Full system model for terahertz generation by optical rectification

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Why is high field THz important?


E_{peak} = 0.7 GV/m

Fleischer, PRL 107 (2011)

Wong, Opt. Exp. 21 (2013)

Palfalvi, Phys. Rev. STAB 17 (2014)
Method: Optical Rectification

- Most efficient method: >1% energy conversion efficiency\(^1\), \(\sim\) mJ THz pulse energy\(^2,3\)

\[
|E_{THZ}(\Omega)|
\]

- Intra-pulse difference frequency generation
- THz bandwidth proportional to optical pulse bandwidth
- Must satisfy phase-matching condition

\[
\mathbf{k}(\omega+\Omega)-\mathbf{k}(\omega)=\mathbf{k}(\Omega)
\]

A General case : Optical Rectification using Tilted Pulse Fronts

- Lithium Niobate : Large bandgap, high damage thresholds.

- THz refractive index $\sim 5$, optical group refractive index $\sim 2.25$ -> Phase matching?

- Intensity fronts tilted with respect to the propagation direction -> 'Tilted Pulse Fronts'

Optical Rectification: Cascading effects

- 100% photon conversion efficiency, energy conversion efficiency
  \[ \eta = \frac{0.3}{300} \approx 0.1\% \]

- Experimentally, energy conversion efficiency > 1% -> greater than 100% photon conversion efficiency

- Repeated down-conversion till phase-matching exists -> Cascading effects

\[ \omega - \Omega \]
\[ \omega - 2\Omega \]
\[ \omega - 3\Omega \]

Small energy change but large change in spectrum!

Motivation : Gap in Theory

• Predicted energy conversion efficiencies > 10%\[^5\], but experimentally \sim (2-3%). **Why?**

• Large spectral reshaping of the optical spectrum -> **Undepleted pump approximation valid?**

• Prior models: **Either 1-D or undepleted.**
Motivation : Requisites for a model

(1) Consider *spatio-temporal distortions* of ultrafast pulses

(2) Consider cascading, self phase modulation.

(3) Solution in 2-D.

Device sizes -> cm scale

*How can you model all this efficiently?*
Simulation Approach: Spatio-temporal distortions

- Propagate spectral component $E(\omega)$ through arbitrary optical setup ->
  Use dispersive ray pulse matrices

- Spatial profile $E(\omega,x,z)$, pre-calculated

$$E_{out}(\omega, x, z) = \sqrt{\frac{\sigma_{in}(\omega)}{\sigma_{out}(\omega, z)}} \cdot E_{in}(\omega, 0, 0)e^{-jk \left[ \frac{(x_0 - x_{out}(\omega, z))^2}{2q_{out}(\omega, z)} + x_{out}(\omega, z)x \right] - j[\phi_0(\omega, z) + \phi_1(\omega, z) + \phi_2(\omega, z) + \phi_3(\omega, z)]}$$

Simulation Approach: THz generation

Initial conditions

\[ E_{in}(\omega,x,0) = 0 \]

Back-propagate optical field from \( z = 0 \)

\[ E_{out}(\omega,x,z) = \left( \frac{\sigma_{in}(\omega)}{\sigma_{out}(\omega,z)} \right) \cdot E_{in}(\omega,0,0) \]

\[ \phi_0(\omega) = k \sum_{i=1}^{N} L_i n_i(\omega) \]

Material dispersion

\[ \sqrt{\frac{1}{A(\omega,z_0) + B(\omega,z_0) / q(\omega)_{in}}} = \frac{\sigma_{in}(\omega)}{\sigma_{out}(\omega,z_0)} e^{-j\phi_1(\omega,z_0)} \]

Angular Dispersion

\[ \phi_2(\omega,z_0) = \frac{k}{2} \left[ x_{in}(\omega) x_{in}(\omega,z_0) - x_{out}(\omega) x_{out}(\omega,z_0) \right] \]

Spatial Chirp

\[ -jk \left[ \frac{(x_0 - x_{out}(\omega,z))^2}{2q_{out}(\omega,z)} + x_{out}(\omega,z).x \right] e^{-j[\phi_0(\omega,z) + \phi_1(\omega,z) + \phi_2(\omega,z) + \phi_3(\omega,z)]} \]

Various other dispersive terms which arise from the setup

\[ \phi_3(\omega,z_0) = \frac{k}{2} \sum_{i=1}^{N} F_i(\omega)x_{outi+1}(\omega) \]
Simulation approach

Initial conditions

\[ P_{op}(\omega, x, z) = \varepsilon_0 \chi^{(2)}_{eff}(x, z) \int_{0}^{\infty} E_{op}(\omega + \Omega, x, z) E_{THz}^{*}(\Omega, x, z) d\omega \]

+ \varepsilon_0 \chi^{(2)}_{eff}(x, z) \int_{0}^{\infty} E_{op}(\omega - \Omega, x, z) E_{THz}(\Omega, x, z) d\omega

\[ \begin{align*}
& - \frac{2k_z \varepsilon_0 c^2}{\omega^2} \mathcal{F}\left\{ \frac{\varepsilon_0 \omega_0 n(\omega_0)^2 n_2(x, z)}{2} |E_{op}(t, x, z)|^2 E_{op}(t, x, z) \right\} \\
& - \frac{2k_z \varepsilon_0 c^2}{\omega^2} \mathcal{F}\left\{ j \frac{\varepsilon_0 \omega_0 n(\omega_0)^2 n_2(x, z)}{2} |E_{op}(t-t', x, z)|^2 \otimes h_r(t') E_{op}(t, x, z) \right\}
\end{align*} \]

Cascading effects

THz + Optical Sum Frequency Generation

Self Phase Modulation

Stimulated Raman Scattering
Simulation Approach: Solving the wave Eqs.

\[ \nabla^2 E(\Omega, x, z) + k^2(\Omega)E(\Omega, x, z) = \frac{-\Omega^2}{\varepsilon_0 c^2} P^{(2)}(\Omega, x, z) \]  

THz

\[ \nabla^2 E(\omega, x, z) + k^2(\omega)E(\omega, x, z) = \frac{-\Omega^2}{\varepsilon_0 c^2} P^{(2)}(\omega; \Omega, x, z) \]  

Optical

Fourier Decomposition

\[ \frac{\partial A(\Omega, k_x, z)}{\partial z} = -\frac{\alpha(\Omega)}{2} A(\Omega, k_x, z) - \frac{j\Omega^2}{(2k_z \varepsilon_0 c^2)} P^{(2)}(\Omega, k_x, z)e^{jk_z z} \]

\[ \frac{\partial \phi_{op}(\omega, k_x, z)}{\partial z} = -\frac{\omega^2}{\varepsilon_0 c^2} P_{op}(\omega, k_x - k_{x0}, z) e^{j \left( k_{z0}(\omega) + \frac{k_{x0}(\omega)k_x}{k_{z0}(\omega)} - \frac{jk_x^2}{2k_{z0}} \right) z} \]

Easily parallelized

Transmitted THz field at a distance \( z_d \) from the crystal

\[ E(\Omega, x, z) = F^{-1} \left\{ A(\Omega, k_x, h \sin \alpha) T(k_x) e^{-j(k_0^2 - k_x^2)^{1/2} z_d} \right\} \]

\[ T(k_x) = \frac{2 \sqrt{\frac{\Omega^2 n(\Omega)^2}{c^2} - k_x^2}}{\sqrt{\frac{\Omega^2 n(\Omega)^2}{c^2} - k_x^2} + \sqrt{\frac{\Omega^2 n(\Omega)^2}{c^2} - k_x^2}} \]

Verification of model

Simulation of Optimal Imaging Conditions

Analytic Calculation

$\gamma = 63^\circ$

$s_1 = 60.06$ cm

$s_2 = 37.1$ cm

$\theta_i = 46^\circ$

$\eta$ (Normalized)

$s_1 = 60.89$ cm

$s_2 = 36.84$ cm

Verification of model

Effects of cascading on THz generation

(a) SPM, GVD-AD, material dispersion, absorption

(b) Cascading Effects, GVD-AD, material dispersion, absorption

- **Without Cascading effects** -> Longer interaction, higher energy efficiency
  
  2.27%

- **With Cascading**, shorter interaction- lower efficiency.
  
  0.85%

Momentum picture

Broadening in Frequency

\[ \chi_{\text{eff}}^{(2)} = 0 \]

\[ \chi_{\text{eff}}^{(2)} = 360 \text{pm/V} \]

Optical Fluence

THz Fluence

Broadening in Momentum

\[ \vec{k}(\omega) \to \vec{k}(\omega - \Omega) \to \vec{k}(\omega - 2\Omega) \]

Energy transfer

Energy transfer

\[ \vec{k}(\omega) \to \vec{k}(\omega - \Omega) \to \vec{k}(\omega - 2\Omega) \]

Broadening in Frequency

Optical Fluence in k-space

\[ \sum |E(\xi, k_x, z)|^2 \]

\[ z = 1.7 \text{mm} \]

\[ z = 3.4 \text{mm} \]

Spectral broadening -> **accompanied by broadening in** \( k_x \)

Time domain picture

\[ \tau_{\text{FWHM}} = 500 \text{ fs} \]
\[ F_{\text{pump}} = 20 \text{ mJ/cm}^2 \]

- Spatio–temporal break-up of the optical pulse
- Large spatial chirp of the THz frequency
Implications: Scaling to large beam sizes

- Conversion efficiency does not scale with beam size.

Implications: Effects of intensity

Different intensities have different optimal beam positions
Implications: Trade-offs of optimizing efficiency

- Higher efficiency can also lead to very large spatial chirp
Conclusion

• Developed a full system model for THz generation

• **Self limiting property**: Group velocity dispersion due to angular dispersion -> causes spatio-temporal break-up of optical pulse.

• Affects THz generation using large pump beams, spatial properties of THz.
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