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Effect of nonlinear absorption on terahertz wave generation via optical rectification in nonlinear crystals



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Paper Reference Number:

Name of the Presenter: Mina Ghaffari

Abstract

We briefly review the theory of terahertz wave generation via optical rectification by using one-dimensional propagation equations of Maxwell in nonlinear media. Then we will use the Drude model to study the effect of nonlinear absorption on optical to terahertz conversion efficiency. Simulation results demonstrate that at optical wavelengths that satisfy phase matching condition, nonlinear absorption limits efficiency and we will need to choose appropriate optical laser wavelength to decrease the effect of nonlinear absorption. According to simulation results, in case of generating 1 THz waves in GaAs, appropriate wavelength for optical intensities lower than 1 GW/cm^2 , is about 1350 nm while for higher intensities, the efficient wavelength will be about 1800 nm .

Key words: Terahertz wave, Optical rectification, Two Photon Absorption

1. Introduction

Terahertz (THz) radiation refers to the region of the electromagnetic spectrum between 10^{11} and 10^{13} Hz. Unique applications of radiation in this least touched region of electromagnetic spectrum in various fields of science and technology such as high-resolution spectroscopy, astronomy, non-destructive materials testing, telecommunication and medical sciences have motivated researchers to develop efficient and compact THz sources.

In the last few decades, optical to terahertz conversions in nonlinear crystals have become of great importance due to their power scalability (which means unlike other methods of THz generation, they do not suffer from saturation of THz output power) and also because of the rapid progress of diode-pumped lasers. Of the many techniques for generating terahertz radiations, optical rectification in various crystals such as LiNbO₃ [1,2], ZnTe, ZnSe, CdS [1] and also zinc-blende GaAs [3,4] and GaP [5], is one of the most explored methods.

In this paper we will discuss the effect of absorption and by using the Drude model, we will consider the effect of two photon absorption (TPA) on optical to terahertz conversion efficiency.

2. Theory of optical rectification

Optical rectification is a second-order nonlinear optics phenomenon which is based on generating a difference frequency ω_3 between two different Fourier components ω_1 and ω_2

of an incident optical pulse on a nonlinear medium: $\omega_3 = \omega_2 - \omega_1$.

The input electric fields of the ultra short optical pulses of the incident light generate a nonlinear polarization in the nonlinear electro-optic crystal which can be written by the following relation:

$$\begin{aligned} \vec{P} &= \vec{P}^{(1)} + \vec{P}^{(2)} + \vec{P}^{(3)} + \dots = \\ &\chi_i^{(1)} \vec{E} + \chi_{ij}^{(2)} \vec{E} \vec{E} + \chi_{ijk}^{(3)} \vec{E} \vec{E} \vec{E} + \dots \\ &= \vec{P}^{(1)} + \vec{P}^{(NL)} \end{aligned} \quad (1)$$

Where $\chi^{(n)}$ is the nth-order nonlinear optical susceptibility tensor and $P^{(n)}$ is the nth-order generated polarization. By using Maxwell's equations in a nonlinear medium, the equation describing the amplitude of the generated THz wave will be as follows [8]:

$$\nabla^2 \vec{E} - \frac{1}{c^2} \frac{\partial^2 \vec{E}}{\partial t^2} = \frac{1}{\epsilon_0 c^2} \frac{\partial^2 \vec{P}}{\partial t^2} \quad (2)$$

Where \vec{E} is the THz electric field, c is the speed of light in vacuum, ϵ_0 is the free space permittivity. When working with materials which are isotropic in the first order susceptibility, we can split off the first order polarization term and then the rest of the polarization will become the source term. In case of optical rectification, we only need the second order nonlinear polarization [9] which can also be written as:

$$P^{(2)} = 2d_{eff} \left| \vec{E} \right|^2 \quad (3)$$

Where $d_{eff} = \frac{1}{2} \chi_{eff}^2$ is the effective nonlinear coefficient for optical rectification and is calculated as:

$$d_{eff} = \frac{(2/\pi)d}{14} \quad (4)$$

Where d_{ij} is the nonlinear coefficient of electro-optic crystal. As we can see, terahertz electric field is proportional to second order derivative of the optical electric field envelope and can be approximated by $-\omega^2 I(t)$ where $I(t)$ is the intensity envelope function of the incident pulse. By solving eq. 2 for a planar wave propagating along z direction, we will have [6]:

$$E_{THz}(z, \Omega) = \frac{I(\Omega)\chi^{(2)}}{n_{THz}} \times \left(\frac{e^{ikz(n_{THz} - n_{g,opt})} - 1}{n_{g,opt} - n_{THz}} + \frac{1 - e^{ikz(n_{THz} - n_{g,opt})}}{n_{g,opt} + n_{THz}} \right) e^{ikn_{g,opt}z} \quad (5)$$

Where n_{THz} and $n_{g,opt}$ are THz phase and optical group indices respectively and k is the free-space wave-vector of the terahertz field which is defined as $k = \frac{\Omega}{c}$. The second term in

this equation is the backward THz wave propagation which is much smaller than the forward wave and can be ignored. In order to simplify the above equation, we can also define $\Delta k = (n_{g,opt} - n_{THz}) \frac{\Omega}{c}$ as the optical and terahertz k-vector mismatch. It is obvious that

terahertz wave electric field amplitude decreases as the mismatch between n_{THz} and $n_{g,opt}$ increases. This means that for efficient optical rectification, the velocity of the optical envelope should be equal to the velocity of the generated terahertz wave. Phase matching condition requires appropriate selection of pump pulse wavelength according to dispersive properties of the electro-optic crystals. Fig. 1 shows the optical group index of GaAs over a range of optical wavelengths in comparison with the terahertz refractive index [7]. We can see that for each THz frequency in the range of 0-6 THz there exists an optical wavelength in range of 1.1-1.4 μm that satisfies the phase matching condition. For example, if we expect generating THz waves at 1 THz in GaAs, the best optical wavelength would be about 1300 nm . Practically it may be impossible to find femtosecond lasers with the exact wavelength required for a specified THz frequency or absorption at that wavelength dominates the phase matching and reduces efficiency. In that case we should choose lasers with wavelengths that lead to smaller values of k-vector mismatch and absorption simultaneously.

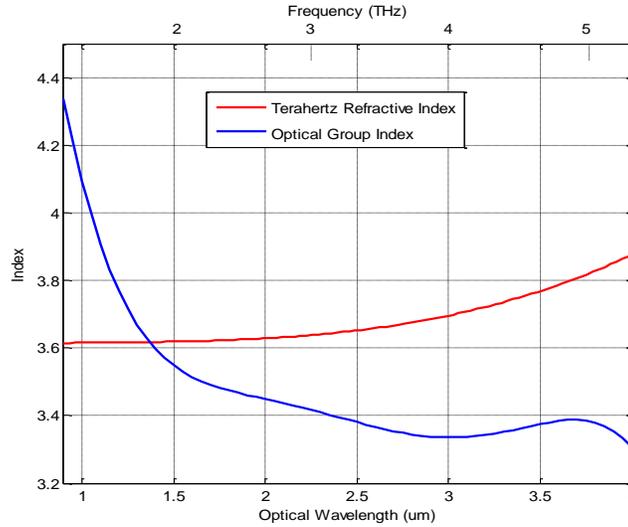


Fig. 1: Optical group and terahertz refractive index of GaAs

We assume that the incident femtosecond laser pulses have Gaussian time envelope electric field [8]:

$$\begin{aligned} E(t) &= \text{Re}[E_0 \exp(-t^2/\tau^2) \exp(i\omega_0 t)] \\ &= \frac{1}{2}[E_0 \exp(-t^2/\tau^2) \exp(i\omega_0 t) + c.c.] \end{aligned} \quad (6)$$

Where ω_0 is the central frequency and τ is its pulse-width. In this case, optical to terahertz conversion efficiency in presence of absorption and k-vector mismatch is defined as follows [9]:

$$\eta_{THz} = \frac{\sqrt{\pi}\Omega_0^2 d_{eff}^2 E_0^2 L^2 \tau^2}{2\sqrt{2}\varepsilon_0 c^3 n_{THz}^2 n_{g,opt}} \exp\left(-\frac{\tau^2 \Omega^2}{4}\right) \exp(\alpha_{THz} L/2) \frac{\sin^2(\Delta k L/2) + \sinh^2(\alpha_{THz} L/4)}{(\Delta k L/2)^2 + (\alpha_{THz} L/4)^2} \quad (7)$$

α_{THz} is THz absorption coefficient that depends on both crystal and incident optical pulse characterizations and will be discussed in more details in next section.

In this paper, we simulate optical rectification in GaAs which is extremely promising for THz generation (because of its intrinsically small THz absorption and high nonlinear coefficient), considering its dispersive properties and also its THz absorption.

3. Nonlinear absorption losses in optical rectification process

Total terahertz absorption in nonlinear crystals consists of two terms and is expressed as:

$$\alpha(\Omega) = \alpha_\varepsilon(\Omega) + \alpha_{fc}(\Omega) \quad (8)$$

The first term in this equation ($\alpha_\varepsilon(\Omega)$) is the intrinsic absorption coefficient which depends on semiconductor's dielectric constant. Fig. 2 shows this term of THz absorption as a function of frequency for GaAs according to data available in [7].

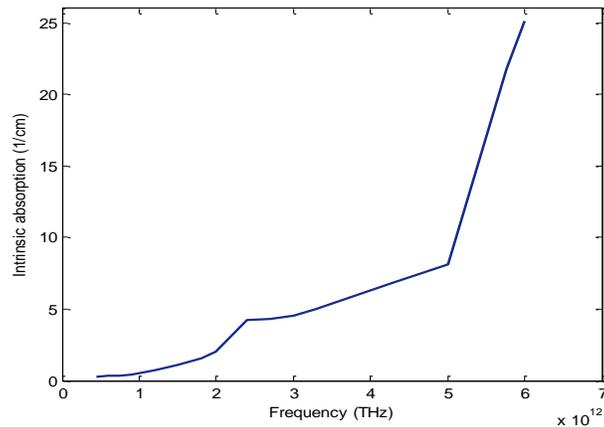


Fig. 2: Terahertz absorption spectra of GaAs

The second term in eq. 8 ($\alpha_{fc}^{(\Omega)}$), called the Free Carrier Absorption coefficient (FCA), is due to free carriers that the incident optical pulse generates in the crystal and depends on both crystal properties and optical pulse characterization.

When an optical pulse with intensity of I , pulse duration of τ and central wavelength of λ_0 is incident to a crystal, some of its photons may get absorbed by electrons in the ground state to go to higher states. In case of THz generation via optical rectification, these absorptions are considered as losses which result in decrease of efficiency. The density of free carriers generated in the crystal is calculated as follows:

$$N_{fc} = \frac{I\tau}{hc/\lambda_0} (\alpha_0 + \frac{1}{2}\beta_2 I + \frac{1}{3}\beta_3 I^2 + \dots) \quad (9)$$

Here h is Planck's constant. α_0 , β_2 and β_3 are linear, two and three-photon absorption coefficients respectively and depend on both crystal type and the wavelength of the optical pulse. Fig. 3 (a,b) illustrates the dependence of α_0 , β_2 on wavelength of the pump pulse [10]. It should be noticed that α_0 , β_2 are both zero, the former for wavelengths greater than crystal's optical wavelength and the latter for twice of that respectively; obviously correspond to energies less than band gap and half of that respectively. Higher orders photon absorption coefficients are low enough to be ignored in most of crystals such as GaAs.

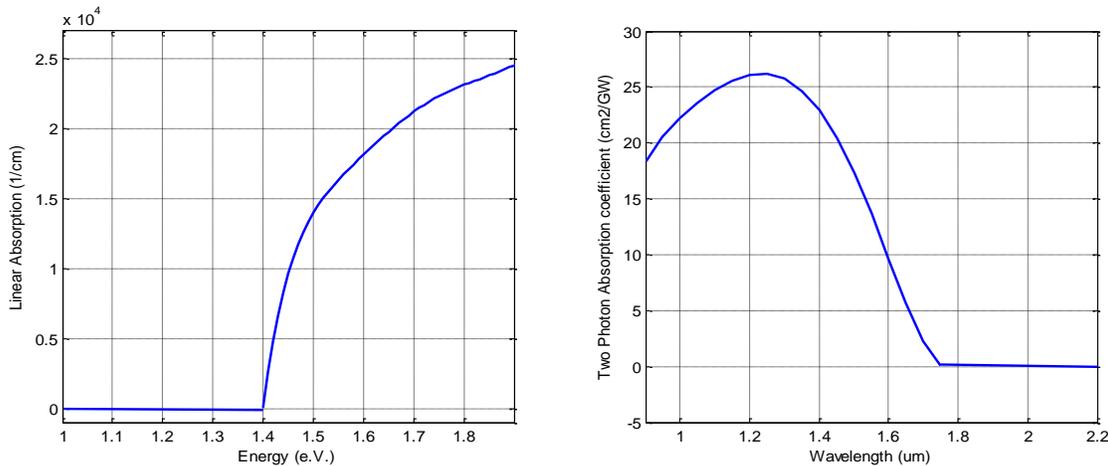


Fig. 3: linear (a) and two photon (b) absorption coefficient spectra of GaAs

In order to reduce absorption losses, we have to choose λ_0 at least twice of the crystal's wavelength, so that the linear absorption in the above equation can be neglected.

Now by using Drude-model [11], we can calculate the free carrier absorption coefficient as follows:

$$\alpha_c(\Omega) = \frac{\epsilon_\infty \omega_p^2 (1/\tau_{sc})}{n_{THz} c [\Omega^2 + (1/\tau_{sc})^2]} \quad (10)$$

Where ϵ_∞ is the high-frequency dielectric constant, τ_{sc} is the electron scattering time and ω_p is the plasma frequency of the free carriers screened by the dielectric constant and is calculated as:

$$\omega_p = \frac{N_{fc} e^2}{m_{eff} m_e \epsilon_\infty \epsilon_0} \quad (11)$$

Where m_{eff} , m_e and e are the effective mass, electron mass and electron charge respectively.

ϵ_∞ , τ_{sc} and m_{eff} for GaAs are 11.1, 200 fs and 0.067 respectively [12]. Fig. 4 demonstrates terahertz absorption coefficient spectra of GaAs when using optical pulses with pulse duration of 100 fs and optical intensity of 1 GW/cm². The generated terahertz frequency is assumed to be 1 THz. As we had expected, for optical wavelengths more than 1.75 μm , there will not be any two photon absorption and hence terahertz absorption decrease to about 1 cm⁻¹ according to Fig. 2.

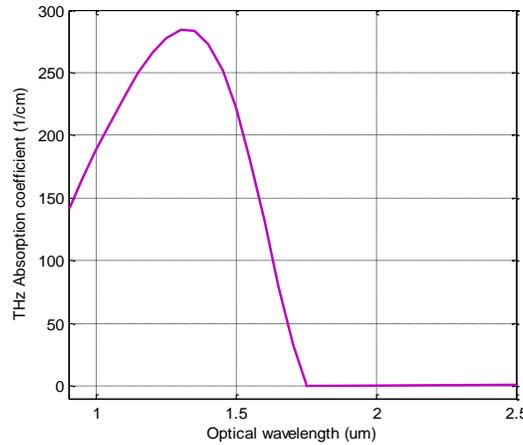


Fig. 4: Terahertz absorption coefficient spectra of GaAs

As it appears in eq. 7, the pump pulse intensity is in the numerator of the fraction and we may conclude that higher intensities result in higher efficiencies, but according to eq. 9, higher intensities for $\lambda < 1.75 \mu\text{m}$, result in more losses in form of free carrier generation and hence higher THz absorption that will reduce the efficiency. In addition, higher absorption restricts crystal length (crystal length can not be longer than $1/\alpha$ [8]) which reduces efficiency even further.

On the other hand, it seems that by increasing optical wavelength and hence decreasing nonlinear absorption coefficient, optical to THz efficiency increases, but according to Fig. 1,

increasing optical wavelength increases optical and terahertz velocity mismatch and hence decreases efficiency. Fig. 5 illustrates relative efficiency for generating terahertz pulses at $f=1$ THz, as a function of optical wavelength for different values of optical intensity when optical pulse duration is 100 fs.

Simulation results demonstrate that for wavelengths less than $1.75 \mu\text{m}$, THz absorption is dominant while for wavelengths more than $1.75 \mu\text{m}$, optical and terahertz velocity mismatch causes efficiency to decrease. They also show that maximum optical to THz efficiency is achieved at wavelengths that have lowest absorption and phase mismatch simultaneously (about 1800 nm in case of generating 1 THz pulses). It also demonstrates that increasing optical pulse intensity increases efficiency for $\lambda > 1.75 \mu\text{m}$ while for $\lambda < 1.75 \mu\text{m}$, efficiency has its maximum when $\frac{\partial \eta_{\text{THz}}}{\partial I} = 0$.

According to these simulations, for low optical pulse intensities and at phase matching wavelengths (about 1300 nm in our case) we can achieve as high efficiencies as for higher intensities when $\lambda > 1.75 \mu\text{m}$. For example, we can see that efficiency at 1300 nm for $I=0.05 \text{ GW/cm}^2$ is almost equal to efficiency at 1800 nm for $I=1 \text{ GW/cm}^2$.

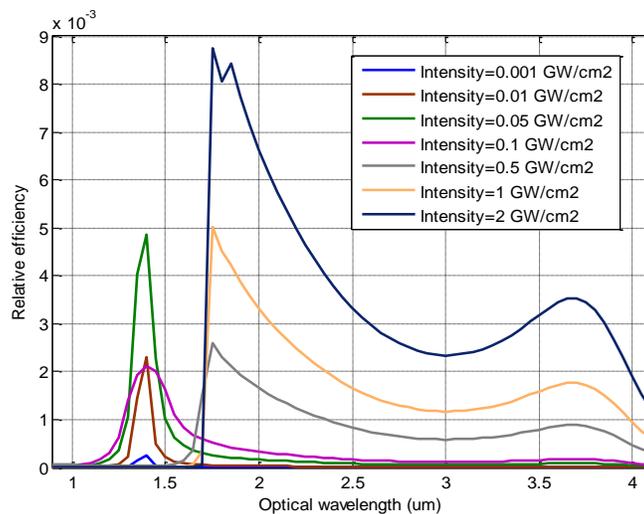


Fig. 5: relative efficiency as a function of optical wavelength for different values of optical intensity

5. Conclusions

We had developed a simulation based study on Terahertz wave generation via optical rectification in electro-optic materials. Simulation results indicate that the main limit in optical rectification process are the difficulty of phase-matching due to the dispersive properties of materials and also THz losses in form of nonlinear absorption. Therefore, the practical pump pulse wavelength should be selected properly to decrease both of these two parameters. Simulation results demonstrated that higher optical wavelengths lead to higher efficiencies when using higher optical pulse intensity.

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