We experimentally demonstrate an efficient broadband second-harmonic generation (SHG) process with a tunable mode-locked Ti:sapphire oscillator. We have achieved a robust broadband and efficient flat conversion of more than 35 nm wavelength by designing an adiabatic aperiodically poled potassium titanyl phosphate crystal. Moreover, we have shown that with such efficient flat conversion, we can shape and control broadband second-harmonic pulses. More specifically, we assign a spectral phase of absolute value and \( \pi \)-step, which allows wavelength tunable intense pump-probe and amplitude modulation of the broadband second-harmonic output. Such spectral phases serve as a proof of concept for other pulse-shaping applications for nonlinear spectroscopy and imaging.

Ultrafast laser sources are at the heart of ultrafast experimental science. In the past two decades, ultrashort pulse laser oscillators and amplifiers have become common equipment in the fundamental scientific exploration, as well as in a handful of industrial applications. Those sources which, by their nature, are broadband and coherent, allow exploring many phenomena that occur at the ultrafast timescale of many scientific processes and dynamical and coherent, allowing femtosecond temporal resolution experiments and ultrafast characterization methods, such as frequency-resolved optical gating (FROG), multiphoton intrapulse interference phase scan and spectral phase interferometry for direct electric-field reconstruction [1,7–9]. Among the various nonlinear conversion processes, three-wave mixing and, especially, second-harmonic generation (SHG), became widely used. Yet, frequency conversion in the ultrashort regime remained quite complicated, as the conventional conversion devices usually exhibit a tradeoff between the conversion bandwidth and the conversion efficiency, rooted in the phase mismatch between the interacting waves, which usually compensates for only a narrowband of frequencies [10,11].
power levels characteristic of high-repetition-rate femtosecond oscillators. We show that with pulse peak energies of nanojoule regime, one can achieve above 50% of energy conversion efficiencies for 70 fs Ti:sapphire pulses. Furthermore, the flat-top conversion frequency response of the presented design allows us to perform broadband pulse shaping manipulations prior to the nonlinear optical conversion, thus not suffering from the spectral limitation that is conventionally imposed by the limited bandwidth of birefringence or regular periodic crystal designs. More specifically, using a spatial light modulator (SLM) in a 4-f pulse shaper, we present a tunable pump-probe apparatus based on a varying absolute spectral phase profile in the frequency domain. In a similar way, we show that when applying a 𝜋-step spectral phase, coherent control of the SHG spectrum can be achieved, imposing a complete dip in the SHG, originating in the complete destructive interference of the second-harmonic field of the fundamental waves [23].

The experimental setup, illustrated in Fig. 1, consists of an 80 MHz repetition rate tunable coherent oscillator between 690 and 1040 nm (Mai-Tai) which served as the pump pulse, delivering 17.5 nm full width half-maximum (FWHM) ~70 fs transform-limited pulses, with energies spanning 10–30 nJ. The pump pulse then passes through JENOPTIK 640D SLM, enabling us to alter the pump pulse spectral phase and temporal shape before it is focused into an adiabatically aperiodically poled potassium titanyl phosphate (adAPKTP) crystal. The residual pump and the generated SHG are separated using a dichroic mirror around 950 nm. The spectrum of the SHG pulse, measured using an Avantes spectrometer, is displayed in the right lower image of Fig. 1, demonstrating efficient conversion over the entire pump-pulse spectrum.

The phase mismatch between the interacting waves is compensated for by using the poling method [10]. In the presented design, the second-order nonlinear susceptibility χ(2)(z) fluctuates between +χ and −χ in batches determined by χ(2)(z) = sign(cos(K(2)(z))z)), where the grating function K(2)(z) is given by K(2)(z) = (118.2z^3 - 45.2z^2 - 997.9z + 7957.1) [cm^{-1}]. and z spans the crystal length z ∈ [-0.5 cm, 0.5 cm].

The crystal grating period Λ = 2π/K(2)(z) adiabatically changes from Λ = 7.1 μm to Λ = 8.9 μm along the optical axis of the nonlinear crystal.

In our analysis, we use the three-dimensional generalization of the fully nonlinear dynamical equations, which dictates the nonlinear conversion of any three-wave mixing and, in particular, the depleted ultrashort SHG case [23]. Two-photon absorption (TPA) of the fundamental pump and the generated SHG was also taken into account as in the case of Ref. [23], likewise found to be of great importance. Since the SHG process occurred within the Rayleigh range of the pump pulse, spatial diffraction has been neglected, eliminating the interaction between different areas of the transverse intensity profile.

The following three-dimensional generalization of the SHG [Eqs. (1) and (2)], is applied for predicting the SHG process behavior:

\[ \frac{dB_{SHG}(r, z, t)}{dz} + iF^{-1}(β(ω + ω_{SHG})B_{SHG}(r, z, ω)) + iF^{-1}(β(ω + ω_{SHG})B_{SHG}(r, z, ω)) 

\]

\[ = -iχ(z)F^{-1}\left(\frac{ω + ω_{SHG}}{n(ω + ω_{SHG})c}F(B_{p}(r, z, t))\right) \]

\[ - \frac{β}{2} B_{SHG}(r, z, t)B_{SHG}(r, z, t), \] (1)

\[ \frac{dB_{p}(r, z, t)}{dz} + iF^{-1}(β(ω + ω_{p})B_{p}(r, z, ω)) = -iχ(z)F^{-1}\left(\frac{ω + ω_{p}}{n(ω + ω_{p})c}F(B_{SHG}(r, z, t))B_{p}(r, z, t))\right), \] (2)

where B_{SHG}(r, z, ω), B_{p}(r, z, t) are the electric field spectral density amplitudes of the pulses, B_{SHG}(r, z, t) and B_{p}(r, z, t) are the electric field modulation around the pulses central frequencies, and β(ω) is the dispersion relation. All physical variables in Eqs. (1) and (2) are carefully defined in Ref. [23].

Incorporation of the measured FROG pump pulse and the measured 40 μm FWHM Gaussian beam profile into the simulations yields great agreement with the experimental results for β = 4 [cm⁻¹]</ref>, the TPA coefficient, in correspondence with the nonlinear coefficient obtained in Refs. [23,24].

The energy conversion efficiency of the crystal is the ratio between the generated SHG pulse energy to the pump pulse energy:

\[ \eta = \frac{E_{SHG}(z_{out})}{E_{p}(z_{in})}, \] (3)

where z_{out} and z_{in} are the locations of the crystal output and input facets, respectively. The conversion efficiency measurements as a function of the central pump wavelength are presented as red dots in Fig. 2(a). Although conversion efficiency measurements were limited in the range of 970–1030 nm due to source limitations, the adiabatic design is capable of an efficient frequency doubling within a bandwidth of 80 nm, shown as blue solid line.

Next, we have added a broadband 4-f pulse shaper based on computer-controlled SLM positioned at its Fourier plane. First, by varying the spectral quadratic phase [25], we have used it as an aligned tunable compressor that can control the group delay dispersion of the fundamental, which also influences SHG output spectral distribution. The architecture for the 4-f pulse...
shaper includes two pieces of 600 grooves/mm ruled grating which disperse the light angularity into a telescope, based on a pair of curved 913.4 mm parabolic mirrors, which fits a 4-f arrangement in a chirped pulse amplification system.

The pump-probe apparatus is implemented by applying an absolute-valued spectral phase centered within the bandwidth of the fundamental spectral phase, denoted by $\omega_{\text{Abs}}$, i.e., $\phi \propto |\omega - \omega_{\text{Abs}}|$. Determined by the sign of the linear phase slope, different parts of the pump spectrum shifted in opposite directions in the time domain, therefore enabling splitting the incoming pump pulse into two localized pump pulses, with an extinction ratio and delay determined by $\omega_{\text{Abs}}$ and the absolute phase slope. The idler’s spectrum dependence with $\lambda_{\text{Abs}} = \frac{2\pi c}{\omega_{\text{Abs}}}$ is presented in Fig. 3 for an input hyperbolic secant pump pulse centered on 1005 nm. When applying the absolute-valued shape spectral phase in the center of the pump pulse $\lambda_{\text{Abs}} = 1005$ nm, as shown in Fig. 3(a-I), the initial pump pulse splits into two pulses with the same peak intensity, resulting in a symmetrical normalized pump probe SHG spectra. When the absolute-valued shape phase is not symmetrical with respect to the pump central frequency, as shown in Fig. 3(a-II), the generated asymmetrical pump pulses result in asymmetrical SHG spectra.

We proceed by applying a $\pi$-step spectral phase. As a result of destructive interference between the different frequency components, the $\pi$-step induces SHG/SFG pulses with spectral dips at different wavelengths. It is worth noting that due to the mathematical similarity between the perturbative solution of a TPA process in a non-resonant two-level quantum system and the instantaneous SHG process (where time scaling $t$ is replaced by $z$, the propagation length in crystal), most of the coherent control schemes that were applied in atomic physics can be adopted to a pulse-shaped SHG using the ASHG non-linear crystal. In order to demonstrate the shaping of the SHG spectra, we have also applied a $\pi$-step function at different locations on the pump spectral phase.

We consider the case of second-harmonic generation of ultrashort pulse with an electric field distribution of $\varepsilon(t)$. From second-order non-resonant time-dependent perturbation theory, the instantaneous second-harmonic spectral field can be found by the autoconvolution of the fundamental spectral field $\tilde{\varepsilon}(\Omega) = \tilde{F}[T(\varepsilon(t))]$ [10], which is described by

$$e_{\text{SHG}}(\omega_0) = \left| \int_{-\infty}^{\infty} \tilde{\varepsilon}(t) \cdot \exp(i\omega_0 t) \, dt \right|^2 = \int_{-\infty}^{\infty} \tilde{\varepsilon}(\Omega) \tilde{\varepsilon}(\omega_0 - \Omega) \, d\Omega,$$

(4)

where $\omega_0$ is the SHG frequency.

If we perform change of variables as $\Omega \rightarrow \Omega + \frac{\omega_0}{2}$, and write explicitly $\tilde{\varepsilon}(\Omega) = A(\Omega) \exp[i\Phi(\Omega)]$, we obtain Eq. (5):

$$e_{\text{SHG}}(\omega_0) = \left| \int_{-\infty}^{\infty} A(\omega_0/2 + \Omega) \left[ A(\omega_0/2 - \Omega) \right]^* \cdot \exp \left[ i \left( \Phi(\omega_0/2 + \Omega) + \Phi(\omega_0/2 - \Omega) \right) \right] d\Omega \right|^2,$$

(5)

where $A(\omega)$ and $\Phi(\omega)$ are the spectral amplitude and spectral phase, respectively.

The equation reflects that the SHG/SFG occurs for all pairs of photons with frequencies which add up to $\omega_0$ and lie within the spectrum of the exciting pulse. It is easy to see that when the phase cancels (i.e., $\Phi(\omega_0/2 + \Omega) + \Phi(\omega_0/2 - \Omega) = 0$) and, for a symmetric amplitude $A(\omega)$ with respect to $\omega_0/2$, the amount of SHG is maximized. This is in agreement with the finding of Meshulach and Silberberg [25,26], where they showed that an anti-symmetrical spectral phase can result in the same TPA rate as transform-limited excitation. The same behavior happens for the instantaneous SHG/SFG case, where the
Based on the broad spectral conversion response of the ASHG nonlinear crystal. We show that the SHG/SFG spectrum can be manipulated by tailoring the shape of the exciting ultrashort pulse. In particular, we investigate the effect of a spectral phase modulation of absolute value and a $\pi$-step spectral phase. We show that spectral shaping, as well as complete destructive interference in the SHG can be achieved. In addition, we show that certain $\pi$-step spectral phase modulation, which leads to long pulses, induces SHG as effective as transform-limited pulses. The basic principles presented here open a wide new area for theoretical and experimental work, as well as possible applications in nonlinear spectroscopy and in atomic and molecular physics.

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**REFERENCES**

1. J.-C. Diels and W. Rudolph, *Ultrashort Laser Pulse Phenomena: Fundamentals, Techniques, and Applications on a Femtosecond Time Scale*, 2nd ed. (Academic, 2006).
2. H. A. Zewail, *Angew. Chem. (Int. Ed.)* 39, 2586 (2000).
3. A. M. Weiner, *Ultrashort Optics* (Wiley, 2008).
4. L. A. Cavalieri, N. Müller, T. Uphues, V. S. Yakovlev, A. Baltus Caronka, B. Horvath, B. Schmidt, L. Blümel, R. Holzwarth, S. Hendel, M. Drescher, U. Kleinberg, P. M. Echenique, R. Kienberger, F. Krausz, and U. Heinzmann, *Nature* 449, 1029 (2007).
5. A. V. Kimel, A. Kirilyuk, P. A. Usachev, R. V. Pisarev, A. M. Balbashov, and T. Rasing, *Nature* 435, 655 (2005).
6. N. Dudovich, D. Oron, and Y. Silberberg, *Ann. Rev. Phys. Chem.* 60, 277 (2009).
7. R. Trebino, *Frequency-Resolved Optical Gating: The Measurement of Ultrashort Laser Pulses* (Springer, 2012).
8. V. V. Lozovoy, I. Pastrik, and M. Dantus, *Opt. Lett.* 29, 775 (2004).
9. C. Iaconis and I. A. Walmsley, *Opt. Lett.* 23, 792 (1998).
10. R. W. Boyd, *Nonlinear Optics*, 3rd ed. (Academic, 2011).
11. H. Suchowski, B. Bruner, A. Arie, and Y. Silberberg, *Opt. Photon. News* 21(10), 36 (2010).
12. H. Suchowski, P. Gorat, and A. Arie, *Laser Photon. Rev.* 8, 333 (2014).
13. C. Heese, C. R. Phillips, B. W. Mayer, L. Gaillmann, M. M. Fejer, and U. Keller, *Opt. Express* 20, 26888 (2012).
14. H. Suchowski, V. Prabhudesai, D. Oron, A. Arie, and Y. Silberberg, *Opt. Express* 17, 12731 (2009).
15. C. Heese, C. R. Phillips, L. Gailmann, M. M. Fejer, and U. Keller, *Opt. Lett.* 35, 2340 (2010).
16. C. Heese, C. R. Phillips, L. Gailmann, M. M. Fejer, and U. Keller, *Opt. Express* 20, 18066 (2012).
17. O. Yaakobi, L. Caspani, M. Clerici, F. Vidal, and R. Morandotti, *Opt. Express* 21, 1623 (2013).
18. H. Suchowski, P. R. Krogen, S. W. Huang, F. X. Kärtnner, and J. Moses, *Opt. Express* 21, 28892 (2013).
19. P. Krogen, H. Suchowski, H. Liang, N. Flemens, K.-H. Hong, F. X. Kärtnner, and J. Moses, *Nat. Photonics* 11, 222 (2017).
20. G. Porat and A. Arie, *J. Opt. Soc. Am. B* 30, 1342 (2013).
21. C. R. Phillips, C. Langrock, D. Chang, Y. W. Lin, L. Gaillmann, and M. M. Fejer, *J. Opt. Soc. Am. B* 30, 1551 (2013).
22. A. Leshem, G. Meshulam, G. Porat, and A. Arie, *Opt. Lett.* 41, 1229 (2016).
23. A. Dahan, A. Levanon, M. Katz, and H. Suchowski, *J. Phys.* 29, 84004 (2017).
24. V. A. Maslov, V. A. Mikhailov, O. P. Shaunin, and I. A. Shcherbakov, *Quantum Electron.* 27, 356 (1997).
25. D. Meshulach and Y. Silberberg, *Nature* 396, 239 (1998).
26. D. Meshulach and Y. Silberberg, *Phys. Rev. A* 60, 1287 (1999).

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**Fig. 4.** ASHG conversion in KTP crystal excited by a pulse with a spectral phase of $\pi$-step, as a function of the step position. (a) Illustration of SLM-induced $\pi$-step phase centered at $\lambda_{\text{step}}$. (b) Experimental results. (c) Simulation results. The simulation results were obtained by inserting the measured spectrum of the original pulse.

**Transform-limited pulse ($\Phi(\Omega) = 0$), and every anti-symmetric phase with respect to $\overline{\pi}_0$, result in the same SHG generation. Therefore, the anti-symmetric phase can significantly affect the shape of the pulse to have much smaller amplitude and a much longer duration, it does not affect the SHG [24].**

Our experimental results of applying an anti-symmetric $\pi$-step phase each time at a different wavelength show different SHG/SFG spectra for each measurement. As expected, this destructive interference appears in the SHG/SFG if the step is not in the middle of the spectrum, and could even lead to complete destructive interference of the SHG/SFG signal at a specific wavelength. In Fig. 4, we show the experimental results, as well as the simulations of broadband ASHG conversion as a function of the $\pi$-step position. The simulation results were obtained by inserting the measured spectrum of the original pulse. Very good agreement is obtained between the experimental results and the numerical simulations.

To conclude, we experimentally investigate the performance of an adiabatic aperiodically poled KTP crystal using a conventional femtosecond high-repetition-rate oscillator in the nanojoule energy regime. We show that efficient broadband ultrashort second-harmonic generation is feasible in a single crystal, experimentally demonstrating an acceptance bandwidth bigger than 40 nm with nanojoule level excitation. Furthermore, we show that the broadband operation of the adiabatic crystal design enables us to perform pulse shaping manipulations on the fundamental pulses. An SLM-dependent tunable pump probe was given as an example, where altering the pump probe spectral and temporal characteristics is possible without the need of realigning the experimental apparatus.