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Coherent control and dark pulses in second harmonic generation

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Abstract

We present the results of our study of the second harmonic generation (SHG) in a nonlinear crystal using phase modulated femtosecond laser pulses. We show experimentally that the SHG process is an analogue of two photon absorption (TPA), yet it provides a better way to study coherent control because it enables one easier control of some important parameters. We demonstrate that for a thick nonlinear crystal pure phase manipulation can result in nontrivial pulses maximizing the second harmonic conversion efficiency and in dark pulses which are not converted at all. With a π step phase modulation applied to the pulse we have measured conversion efficiency for a set of crystals with different thicknesses to explore the effect of a finite spectral width of the TPA transition.

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Keywords: Second harmonic; Dark pulses; Coherent control

1. Introduction

The coherent control has been proposed for the first time two decades ago by Brumer and Shapiro [1] who suggested that two optical fields can be used to control the photodissociation of a molecule. The scheme they considered is an analogy of a two slit interference experiment in which the outcome depends on a phase between two pathways leading to the final state. In another theoretical analysis they showed that by employing a laser with frequency ω and its second harmonic at 2ω one can control the direction of the photoinduced current in a semiconductor [2]. At the beginning of the 1990's Zewail group took advantage of localized wave packed excitation to access two different electronic states of iodine molecule [3] in a femtosecond pulse-probe experiment. As a result of those experiments a new field of research called coherent control emerged. Usually in coherent control the prime objective is to move a quantum system from its initial state to a predetermined final state exerting as little effort (energy) as possible. Successful experimental realizations of coherent control of different processes including: ionization and dissociation of molecules [4,5], molecular fluorescence excitation [6,7] or current excitation in semiconductors [8,9] were reported and excellent review articles [10–12] are available.

As early as 1992 Broers et al. published the first paper on, what we would call today, quantum control of two-photon processes [13,14]. They did not use this term explicitly – instead they wrote about "energy focusing" to describe enhancement of a narrow-band SHG and TPA efficiency by binary spectral amplitude modulation and quadratic phase modulation. This effect bears very close resemblance to Fresnel diffraction as shown by the increase of SHG power in a given narrow band of frequencies for the amplitude mask equivalent to Fresnel zones.

One of the simplest yet very profound demonstrations of coherent control was the work by Meshulach and Silberberg [15]. They presented a theoretical analysis and performed an experiment on TPA in alkali atoms illuminated by phase modulated ultrashort laser pulses. The most striking effect of their work was, probably, the demonstration that pulses other than the transform limited ones can also maximize TPA. Later, it was also shown that,

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when an intermediate state between the ground and excited states is involved, phase modulated pulses can produce up to a 7 times higher TPA rate than the transform limited ones [16]. Two years after the initial experiment with coherent two photon absorption, Zheng and Weiner noticed that the equations governing TPA and the SHG in a nonlinear crystal have the same structure [17].

A series of experiments on various aspects of coherent control of multiphoton processes with phase modulated pules was conducted by Dantus and co-workers [18–24]. Among other things, they have developed a new method for ultrashort laser pulse characterization and improved excitation selectivity through binary phase shaping in two-photon microscopy. Motzkus and co-workers have reported an experiment on a nonresonant TPA in alkali atoms with phase shaped pulses. They have used evolutionary algorithm to coherently control the transition probability demonstrating both increase and decrease of the transition probability [25–28]. Yet another experiment on two photon transition in sodium atoms led to the development of a new, simple and very sensitive method for the calibration of the spatial light modulators [29].

In the SHG process involving a monochromatic light source two photons from the fundamental beam with a frequency of $\omega/2$ are converted into one photon in the second harmonic beam with a frequency of ω as a result of nonlinear interaction in a nonlinear crystal. When the fundamental beam is formed of short pulses with finite bandwidth the second harmonic at a given frequency ω can result from any photon pair whose frequencies add up to the desired frequency. Since, for coherent input pulses, the phases of all spectral components are well defined one has to consider a coherent superposition of all possible paths leading to generation of the second harmonic field. Because of this, coherent control of the SHG process is possible. Classically, an expression for the electric field of the second harmonic can be derived assuming wave optics and the nonlinear response of the medium to the oscillating electric field at the fundamental frequency. If the depletion of the fundamental beam is negligible the second harmonic field in the frequency domain is given by [30]:

$$\tilde{\epsilon}_2(\omega) = \int_{-\infty}^{\infty} \tilde{\epsilon}_1(\omega/2+\Omega)\tilde{\epsilon}_1(\omega/2-\Omega)\mathrm{d}\Omega \times D(\omega), \qquad (1)$$

where $\tilde{\epsilon}_1$ and $\tilde{\epsilon}_2$ are the spectral amplitudes of the fundamental and second harmonic waves respectively. Each of them can be decomposed into its real positive magnitude $A(\omega)$ and phase $\Phi(\omega)$: $\tilde{\epsilon}(\omega) = A(\omega)\exp[\mathrm{i}\Phi(\omega)]$. The last factor in Eq. (1) describes a phase matching condition in a nonlinear crystal: $D(\omega) = \Gamma L \mathrm{sinc}[\alpha(\omega-\omega_0)L/2]$, with L being the crystal length, Γ – the nonlinear coupling coefficient and $\alpha = 1/v_{g1} - 1/v_{g2}$ Group Velocity Mismatch (GVM) between the fundamental and doubled waves. We assume here that at the frequency $\omega_0/2$ a perfect phase matching is achieved in the crystal. $D(\omega)$ can be regarded as a frequency dependent filter which defines SHG efficiency for different frequencies [31].

It was pointed out by Meshulach [15] that the ability to coherently control TPA strongly depends on the spectral width of the two-photon transition. With narrow-band transitions characteristic of atomic gases it is possible to maximize TPA with an infinite number of nontrivial phase shapes whereas with broadband transitions often encountered in condensed media, e.g., a solution of the Coumarin 6G in methanol, a flat spectral phase corresponding to the shortest, i.e., a Fourier limited pulse is the only solution. The two extreme cases are easily quantified by providing the ratio of the spectral width of the laser pulse to the spectral width of the two-photon transition. In the first case this ratio is very large and in the second case small. One would expect that for the intermediate values of this parameter a continuous transition between the two regimes can be achieved. However, it is very difficult to explore the intermediate range using two photon absorption as this would require either a collection of pulsed lasers with widely different bandwidths or a collection of absorbing media with different widths of two-photon transitions. Neither of the two approaches is appealing. Still, one can use the formal correspondence between TPA and SHG [17] to explore coherent quantum control of two-photon absorption in the intermediate regime by studying second harmonic generation in a nonlinear crystal.

The basic idea of our approach relies on the observation that in the SHG process one can continuously tune between the two extreme regimes by taking advantage of the phase matching part $D(\omega)$ in Eq. (1). If one takes a thick nonlinear crystal with a significant GVM the phase matching part $D(\omega)$ responsible for frequency dependent conversion efficiency becomes very narrow and can be well approximated by a $\delta(\omega-\omega_0)$ function. It is convenient to introduce the interaction length given by $L_i = \tau/|\alpha|$, where τ is the duration of a transform limited fundamental pulse. Then, in the thick crystal regime with $L \gg L_i$ the SHG spectrum is limited by the spectral filtering due to the phase matching in a nonlinear crystal. When a thin ($L \ll L_i$) crystal is used a broadband generation of the second harmonic is possible and the spectral width of the output field is limited solely by that of the input.

For a thick nonlinear crystal $D(\omega) \approx \delta(\omega-\omega_0)$; the filter frequency ω_0 can be tuned, e.g., by changing crystal orientation. Then Eq. (1) gives the power of the quasi-monochromatic second harmonic:

$$P_{\mathrm{SH}}^{\mathrm{Thick}} \sim \left| \int_{-\infty}^{\infty} A(\omega_0/2+\Omega)A(\omega_0/2-\Omega) \times \exp[\mathrm{i}\{\Phi(\omega_0/2+\Omega)+\Phi(\omega_0/2-\Omega)\}]\mathrm{d}\Omega \right|^2. \qquad (2)$$

The effect of the spectral filtering is now evident: the second harmonic field at a frequency of ω_0 results from summing up all frequency pairs ω_i, ω_j in the fundamental beam fulfilling the energy conservation condition $\omega_0 = \omega_i + \omega_j$. The value of the integral depends on the spectral phase of the input pulse and thus one can achieve coherent control by the pure phase shaping of the input pulses. In particular,

as we demonstrate experimentally, there are spectral phase shapes for which the second harmonic at a given frequency is zero. It should be mentioned that, contrary to SHG or TPA, one photon transition probability is not phase dependent at all, it depends only on the spectral amplitude.

In the other limit of the SHG – a thin crystal regime – $D(\omega)$ can be approximated by a constant value over the whole input pulse spectrum – there is no filtering due to the phase matching in a crystal and all input frequencies are converted with the efficiency proportional to the product of the respective spectral amplitudes. This results in a spectrum of the second harmonic limited by the spectrum of the input pulse only [32]. Again, using Eq. (1) one can write the power of the second harmonic as:

$$P_{\mathrm{SH}}^{\mathrm{Thin}} \sim \int_{-\infty}^{\infty} \left| \int_{-\infty}^{\infty} \tilde{\epsilon}_1(\omega/2+\Omega)\tilde{\epsilon}_1(\omega/2-\Omega)\mathrm{d}\Omega \right|^2 \mathrm{d}\omega. \tag{3}$$

Eq. (3) can be cast into a more intuitive form if the convolution theorem is used:

$$P_{\mathrm{SH}}^{\mathrm{Thin}} \sim \int_{-\infty}^{\infty} I_1^2(t)\mathrm{d}t. \tag{4}$$

The result for a thin crystal regime is the same as the one for TPA in condensed matter (e.g., Rhodamine 6G). It is clear from Eq. (4) that in order to maximize either the second harmonic signal in a thin nonlinear crystal or the TPA in Rhodamine 6G one has to use Fourier-limited and thus the shortest possible pulses.

The filter function $D(\omega)$ opens a convenient way of studying coherent control of TPA processes (utilizing the analogy between TPA and SHG) as it provides an easy method to change the relative spectral widths of the laser pulse and the two-photon transition. We use this approach to demonstrate experimentally the effect of finite two-photon transition bandwidth on the effectiveness of coherent control in the TPA process. Instrumentally, we change the spectral width of the TPA line by either changing the thickness of the SH crystal or spectral filtering of the SH signal.

2. Experiment and results

The experimental set-up for the second harmonic generation with phase modulated pulses is shown in Fig. 1. A beam of pulses from a home-built Ti:Sapphire femtosecond oscillator was passed through a standard 4f pulse shaper [33] equipped with a Liquid Crystal Spatial Light Modulator with 640 independent sectors (JenOptik SLM-S640d).

The phase-voltage dependence of our SLM was obtained with a standard procedure: the light modulator was placed between two crossed polaroids and illuminated with a semiconductor laser with a known wavelength. Transmission versus voltage was measured and used to retrieve the phase-voltage calibration. Calibration for other wavelengths was then calculated using Sellmeier formula for liquid crystals. The results of this procedure are in excellent agreement (within 2%) with the data provided by the manufacturer.

The spectral window of the pulse shaper was 124 nm resulting in the spectral resolution of 0.2 nm/pixel. The shaped pulses were passed through a nonlinear crystal and the second harmonic signal was registered with an either a photodiode or a spectrometer. This way both the total SH power and the SH spectrum could be recorded.

Prior to any measurements involving phase modulation the pulses at the output of the shaper were compressed to remove the residual chirp from the laser as well as the phase distortions introduced by the shaper itself. This was achieved by running a standard evolutionary algorithm [34] with the feedback signal generated by a two-photon photodiode placed in the position of the second harmonic crystal. The evolutionary algorithm increased the feedback signal approximately 3 times, however, it did not deliver a correct spectral phase in the full spectral range (see Fig. 1b). As might be expected, for frequencies for which the spectral amplitude is small phase modulation had little effect on the feedback signal and its result was hidden in the experimental noise. Because of that the resultant phase acquired random values in the spectral wings.

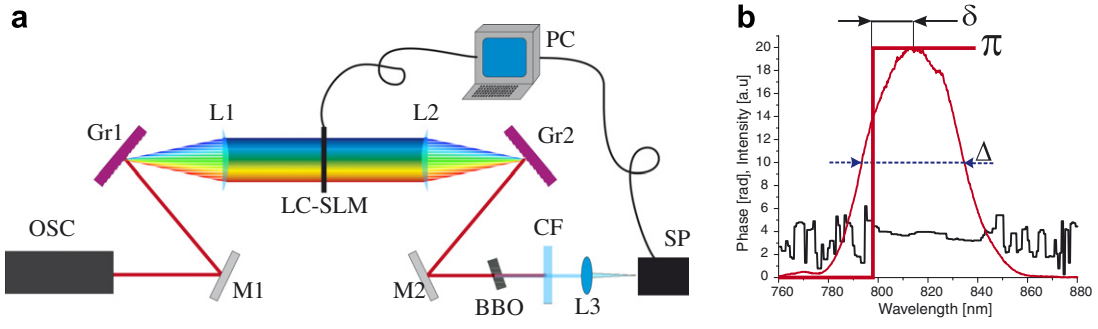

Fig. 1. (a) Scheme of the experimental set-up for SHG generation with phase modulated pulses. OSC – Ti:Sapphire femtosecond oscillator, M1, M2 – flat mirrors, Gr1, Gr2 – diffraction gratings 1200 lines/mm, L1, L2 – cylindrical lenses $f = 30$ cm, LC-SLM – spatial light modulator, SP – spectrometer (Ocean Optics USB2000), CF – blue color glass filter, BBO – β-barium borate nonlinear crystal (thickness ranging form 20 μm to 2 mm). (b) Measured laser spectrum with FWHM width of Δ and the compensating phase (see text). The step function shown in the figure indicates the position of the π step phase modulation imposed on the pulses by the pulse shaping system. The position of the step function is offset by δ from the center of the laser spectrum.

Such phase "background" was added to a desired phase shape function in all the subsequent measurements.

In order to compare our results with those for the TPA process in atoms [35] we applied the same phase modulation to the laser pulses – a single π step with the position of the step being the only control parameter. The SH intensity was recorded versus the position of the phase step.

The results of our measurements – SH signal versus the π step position – for BBO crystals with four different thicknesses are shown in Fig. 2. Not surprisingly, our experimental results very closely resemble the available data for TPA in atoms. For a thick crystal (Fig. 2a), the second harmonic signal is high for the π step positions which are far from the central frequency. In this case, the phase modulation affects just the spectral wings of the pulse and the pulse can still be well approximated by a transform limited one. When the π step position approaches $\omega_0/2$ the SH signal drops to almost zero at $\delta/\Delta = \pm 0.31$. For δ/Δ close to zero the SH signal revives rapidly reaching the maximum at $\delta/\Delta = 0$. The results shown in Fig. 2a can be explained if we recall Eq. (2). The phase part in the integral reads as: $\phi(\Omega) = \Phi(\omega_0/2 + \Omega) + \Phi(\omega_0/2 - \Omega)$. The first obvious solution maximizing SHG is $\phi(\Omega) = 0$ which is a well known case of transform limited pulses. However, there are other nontrivial solutions: any phase antisymmetric around $\omega_0/2$, i.e., fulfilling the condition: $\Phi(\omega_0/2 + \Omega) = -\Phi(\omega_0/2 - \Omega)$ should also maximize the second harmonic signal, giving the same power as transform limited pulses. It is worth noting that the π step is just one example of the phase modulation which maximizes the SH power. A pulse with any antisymmetric phase will give the maximum SH efficiency. In the case of a thin crystal ($D(\omega) \approx$ const) the SH power depends on the temporal pulse profile only (see Eq. (4)). It is maximized for the shortest – Fourier limited pulses and any spectral phase other than a flat one leads to pulse lengthening and a decrease in the SHG efficiency. This has been verified experimentally with a 20 μm BBO crystal and laser pulses with the π step phase modulation (Fig. 2d). According to the dispersion data for the BBO material [36] the ratio L_i/L is ≈6 in the case of 20 μm BBO crystal and therefore the limit of a very broadband TPA should be well modeled. The results obtained for the SHG in a thin crystal are very similar to those for the TPA with a broad transition line. In this case there should be no revival of the SH signal when the phase step is centered at the maximum of the laser spectrum. The remnants of a revival observed in the experimental data show that even with a very broad line of the TPA transition – in this case it is 6 times broader than the laser spectrum – some coherent control is still possible.

Using the second harmonic generation in a nonlinear crystal instead of two-photon absorption enables one to study phase modulation effects in different regimes. In particular, one can vary the ratio of the filter function width to

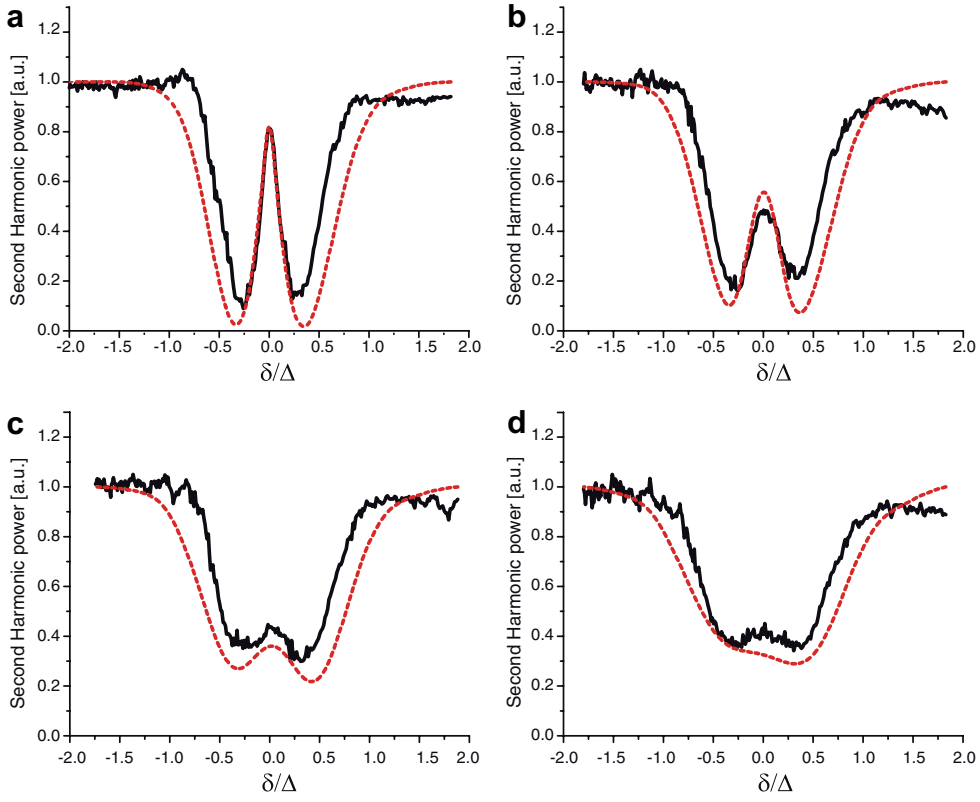

Fig. 2. Second harmonic power as a function of the phase π step position for different BBO crystal lengths. (a) 2 mm, (b) 0.5 mm, (c) 0.2 mm, (d) 0.02 mm. Solid line – experiment, dashed line – simulation. The ratio L_i/L is 0.061, 0.24, 0.61, 6.1 for (a), (b), (c) and (d), respectively.

the width of the pulse spectrum. This is illustrated in Fig. 2 where the experimental data for four crystals with thicknesses from 20 μm to 2 mm are presented together with the results of the numerical modeling. The numerically computed curves were obtained as follows. In order to calculate efficiently the integral in Eq. (1) we use the convolution theorem: we take Fourier transform of the fundamental field, square it and back-transform it to the frequency domain to obtain the SH field:

$$\tilde{\epsilon}_2(\omega) = \mathscr{F}\left(\left[\mathscr{F}^{-1}\tilde{\epsilon}_1(\omega/2)\right]^2\right) \cdot D(\omega). \tag{5}$$

The filter function $D(\omega)$ was a sin$c$ function scaled properly to account for a given crystal thickness. This procedure was repeated for a range of the phase step positions δ used in the experiment. The overall agreement between the experiment and numerical modeling is very good. All the basic features present in the experimental data are reproduced in the modeling. The main discrepancy is the width of the double well structures – the experimental ones are narrower than their calculated counterparts. There are several possible reasons for this discrepancy. Firstly, one should consider possible errors due to imperfect phase-voltage calibration of the SLM. As described in the experimental section, this calibration is correct within a few percent. Anyway, the possible error in this calibration would only decrease the modulation depth of the curves shown in Figs. 2 and 3 without changing their widths. Secondly, it is well known that liquid crystal SLM can change the pulse spectrum significantly when sharp phase modulation is applied [37] due to diffraction effects. However, in our experimental set-up, the spectral hole created by a single sharp phase jump has a width of just 0.4 nm so its influence can not explain visible discrepancies. Last but not least, the spectral phase of the input pulse is not perfectly flat. This is due to the finite accuracy of the evolutionary algorithm used to flatten the phase of the pulses prior to applying the step function. As already discussed, the algorithm produced random phases in the spectral wings of the pulse and any additional phase modulation in those regions should have no effect on the SHG efficiency. We have verified this by running our numerical code that simulates SHG process and assuming random phase in the spectral wings. The results of the modeling show that the SHG spectrum is narrower than the one obtained with a flat spectral phase. Thus such a phase randomization is equivalent to the spectral narrowing as observed in our experimental data.

In principle, one could apply another method to flatten the spectral phase of the input pulse. The pulses after 4f setup could be characterized with either FROG or SPIDER. The retrieved spectral phase could then be applied (with opposite sign) to a well calibrated SLM to get transform limited pulses at the shaper output. This has not been done in our experiment as a full input pulse characterization would result in a complex experimental set-up.

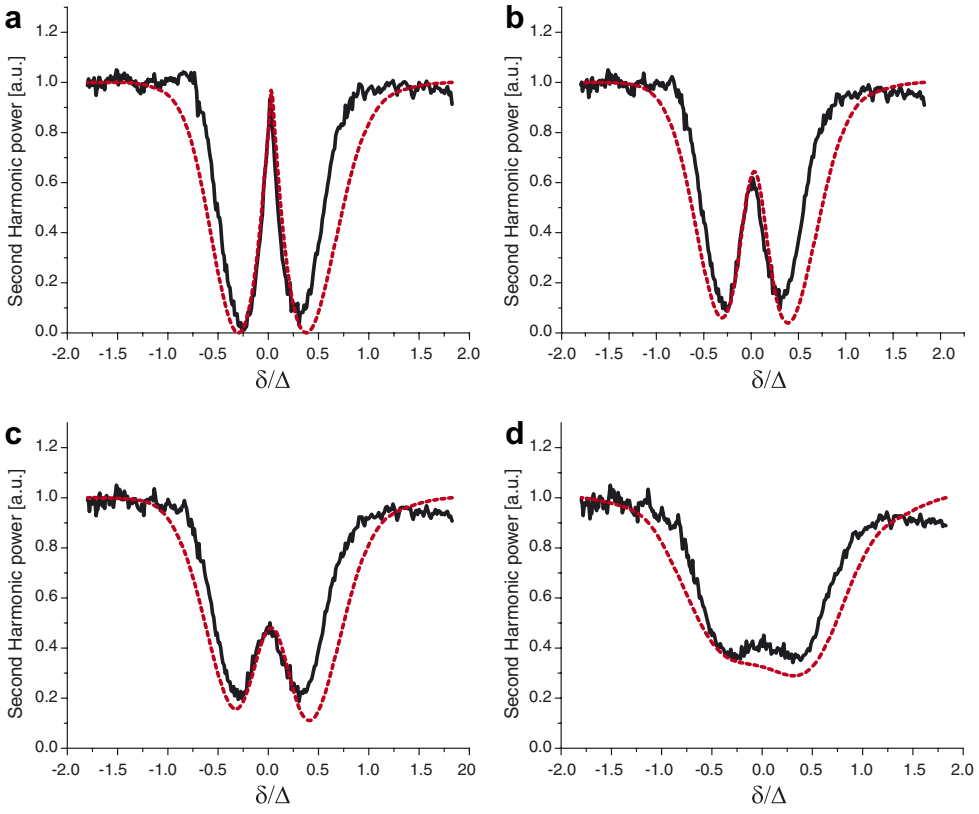

Fig. 3. Second harmonic power as a function of the phase π step position for 20 μm BBO crystal and different widths of the gaussian spectral filter $D(\omega)$. The FWHM width of the filter is: (a) 0.2 nm, (b) 3 nm, (c) 6 nm, (d) infinite. Solid line – experiment, dashed line – simulation.

The data shows that the transition from a thick crystal enabling coherent control of the SHG process to a thin one where there are no nontrivial pulses maximizing the efficiency is smooth. It should be noted that no such data for TPA exists while with the method presented here it is rather straightforward to obtain it.

It should also be pointed out that the nature of the spectral filter $D(\omega)$ involved in the second harmonic generation is not important. The results shown in Fig. 2 can also be obtained with other methods. For example, one can use a very thin crystal followed by a spectrometer. We applied this approach – with a 20 μm BBO crystal we registered the spectra of SH for different positions of the π phase step. Then each spectrum has been numerically filtered with a gaussian filter and integrated to provide a number which is proportional to the signal given by Eq. (1). The results are shown in Fig. 3 together with the results of numerical modeling. With this procedure one can easily change the width of the spectral filter by a simple manipulation of a computer code analyzing the experimental data. Moreover, a single set of experimental data (a set of SH spectra) is sufficient to retrieve the results for different values of the filter width. The results obtained using this method are consistent with the data shown in Fig. 2.

It is instructive to consider the thick crystal case and look at the temporal profiles of three pulses with different positions of the π step – a transform limited one, a pulse with a phase which is antisymmetric with respect to $\omega_0/2$, and a pulse with the π step at $\delta = \pm 0.31\Delta$. The last one is not converted into the second harmonic and we call it *a dark pulse* by analogy to dark states in atoms [38]. To gain a better insight into the nature of a dark pulse we numerically modeled the temporal profiles of the three pulses. We assumed a gaussian pulse spectrum with FWHM equal to the spectral width of our laser and a π step phase. The results are shown in Fig. 4. Even though the peak intensity of the pulse with anti-symmetric phase is more than 2 times smaller than the intensity of the transform limited pulse they produce the same SH power. At the same time the temporal profile of a dark pulse is quite similar to that of the pulse with antisymmetric phase but they are converted to SH with drastically different efficiency. One can consider this case as an illustration of the power of coherent control.

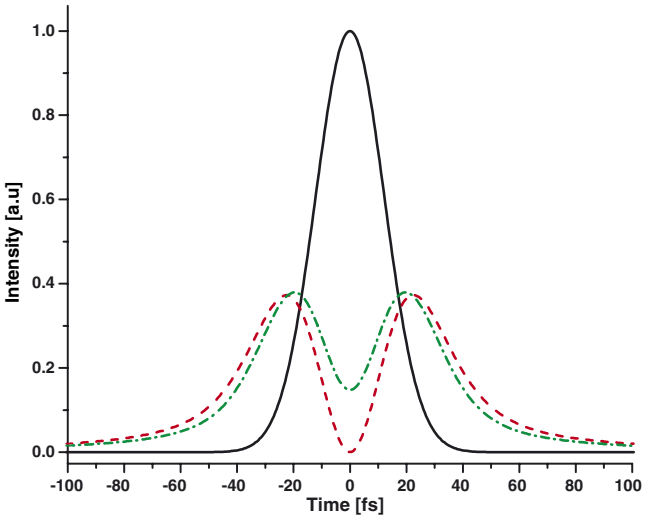

Fig. 4. Temporal pulse profiles: solid line – a transform limited pulse, dashed – a pulse with antisymmetric phase, dash–dot – a dark pulse.

3. Conclusions

In summary, we have experimentally demonstrated some effects of the phase modulation in the second harmonic generation process. We have built an analogy between the second harmonic generation and the well studied two-photon absorption processes and emphasized one-to-one correspondence between the two. We have demonstrated an experimental method that makes studies of coherent control in TPA using a well established and relatively simple method of SHG in nonlinear crystals possible. It enables one to explore the continuous transition between different regimes of either TPA or SHG. For a thick crystal regime we have shown that a pure phase manipulation of the laser pulse can lead to a dramatically different efficiency of the SHG process. In particular, we have shown the existence of dark pulses that are not converted to the second harmonic even though their temporal profiles are quite similar to the temporal profiles of the pulses that are converted with the maximum achievable efficiency. Good agreement with analytical predictions and previous experimental result on coherent control in TPA in atoms was achieved.

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