Cascaded quadratic optical nonlinearities are well known for producing effective third-order nonlinear optical effects such as self-phase modulation or self-steepening. As a result, they have been extensively applied in areas such as mode-locking and pulse compression. In this article, a regime of cascaded quadratic nonlinearities involving highly phase-matched second-order interactions is introduced, which produce an effective third-order nonlinearity analogous to Raman shifting rather than the typical case of self-phase modulation. This results in a continuous red-shift of the optical pump frequency rather than spectral broadening. This phenomenon is particularly relevant to terahertz generation, where a continuous red-shift of the pump frequency resolves current issues of dispersion and laser-induced damage. In the absence of absorption or dispersion, the presented method results in optical-to-terahertz energy conversion efficiencies that approach 100%, which is not possible with conventional cascaded difference-frequency generation. Designs of aperiodically poled lithium niobate structures are presented, which result in energy conversion efficiencies of 35% even in the presence of dispersion and absorption. The presented work thus addresses an important bottleneck in terahertz generation, which paves the way for the development of compact particle accelerators, X-ray free-electron lasers, advanced electron-beam diagnostics, and various experiments in condensed-matter physics and chemistry.

1. Introduction

Nonlinear optical effects have enabled the generation of light frequencies from the microwave to the X-ray regions of the electromagnetic spectrum. When one nonlinear optical effect initiates further nonlinear optical effects and their evolution is coupled, they may be collectively referred to as cascaded nonlinear optical effects. An inexhaustive list includes refs. [1–8].

In the context of cascaded nonlinear optical effects, cascaded second-order processes in quadratic media have garnered significant interest. [7] Of particular interest, has been the use of phase-mismatched second harmonic generation (SHG) to produce controllable effective third-order nonlinear optical effects on the fundamental wave. [1–6] An extensive set of applications ranging from soliton pulse compression to frequency comb generation and mode-locking [7, 9] have been realized using these phase-mismatched second-order processes.

In contrast, another class of cascaded second-order nonlinearities occur when a large number of highly phase-matched second-order processes evolve in concert. This is of great interest to terahertz generation, [10–14] where the repeated energy down-conversion of an optical or near infrared (NIR) pump photon to multiple terahertz photons via cascaded difference-frequency generation (DFG) was proposed to surmount the single-photon conversion efficiency or Manley-Rowe limit. [15] The approach has already resulted in the highest optical-to-terahertz energy conversion efficiencies [16, 17] to date.

The generation of terahertz radiation is accompanied by a dramatic broadening of the optical pump spectrum. Energy conservation requires the center frequency of the optical spectrum to be red shifted in the case of efficient terahertz generation. However, due to dramatic spectral broadening, both red (Stokes) and blue shifted (Anti-Stokes) components are observed in the emergent optical spectrum. Therefore, cascaded DFG in the terahertz range has often been considered to effectively be a self-phase modulation (SPM) effect. [2, 18, 19]

While cascaded DFG promises to yield optical-to-terahertz conversion efficiencies of several percent, [11, 12] further enhancement of conversion efficiencies are inhibited by the aforementioned spectral broadening. This is due to the following reasons. First, there is an annihilation of terahertz photons via the generation of Anti-Stokes spectral components. Second, the spectral broadening impedes further terahertz generation due to dispersion. [11] Third, the spectral broadening causes an
increase in the pump intensity which exacerbates laser-induced damage.\cite{14}  

In this paper, we introduce a new regime of cascaded second-order nonlinearities, particularly relevant to narrowband terahertz generation which addresses these issues. We show that in conditions satisfying broadband phase matching, cascaded DFG with an optical pump spectrum comprised of a series of narrowband lines separated by the desired terahertz frequency (henceforth referred to as a multi-line format) is equivalent to a Raman shifting process rather than an SPM process. In the time domain, the multi-line format is equivalent to a sequence of short pulses separated by the time period of the desired terahertz wave\cite{11} (2 ps for 0.3 THz). In contrast to intra-pulse DFG, a sequence of short pulses coherently circumscribes walk-off between the optical and terahertz waves and coherently boosts conversion efficiency. Such a sequence of short pulses can be generated by superposing a series of long optical pump pulses ($r = 200$ ps each corresponding to the linewidth), each with a different center frequency. The various center frequencies of these long pulses are separated by the desired terahertz frequency (e.g., 300, 300.5, 301 THz). Furthermore, this sequence of short pulses repeats with the repetition rate of the laser, which is expected to be in the kHz range (or every milli-second). For an illustrative example, the reader is referred to Figures S5 and S6, Supporting Information.

In this regime, a continuous red shift of the pump frequency accompanied by spectral narrowing of the pump rather than spectral broadening is observed. In the limit of zero absorption and dispersion, this produces optical-to-terahertz energy conversion efficiencies approaching 100%. To illustrate the feasibility of the approach under practical conditions, we numerically design an aperiodically poled Lithium Niobate crystal pumped by the aforementioned multi-line format centered at a pump wavelength, $\lambda_0 = 1.3$ $\mu$m, and cryogenic temperature of $T = 10$ K to produce conversion efficiencies of $\approx 35\%$.

2. Analytic Proof of Raman Shifting

We begin with an analysis of normalized equations (see Supporting Information) for cascaded DFG of terahertz radiation in Equations (1) and (2).

\[
\frac{dA_{op}(\omega, z)}{dz} = -j\omega \chi^{(2)}_{eff} \int_0^\infty A_{op}(\omega - \Omega, z) A_{THz}(\Omega, z) e^{i\Delta k z} d\Omega
\]

\[
\frac{dA_{THz}(\Omega, z)}{dz} = -\frac{\alpha(\Omega)}{2} A_{THz}(\Omega, z)
\]

\[
\frac{dA_{op}(\omega - \Omega, z)}{dz} = -j\omega \chi^{(2)}_{eff} \int_0^\infty A_{op}(\omega - \Omega, z) A_{THz}(\Omega, z) e^{i\Delta k z} d\Omega
\]

Equation (1) describes the evolution of the normalized spectral amplitude, $A_{op}(\omega, z)$, of the optical pump at angular frequency $\omega$ in a quadratic nonlinear medium with an effective second-order susceptibility $\chi^{(2)}_{eff}$. The use of normalized equations make it easier to obtain quantities such as the average frequency shift, as shall be shown subsequently. The first term on the right hand side of Equation (1) represents the generation of $A_{op}(\omega, z)$ by sum-frequency generation (SFG) between optical and terahertz spectral components, while the second term represents DFG between optical and terahertz components. The phase-mismatch is given by $\Delta k$. Equation (2) represents the evolution of the terahertz spectral component, $A_{THz}(\Omega, z)$ at angular frequency $\Omega$. The first term in Equation (2) represents attenuation of the terahertz spectral component at a rate given by the absorption coefficient $\alpha(\Omega)$. The second term in Equation (2) represents DFG processes between all spectral components of the optical pump. In the absence of absorption, Equations (1) and (2) conserve energy per unit area (or fluence) to unity under present normalization conditions. As a result, $X_{eff}^{(2)} = \chi^{(2)} F_{pump}(2c'\epsilon_0 n_{THz} n_{op}(\omega_0)^2)^{1/2}$, where $\chi^{(2)}$ is the conventional second-order susceptibility in SI units. $F_{pump}$, $n_{THz}$, $n_{op}(\omega_0)$ are the pump fluence, terahertz refractive index and optical refractive index at the central angular frequency, $\omega_0$, of the pump respectively. In Equation (1), third-order nonlinearities are not considered since our previous work\cite{11} has revealed their negligible effects on multi-line pump formats in lithium niobate. Equations (1) and (2) may also be interpreted in the time domain as shown in ref. [20]. However, for the remainder of this paper, we will retain the frequency-domain viewpoint for the sake of consistency.

We will analytically show that Equations (1) and (2) are equivalent to a Raman shifting process. Using multiple-scale analysis\cite{21,22} or adiabatic elimination,\cite{23} Equations (1) and (2) can be reduced to a single self-consistent equation for $A_{op}(\omega, z)$ in Equations (3)–(5) for the case of perfect and broadband phase matching, that is, $\Delta k = 0$ ($V \omega$) and finite absorption, that is, $\alpha > 0$ (see Supporting Information).

\[
\frac{dA_{op}(\omega, z)}{dz} = -\int_0^\infty \frac{2\omega_0 \Omega F_{pump}(2c'\epsilon_0 n_{THz} n_{op}(\omega_0)^2)^{1/2}}{\alpha(\Omega)} (A_{op}(\omega - \Omega, z) R(\Omega, z)) - A_{op}(\omega + \Omega, z) R^*(\Omega, z) d\Omega
\]

\[
R(\Omega, z) = \int_0^\infty A_{op}(\omega' + \Omega, z) A_{op}^*(\omega', z) d\omega'
\]

By performing an inverse Fourier transform on Equation (3), it is reduced to the familiar Raman-scattering-like term in the time domain in Equation (5).

\[
\frac{dA_{op}(t, z)}{dz} = -\chi''(\omega) A_{op}(t, z) A_{op}(t, z)
\]

In Equation (5), $\chi''(\omega)$ is used to denote the imaginary part of the Raman gain spectrum and corresponds to the inverse Fourier transform of the first term within the integral of Equation (3).

From Equation (3), we notice that the first term inside the integral on the right-hand side is purely real. This is contrary to the situation for phase-mismatched cascaded SHG, where it is purely imaginary. The latter results in spectral broadening and is tantamount to SPM-like effects. Here, the purely real nature of the first term in Equation (3) delineates a transfer of energy from the optical to the terahertz spectrum. Naturally, the rate of this transfer is inversely proportional to the absorption coefficient.
Equation (3) is of the same form as the soliton frequency-shift equation presented in ref. [24].

We can further verify this by evaluating the rate of change of the average angular frequency of the pump spectrum, $\langle \omega \rangle = 2\pi f$ (see Supporting Information) using Equation (3).

$$\frac{d\langle \omega \rangle}{dz} = \frac{-8\omega_0 \chi^{(2)}_0}{\alpha} \int_{0}^{\infty} \Omega^2 |R(\Omega, z)|^2 d\Omega$$  \hspace{1cm} (6)

Since the integral in Equation (6) is obviously $>0$, it is clear that the $d\langle \omega \rangle/dz < 0$, which is proof that there is a red shift of the average pump frequency.

Next, we evaluate Equation (6) at $z = 0$ for the case of a multi-line pump spectrum containing $N_w$ lines of equal amplitude as shown in Equation (7). The linewidth of each line is assumed to correspond to a duration $\tau$.

$$A_{op}(\omega) = \left( \frac{r}{N_w \sqrt{2\pi}} \right)^{1/2} \sum_{n=0}^{N_w-1} e^{-\frac{\omega - \omega_0 - n\Omega}{\tau}}$$  \hspace{1cm} (7)

In Equation (7), $\Omega$ is the terahertz angular frequency to be generated. The expression for $d\langle \omega \rangle/dz$ is presented below in Equation (8).

$$\frac{d\langle \omega \rangle}{dz} \bigg|_{z=0} = \frac{-8\pi^{1/2} \omega_0 \Omega^2 \chi^{(2)}_0}{\alpha \tau} \left( \frac{N_w - 1}{N_w} \right)^2$$  \hspace{1cm} (8)

From Equation (8), it is evident that the rate of change of $\langle \omega \rangle$ is negative with a magnitude that increases with $N_w$, for a constant pump fluence. A corollary of this is that the average blue-shift is larger for smaller $N_w$. For the case of cascaded optical parametric amplification, $N_w \approx 1$ and therefore $d\langle \omega \rangle/dz \bigg|_{z=0} \approx 0$, which is consistent with refs. [12, 13]. In fact, simulating different input pump spectra using Equation (3) predicts effects which are qualitatively identical to a direct numerical solution of Equations (1) and (2) (Supporting Information). Now, consider an input pump spectrum of the form given by Equation (9).

$$A_{op}(\omega) = \left( \sum_{n=0}^{N_w-1} e^{-\frac{\omega - \omega_0 - n\Omega}{\tau}} \right)^{-1/2} \left( \frac{r}{\sqrt{2\pi}} \right)^{1/2} \times \left( \sum_{n=0}^{N_w-1} e^{-\frac{\omega - \omega_0}{\tau}} \right)^{-1}$$  \hspace{1cm} (9)

In Equation (9), an amplitude that decays as $e^{-\omega^2/N_w^2}$ is considered. A finite sum of $2N$ terms is used to account only for positive frequencies greater than the terahertz frequency. Using Equation (9), we obtain a value for $d\langle \omega \rangle/dz$ given by Equation (10).

$$\frac{d\langle \omega \rangle}{dz} \bigg|_{z=0} = \frac{-8\pi^{1/2} \omega_0 \Omega^2 \chi^{(2)}_0}{\alpha \tau} \left( \sum_{n=0}^{N_w-1} e^{-\frac{\omega - \omega_0 - n\Omega}{\tau}} \right)^{-2} \times \left( \sum_{n=0}^{N_w-1} e^{-\frac{\omega - \omega_0}{\tau}} \right)^{-1/2}$$  \hspace{1cm} (10)

Equation (10) changes much more dramatically with $N_w$ and is always larger compared to Equation (8) (see Supporting Information). Therefore, it is preferable to use a multi-line format whose amplitudes follow Equation (9) rather than Equation (7).

3. Numerical Results

Having established the physical concept, we present simulation results for the case of multi-cycle or narrowband terahertz generation. To alleviate computational cost, we use a version of Equations (1) and (2) which approximate the optical spectrum as a series of discrete lines separated by the generated terahertz angular frequency, $\Omega$ (Supporting Information). In our initial set of simulations, we exclude dispersion and absorption to show that Raman-shifting will asymptotically approach perfect energy transfer as $N_w$ in Equation (9) increases. The evolution of the optical spectra in S1 units, $\eta_{in}$, is presented in Figure 1 for input pump spectra following Equation (9) for various values of $N_w$. A fluence four times below the damage threshold, that is, $F_{pump} = 0.35 \text{ J cm}^{-2}$ is considered. Additionally, $\chi^{(2)} = 2\pi \chi^{(2)}_0 = 213 \text{ pm V}^{-1}$ and $\tau = 200 \text{ ps}$. Here, $\chi^{(2)}_0$ is the bulk second-order susceptibility.

In Figure 2a, we depict the conversion efficiency, $\eta$. In Figure 2b, we numerically calculate the evolution of the center frequency, $f$. For $N_w = 1$ in Equation (9), we notice a significant blue shift in Figure 1a. The simultaneous red and blue shift results in spectral broadening, which is not desired as stated at the outset. In Figure 2a, we see that even in the absence of dispersion or absorption, the conversion efficiency reaches only a maximum value of $\approx 20\%$ at $z = 5 \text{ cm}$ and then drops. The case presented in Figure 1a is the case of conventional cascaded different frequency generation. Correspondingly, in Figure 2b, we see that there is an increasing red shift in the pump frequency up to $z = 5 \text{ cm}$. However, this red shift subsequently reduces and is accompanied by the reduction in conversion efficiency observed in Figure 2a.

In Figure 1b, $N_w = 5$. This results in a continuous red shift over a longer distance of $z \approx 7 \text{ cm}$ before spectral broadening accompanied by blue shift of the pump frequency begins to occur. This is reflected in the conversion efficiency, which rises to nearly $80\%$ in Figure 2a before declining. Since absorption is not considered here, the decline in efficiency corresponds to the circulation of energy between the terahertz and optical spectra. Figure 2b thus shows a reduction in the red shift of the pump frequency beyond $z = 7 \text{ cm}$.

Figure 1c,d depicts the evolution of optical spectra for the cases of $N_w = 10, 25$, respectively. The trend of continuous red shift and reduced spectral broadening and blue shift of the pump frequency is observed in these cases. The conversion efficiency in Figure 2a for these cases is correspondingly higher compared to the $N_w = 1, 5$ cases. For $N_w = 10$, a conversion efficiency as large as $90\%$ is seen at $z = 8.5 \text{ cm}$. However, beyond this distance, energy returns to the optical domain as evident by the reduction in the red shift of the pump frequency in Figure 2b. For $N_w = 25$, the optical spectrum not only exhibits a continuous red shift but also results in spectral narrowing, which is not seen as distinctly in the previous cases. The spectral narrowing is a consequence of the continuous energy transfer to the terahertz field as evident in Figure 2a. Naturally, an incessant red shift of the pump...
frequency is seen for this case in Figure 2b. We have also included efficiency and average frequency calculations for $N_w = 150$ in Figure 2. Based on these cases, it is evident that the conversion efficiency asymptotically approaches 100% as $N_w$ increases. In addition to the continuous red-shift, a continuous reduction in the peak intensity of the pump also occurs as can be seen in Figures S5 and S6 of Supporting Information.

4. Design of Aperiodically Poled Structures for Raman Shifting

We now demonstrate the feasibility of cascaded Raman shifting in conditions with both dispersion and absorption. For illustrative purposes, we assume Magnesium Oxide doped lithium niobate due to their availability in cm² aperture crystals. The crystal is assumed to be cooled to $T = 10$ K to minimize absorption. For a terahertz frequency $\Omega_0/(2\pi) = 0.5$ THz, the absorption coefficient is $\alpha = 0.25$ cm⁻¹. Material dispersion and absorption are the same as in ref. [11]. Only red shifting of the pump spectrum in the transparency window of Lithium niobate between $\lambda_0 = 1$ and $\approx 3 \mu$m is considered. We implemented a gradient-based optimization algorithm (see Supporting Information) to determine the poling period as a function of distance $z$. The resultant center frequency used for obtaining the poling period is, $f_p(z) = [2.3991 - 0.0591(z/5\text{ cm}) - 2.1285(z/5\text{ cm})^2 + 1.9987(z/5\text{ cm})^3] \text{ THz}$. For an input pump spectrum corresponding to $N_w = 5$ in Equation (9) and centered at $\lambda_0 = 1.3 \mu$m, we present results in Figure 3. The choice of $\lambda_0$ was based on the ease of optimization procedures and maximizing conversion efficiency.

In Figure 3a, we show the conversion efficiency for the case when dispersion (absorption is however non-zero) is excluded (black, dashed line). The conversion efficiency for the case of optimized aperiodic poling is depicted in the red curve. As is evident, almost perfect agreement is obtained. The spectral evolution for the aperiodically poled structure is presented in Figure 3b which depicts the desired continuous red shift as seen in the reducing center frequency, $f$, of the pump in Figure 3c. The inset in Figure 3c delineates the input optical spectrum at $z = 0$. In addition to the optimized aperiodic poling case, conversion efficiency for a periodically poled lithium niobate (PPLN) crystal with wavelengths $\lambda_0 = 1.03 \mu$m (blue) and $2 \mu$m (green) are also plotted in Figure 3a. The former represents a well developed laser technology while the latter represents a region where the group-velocity dispersion is $\approx 0$ for Lithium Niobate. Even in these cases, one obtains high conversion efficiencies of 10% and 25%, respectively. The drop in conversion efficiency occurs due to the phase mismatch that sets in as spectral broadening of the pump occurs.

Corresponding to the spectral evolution in Figure 3b for the aperiodically poled structure, a reduction in the time-domain peak intensity ($I_{\text{max}}$) of the pump for $z \leq 4$ cm is evident in Figure 4a. For $z > 4$ cm, an increase in $I_{\text{max}}$ occurs due to the onset of spectral broadening of the pump as evident in Figure 3b. For temporal snapshots of the optical pump, the reader is referred
Figure 3. Performance in the presence of dispersion and absorption at $T = 10$ K for lithium niobate phase matched for the generation of 0.5 THz. a) For optimized aperiodic poling (red), the conversion efficiency is approximately the same as the dispersion-free case (black, dashed). High conversion efficiencies can also be obtained using periodically poled structures with the pump centered at $\lambda_0 = 1.03 \, \mu\text{m}$ (blue) and $2 \, \mu\text{m}$ (green). b) Continuous red shift of the pump frequency in the case of optimized aperiodic poling, which validates its functionality. c) The numerically evaluated center frequency of the optical spectrum corresponding to (b). The input optical spectrum with $N_w = 5$ in Equation (9) is depicted in the inset.

Figure 4. a) The peak time-domain intensity of the optical pump as a function of distance corresponding to the spectral evolution presented in Figure 3b for the aperiodically poled structure. b) Corresponding maximum free-electron densities remain below the damage threshold value.

to Supporting Information. The corresponding maximum free-electron densities $N_{\text{max}}$ were calculated along the lines of ref. [14] and are depicted in Figure 4b as a function of $z$. The spatial evolution of $N_{\text{max}}$ follows the trends of $I_{\text{max}}$ from Figure 4a and remain below the laser-induced damage threshold density (red, dashed). The damage-threshold density is obtained from a single Gaussian pulse with $e^{-2}$ duration of $\tau = 200$ ps and a laser-induced damage fluence of 1 J cm$^{-2}$.[11]

When optical-to-terahertz conversion efficiencies reach levels as large as those simulated above, nonlinear optical effects due to the terahertz field are expected to become important. The consideration of these effects might indeed limit conversion efficiencies and would require further study, which is out of the scope of this paper. However, irrespective of such considerations, the ability to minimize spectral broadening of the pump and maximize red-shifting of the pump frequency is a powerful tool to significantly improve optical-to-terahertz conversion efficiencies.

In conclusion, we introduced a new regime of cascaded quadratic or second-order nonlinearity. We find that under broad-band phase matching conditions, cascaded difference frequency generation is akin to a Raman shifting process. In particular, a continuous red shift accompanied by complete energy transfer to the terahertz spectrum can be realized for multi-line input optical spectra in the absence of absorption or dispersion. We presented a design which can produce a continuous red shift in the presence of absorption and dispersion by an optimized aperiodically poled profile in lithium niobate. Conversion efficiencies as high as 35% can be achieved in this case. Furthermore, since the peak intensity of the optical pulse reduces as terahertz radiation is generated, laser-induced damage can be circumvented. The approach may be extended to materials such as Potassium Titanyl Phosphate (KTP), which have shown to possess higher damage threshold fluences.[26] Thus, a new physical mechanism for terahertz generation which can result in very high conversion efficiencies while alleviating limitations due to dispersion and laser-induced damage has been presented.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

Conflict of Interest

The authors declare no conflict of interest.

Keywords
electron beams, free-electron lasers, nonlinear optics, terahertz generation
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