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Ultrafast adiabatic second harmonic generation

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Abstract

We introduce a generalization of the adiabatic frequency conversion method for an efficient conversion of ultrashort pulses in the full nonlinear regime. Our analysis takes into account dispersion as well as two-photon processes and Kerr effect, allowing complete analysis of any three waves with arbitrary phase mismatched design and any nonlinear optical process. We use this analysis to design an efficient and robust second harmonic generation, the most widely used nonlinear process for both fundamental and applied research. We experimentally show that such design not only allows for very efficient conversion of various of ultrashort pulses, but is also very robust to variations in the parameters of both the nonlinear crystal and the incoming light. These include variation of more than 100 °C in the crystal temperature, a wide bandwidth of up to 75 nm and a chirp variation of 300 fs to 3.5 ps of the incoming pulse. Also, we show the dependency of the adiabatic second harmonic generation design on the pump intensity and the crystal length. Our study shows that two photon absorption plays a critical role in such high influence nonlinear dynamics, and that it must be considered in order to achieve agreement with experimental results.

Keywords: nonlinear optics, ultrafast phenomena, adiabatic processes, frequency conversion

(Some figures may appear in colour only in the online journal)

1. Introduction

Second harmonic generation (SHG) is the most basic nonlinear optical process, yet the most powerful and widely used. Since the first demonstration of the SHG process right after introducing the first laser [1], it revolutionized the field of light–matter interactions, allowing a new way for exploring atomic, molecular and condensed matter systems. Due to its simplicity, requiring solely shining a single laser beam into a nonlinear crystal, it gained popularity among diverse applications from nonlinear spectroscopy, metallurgy, photoinduced dynamics in 2D and condensed matter dynamics, noninvasive background free diagnostics, and the generation of new color sources [2–5]. It is, however, generally difficult to obtain efficient and robust conversion from a pump pulse to its harmonics for a broad range of colors. This is mainly due to the lack of phase mismatch (lack of momentum conservation) between the interacting waves, which conventionally can be compensated only for a narrow band of frequencies. This is mainly due to the lack of phase mismatch (lack of momentum conservation) between the interacting waves, which conventionally can be compensated only for a narrow band of frequencies. In the fully nonlinear regime, where none of the interacting waves can be considered to be undepleted, efficient broadband conversion is even more difficult due to its complex dynamics. Although, in the past, several methods were suggested to deal with the conversion of a broadband source, such as short birefringent crystals [6], multi-periodic modulation [7], chirp patterns [8, 9], temperature gradient manipulations [10, 11]
and random oriented crystal [12]. Those indeed achieved very broadband conversion, but at the expense of limited conversion efficiencies.

In recent years, a new direction in frequency conversion has emerged—adiabatic frequency conversion—a method that is based on adiabatic evolution of the nonlinear optics dynamics. The suggested method enables to overcome the tradeoff between conversion efficiency and bandwidth [13, 14]. Although it was first considered theoretically for SHG by Baranova [15], the initial extensive experimental research was performed in sum/difference frequency conversion (SFG/DFG) in the undepleted pump approximation, offering the use of SU(2) dynamical symmetry with the analogous mechanism of Landau–Zener transition [16].

In the past few years, the fully nonlinear regime received special attention. Research on adiabatic interactions with nonlinear dynamics as adiabatic OPA and OPO was conducted by Phillips et al. [17, 18], Heese et al. [19–21] and Yaacobi [22]. In parallel, adiabatic DFG allowed an efficient conversion of near-IR few cycle pulses to octave-spanning mid-IR pulses [23, 24]. A general physical model of adiabatic frequency conversion in the fully nonlinear dynamics regime, was presented recently by Porat and Arie [25]. In the past year, a first proof of adiabatic SHG was demonstrated with nanosecond pulses [26], exhibiting an unmatched temperature robustness. Yet, the efficient SHG conversion of ultrashort pulses is still lacking. Here, the adiabatic conversion method is generalized to include efficient SHG conversion of ultrashort pulses. In our analysis, we do not only solve the full nonlinear dynamics with arbitrary interacting ultrashort pulses and nonlinear phase-mismatched design, but also take into account Kerr effect and two photon absorption of the generated SHG. This will be shown to be a critical aspect in our experimental observations that was lacking in all previous research of adiabatic frequency conversion.

2. Theoretical background

Let us start by developing the fully nonlinear dynamical equations that dictate nonlinear conversion of any three wave mixing, and in particular of SHG. We will present the full nonlinear equations in the time domain, including two photon absorption (TPA) and Kerr effect, and will discuss the adiabaticity criteria for an efficient frequency conversion process.

2.1. Nonlinear dynamics in three wave mixing

The electric field wave equation, in the presence of nonlinear polarization is given by

\[
\nabla^2 \tilde{E}(\vec{r}, t) - \mu \varepsilon_0 \gamma \frac{\partial^2 \tilde{E}(\vec{r}, t)}{\partial t^2} = \mu \frac{\partial^2 \tilde{P}_{NL}(\vec{r}, t)}{\partial t^2} \tag{1}
\]

where \( \mu \) is the material permeability, \( \varepsilon_0 \) is the vacuum permittivity, \( n \) is the material refractive index and \( \tilde{P}_{NL}(\vec{r}, t) \) is the nonlinear polarization [27]. In our analysis we assume that the electric field is planar. One can therefore write:

\[
\tilde{E}(\vec{r}, t) = \frac{1}{2\pi} \int_0^{\infty} \frac{1}{2} (\tilde{A}(\omega, \omega) e^{i(\omega_1 t - \beta \omega_1 t)} + \text{c.c.}) d\omega \tag{2}
\]

where \( \tilde{A}(\omega, \omega) \) is the field amplitude spectral density and \( \beta(\omega) = \frac{\omega(\omega)}{c} \) is the material wave vector. By changing the integral limits one can deduce the electric field:

\[
E(\zeta, \omega) = (\tilde{A}(\omega, \omega) + \tilde{A}^*(\omega, -\omega)) e^{-i\beta(\omega) \zeta}. \tag{3}
\]

By applying Fourier transform to equation (1), substitute \( \tilde{E}(\zeta, \omega) \), and assuming the slowly varying amplitude approximation \( |\frac{\partial^2 \tilde{A}(\omega)}{\partial \omega^2}| \ll |\beta(\omega)\tilde{A}(\omega, \omega)| \), equation (1) takes the form:

\[
\left( \frac{\partial \tilde{A}(\omega, \omega)}{\partial \zeta} + \frac{\partial \tilde{A}^*(\omega, -\omega)}{\partial \zeta} \right) e^{i\beta(\omega) \zeta} = -\mu \omega^2 \tilde{P}_{NL}(\omega, \omega). \tag{4}
\]

Without loss of generality, we will assume that the electric field is polarized along the \( x \) direction. By applying Fourier transform to the nonlinear polarization term \( \tilde{P}_{NL}(\omega, \omega) = 2c_0 \chi(\omega) \tilde{E}^2(\zeta, t) \tilde{E}^* \) and substituting it into equation (4), the master equation for second order nonlinear interactions is deduced:

\[
\left( \frac{\partial \tilde{A}(\omega, \omega)}{\partial \zeta} + \frac{\partial \tilde{A}^*(\omega, -\omega)}{\partial \zeta} \right) e^{i\beta(\omega) \zeta} = -2i\frac{\omega \chi(\omega)}{cm(\omega)} E^*(\omega, \omega) * E(\omega, \omega) \tag{5}
\]

where ‘*’ stands for convolution. We continue the derivation by expanding the electric field in terms of the interacting pulses: \( E(\zeta, \omega) = E_1(\zeta, \omega) + E_2(\zeta, \omega) + E_3(\zeta, \omega) \), where \( E_j(\zeta, \omega) = (A_j(\zeta, \omega) + A_j^*(\zeta, -\omega)) e^{-i\beta(\omega) \zeta} \) for \( j = 1, 2, 3 \). \( A_1(\omega, \omega), A_2(\omega, \omega) \) and \( A_3(\omega, \omega) \) are centered around frequencies \( \omega_1, \omega_2 \) and \( \omega_3 \) respectively, obeying \( \omega_1 + \omega_2 = \omega_3 \). Inserting the electric fields into equation (4) and observing the spectral shape of the convoluted signals, one can deduce the following set of equations:

\[
\begin{align*}
\frac{\partial B_1(\omega, \omega)}{\partial \zeta} + i\beta(\omega) B_1(\omega, \omega) &= -i\frac{\omega \chi(\omega)}{cm(\omega)} B_3(\omega, -\omega) * B_3^*(\omega, -\omega) \\
\frac{\partial B_2(\omega, \omega)}{\partial \zeta} + i\beta(\omega) B_2(\omega, \omega) &= -i\frac{\omega \chi(\omega)}{cm(\omega)} B_3(\omega, -\omega) * B_1^*(\omega, -\omega) \\
\frac{\partial B_3(\omega, \omega)}{\partial \zeta} + i\beta(\omega) B_3(\omega, \omega) &= -i\frac{\omega \chi(\omega)}{cm(\omega)} B_1(\omega, \omega) * B_2(\omega, \omega)
\end{align*} \tag{5}
\]

where \( B_{1,2,3}(\omega, \omega) = A_{1,2,3}(\omega, \omega) e^{-i\beta(\omega) \zeta} \). In the continuous wave (CW) case, the electric field is monochromatic: \( B_j(\omega, \omega) = B_j(\delta(\omega - \omega)) \) where \( j = 1, 2, 3 \). Substitution of the monochromatic amplitudes into equations (5) yields the conventional set of equations (in terms of \( A_j \)) as presented in [27]:

\[
\begin{align*}
\frac{\partial A_1}{\partial \zeta} &= -i\gamma A_1 A_2 e^{-i\Delta(\omega)} \\
\frac{\partial A_2}{\partial \zeta} &= -i\gamma A_1 A_2 e^{-i\Delta(\omega)} \\
\frac{\partial A_3}{\partial \zeta} &= -i\gamma A_1 A_2 e^{i\Delta(\omega)} \tag{6}
\end{align*}
\]
where $\gamma = \frac{\omega \lambda \theta}{c \omega_0}$ is the coupling coefficient and $\Delta \beta = (\beta(\omega_1) - \beta(\omega_2) - \beta(\omega))$ is the phase mismatch between the interacting waves. Using equations (6) it is possible to design the nonlinear second susceptibility $\chi(2)$ in order to compensate the inherent phase mismatch $\Delta \beta$. The obtained design will work fine even in the case of quasi-monochromatic waves, having their bandwidth much smaller than their central frequency, thus having an inherent phase mismatch $\Delta \beta$ which is nearly the same as in the CW regime.

It is possible to use equations (6) also for ultrashort pulses whenever group-velocity mismatch (GVM) between the interacting waves can be neglected. For example, in the case of 10 mm SHG potassium titanyl phosphate (KTP) crystal designed to convert 1030 nm pump pulse into 515 nm SHG pulse, the maximum GVM along the crystal can be at most $2\tau_{GVM} = L(v_g^{-1}(1/\lambda_{SHG}) - v_g^{-1}(1/\lambda_{pump})) = 6.8$ ps. It is therefore not possible to use the conventional equations (6) to calculate the dynamics in the picosecond regime.

For the ultrashort pulse case, we shall continue by describing every pulse centered around central frequency $\omega_1$ as a multiplication of slowly varying time domain amplitude function modulated with the central pulse frequency: $B_z(z, \omega) = \mathcal{F}(B(z, t)e^{i\omega_1 t})$. We will use the following identities:

$$
\mathcal{F}^{-1}(B(z, \omega - \omega_1)) \equiv e^{i\omega_1 t}B_1(z, t)
$$

$$
\mathcal{F}^{-1}(B(z, \omega - \omega_1) + B(z, \omega - \omega_1))_\omega + i\omega_1 = e^{i\omega_1 t}B_1(z, t)B_2(z, t)
$$

Equations (7) take into account nonlinear processes that are proportional to the real and imaginary parts of the third order nonlinearity $\chi(3)$, namely Kerr effect and TPA. When the optical material experiences TPA, an intensity density dependent term is added to the linear absorption, constituting an absorption coefficient $\alpha = \alpha_0 + \beta I$, where $\beta(mW^{-1})$ is the two photon absorption coefficient [27]. KTP absorbs in the range $\lambda < 350$ nm, hence, two photon absorption may limit the performance of the SHG crystal if the generated SHG wavelength is smaller than 700 nm. The second effect we take into account is the Kerr effect, which is the physical process constituting an intensity dependent refractive index term [27] $n = n_0 + n_2 I$, where $n_0$ is the weak-field refractive index and $n_2(m^2 W^{-1})$ is the second-order index of refraction.

The incorporation of both TPA and Kerr effect to the simulation is done in the time domain:

$$
\frac{\partial B(z, t)}{\partial \tau} + i\mathcal{F}^{-1}(\beta(\omega_1 + \omega)B_1(z, t))
$$

$$
= -i\chi(z)\mathcal{F}^{-1}\left(\frac{\omega + \omega_1}{c m(\omega + \omega_1)}\mathcal{F}(B(z, t)B_2(z, t))\right)
$$

$$
\frac{\partial B_2(z, t)}{\partial \tau} + i\mathcal{F}^{-1}(\beta(\omega + \omega_2)B_2(z, t))
$$

$$
= -i\chi(z)\mathcal{F}^{-1}\left(\frac{\omega + \omega_2}{c m(\omega + \omega_2)}\mathcal{F}(B(z, t)B(z, t))\right)
$$

$$
\frac{\partial B_3(z, t)}{\partial \tau} + i\mathcal{F}^{-1}(\beta(\omega + \omega_3)B_3(z, t))
$$

$$
= -i\chi(z)\mathcal{F}^{-1}\left(\frac{\omega + \omega_3}{c m(\omega + \omega_3)}\mathcal{F}(B_3(z, t)B_2(z, t))\right)
$$

The left-hand side term $\beta(\omega_1 + \omega)B_1(z, \omega)$ accounts for the dispersion the pulses experience through propagation along the crystal, while the right-hand side accounts for the nonlinear interaction between the pulses. In the case of narrowband pulses, where the pulse bandwidth is much smaller than their central frequency, $(\omega + \omega_1) \approx \omega_1$ and equations (8) takes the form:

$$
\frac{\partial B(z, t)}{\partial \tau} + i\mathcal{F}^{-1}(\beta(\omega_1 + \omega)B_1(z, t))
$$

$$
= -i\chi(z)\mathcal{F}^{-1}\left(\frac{\omega + \omega_1}{c m(\omega + \omega_1)}\mathcal{F}(B(z, t)B_2(z, t))\right)
$$

$$
\frac{\partial B_2(z, t)}{\partial \tau} + i\mathcal{F}^{-1}(\beta(\omega_1 + \omega)B_2(z, t))
$$

$$
= -i\chi(z)\mathcal{F}^{-1}\left(\frac{\omega + \omega_1}{c m(\omega + \omega_1)}\mathcal{F}(B(z, t)B(z, t))\right)
$$

$$
\frac{\partial B_3(z, t)}{\partial \tau} + i\mathcal{F}^{-1}(\beta(\omega_1 + \omega)B_3(z, t))
$$

$$
= -i\chi(z)\mathcal{F}^{-1}\left(\frac{\omega + \omega_1}{c m(\omega + \omega_1)}\mathcal{F}(B_3(z, t)B_2(z, t))\right)
$$

The equations for the SHG process can be easily deduced from equations (8). The pump pulse, centered around $\omega_1 = \omega_2 = \omega_p$ interacts with itself and generates the SHG pulse around $\omega_{SHG} = \omega = 2\omega_p$.

$$
\frac{\partial B_{SHG}(z, t)}{\partial \tau} + i\mathcal{F}^{-1}(\beta(\omega + \omega_{SHG})B(z, t))
$$

$$
= -i\chi(z)\mathcal{F}^{-1}\left(\frac{\omega + \omega_{SHG}}{c m(\omega + \omega_{SHG})}\mathcal{F}(B_3(z, t))\right)
$$

$$
\frac{\partial B_p(z, t)}{\partial \tau} + i\mathcal{F}^{-1}(\beta(\omega + \omega_p)B_p(z, t))
$$

$$
= -i\chi(z)\mathcal{F}^{-1}\left(\frac{\omega + \omega_p}{c m(\omega + \omega_p)}\mathcal{F}(B_{SHG}(z, t)B_p(z, t))\right)
$$

In our analysis we also take into account nonlinear processes that are proportional to the real and imaginary parts of the third order nonlinearity $\chi(3)$, namely Kerr effect and TPA. When the optical material experiences TPA, an intensity density dependent term is added to the linear absorption, constituting an absorption coefficient $\alpha = \alpha_0 + \beta I$, where $\beta(mW^{-1})$ is the two photon absorption coefficient [27]. KTP absorbs in the range $\lambda < 350$ nm, hence, two photon absorption may limit the performance of the SHG crystal if the generated SHG wavelength is smaller than 700 nm. The second effect we take into account is the Kerr effect, which is the physical process constituting an intensity dependent refractive index term [27] $n = n_0 + n_2 I$, where $n_0$ is the weak-field refractive index and $n_2(m^2 W^{-1})$ is the second-order index of refraction.
where \( I_p(z, t) = \frac{|B_p(z, t)|^2}{\eta_0} \) and \( I_{\text{SHG}}(z, t) = \frac{|B_{\text{SHG}}(z, t)|^2}{\eta_0} \).

### 2.2. Adiabaticity condition in the nonlinear regime

In an efficient second harmonic generation process, most of the pump energy is transferred to the SHG pulse energy. The pump is therefore by definition depleted. The comprehensive theory for an adiabatic frequency conversion process, involving three wave mixing (TWM) was developed by Porat and Adie [25]. The adiabacity criteria for the general case, where the interacting waves may be depleted, was developed in the CW regime. Start with defining the relative strength of the phase mismatch compared to the nonlinearity

\[
\Delta\Gamma = \Delta\beta\sqrt{\gamma_1 \gamma_2},
\]

and continue with defining the scaled propagation length

\[
\xi = z\sqrt{\gamma_1 \gamma_2}.
\]

The adiabaticity criteria takes the form:

\[
\left| \frac{d(P_3^2)/d\Gamma}{d\Delta\Gamma/d\xi} \right| \ll \nu
\]

(12)

where \((P_1^i, Q_1^i)\) are the canonical coordinates of the two stationary states of the system, \(P_1\) is the photon flux excess of the two low-frequency waves over the high-frequency wave, \(P_3\) is the total photon flux and \(\nu = \sqrt{\frac{\partial^2 H}{\partial Q_1^i \partial Q_1^j} \frac{\partial^2 H}{\partial P_1^i \partial P_1^j}}\) is the frequency in which the system orbits its fixed point in the phase space. The adiabacity condition in equation (12) is suitable for the CW regime or to a narrow-band pulses, and cannot be used when analyzing ultrashort pulses with bandwidth not negligible compared to their central frequency. Adiabatic evolution theory for ultra-short pulses is still absent, and numerical simulations are therefore required.

### 3. Numerical approach and predictions

The numerical simulations of equations (10) were performed by implementing the split-step Fourier method for the linear part of the equations, while the numerical integration for the nonlinear part is implemented using the 4th order Runge–Kutta method. The simulation procedure for every integration step \(\Delta z\) starts with accounting to the linear term. The pulses are first propagated:

\[
L_{\text{SHG}p}(\zeta, \omega) = \beta(\omega + \omega_{\text{SHG}p}) - \frac{(\omega + \omega_{\text{SHG}p})}{\nu(\omega_p)}.
\]

(13)

All pulses are simulated relative to a time reference that moves together with the pump pulse, hence, a time domain translation term \(\frac{(\omega + \omega_{\text{SHG}p})}{\nu(\omega_p)}\) is added to the dispersion.

The second step is to account for the nonlinear term, which is numerically integrated in the time domain. Using the amplitudes \(C_{\text{SHG}p}(\zeta, \omega), B_{\text{SHG}p}(\zeta + \Delta z, \omega)\) are calculated. The numerical procedure is iteratively repeated until the numerical integration is done all over the crystal optical axis. An example of the simulated amplitudes in the time domain is shown in figure 1.
We would like to numerically examine the robustness of the designed SHG crystal without taking into account undesired parasitic effects as TPA. The crystal grating function $K_g(z)$ is $K_g(z) = \frac{\chi(z)}{\chi(0)}$ where the second order nonlinear susceptibility is given by $\chi(z) = \chi_{\text{cg}}(\text{cos}(K_g z))$, thus fluctuating between $+\chi$ to $-\chi$ in batches spanning between 3.5 $\mu$m to 4.5 $\mu$m. The crystal dimensions are depicted in figure 3. The crystal grating period $\Lambda = \frac{2\pi}{K_g(c)}$ adiabatically changes from $\Lambda = 7.4$ $\mu$m to $\Lambda = 8.4$ $\mu$m along the optical axis of the nonlinear crystal.

We define the energy conversion efficiency of the crystal to be the ratio between the generated SHG pulse energy to the input pump pulse energy:

$$\eta = \frac{E_{\text{SHG}}(z_{\text{final}})}{E_{\text{p}}(z_{\text{initial}})}$$

In figure 2(A) the crystal conversion efficiency is simulated for an input Gaussian pump pulse with $\Delta \lambda_{\text{FWHM}} = 1$ nm, $\Delta T_{\text{FWHM}} = 4$ ps, $T = 20 \, ^\circ\text{C}$ and mode field diameter of 0.5 mm for several energies. It is therefore shown that efficient wideband ultrashort second harmonic generation is feasible in a single KTP crystal, exhibiting a flat conversion efficiency curve with a bandwidth of 75 nm, spanning from 980 to 1070 nm. It is shown that decreasing the energy lowers the typical conversion efficiency, while maintaining the plateau in the conversion efficiency curve.

We also added to figure 2(A) the conversion efficiency for $T = 100 \, ^\circ\text{C}$ in the dashed line. It is clear that the design is not prone to temperature changes. The reason for its robustness is the fact that the conversion efficiency will be dramatically altered only for wavelengths having their rapid adiabatic passage (RAP) near the crystal edges, thus sensitive to refractive index change that might change the RAP position outside or inside the crystal. Refractive index dependence with temperature was taken from [29].

We have also examined the robustness of the adiabatic design to different pulse chirps. In figure 2(B), the conversion efficiency is plotted for the same Gaussian pulse as in figure 2(A), being chirped in the time domain. $\tau / \tau_{\text{TL}}$ is the ratio between the chirped pulse full width half maximum (FWHM) in the time domain and the transform limited pulse FWHM. The interaction term generating the SHG pulse is proportional to $B_g^2(z,t)$, hence proportional to the pump intensity. It is therefore clear why stretching the pulses in the time domain has the same effect as lowering the pulse energy, leaving the pump intensity unchanged.

4. Experimental results

The experiment setup is depicted in figure 3. The second output of an optical parametric chirped pulse amplification (OPCPA) system (Venteon dual) served as the front end, delivering 10 $\mu$ J, 2 Mhz pump pulses. The CPA beam is then focused on an adiabatically aperiodically poled KTP (adAPKTP) crystal, extraordinary-wave (e-wave) polarized along the crystal $c$-axis. The residual pump and the generated SHG pulse was measured using a HORIBA spectrometer and displayed in figure 3, thus demonstrating an efficient conversion efficiency all over the pump spectra between 1020–1040 nm.

The adAPKTP domain structure was fabricated in uncoated, 1 mm thick z-cut, flux grown KTP plates by ‘Raicol Crystals’. Conventional photolithographic process and electric-field poling were used to create the designed domain structure [30, 31].

It was possible to control both the temporal and the spectral properties of the pump pulse using a mechanism located inside the venteon CPA module. By spatially stretching the beam spectra using a grating, it was feasible to truncate parts of the scattered beam thus controlling its bandwidth, altering the measured 20 nm pump spectra presented in figure 3 into a Gaussian-shape pump pulse centered around 1036 nm with a bandwidth of 10 nm. An adjustable mirror, placed after

**Figure 2.** Simulated conversion efficiency as a function of central pump wavelength, for different energies and pump chirp. (A) The conversion efficiency is simulated for several energies for both $T = 20 \, ^\circ\text{C}$ and $T = 100 \, ^\circ\text{C}$. (B) The conversion efficiency curve is simulated for several input pump chirp.
the grating, enabled us to change the relative phase between the pulse wavelengths, hence changing its duration between 350 fs to 3.5 ps in the full-bandwidth case, or between 600 fs to 2.6 ps in the truncated-bandwidth case. Measurements of the pump pulse duration were done using frequency resolved optical grating (FROG).

Conversion efficiency measurements were conducted for a large span of pump energies and presented in figure 4. For low pump energies, a good correspondence between numerical prediction and experimental results was achieved, even without taking into account parasitic effects such as TPA. For the strong pump regime, we have examined several mechanisms in order to explain the early saturation in the conversion efficiency curve. We examined through numerical simulation back-conversion of the generated SHG and other nonlinear cascaded effects through TWM, and found that such effects are not occurring in our adiabatic design. Possible degenerate four wave mixing (FWM) interactions were also
Figure 5. Temperature robustness. (A) Temperature barely affects the conversion efficiency for a very wide span of temperatures. The robustness is not altered with pump energy, as discussed in figure 2(A). Measurements of the conversion efficiency with the crystal cut in half give better results than those obtained with the whole crystal (green dots versus red dots), attributed to shorter possible absorption length. (B) SHG spectra is measured at 0.1 μJ for different temperatures. As seen, the different spectra are hardly affected by temperature.

Figure 6. Pulse duration effect on conversion efficiency. Inset (A) FROG measurement of 660 fs pulse duration. Inset (B) FROG measurement of 2.58 ps pulse duration.
dismissed due to phase mismatch considerations. We inserted the pump pulse into a bulk-KTP and experienced no absorption at all, giving rise to the fact that the generated green is absorbed during propagation along the crystal optical axis, decreasing the measured conversion efficiencies. Assuming that the absorption is attributed to TPA of the generated SHG pulse and incorporating it into the simulation, enabled us to fit the numerical predication to the experimental results at the full nonlinear regime with $\beta = 4(\text{cm GW}^{-1})$, in very good agreement with the nonlinear coefficient measurements of $\beta(532 \text{ nm})$ [32]. Kerr effect included in the simulation had no significant effect. The performance of the adiabatic SHG design has been validated for both the broader spectrum of 20 nm as well as to the 10 nm truncated one.

We also measured the dependence of the conversion efficiency as a function of the temperature of the nonlinear crystal. The results, which are presented in figure 5, exhibit a complete robustness of the designed crystal and experimentally validate the numerical prediction presented in figure 2(A). It is shown that temperature barely affects the adAPKTP performance. The conversion efficiency along with the SHG spectra are indifferent to temperature over a very wide range of more than 100 °C.

In order to examine whether the conversion efficiency is bounded due to TPA as predicted by simulation, the adAPKTP crystal was cut into two, hence decreasing the length in which SHG can be absorbed throughout. The saturation plateau in the conversion efficiency curve was now measured to be in the range of 40%–50%, justifying our assumption.

The effect of stretching the pump pulse on the conversion efficiency was also examined for the truncated bandwidth case. The pump energy was measured to be $0.7 \mu J$. The pulse durations, defined by 10%–90% knife edge and FROG measurement, varied between 660 fs (figure 6 inset (A)) to 2.58 ps (figure 6 inset (B)). In our measurements we observe that although the intensity density is decreased by $\sim 4$, the conversion efficiency did not change dramatically, as could be naively expected when comparing to the CW regime where $\eta \propto f_p^2$ [27]. The robustness shown in figure 2(B) is experimentally validated. Numerical predication with $\beta = 4(\text{cm GW}^{-1})$ is in agreement with experimental results. Measurements in the full bandwidth case were also conducted, with pump energy of 0.9 $\mu J$, exhibiting a decrease in conversion efficiency from $\eta = 27\%$ to $\eta = 9\%$ as pulse duration increases from 290 fs to 3.23 ps.

5. Conclusions

To conclude, we have experimentally demonstrated a robust and efficient SHG of ultrashort pulses based on adiabatic frequency conversion. We have shown that the conversion from near-IR to visible in a SHG process is not sensitive to the bandwidth of the fundamental pulse and to the temperature of the nonlinear crystal. It was also demonstrated that stretching the pump does not have a critical effect on the conversion process. A time domain simulation for any TWM process was developed, and good correspondence with experimental results has been obtained. The frame work of the simulation enables us to easily incorporate parasitic effects, both in the time or in the spectral domain, and can be generalized to include high-order nonlinear effects such as FWM for further research on ultrashort frequency conversion. Such an achievement can be useful in the design of extremely stable frequency conversion optical elements, aimed to perform at harsh environmental conditions as adverse temperatures, shocks, tensile stress and external pressure, as well as in fundamental research in second harmonic imaging microscopy and plasmonic nanostructures. Thus, appealing for a wide range of applications in medical procedures, avionics, satellites, and field-deployable communications systems.

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