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Generation of Octave-Spanning Multiple Harmonics for Ultrafast Waveform Synthesis

Wei-Chun Hsu[1,2], Zhi-Ming Hsieh[1,3], Ying-Yao Lai[4], Chien-Jen Lai[5], Wei-Ting Chen[1], Lung-Han Peng[4], Ci-Ling Pan[2], and A. H. Kung[1,2]

[1]*Institute of Atomic and Molecular Sciences, Academia Sinica, Taipei 10617, Taiwan,China*

[2]*Department of Photonics, National Chiao Tung University, Hsinchu 30010, Taiwan,China*

[3]*Physics Department, National Taiwan University, Taipei 10617, Taiwan,China*

[4]*Institute of Photonics and Optoelectronics, National Taiwan University, Taipei 10617, Taiwan,China*

[5]*Department of EECS, MIT, Cambridge, MA 02139, USA*

E-mail: *sunday2016@gmail.com*

Abstract: Up to seven laser harmonics covering more than two octaves in frequency have been generated efficiently in a single PPLT crystal, permitting the syntehsis of 1.5 femtosecond pulses in a stable and compact setting.

Keywords: waveform synthesis, quasi-phase-matching

1. INTRODUCTION

Ultrashort subfemtosecond pulses with attosecond timing are essential for probing the evolution of electronic processes in atoms and molecules. The ultimate goal of these studies is to achieve coherent control of electronic motion in matter with light. To reach this goal it requires developing the ability to generate light pulses with an octave-spanning spectrum and having the ability to shape the pulses. There have been two approaches that can accomplish this. In one approach single-cycle to sub-cycle pulses are synthesized from a wide comb of equidistant frequencies generated by adiabatically driving a Raman coherence with two nanosecond high power lasers [1]. Another approach produces phase-coherent optical pulses by phase-locking two femtosecond lasers [2]. An improved version of the latter approach mixes the pulses from a femtosecond laser with pulses from a synchronously-pumped OPO to produce a wideband frequency spectrum [3]. These approaches are all quite complex and have yet to produce well-controlled and long lasting waveforms that are needed for applications.

Since the frequency ω_q and phase ϕ_q of the frequency comb that Fourier synthesizes to periodic waveforms would satisfy the conditions:

$$\omega_q = \omega_{ceo} + q\omega_m, \quad \phi_q = \phi_0 + q\phi_m \qquad (1)$$

where ω_{ceo} is the carrier-offset frequency, ω_m is the comb frequency spacing, ϕ_0 is a static offset, and ϕ_m is a linear phase difference between adjacent comb components, it is clear that all the components of the comb are harmonics of the comb frequency spacing. Thus by starting with a fundamental frequency at ω_m the problem of generating a wide comb is reduced to that of generating as many harmonics of ω_m as is practical. The solution is simple as these harmonics can be generated by nonlinear mixing. As such they naturally will all be phase stable and coherent relative to each other and could synthesize to long lasting ultrafast waveforms [4].

In this paper we report the generation of up to seven phase-coherent harmonics in a single periodically-poled lithium tantalate crystal. Harmonics 2 to 5 were generated by efficient quasi-phase-matched (QPM) cascaded sum mixing from a fundamental wavelength while harmonics 6 and 7 were from random-quasi-phase-matching of the lower harmonics. These harmonics would produce stable periodic sub-cycle waveforms in the femtosecond and subfemtosecond time scale. Note that although multiple harmonics can be generated using an array of nonlinear crystals, the discrete arrangement will involve splitting and combining beams and adjusting beam polarizations several times so that it will lead to disastrous phase fluctuations and inefficient energy conversion. Cascaded QPM generation in a single crystal is compact and efficient. It can be an extremely effective source for the production of stable ultrafast waveforms.

2. EXPERIMENTAL AND RESULTS

Figure 1 is a schematic depicting the processes used in the present approach for multiple harmonics generation. For convenience the fundamental wavelength was chosen at 2406 nm. A nanosecond source at 2406 nm had been constructed and was available for this experiment. The key component in this set up is the periodically-poled lithium tantalate (PPLT) crystal that has been fabricated in house to satisfy sequential QPM of all four processes inside the crystal. The poled domain periods in the crystal are 34.09 μm, 22.33 μm, 13.18
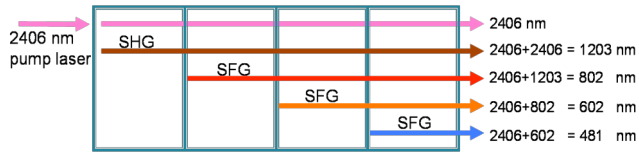
Figure 1. Diagram showing scheme of multiple harmonics generation. Input fundamental wavelength is 2406 nm.

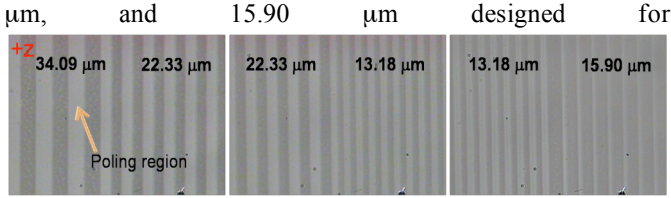μm,   and   15.90   μm   designed   for

Figure 2. Micrographs of poled domains in the PPLT crystal, showing etched domains at the boundaries where there is a period change. The corresponding periods are shown in the inside top of the micrograph.

first order QPM for processes 1-3 and second order QPM for process 4 at 100 °C respectively for the four processes. Figure 2 shows micrographs of different poled domain regions of the crystal showing the arrangement of periodic domains at the transition boundaries.

The PPLT crystal was 2 cm long, 1.3 cm wide and 0.5 mm thick. The pump beam at 2406 nm was 3.5 ns in duration. When it was focused through the crystal the input and the generated harmonics exit the crystal collinearly as a beam of white light. The beam consisted of primarily the first five harmonics of the input frequency. The temperature of the crystal was scanned to optimize the output at the fifth harmonic of 481 nm. The designed temperature for QPM was 100 °C. Experimentally it was found to occur at 120 °C. Such a difference is quite common with periodically-poled crystals mainly because of insufficient refractive index data to produce a consistent Sellmeier equation for the material.

Figure 3 shows the measured average power at the second to the fifth harmonic that were monitored as a function of the input power. At maximum input, corresponding to an incident intensity of 250 MW/cm^2, as much as 30% of the pump was converted to the higher harmonics. About 25% was into the second harmonic while the balance 5% was distributed among the rest. Figure 4 is a picture of the harmonics taken after they were dispersed by a prism. The fundamental at 2406 nm cannot be recorded. In addition to the four recordable harmonics, the sixth and the seventh harmonics at 401 nm and 344 nm could also be observed. The strength of these two are quite small as they were generated only through random phase-matching of the lower harmonics.

3. DISCUSSION

The harmonics thus generated naturally satisfy the frequency condition of equation (1). By installing a phase
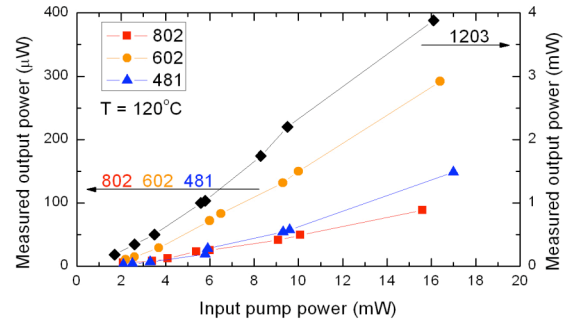

Figure 3. Average power of the harmonics vs input power.

Figure 4. Picture taken with a CCD camera of the images of the second to the sixth harmonic from the near-ir to the uv dispersed by a prism. The ir and uv images are those from the phosphorescent sensors.

compensator the phase condition of (1) can also be met. Consequently it will be possible to Fourier synthesize waveforms that have stable and controllable carrier-envelope phases with these outputs. The generated relative intensities match quite well with those required to synthesize a sawtooth waveform. Transform-limited pulses synthesized from these will reach 0.6 cycles with a 500 attosecond field width and 1.4 fs envelope width. In order to reach these durations it will require modifying the amplitudes of the harmonics.

This cascaded QPM generation and the adiabatic Raman generation are two techniques that allow full control of the carrier-envelope phase in waveform synthesis. With the Raman technique a large number (more than 10) of harmonics can efficiently be produced in one pass. It is thus most suitable for generating waveforms with the shortest durations. The QPM technique we have demonstrated here offers complementary advantages. It is not locked to a Raman transition. So the technique can work for any desired input frequency. It is tunable to the extent allowed by QPM of the highest harmonic. It requires a rather low input power so that there is the potential to construct a quasi-CW train of periodic pulses. The pump duration can be from femtoseconds to the CW. So this QPM approach offers many new possibilities of optical waveforms for explorations in ultrafast science and quantum control.

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