Cascaded second-order processes for the efficient generation of narrowband terahertz radiation

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Abstract

The generation of high-energy narrowband terahertz radiation has gained heightened importance in recent years due to its potentially transformative impact on spectroscopy, high-resolution radar and more recently electron acceleration. Among various applications, such terahertz radiation is particularly important for table-top free electron lasers, which are at the moment a subject of extensive research. Second-order nonlinear optical methods are among the most promising techniques to achieve the required coherent radiation with energy >10 mJ, peak field >100 MV/m, and frequency between 0.1 and 1 THz. However, they are conventionally thought to suffer from low efficiencies $<\sim 10^{-3}$, due to the high ratio between optical and terahertz photon energies, in what is known as the Manley-Rowe limitation. In this paper, we review the current second-order nonlinear optical methods for the generation of narrowband terahertz radiation. We explain how to employ spectral cascading to increase the efficiency beyond the Manley-Rowe limit and describe the first experimental results in the direction of a terahertz-cascaded optical parametric amplifier, a novel technique which promises to fully exploit spectral cascading to generate narrowband terahertz radiation with few percent optical-to-terahertz conversion efficiency.

1. Introduction

The past decades have seen an increasing interest of the scientific community in the production of terahertz radiation for various purposes, some of the main ones being linear spectroscopy [1] and imaging [2]. An important reason for this application is that the terahertz frequency matches the low-energy excitations in solids and rotational spectra of liquids and gases. The non-ionizing nature of terahertz radiation has also made it an interesting prospect as a substitute for X-rays (T-Rays) in pump-probe type experiments [3-4] and for imaging. The distinct terahertz spectral signatures of several materials make it amenable for their identification.

Terahertz radiation is defined as the electromagnetic radiation whose frequencies lie between the infrared and the microwave spectral regions, i.e., between 0.3 THz and 3 THz, corresponding to wavelengths between 1 mm and 0.1 mm, as defined by the International Telecommunication

Union (ITU) [5]. This official definition of terahertz radiation is often extended to also include the 0.1-10 THz frequency range.

With growing interest in nonlinear spectroscopic applications, extensive research into developing intense single-cycle terahertz radiation was conducted in the recent years [6]. However, there has not been a lot of effort devoted to developing highly energetic *narrowband* terahertz sources. The past few years, however, have seen the conception of and initial experiments towards compact, table-top free-electron lasers (TTFELs) which promise to produce ultrashort, highly energetic coherent X-ray pulses [7]. In this direction, acceleration of electrons driven by terahertz radiation has been demonstrated experimentally [8].

In conventional X-ray free-electron lasers (FELs) such as Flash, Flash 2, LCLS and LCLS-II, Sacla, Fermi etc., electrons are accelerated with a radio-frequency (RF) accelerator. The typical RF wavelength used is on the order of 10 cm, and the total length of the accelerator is on the order of 100 m. The minimum length of the accelerator is primarily limited by the maximum permissible electric field (~ 10 MV/m), or acceleration gradient in the accelerating structures, which in turn is limited by material damage. It has been found empirically that the damage threshold increases with the square root of the frequency and the inverse of the fourth root of the pulse duration of the accelerating field [9]. Consequently, terahertz drivers can be of significantly larger field strengths compared to RF counterparts, while still possessing large enough wavelengths to support electron bunches of pC-level charge in the accelerating field. In a TTFEL, the acceleration of electrons may be achieved with a high-field, multi-cycle terahertz beam (~ 100 cycles) in a waveguide which shrinks the size of such an accelerator to ~ 1 m. The high field gradients help to overcome the effects of electron-electron Coulomb repulsion (or space-charge effects), allowing electron bunches to be compressed to shorter durations than in traditional FELs resulting in electron beams of superior quality. However, in order to achieve electron acceleration up to ~ 10 -100 MeV, narrowband, multi-cycle (~ 100 optical cycles) coherent terahertz radiation with energy > 10 mJ, peak field > 100 MV/m, frequency between 0.1 and 1 THz is required [10]. The necessity of such a large terahertz field has triggered a widespread effort towards developing new techniques for the generation of terahertz radiation with high energies and efficiencies. In addition, nonlinear spectroscopic applications necessitate the development of efficient and robust high-field terahertz sources [11]. At the outset, we would like to emphasize the focus of this article on *narrowband* or *multi-cycle* terahertz radiation.

There exist several techniques for the production of terahertz radiation, namely solid-state devices, vacuum electronic devices, quantum cascade lasers (QCLs) and optical techniques [11-12]. Solid-state devices like photoconductive switches [13] or antennas can be used for both generating and detecting terahertz radiation. While being efficient, they can be challenging to scale up in energy. Vacuum electronic devices like gyrotrons typically produce sub-terahertz radiation [14] and can even surpass 1 THz but with low peak power [15], while FELs, which can also produce terahertz radiation, are expensive, large-scale facilities. QCLs are efficient at high terahertz frequencies but are limited to very low peak powers.

Nonlinear optical techniques are based on frequency down-conversion of visible or infrared laser radiation [16-17]. They are currently under development and are among the most promising techniques to generate the large peak fields and pulse energies at THz frequencies which are necessary for TTFELs. The basic idea of nonlinear optical techniques for terahertz generation is the conversion of photons with high energy (on the order of $\sim$ eV) into terahertz photons with lower energies ($\sim$ meV) through second-order nonlinear optical processes. The crystals that are typically used for terahertz generation are lithium niobate, LiNbO_3 (LN) [18], lithium tantalate, LiTaO_3 (LT), gallium arsenide (GaAs) [19], gallium phosphide (GaP) [20], gallium selenide (GaSe), zinc telluride (ZnTe), cadmium selenide (CdSe) and organic materials such as DAST. An efficient means to achieve narrowband phase-matching consists of poling the crystal [21] or orientation-patterning it [22] along the beam propagation axis, thereby inducing a periodic modulation of the effective second-order nonlinearity that allows quasi-phase matching.

Due to the large ratio between optical and terahertz photon energies, a 1:1 photon conversion efficiency between each optical pump photon and each terahertz photon (both interacting with a third photon at the sum or difference frequency of the other two photons, i.e., in the optical domain) would correspond to only a $\sim 10^{-3}$ energy conversion efficiency (Manley-Rowe limit) for 300 THz to 0.3 THz conversion. To achieve higher efficiencies, the optical photons each need to produce more than one terahertz photon, which requires exploiting cascading effects [23-24]. However, the realization or proposal of physical implementations that can feasibly produce significant cascading to achieve very high conversion efficiencies for multi-cycle terahertz generation is still an outstanding problem. The problem is further compounded by the requirement of long terahertz pulse durations, comprising hundreds of cycles, which limits the permissible optical pump intensity due to laser fluence-induced damage limitations. While $>1\%$ optical-to-terahertz conversion efficiencies have already been achieved for single-cycle terahertz pulses [25-27], the highest conversion efficiency achieved so far with multi-cycle pulses in the terahertz range (0.5-THz) is 1.2×10^{-3} [28]. This result was achieved through optical rectification (OR) of short, broadband Ti:sapphire pulses, where the nonlinear interaction occurred between two optical frequencies, 0.5-THz apart from each other, contained within the spectrum of a single incident pump laser pulse. Although attractive for its simplicity, the use of a single short pulse as a driver for multi-cycle terahertz generation is not ideal in several important ways, which will be discussed in section 2. A more optimized configuration consists of using two separate pulses, allowing independent control of the frequencies, bandwidths and pulse-durations of the two interacting spectral components, as is often done for difference-frequency generation (DFG), optical parametric oscillation (OPO), and optical parametric amplification (OPA). Of these, OPOs are unlikely to permit significant cascading due to limitations imposed by the cavity resonance condition. A new, highly promising technique using a two-pulse configuration for the efficient generation of desired terahertz pulses is the terahertz cascaded OPA (THz-COPA), which has been theoretically shown to reach up to a few percent optical-to-terahertz conversion efficiency [29]. The feasibility of this technique has been demonstrated in first proof-of-principle experiments [30-34].

This paper provides a review of past developments of optical frequency conversion techniques employing second-order nonlinear processes for the efficient production of multi-cycle coherent terahertz radiation, with particular focus on our recent work on cascaded OPAs for efficient terahertz generation.

2. Second-order schemes for terahertz generation

The conversion of optical frequencies into multi-cycle terahertz radiation can be achieved with different techniques, which are pictorially represented in Fig. 1. The underlying process is always the frequency mixing of photons at two frequencies, ν_1 and ν_2 , generating photons at their difference frequency, $\nu_1 - \nu_2$, which lies in the terahertz region. The first two techniques are cavity based and parametric, in which one of the two optical frequencies is either not present at the input or has a much lower number of photons than the other one. Fig. 1a illustrates the OPO, also known in the literature as terahertz parametric oscillator (TPO), where the pump at ν_1 transfers its energy to both photons at ν_2 and to photons at the terahertz frequency $\nu_1 - \nu_2$, which are both stimulated by optical noise and whose coherence is built up by means of the cavity [23,35-40]. Fig. 1b illustrates the injection-seeded TPO (is-TPO), which is similar to the TPO but with seed injection at ν_2 [36,41]. The other four techniques are single-pass methods, i.e., they do not use a cavity. Fig. 1c illustrates DFG, or inter-pulse DFG, which consists of the interaction of two distinct pulses at ν_1 and ν_2 with comparable number of photons [24,42-43]. In OR (Fig. 1d), also known as intra-pulse DFG, the interacting optical frequencies ν_1 and ν_2 are co-present in the injected broadband pulse [28, 44-46]. The last two techniques are OPAs: single-pass parametric techniques in which the number of pump photons at ν_1 is initially much larger than the number of

photons of the injected seed. The seed can be either in the terahertz region, at $\nu_1 - \nu_2$ (THz-seeded OPA, Fig. 1e) [47] or in the optical region, at ν_2 (optically seeded OPA, Fig. 1f), also known as injection-seeded terahertz parametric generator (is-TPG) [29-34, 48-51].

Fig. 1: Pictorial representation of the second-order nonlinear optical methods for terahertz generation, as described in the text. In each process, the full colors represent the input pulses, and the empty ones represent their growth due to amplification. The blue pump pulses at frequency ν_1 and the red signal pulses at ν_2 interact and generate the terahertz idler pulses (black) at $\nu_1 - \nu_2$. In (a) and (b) the curved mirrors represent resonant cavities. In (c), the green pulse contains both ν_1 and ν_2 frequencies.

The method that has experimentally given the highest conversion efficiency so far is OR, as mentioned in the introduction. Carbajo *et al.* [28] used an amplified Ti:sapphire laser and studied the dependence of the optical-to-terahertz conversion efficiency towards various parameters, including the input intensity, bandwidth and temperature. The highest achieved efficiency was 1.2×10^{-3} at 100 K (Fig. 2).

Fig. 2. Influence of pump pulse format in OR (crystal length 5 mm; poling period 212 μm ; temperature 100 K) measured in [28]. (a) Extracted efficiency as a function of pump intensity for bandwidths ranging from 3.4 to 10 nm; (b) efficiency as a function of pump bandwidth for a pump intensity of 50 GW/cm^2 ; (c) terahertz energy as a function of pump energy and intensity. Figure taken from [28].

OR is the simplest of the described methods, because it requires only one broadband pulse and a crystal (no cavity, no synchronization, no mode matching between beams are necessary). This method, however, is not optimized in several ways. First, the difference in group-velocity between terahertz and optical frequencies causes the short pump-pulse envelope (0.1-1 ps) to walk-off quickly from the generated long terahertz radiation (10-100 ps). Second, since the available aperture of nonlinear crystals is usually limited, damage caused by high peak intensities of short pulses limits the pump energy which can be used in the interaction. Finally, the broadband spectral content of the short pulses is not well tailored for the generation of a single, narrow-bandwidth spectral component comprising the multi-cycle terahertz pulse. These drawbacks can be avoided when using two independent inputs, like in OPAs.

Tripathi *et al.* [47] pursued a combination of both OPA techniques (is-TPG and THz-seeded OPA), in a two-stage setup and demonstrated seeding of a terahertz OPA by terahertz radiation for the first time. In their system, the first stage consisted of an is-TPG setup, in which a 14-mJ, 1064-nm pump interacts with a 5.5-mW, 1072-nm continuous-wave seed to produce 12.1-nJ of 2-THz radiation in an LN crystal. In the second, terahertz-seeded OPA stage, the optical pump at 1064 nm (7.2 mJ) interacted with the terahertz wave, which had been previously generated, in a second LN crystal at room temperature. The authors studied the output energy behavior versus input terahertz energy (Fig. 3). The gain was high when the injected seed was low, but tended to decrease to unity or less when the injected seed energy was $>1\text{nJ}$. The maximum achieved efficiency was $\sim 2 \times 10^{-7}$.

Fig. 3 (a) Input/output characteristics of the terahertz amplification system in [47]. (b) Gain plotted as a function of input terahertz energy. Figure taken from [47].

3. Cascading in second-order processes and cascaded OPA for terahertz generation

Cascading of a second-order process, illustrated in Fig. 4, is a regime in which a first 3-photon interaction (0) triggers a second 3-photon interaction (1) such that two of the photons from (0) generate a photon at a new frequency in (1). The second interaction in turn uses the new frequency to trigger a third interaction (2) which triggers a fourth interaction (3), and so on, generating new frequencies each time. More precisely, if the frequencies of the photons in (0) are $\nu_{1,2,3}$, satisfying $\nu_3 = \nu_1 \pm \nu_2$, then the three frequencies in (1) will be $\nu_{2,3,4}$, satisfying $\nu_4 = \nu_2 \pm \nu_3$. All interactions must also simultaneously satisfy the phase-matching conditions to occur efficiently.

Fig. 4: Representation of cascaded second-order nonlinear processes. Each process (0, 1, 2, 3...) involves three photons. Starting from (1), two frequencies are each time inherited from the previous process, whereby a new frequency is generated.

If the frequency of one of the three interacting photons, e.g., v_3 , remains constant in every one of the processes 0, 1, 2, 3..., the photons at this frequency can interact nonlinearly with the other photons several times. If v_3 lies in the terahertz region, and all other photon frequencies lie in the optical region, the terahertz photons can be effectively amplified with total quantum efficiency much higher than unity, thus breaking the Manley-Rowe efficiency limit.

Although the processes described in section 2 can all be driven into a cascaded regime, the extent of cascading that can be attained with each technique is not the same.

Cascading has been observed experimentally and/or studied theoretically in DFG [24, 42-43], OR [28], OPO [52], optically seeded OPAs [29-34, 51].

Saito *et al.* [52] demonstrated theoretically that OPOs can be cascaded. The photon conversion efficiency of a cascaded OPO for terahertz generation can slightly exceed the Manley-Rowe limit of 1 (1.086), as shown in Fig. 5. OPOs, though, are limited in the amount of cascading that can occur by the finite bandwidth of the cavity which does not allow for many cascaded orders.

Fig. 5. Theoretical optical-to-terahertz photon conversion efficiency as a function of the pump wavelength for various terahertz frequencies in an OPO. The efficiency exceeds unity, indicating that cascading is involved. Figure taken from [52].

For OR, on the other hand, the extent of cascading is limited by the interaction length, which is set by group-velocity walk-off. DFG, although it can be strongly cascaded, requires the presence of two narrowband pulses with wavelengths close to each other and similar number of photons. This would be possible with two synchronized laser gain materials with central frequency shifted by the desired terahertz frequency. Unfortunately, finding two such materials with the correct frequency difference in the gain bandwidth is challenging, particularly at high energies. OPAs, by contrast, can be strongly cascaded without such requirements on photon number, because a weak seed gets amplified by a strong pump, allowing greater asymmetry between the optical pulses and thus greater flexibility on the laser system requirements.

In a cascaded OPA for terahertz generation (THz-COPA), several OPA processes are co-present in the nonlinear crystal. A pump pulse at frequency v_p and a signal pulse at frequency v_s are injected into the nonlinear crystal. The strong pump then parametrically amplifies the weak injected signal and generates a third pulse at frequency $v_i = v_p - v_s$ (idler). The pulses at v_s (now

acting as a pump) and at ν_i interact again and generate the first cascaded order at $\nu_{c1} = \nu_s - \nu_i$. The process occurs again between ν_{c1} (new pump) and ν_i , so that photons at $\nu_{c2} = \nu_{c1} - \nu_i$ are generated. By repeating this process multiple times, several cascaded orders are generated coherently within the same crystal. Fig. 6 shows the photon energies in a THz-COPA.

Fig. 6: Photonic view of a THz-COPA. The photons at energies $h\nu_p$ (pump, blue arrow) and $h\nu_s$ (signal, green arrows) are injected into the nonlinear crystal and generate the idler at $h\nu_i$ (red arrows) in the terahertz range in OPA 0. The signal of OPA 0 acts as a pump in OPA 1 and amplifies the idler generated in OPA 0, while producing the first cascaded order at energy $h\nu_{c1}$ (black arrows, like all cascaded orders). The first cascaded order pumps again the idler in OPA 2 and so on.

Significant cascading can only occur if multiple cascaded OPA processes remain simultaneously within the phase-matching bandwidth of the crystal, a condition that is possible if the energies of all photon triplets are close to each other, i.e., if $\nu_p \gg \nu_s \gg \nu_{c1} \gg \nu_{c2} \dots \gg \nu_i$. Such is the case with the THz-COPA, where the frequency ν_i lies in the terahertz range, and all other frequencies lie in the optical range.

Note that cascading effects in OPAs have been observed in the more conventional case of visible or infrared OPAs [53-54]. In this case, however, the large frequency disparity of the interacting photons means that the cascaded interactions occur outside of the phase-matching bandwidth and produce photons far from the targeted frequency, and are thus considered parasitic. In addition, some signatures of cascading in an is-TPG, i.e., an OPA for terahertz generation, have been observed in [51].

We have for the first time fully developed the concept and theoretically analyzed the THz-COPA [29] and performed preliminary experiments [30-34]. We have studied the case in which ν_p , ν_s , ν_{c1} , $\nu_{c2} \dots$ are in the infrared ($\lambda \sim 1 \mu\text{m}$, $\nu \sim 290 \text{ THz}$), and ν_i is at 0.5 THz ($\lambda \sim 600 \mu\text{m}$). The first process, OPA 0 in Fig. 6, is both pumped and seeded by infrared photons, which generate the idler in the terahertz range. The amplified signal of OPA 0 acts as a pump in OPA 1, which is seeded by the terahertz radiation generated in OPA 0. The same is true for all successive OPAs. Only OPA 0 is seeded optically, all other OPAs are seeded by terahertz. In the experiment, a stretched Yb:KYW regenerative amplifier [55], whose spectral range is 1026-1032 nm foot-to-foot (-20 dB), was split into two parts: the first one, $\sim 40 \mu\text{J}$, of which only $\sim 400 \text{ nJ}$ were within the gain bandwidth at 1031.2 nm wavelength, was used as a seed for the THz-COPA; the other part, at a wavelength of 1029.45 nm, was first amplified up to 15 mJ in a cryogenically cooled 4-pass amplifier [56] and then used as a pump for the THz-COPA. The pump and seed pulse durations were $\sim 170 \text{ ps}$.

When pump and seed were temporally and spatially overlapped in a periodically poled LiNbO₃ (PPLN) crystal with 212- μm poling period, quasi-phase-matched for the generation of 0.5-THz radiation, the seed energy was parametrically amplified. Fig. 7 shows the measured optical gain at the signal wavelength in one case without cascading (black line) when the pulses interacted in a single crystal, in the other case (blue line) with cascading, which was obtained by using a combination of 2 consecutive crystals. At the pump fluence for which the cascading begins, the optical gain of the signal starts to saturate. The saturation of the signal gain is caused by the onset of additional nonlinear processes which generate cascaded orders, that compete with the signal for photons.

Fig. 7: Optical gain at the seed line of a THz-COPA versus pump fluence for a 2-cm PPLN crystal (black line) and for a combination of 2-cm + 1-cm PPLN crystals (blue line). The gain saturation of the signal corresponds to the onset of cascading, since the first cascaded order also starts to experience gain. The pump intensity was in the order of GW/cm^2 .

Fig. 8 shows a typical measured spectrum of the optical output of the THz-COPA compared with the pump alone. At least 5 cascaded orders at longer wavelengths than the pump and the seed can be clearly resolved in the spectrum, and further orders may be below the noise level of the spectrometer.

Fig. 8: Spectra of the pump (blue line) and of the optical output of the THz-COPA, when both pump and seed are present (black line), showing at least 5 cascaded orders.

We measured the spectrally-integrated optical gain, the ratio between the total output energy, (composed of amplified signal and generated cascaded orders and excluding the pump), and the input seed energy injected into the crystal. As shown in Figure 9, the optical gain varies exponentially with the pump fluence.

Fig. 9: The spectrally integrated optical gain, i.e., the total energy of amplified signal and all cascaded peaks, versus pump fluence, shows an exponential behavior.

These proof-of-principle experiments were performed with un-optimized crystal lengths and at room temperature, where absorption of the terahertz in the periodically-poled lithium niobate crystal was strong (absorption coefficient $\sim 7 \text{ cm}^{-1}$). As a result, terahertz radiation was detected with a pump-to-terahertz conversion efficiency of 1.5×10^{-5} that was low but in good agreement with our theoretical predictions. However, a conversion efficiency of few percent has been predicted for optimized crystal, temperature and optical parameters [29,57]. These first results therefore give confidence that the THz-COPA will signify an important technological leap forward for efficient generation of coherent multi-cycle terahertz radiation, and consequently for the construction of TTFELs producing fully coherent X-rays.

4. Conclusion

The generation of high-energy coherent narrowband terahertz radiation will acquire continued importance in optical physics due to its significance for the construction of TTFELs. In this paper, we reviewed the schemes implemented so far for multi-cycle terahertz generation using nonlinear optical techniques of the second order. Such techniques, though not yet fully developed, constitute the most promising tools to realize the required multi-mJ-level terahertz sources.

So far, a maximum optical-to-terahertz conversion efficiency of 1.2×10^{-3} has been achieved with OR. To achieve higher efficiencies, it is necessary to utilize cascaded processes, thus overcoming the Manley-Rowe limitation. The process that is most favorable for strong cascading using high-energy laser systems, that do not have sufficient bandwidth for direct DFG, is the optically-seeded THz-COPA. First experiments in this direction confirm that the principle works as predicted by theory, and simulations predict that optimized efficiencies for narrow-band terahertz generation of few percent should be possible. Therefore, currently existing laser systems producing 200-mJ of optical energy should be sufficient to produce the required 10-mJ-scale terahertz radiation needed to accelerate electrons to relativistic energies and to serve as TTFEL drivers.

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