FM: flap mirror; G: diffraction grating; LC SLM: liquid-crystal spatial light modulator; M: mirror; P: pinhole; SHG: second harmonic generation; SM: spherical mirror; THG: third harmonic generation. *Dotted lines*: optional beam lines

thick BBO crystal. Shaped SH pulses were characterized either by a spectrometer or a second cross-correlator. This intensity cross-correlator consisted of a 300-μm-thick BBO crystal in which the shaped SH pulses and the reference fundamental pulses were mixed to generate third harmonic radiation as a cross-correlation signal. The thicker frequency-doubling crystal was used for the generation of shaped SH pulses with sufficient pulse energy to permit subsequent characterization by cross-correlation.

4 Experimental results

To prove the validity of (7), the spectra of frequency-doubled, sinusoidally phase-modulated fundamental pulses were first measured as a function of the spectral modulation frequency Δt. To achieve perfect modulation, the modulation amplitude Φ was set to 1.2 rad, which corresponds to the first zero of the Bessel function J_0. Figure 2a and b show the measured SH spectra obtained for sine and cosine modulations, which correspond to relative phases in the exponent of (3) of $\psi = \pi/2$ and $\psi = 0$, respectively. The spectra show complete modulation as expected theoretically. For the sine modulation, the intensity of the center-frequency component is always maximal as theoretically expected, while for the cosine modulation the center frequency is only a clear minimum for $\Delta t > 100$ fs. Obviously, the theoretical description fails for smaller spectral modulation frequencies. This is in excellent agreement with (8), which predicts the validity of (7) for $\Delta t > 100$ fs for a spectral bandwidth $\Delta\omega_{\mathrm{FWHM}} = \sqrt{2 \ln 2} \cdot \Delta\omega_1$ of 25 nm at a center wavelength of 800 nm and $p = 0.01$. The decreasing modulation contrast at high modulation frequencies is attributed to the finite resolution of the spectrometer, because even at very high modulation frequencies ($\Delta t > 10$ ps), where spectral modulations were no longer visible, pulse trains were still detectable using the cross-correlator.

If we vary the modulation amplitude Φ for a sinusoidal phase modulation at a constant spectral-modulation frequency Δt we obtain the SH spectra shown in Fig. 3a. The dependence of the spectral intensity at the center of the SH spectrum (405 nm) is shown in Fig. 3b and is superposed with the theoretically expected curve calculated using (7). As predicted, a full modulation of the SH spectrum is achieved only using phase-modulation amplitudes Φ which correspond to zeros of the Bessel function J_0. The occurrence of well-defined minima at different Φ can be utilized for testing directly the phase calibration of the phase-modulating device.

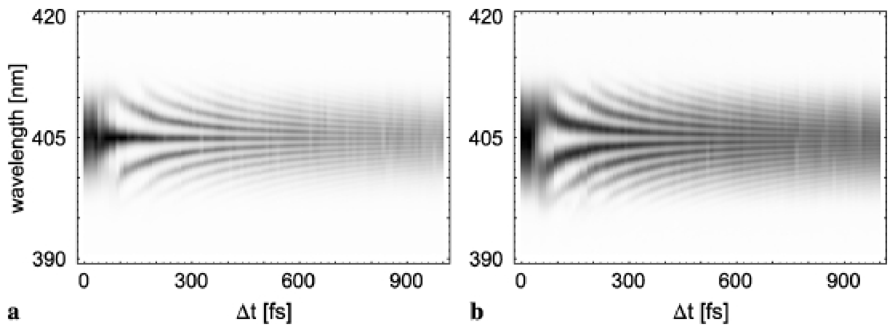

FIGURE 2 Measured spectra of frequency-doubled fundamental pulses with **a** antisymmetric sine and **b** symmetric cosine phase modulation as a function of the modulation frequency $\Delta t = 0$–1000 fs with $\Phi = 1.2$ rad: **a** $\psi = \pi/2$; **b** $\psi = 0$. A *white pixel* corresponds to a normalized spectral intensity of 0 and a *black pixel* to 1

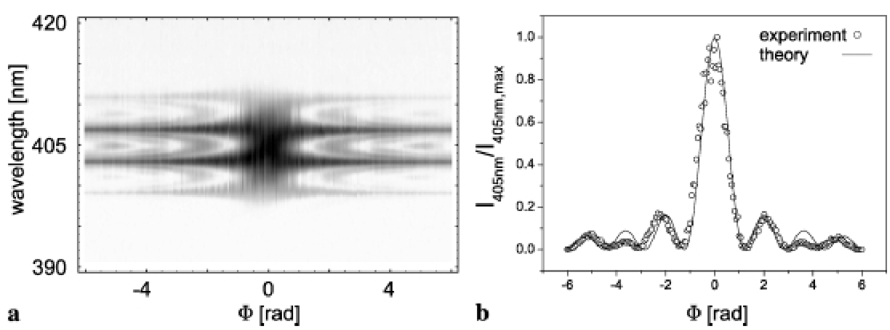

FIGURE 3 **a** Measured spectrum of frequency-doubled, sinusoidally phase-modulated fundamental pulses as a function of the modulation amplitude Φ for $\psi = 0$ and $\Delta t = 130$ fs. A *white pixel* corresponds to a normalized spectral intensity of 0 and a *black pixel* to 1, respectively. **b** Intersection through **a** at the central spectral component, i.e. 405 nm. *Circles*: experimental results; *solid line*: function $J_0\left(2\Phi \cos\left(\frac{\Delta t \cdot \omega_2}{2} + \psi\right)\right)^2$

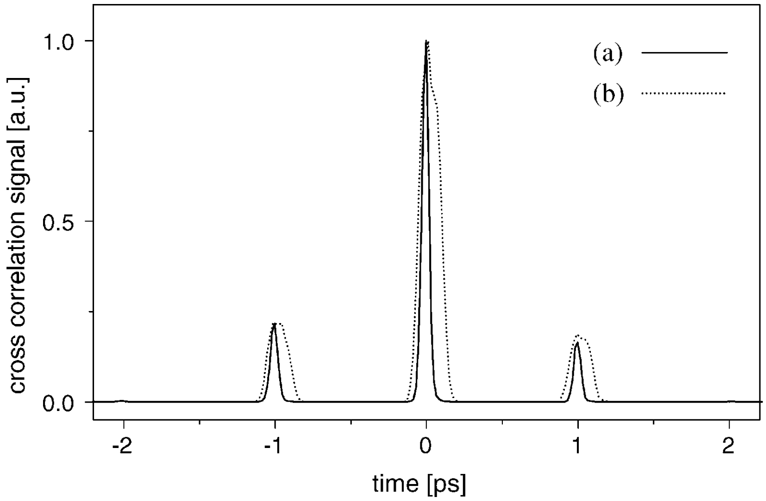

FIGURE 4 Measured intensity cross-correlation traces of the unmodified fundamental reference pulse with a sinusoidally phase-modulated fundamental pulse (a) and the corresponding frequency-doubled pulse (b). Note that *curve (a)* represents the square of the measured values. The phase-modulation amplitude was set to $\Phi = 1.2$ rad, the relative phase to $\psi = 0$ rad and the spectral modulation frequency to $\Delta t = 1000$ fs

The dependence shown in Fig. 3b is equivalent to that of the probability of a non-resonant two-photon transition in an atomic system excited by sinusoidally phase-modulated femtosecond laser pulses [5]. This problem was solved in [5] for the special case of a squared-uniform input spectrum. The results presented here therefore provide a generalization for the non-resonant two-photon transition. Both the center-frequency component and the two-photon transition probability are basically determined by a self-convolution of the input pulse spectrum. In principle, the authors of [5] take advantage of the possibility of creating constructive or destructive interference at the center-frequency component of a virtual SH spectrum to control the two-photon transition probability, depending on whether a symmetric (cosine) or antisymmetric (sine) phase modulation is applied to the input pulse at the fundamental frequency.

To show that (2), i.e. the description of shaped SH pulses in time, is correct, we temporally characterized both phase-modulated fundamental pulses and the corresponding frequency-doubled pulses by means of cross-correlations. Fig. 4 shows intensity cross-correlation traces of an unmodified fundamental reference pulse with a sinusoidally phase-modulated fundamental pulse (curve (a)) and the corresponding frequency-doubled pulse (curve (b)). To allow for a comparison of the two curves, the values of the cross-correlation of the shaped fundamental pulse are squared. The phase-modulation amplitude was again set to $\Phi = 1.2$ rad, to yield the maximum modulation contrast, and the spectral-modulation frequency to $\Delta t = 1000$ fs. The relative phase has no effect on these curves and was set to $\psi = 0$ rad.

In agreement with the applied spectral-modulation frequency, the resulting triple pulses show a temporal separation between the peaks of exactly 1000 fs.

Equation (2) implies that the temporal amplitudes within a shaped SH pulse are equal to the square of that within the incident shaped fundamental pulse. This is clearly illustrated by comparing curves a and b, considering that curve a shows the squared cross-correlation of the shaped fundamental pulse. The increased widths of the SH pulse replica can be assigned to a group-velocity mismatch of the fundamental and the SH in the 500-μm-thick BBO crystal used for SHG and its reduced spectral acceptance. In contrast to the 100-μm-thick crystal, which can easily accept the whole fundamental spectrum, the 500-μm-thick crystal can only accept about half of it. Nevertheless, it was employed to provide sufficient SH power for the cross-correlation measurements, and the general temporal shape of the modulated pulse is still in excellent agreement with the theoretical predictions.

5 Conclusion

We have shown that frequency doubling of phase-modulated fundamental pulses in nonlinear crystals of a moderate thickness is an appropriate way to achieve shaped SH pulses. While the temporal shape of a generated SH pulse may be approximated very well by the squared temporal shape of the incident fundamental pulse, the spectral amplitudes and phases of the resulting SH spectrum can show drastic alterations compared to the spectral amplitudes and phases of the incident fundamental pulse.

As an example, it was shown that sinusoidal phase modulations are transferred into amplitude modulations of the SH spectrum, with modulation depths depending on the phase-modulation amplitude. We have presented an analytical expression for the spectrum of frequency-doubled sinusoidal phase-modulated fundamental pulses which perfectly matches the experimental results. An important implication of this analytical expression is that the spectral resolution of the periodic amplitude modulation is determined only by the spectral resolution of the phase-only shaper.

By employing a common liquid-crystal spatial light modulator (LC SLM) with a transparency range down to 420 nm, in combination with a BBO crystal for SHG which supports phase matching down to 410 nm, the presented technique should be able to generate shaped ultraviolet pulses down to 210 nm.

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