Cascaded optical parametric chirped-pulse amplification for multi-cycle THz-wave generation

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Abstract: We experimentally demonstrate cascaded optical parametric chirped-pulse amplification, an efficient method for the amplification of multi-cycle THz-frequency waves. Experimental results are supported by numerical simulations and promise THz efficiencies at the percent level.

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Significant knowledge of the chemical and biological world at the atomic and molecular level – both structure and dynamics – originates from investigations performed using X-ray radiation from synchrotron and free-electron laser (FEL) facilities. Owing to the financial cost of these facilities, only a limited number are available worldwide, which creates a bottleneck for scientific progress. There is therefore a strong demand for tabletop, high-flux X-ray sources with output parameters enabling a range of experiments currently performed at synchrotron and FEL facilities.

Laser-driven particle accelerators have recently emerged as a promising pathway toward low-cost, laboratory-size coherent X-ray sources, yet a number of technical challenges must still be overcome. In particular, the use of optically generated multi-millijoule multi-cycle THz-waves as the charged-particle accelerating field appears as an attractive solution to downsize standard accelerators. However, this approach is currently hampered by the limited energy conversion efficiency (or conversion efficiency) of down-conversion processes employed to derive THz frequency fields from near-IR optical pulses. The generation of high-energy THz-frequency pulses therefore requires either dramatic scaling of the near-IR driving laser systems toward energies far exceeding the current state-of-the-art or increasing the conversion efficiency of the frequency down-conversion process.

Over the past few years, our group has been pursuing both approaches to tackle this problem. For instance, we recently reported record-high 0.1% conversion efficiencies for narrowband terahertz generation via optical rectification of optical pulses derived from a Ti:sapphire laser [1] in a periodically poled lithium niobate crystal at cryogenic temperatures. We are simultaneously developing high-energy and high-average power cryogenically cooled Yb:YAG laser systems that largely exceed the capabilities of Ti:sapphire systems [2,3]. The favorable scaling in output energy obtained using cryogenically cooled Yb:YAG systems, however, comes at the expense of available amplified bandwidth, therefore preventing the use of optical rectification as a frequency down-conversion method to generate terahertz waves in the frequency range of few hundred GHz [1].

Here, we present an alternative method that paves the way toward efficient down-conversion of high-energy near-IR pulses into the THz-frequency range, relying on cascaded optical parametric amplification (COPA). This method enables (i) taking full advantage of high-energy narrowband driving lasers (ii) increased design flexibility and multi-stage modularity and (iii) promises conversion efficiencies beyond the Manley-Rowe limit – particularly under cryogenic cooling – since it favors efficient cascading of the parametric amplification process. Based on initial experiments accompanied by numerical simulations, conversion efficiencies >1% at cryogenic temperatures – to realize multi-millijoule terahertz frequency pulses – appear feasible.

1. Experimental setup

A key aspect of our method is the use of few-hundred picosecond pump pulses with sub-nanometer spectral bandwidth and multi-millijoule energy level – ideally matching the output parameters of cryogenically cooled Yb:YAG amplifiers – and of few-nanoseconds, spectrally chirped seed pulses with a few nanometers of bandwidth. Resorting to relatively long pump pulses relaxes temporal walk-off limitations and strongly mitigates the accumulation of nonlinear phase on the pump beam. Combining these two advantages enables resorting to long –

several centimeters – nonlinear gain media, which favors parametric gain and, in the case of THz generation, also favors the cascading process, which ultimately enhances the conversion efficiency.

In our implementation, we use ~ 1.5 mJ energy optical pulses with ~ 6 nm bandwidth around 1029 nm, temporally stretched to ~ 2 ns full width duration as input to our setup. Of this 1.5 mJ, 320 μ J of energy is directly sent to the COPA to serve as signal beam while the remaining energy is directed toward a home-built, 4-pass, cryogenically cooled Yb:YAG amplifier [4]. Pulses with up to 50 mJ energy at 100 Hz repetition rate, spectrally narrowed – via spectral gain narrowing – to ~ 0.35 nm bandwidth (300 ps duration) with center wavelength around 1029.5 nm emerge from this amplifier and are directed toward the COPA to serve as pump pulses. A delay line is installed that allows compensating for the extra beam path experienced by the pump beam in the Yb:YAG amplifier and to enable tuning of the temporal overlap between pump and signal pulses in the parametric amplifier. Pump and seed pulses are vertically polarized and spatially overlapped collinearly (Fig. 1). The COPA takes place in a series of four consecutive parametric amplifiers consisting successively of a 2 cm-thick, 0.3×0.3 cm² clear aperture MgO:PPLN crystal (MgO:PPLN), a 1 cm-thick, 0.3×1 cm² clear aperture MgO:PPLN crystal, and two consecutive 0.5cm-thick, 0.3×1 cm² clear aperture MgO:PPLN crystals. All crystals are poled with the same period to ensure phase-matching at 515 GHz frequency. Notice that in this proof-of-principle experiment no detailed optimization of the fluence on each crystal was performed and the fluence was capped at 1 J/cm² (7 mJ energy, 3.5 GW/cm² peak intensity) in each COPA stage to ensure safe operation far away from damage threshold. In addition, only one surface of one of the 0.5cm-thick COPA crystal featured an AR-coating while all other surfaces were left uncoated. An imaging setup monitored the beam profile at the exit surface of the last COPA stage. The aim of this imaging setup is to ensure that neither beam break-up due to cascading or catastrophic beam collapse due to self-focusing occurs during amplification.

Fig.1. Layout of the parametric amplifier along with relevant beam parameters. W: wedged glass piece; BD: beam dump; ND: neutral density filter; BPF: bandpass filter; PD: silicon photodiode; OSA: optical spectral analyzer; PPLN: periodically poled lithium niobate crystal.

The characterization of the amplifier is performed via monitoring the signal beam of the parametric amplifier. The signal wavelength is centered at 1031.25 nm, a wavelength set by both the fixed period of the PPLN crystals and the fixed emission wavelength of cryogenic Yb:YAG used to pump the COPA. A small fraction of the collinearly propagating pump and signal beams is picked by an uncoated wedged piece of glass and further attenuated by neutral density filters before being sent onto a narrow-bandpass dielectric filter with ~ 1.8 nm full-width transmission at 1029.45 nm. Upon spectral filtering of the pump beam, the remaining amplified signal beam is reflected off the filter either to an optical spectral analyzer (0.1 nm resolution) or toward a silicon photodiode. The optical losses generated by each attenuator between the OPAs and the detection were tracked to enable an energy calibrated measurement of the spectral behavior of the amplifier.

2. Results and discussion

The spectrally integrated gain was first measured by directing the beam toward the photodiode detector. We define the gain as the ratio of the measured signal at the photodiode with no pump to the signal measured with a set value of pump energy. For this measurement, care was taken to subtract the small leftover contribution from the pump beam – leaking from the narrow-bandpass filter – to the overall photodiode signal to provide an accurate value of the gain experienced by the signal beam. We measure a gain as high as 200 (Fig. 2 – left, black curve) and observe an exponential growth of the gain (Fig. 2 – left, red fit curve) in this proof-of-principle experiment. Equipping the COPA crystals with AR-coating or using a single-stage COPA with a contiguous 4-cm-long crystal would result in a drastic increase in optical gain. We estimate that owing to the Fresnel losses alone, the maximum pump energy is reduced from 7 mJ incident on the first OPA crystal to < 2.8 mJ incident on the last COPA crystal. The total seed

energy for this experiment was – as mentioned above – set to 320 μJ , yet only a ~ 0.25 nm spectral band centered around 1031.25 nm of these 5 nm bandwidth pulses effectively seeds the parametric amplifier. The in-band energy was estimated to be 2.3 μJ and therefore the amplified energy at the signal wavelength to be 460 μJ .

Fig. 2. (Left) Measured spectrally integrated optical gain at the output of the 4-stage cascaded optical parametric amplifier showing gain up to 200 at $1 \text{ J}/\text{cm}^2$ fluence incident on the input crystal; (right - top) measured seed (grey curve, shaded area) and amplified (black curve) spectra at the output of the amplifier for a pump fluence of $1 \text{ J}/\text{cm}^2$, showing up to 10 cascaded orders. Numerical simulations considering Fresnel reflections (blue vertical lines) show qualitative agreement with experimental data. The experimental spectra have been normalized to their respective energy content to provide qualitative spectral energy density value for each cascaded peak; (right - bottom) Numerical simulation using 100 times higher in-band seed energy as compared to the current setup in a single 4 cm-thick crystal without Fresnel losses. All other parameters were identical.

The spectral behavior of the parametric amplifier was also investigated (Fig. 2 – top right). For the maximum fluence we allowed on the COPA crystals ($1 \text{ J}/\text{cm}^2$) and the maximum seed that was available at the time of the experiment, we observed a total of up to 10 cascaded peaks separated by ~ 1.8 nm – corresponding to the energy of 516 GHz frequency idler photons. These spectral peaks, with spectral width of ~ 0.25 nm, were both blue and red-shifted with respect to the seed wavelength and extended from 1020 to 1040 nm. We observed no spatial beam break-up despite the numerous cascaded cycles undergone by the pump photons. Since the detection system was arranged to permit energy calibration of the spectra, we retrieve the spectral energy density of each peak and find that the maximum spectral energy density is as high as 10-100 $\mu\text{J}/\text{nm}$ for the first 6 peaks. Numerical simulations considering simultaneous nonlinear evolutions of IR and THz radiation were performed that qualitatively match the experimental data and revealed a computed conversion efficiency from near-IR pump to THz-frequency wave of $5 \times 10^{-3}\%$. Further simulations were performed for a single 4 cm-thick crystal devoid of the Fresnel losses present in the experiment (Fig. 2 – bottom right) while leaving all pump parameters unchanged. The seed energy was, however, scaled by two orders of magnitudes, which resulted in a dramatic increase in cascading with favored red-shifting and a conversion efficiency of $4 \times 10^{-2}\%$. Reducing the temperature of the crystal to 100 K showed further scaling of the conversion efficiency to 1.6%.

3. Conclusion and future work

We have demonstrated and characterized the first implementation of a cascaded optical parametric amplifier (COPA), specifically designed to enhance frequency down-conversion from near-IR to THz frequency via favored cascading. Experimental measurements confirm that this proof-of-principle implementation resulted in cascading in the optical spectrum and high gain at limited peak intensity. Future work includes the direct characterization of the generated THz-wave and management of the strong room-temperature absorption of the THz wave.

4. References

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