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# Cascaded Third-Harmonic Generation Approaching Full Efficiency through an Unconventional Pathway

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## ABSTRACT

Third harmonic generation (THG) achieved by the simultaneous phase matching of multiple quadratic nonlinear processes in a single crystal has been studied previously both theoretically and experimentally, achieving modest conversion efficiencies.[1] As opposed to the common practice of using second harmonic generation (SHG) and sum frequency generation (SFG) stages in series, performing these processes in parallel results in a narrow material parameter range in which the transfer of energy to the third harmonic can be fully efficient, even under conditions of perfect phase matching.[2] This is largely due to the presence of back-conversion in the conversion dynamics, which limits the efficiency for spatio-temporally non-uniform beams. Here we demonstrate an unconventional method of THG where the fundamental wave is first frequency-quadrupled through cascaded SHG before being down-converted to the third harmonic. When the three associated phase matching conditions are met simultaneously, we find that energy is transferred robustly and efficiently from the fundamental wave to the third harmonic with inhibited back-conversion. This is an example of convergent dynamical behavior emerging from hybridized nonlinear optical processes as recently observed in hybridized parametric amplification.[3] These dynamics are achievable over a broad parameter range that correspond to common nonlinear crystals. Using a spatio-temporal numerical investigation, we find that 25-ps $CO_2$ laser pulses with Gaussian spatio-temporal profiles and 10-$\mu m$ wavelength can be converted to their third harmonic with over 80% conversion efficiency in a realizable monolithic orientation-patterned GaAs device. High performance realizable domain-poled KTP and LNB devices are also investigated.

**Keywords:** Frequency conversion, Hybridized nonlinear optics, Third-harmonic generation, Quasi-phase matching.

## 1. INTRODUCTION

Third-harmonic generation (THG) is a commonly used nonlinear wave-mixing process for extending the frequency range of high-powered, ultra-fast lasers.[2,4–7] The two prominent methods of THG using $\chi^{(2)}$ processes involve sum-frequency generation (SFG) and second-harmonic generation (SHG), performed either simultaneously in a single crystal, or sequentially with two crystals. The simultaneous method is challenging. With few exceptions, simultaneous processes result in cyclic conversion dynamics, which ensures spatio-temporally non-uniform pulses cannot be fully converted[2] (Fig. (2)). Moreover, the peak THG conversion efficiency obtained during a cycle is usually less than one. As a result, conversion efficiencies are modest, even when the device is optimized.[1] Thus, sequential processes are typically employed when high THG efficiency is required. In addition to being relatively cumbersome compared to a monolithic device, achieving a high conversion efficiency in this case relies on having initial conditions in the SFG stage of equal second-harmonic and fundamental photon numbers (Fig. (1)). With this condition met, 80% efficiency has been achieved with large beams and pulse durations $\geq$ 100 ps,[8] an important technology for inertial confinement fusion research facilities.[7]

Here we propose a new method for THG using hybridized nonlinear optics that has the convenience of being monolithic while allowing very high conversion efficiency due to its having convergent rather than cyclic evolution dynamics. This previously unexplored process involves cascaded SHG to quadruple the fundamental frequency, and subsequent down-conversion to the third-harmonic through difference-frequency generation (DFG), a process we will call Fourth-to-Third-Harmonic Generation (FTHG) (Fig. 3). Using numerical solutions of the coupled

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amplitude equations, we illustrate the dynamics of this method when all processes are phase matched. Through the use of spatio-temporal pulse propagation simulations, we predict the viability of this method to produce high conversion efficiency in manufacturable quasi-phase matching (QPM) devices. We consider three applications of FTHG with relevance to common lasers having frequencies in the mid-IR range: $2.06\mu m$ fundamental wavelength in quasi-periodically poled LiNBO and KTP, and $10\mu m$ fundamental wavelength in orientation-patterned GaAs. We find that these realistic structures can robustly produce third-harmonic intensities with upwards of 80% efficiency. In all cases, spatio-temporally Gaussian fundamental wave intensity profiles are assumed.

## 2. CONVENTIONAL THG IN $\chi^{(2)}$ NONLINEAR MEDIA

### 2.1 Two-Stage THG

As discussed above, the most common configuration for achieving high THG conversion efficiency requires the use of two separate nonlinear stages. Under this configuration, the input field at the fundamental harmonic is split between the two stages. The first stage performs SHG to double a portion of the fundamental field to its second harmonic. The second stage performs SFG of the remaining fundamental field with the second harmonic field from the previous stage to generate the third harmonic field. This is illustrated in Fig. 1a.

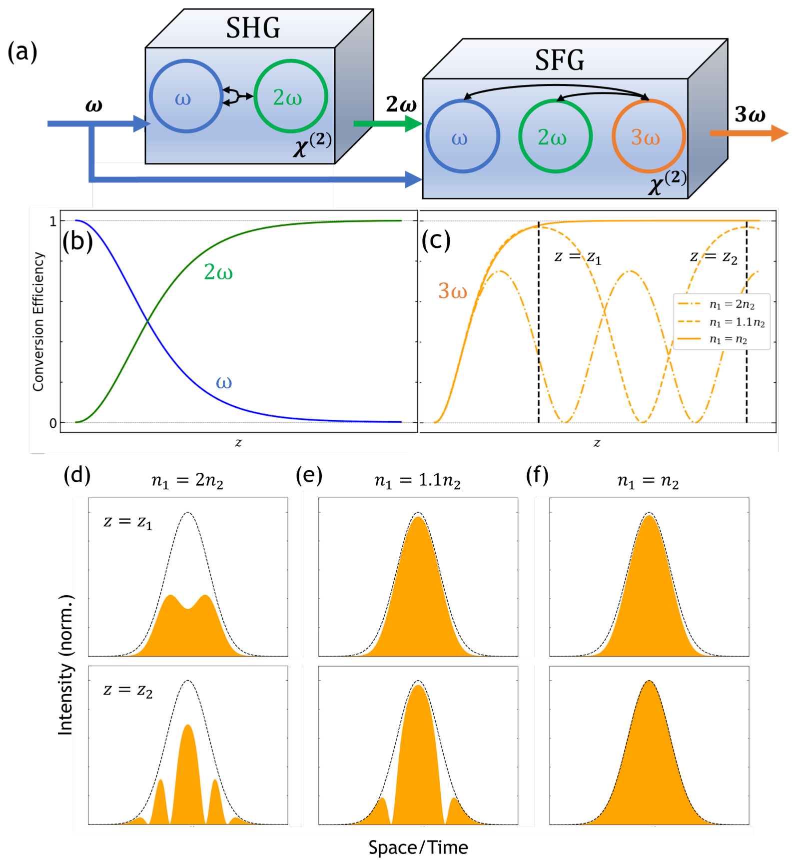


Figure 1: (a) Schematic diagram of conventional two-stage THG setup. (b) Monochromatic plane wave dynamics for the SHG stage. Full, non-cyclic intensity conversion from the fundamental harmonic (blue) to the second harmonic (green) is observed. (c) Monochromatic plane wave dynamics for the SFG stage. The third harmonic intensity (orange) is depicted under different initial values of the fundamental and second harmonic flux densities given by $n_1$ and $n_2$, respectively. Non-cyclic and asymptotically full conversion efficiency to the third harmonic occurs only when $n_1 = n_2$ (solid orange). (d-f) Spatiotemporal conversion to the third harmonic at the blue dashed lines in (c) corresponding to $z = z_1$ and $z = z_2$ for initial flux densities $2n_1 = n_2$, $1.1n_1 = n_2$, and $n_1 = n_2$, respectively. All simulations assume conditions of perfect phase matching.

In practice, the SHG process tends to be very efficient with full asymptotic conversion possible for monochromatic plane waves when perfectly phase matched (Fig. 1b). A potential bottleneck in efficiency comes from the SFG process. Two limitations on SFG efficiency arise when the input photon flux densities, $n_1$ and $n_2$, of the fundamental and second harmonic are unequal. First, one of the lower harmonics will deplete before the other, effectively capping the maximum conversion efficiency achievable (as seen in the two dashed curves of Fig. 1c). Second, the SFG dynamics will oscillate, resulting in asynchronous conversion along the spatiotemporal distribution of the mixing harmonics. Figs. 1d,e illustrate both the cap on maximum THG conversion efficiency and the consequence of asynchronous conversion: no device length can be chosen at which all spatiotemporal coordinates are maximally converted.

As is well known, these limitations on efficiency can be mitigated by choosing $n_1$ and $n_2$ to be equivalent. As seen in Figs. 1c,f, this results in full asymptotic conversion to the third harmonic field across the entire spatiotemporal distribution. If the SHG stage is operated with unity efficiency, this amounts to splitting the fundamental in a 2:1 ratio between the first and second stage and then ensuring identical pulse duration and beam shape between the first and second harmonic inputs to the second stage.

## 2.2 Simultaneous (Single-Stage) THG

An alternative to using two separate nonlinear stages is to perform both nonlinear interactions simultaneously in a single device (Fig. 2a). Simultaneous THG can be achieved using one of a number of multi-process QPM techniques that allow for simultaneous phase matching of the SHG and SFG processes. This makes for a simple and compact implementation of THG. However, as SFG begins as soon as any second harmonic photons are generated, the initial fundamental and second harmonic field photon flux densities cannot be equal. This leads to the same limitations in THG efficiency due to inhibited and oscillatory dynamics driven by a ”mismatch” in the photon flux densities described in the previous section (Fig. 2b).

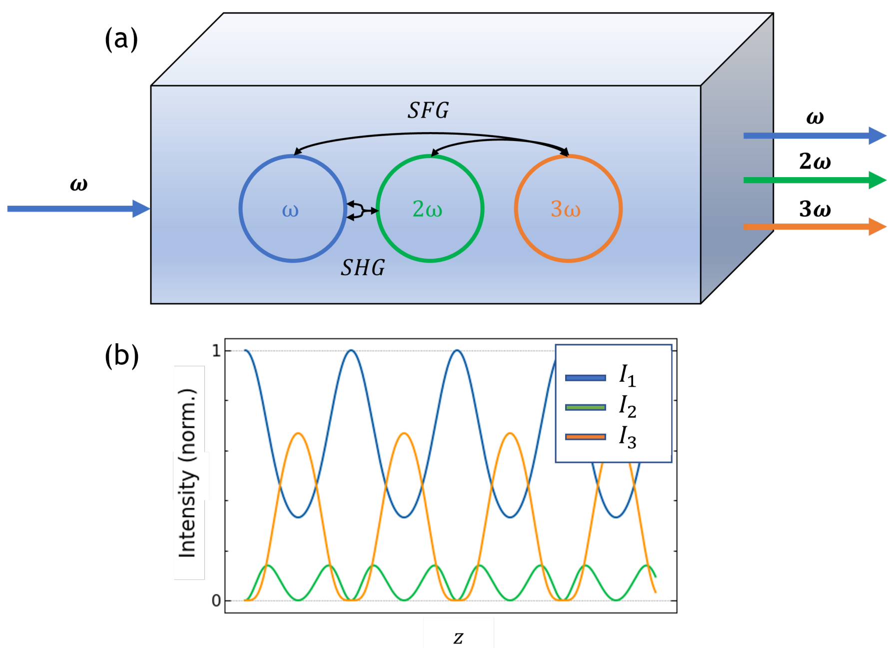


Figure 2: (a) Schematic diagram of a conventional ‘simultaneous THG’ stage, in which SHG and SFG processes are simultaneously phase matched. (b) Representative monochromatic plane-wave dynamics of such a device. The lack of control over the relative photon flux densities of the mixing harmonics leads to oscillatory evolution and a reduced maximum conversion efficiency.

## 3. FOURTH-TO-THIRD-HARMONIC GENERATION

Here, we investigate a novel route toward achieving efficient THG in a single nonlinear stage. Our approach, illustrated in Fig. 3a, involves converting a fundamental harmonic field to its third harmonic by three simultaneously occurring nonlinear processes. The first two nonlinear processes serve to convert the fundamental harmonic to its fourth harmonic through two cascaded SHG processes. The third and final process performs difference frequency generation (DFG) on the fourth harmonic to generate the third harmonic and replenish the fundamental harmonic. The energy in the fundamental harmonic is then re-upconverted to the fourth harmonic, restarting the cycle and resulting in full asymptotic conversion to the third harmonic (Fig. 3b).

We find under this configuration that the THG process takes on the same damped-cyclic behavior observed for parametric amplification hybridized with SHG,[3,9,10] leading to asymptotically full conversion. As these convergent dynamics are independent of the initial intensity, asymptotically full THG conversion dynamics occur across all spatiotemporal coordinates (Fig. 3c). Thus, as long as the phase matching conditions for each of the three nonlinear processes can be simultaneously met, efficient THG under the FTHG approach becomes as simple as aligning a laser beam into a single device.

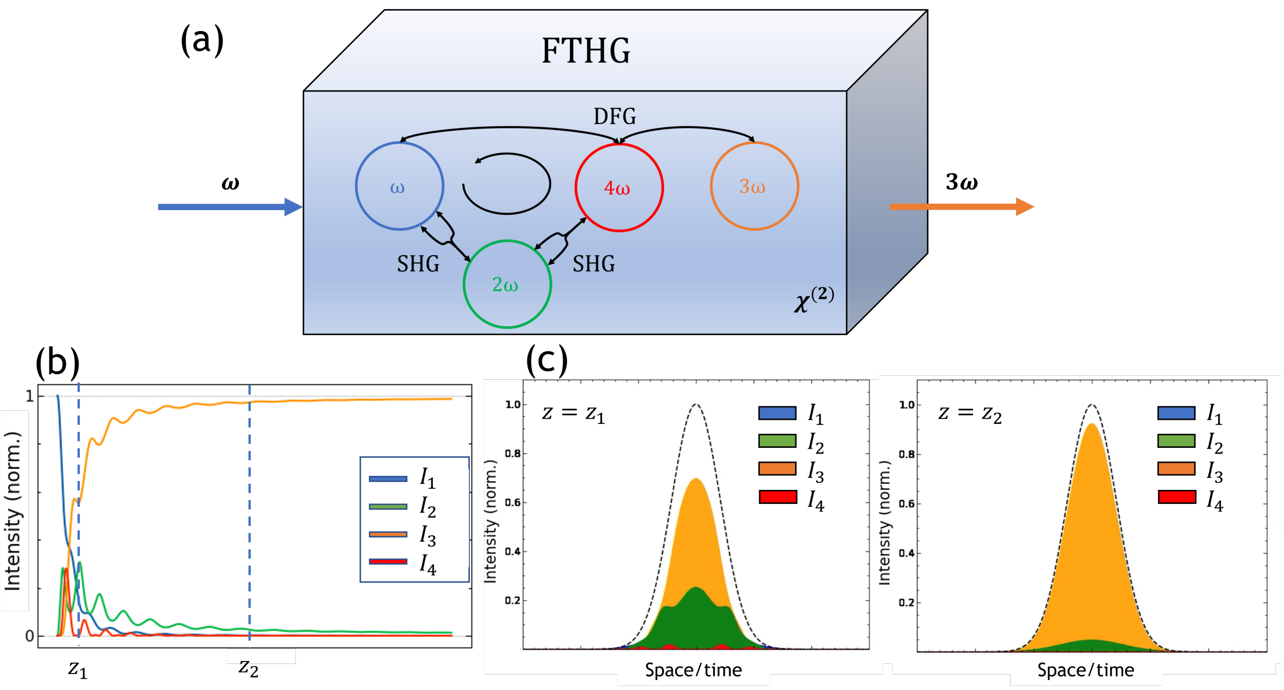


Figure 3: (a) FTHG schematic diagram. A monolithic device achieves THG by three simultaneously phase-matched processes: $\omega + \omega \rightarrow 2\omega$ (SHG), $2\omega + 2\omega \rightarrow 4\omega$ (SHG), $4\omega - \omega \rightarrow 3\omega$ (DFG). (b) The dynamics of FTHG are rapidly damped cycles, with asympotically full conversion to the third harmonic field. (c) Asymptotic convergence as propagation approaches infinity allows spatiotemporally uniform conversion to the third harmonic and thus high THG efficiency, as illustrated by spatiotemporal intensity plots corresponding to $z = z_1$ and $z = z_2$ in (b).

### 3.1 Simulations

Figure 4 depicts numerical pulse propagation simulations of the FTHG process in aperiodically poled $LiNbO_3$ designed to convert a 2.06 µm wavelength fundamental harmonic to a 690 nm third harmonic. The input fundamental harmonic has an initial intensity of 0.25 $\frac{GW}{cm^2}$, and is 100 ps in duration. The phase matching is achieved using the multiprocess QPM technique described in[11] with amendments to the domain sizes and resolution to ensure manufacturability. While the energy appears to oscillate asynchronously across the temporal extent of the first, second, and fourth harmonic pulses, the third harmonic grows asymptotically, acquiring the majority of the energy in the system.

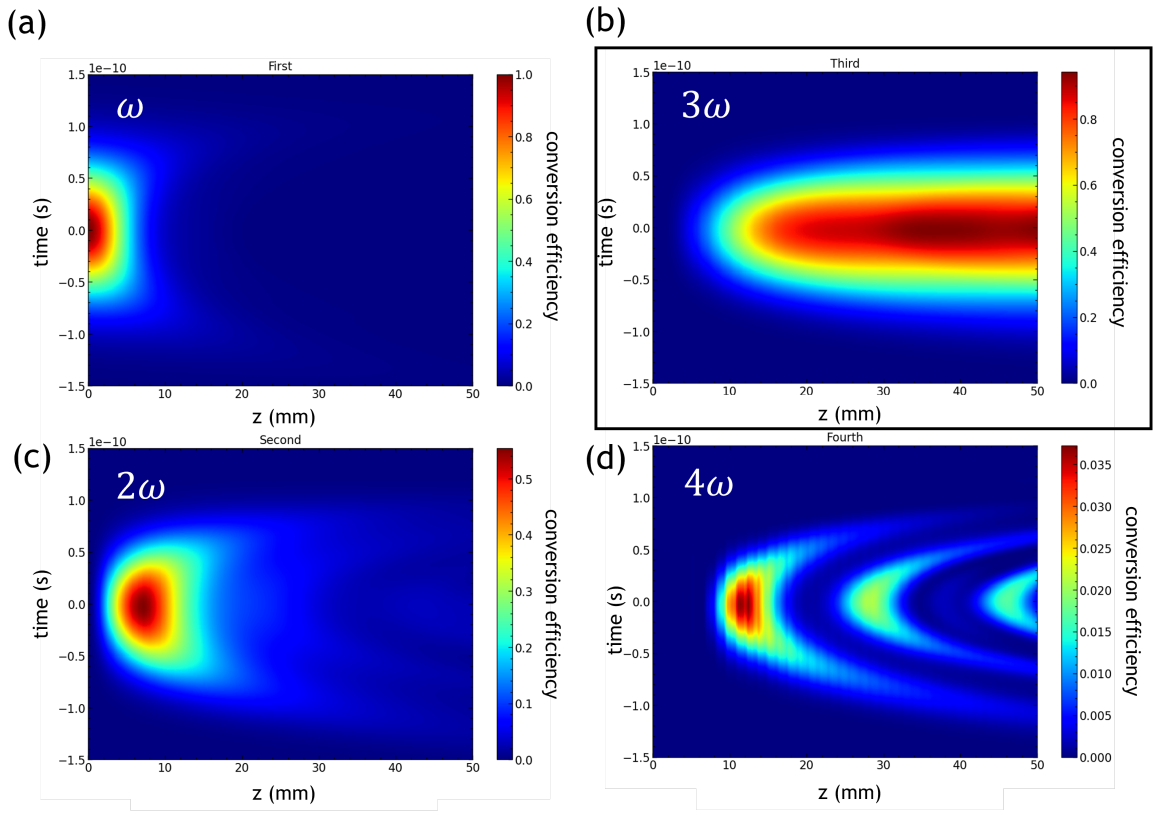


Figure 4: Numerical simulation of a 2.06 µm, 0.25 $\frac{GW}{cm^2}$, 100 ps incident fundamental harmonic pulse undergoing FTHG in aperiodically poled $LiNbO_3$.

To predict the total FTHG conversion efficiency possible in the $LiNbO_3$ device, we perform a spatiotemporal simulation of 1+2D Gaussian pulse and beam shapes. We find that the integrated conversion efficiency of the fundamental to the third harmonic asymptotically approaches 87% after 5 cm of propagation. Similarly, for a 5 cm aperiodically poled KTP device we find 82% conversion efficiency possible. For FTHG of 10 µm pulses to 3.33 µm, we find orientation patterned GaAs can accomplish the task with upwards of 81% conversion efficiency. Thus, the FTHG method provides a general approach for achieving THG with conversion efficiencies exceeding 80% in a monolithic device with a spatiotemporally non-uniform incident pulsed laser of 100-ps duration.

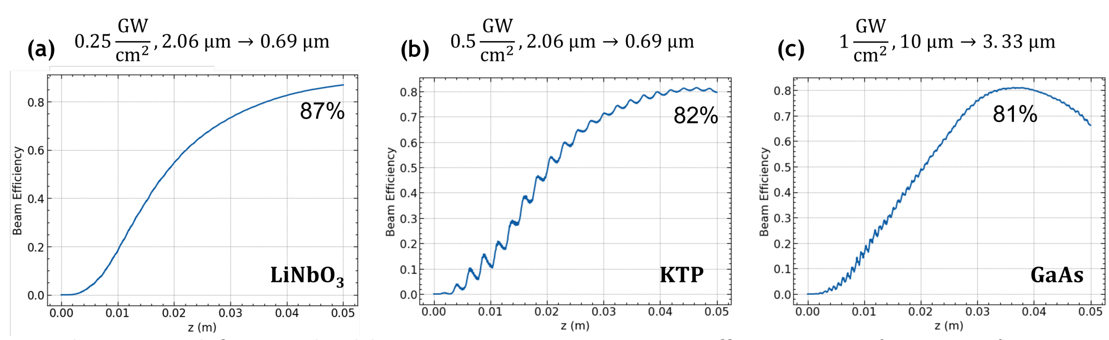


Figure 5: Total integrated first-to-third-harmonic energy conversion efficiency as a function of propagation for FTHG of 2.06 µm light in aperiodically poled $LiNbO_3$ or KTP and 10 µm light in orientation patterned GaAs. Efficiencies exceeding 80% can be consistently achieved.

## 4. DISCUSSION AND OUTLOOK

We have investigated an unexplored pathway for third-harmonic generation, fourth-to-third harmonic generation (FTHG), involving the use of 3 different nonlinear, simultaneous processes. We have found that FTHG has damped-cyclic behavior with full convergence to the third harmonic, which is ideal for converting nonuniform spatio-temporal pulsed lasers in a monolithic device with high efficiency.

Comparing this method to existing methods for generating the third harmonic, our propagation simulations predict performance as good as two-stage THG, the most efficient known method for THG. While the devices we explored are based on multi-process QPM, and thus do not easily scale to very high energy applications due to the limited apertures of polable quadratic materials, for applications at lower energy, the convenience of performing FTHG in a single crystal may be desirable.

From a broader perspective, the discovery of another multi-process nonlinear conversion scheme with convergent, damped-cyclic dynamics – in addition to the recently discovered hybridized parametric amplification approach[3,9,10] – suggests that additional devices for high efficiency frequency conversion of spatiotemporally nonuniform waves may be found for still more applications.

## ACKNOWLEDGMENTS

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