# THz GENERATION BY OPTICAL RECTIFICATION FOR A NOVEL SHOT TO SHOT SYNCHRONIZATION SYSTEM BETWEEN ELECTRON BUNCHES AND FEMTOSECOND LASER PULSES IN A PLASMA WAKEFIELD ACCELERATOR\*

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## Abstract

We investigate the influence of the optical properties and of the theoretical description of the THz generation on the conversion efficiency of the generation of short THz pulses. The application is a feedback system for SINBAD with a time resolution of less than 1 fs for the synchronization of the electron bunch and of the plasma wake field in a laser driven plasma particle accelerator. Here stable THz pulses are generated by optical rectification of a fraction of the plasma generating high energy laser pulses in a nonlinear lithium niobate crystal. Then the generated THz pulses will energy modulate the electron bunches shot to shot before the plasma to achieve the required time resolution. In this contribution we compare different approximations for the modeling of the generation dynamics using second order or first order equations as well as considering pump depletion effects. Additionally, the dependence of the efficiency of the THz generation on the choice of the dielectric function has been investigated.

## **INTRODUCTION**

Achieving new discoveries, e.g. the Higgs boson or the strong interacting Quark Gluon Plasma, or cancer treatment with the high-energy particle beams are two of several application fields of particle accelerators. Because of their extremely large accelerating electric fields [1], plasma-based particle accelerators driven by laser beam can overcome the limit of the standard accelerators given by the physicalchemical properties of the material used for the construction as well as by the huge size and the financial costs. In fact, the acceleration gradients of conventional linear accelerators are limited to 10 MVm<sup>-1</sup> [2], whereas for laser-driven particle accelerators values in the order of 1 TVm<sup>-1</sup> can be achieved. Nevertheless, the period of the fields in this method known as plasma wakefield acceleration (PWA) is in the range of 10 fs. Consequently, an optimal acceleration requires stable synchronization of the electron bunch and of the plasma wakefield in the range of few femtoseconds.

For this purpose, we are developing a new shot to shot feedback system for SINBAD with a time resolution of less than 1 fs. We plane to generate stable THz pulses by optical rectification of a fraction of the plasma generating high energy laser pulses in a periodically poled lithium niobate crystal (LiNbO<sub>3</sub>) (PPLN). These pulses allow to energy modulate the electron bunches shot to shot before the plasma in order to achieve the time resolution [3].

This paper focuses on the generation of THz pulses. We present our investigation of the influence of the optical properties as well as the theoretical description of the THz generation on the conversion efficiency as well on the optimum crystal length for the generation length of short THz pulses.

The paper is organized as follows. First, we derive the general equations for the description of the THz generation and then we introduce two different methods in order to include the effects of the laser pump on the crystal. Therefore we compare the corresponding results for the efficiency as well as for optimum crystal length of the THz generation. The conclusions finalize this work.

## **MODELING THE THZ GENERATION**

Starting from the Maxwell equations the following one dimensional system of coupled differential equations for the laser pulse  $E_{\rm L}(\omega_{\rm L}, z) = A_{\rm L}(\omega_{\rm L})e^{-ik(\omega_{\rm L})z}$  and for the THz wave  $E_{\rm T}(\omega_{\rm T}, z) = A_{\rm T}(\omega_{\rm T})e^{-ik(\omega_{\rm T})z}$ 

$$\begin{pmatrix} \frac{\partial^2}{\partial z^2} + \frac{\omega_{\rm T}^2}{c^2} \varepsilon(\omega_{\rm T}) \end{pmatrix} E_{\rm T}(\omega_{\rm T}, z) = G_{\rm T}(\omega_{\rm T}, \omega_{\rm L}, z) (1)$$
$$\begin{pmatrix} \frac{\partial^2}{\partial z^2} + \frac{\omega_{\rm L}^2}{c^2} \varepsilon(\omega_{\rm L}) \end{pmatrix} E_{\rm L}(\omega_{\rm L}, z) = G_{\rm L}(\omega_{\rm L}, \omega_{\rm T}, z), (2)$$

can be derived [4–7]. Here  $\varepsilon(\omega)$  denotes the generalized (complex) dielectric function and the inhomogeneous term  $G_{\rm L}$  and  $G_{\rm T}$  are related to the nonlinear polarizations in the optical and THz frequency range respectively.

A simplification for this system is the slope varying approximation (SVA), where the second spatial derivatives of the amplitudes  $A_m$  with  $m \in L$ , T are neglected. This approach is used in almost all investigations [4,8–10] and leads to coupled system of linear differential of the first order,

$$\left(\frac{\partial}{\partial z} + \frac{\alpha_{\rm m}(\omega_{\rm m})}{2}\right) A_{\rm m}(\omega_{\rm m}, z) = G_{\rm m}^{\rm SVA}(\omega_{\rm T}, \omega_{\rm L}, z) \qquad (3)$$

where  $\alpha_m$  indicates the adsorption coefficient in the optical (m = L) and in the THz range (m = T) respectively and the inhomogeneous terms become [11]

$$G_{\rm m}^{\rm SVA} = \iota G_{\rm m} \frac{e^{\iota k(\omega_{\rm T})z}}{2k(\omega_{\rm m})}.$$
 (4)

The assumption  $G_{\rm L} = 0$  leads to the decoupling of the system and only Eq. (1) or Eq. (3) with m = T has to be

<sup>\*</sup> The work of S. Mattiello is supported by the German Federal Ministry of Education and Research (BMBF) under contract no. 05K16ROA.

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8th Int. Beam Instrum. Conf. ISBN: 978-3-95450-204-2

$$P_{\rm NL}(\omega_{\rm T},z) = \varepsilon_0 \chi^{(2)}(z) \int_0^\infty d\omega E_{\rm L}(\omega_{\rm T}+\omega,z) E_{\rm L}^*(\omega,z).$$
(5)

Here  $\varepsilon_0$  and  $\chi^{(2)}(z)$  indicate the vacuum dielectric constant author(s). and the second order nonlinear susceptibility respectively.

The optical properties of the material are encoded in the complex dielectric function  $\varepsilon(\omega)$ , which is related to the  $\stackrel{\text{o}}{=}$  wave vector  $k(\omega)$ , the refractive index  $n(\omega)$  and the adsorp-

$$n(\omega) = \frac{k(\omega)c}{\omega} = \Re\sqrt{\varepsilon(\omega)} \quad \alpha(\omega) = \frac{2\omega}{c}\Im\sqrt{\varepsilon(\omega)}.$$
 (6)

If wave vector  $k(\omega)$ , the respectively the formula  $\alpha(\omega)$  by  $\frac{k(\omega)c}{\omega} = \Re_{\mathbf{V}}$ For the polarization of the frequency region in the frequency region is needed. For this purpose For the polarization only the knowledge of the wave vector in the frequency region around laser central frequency  $\omega_0$ is needed. For this purpose, we describe the refractive index must squared as function of the frequency using the Sellmeier work equation given in [12]. Nevertheless, performing a Taylor expansion of the wave vector for the term  $k(\omega_{\rm T} + \omega) - k(\omega)$ , this which enters in the polarization integral, a linear approximation, tion, tion, tion,

$$k(\omega_{\rm T} + \omega_{\rm L}) - k(\omega_{\rm L}) \approx \frac{n_{\rm opt}^{\rm gr}}{c} \omega_{\rm T}$$
 (7)

can be used [8–10]. In this manuscript we will utilize and Ŋ compare both descriptions in order to evaluate the effects of the whole dispersion relation. <u>(</u>61

For the THz region we use a physical motivated description for  $\varepsilon(\omega_{\rm T})$  based on the oscillator model given in [13].

## **INFLUENCE OF THE FREE CARRIERS**

An important effect is given by the free carries (FC), which are generated by the pump adsorption in the material [5,9, 10, 14] and lead to a decreasing of the pump intensity in the crystal. In this manuscript we follow two different strategies in order to systematically consider the influence of the FC.

#### (a) Modification of the Dielectric Function

In this first approach (a) we decoupling the differential equations by setting  $G_L = 0$  and we systematically modify the dielectric function, which encodes the FC contributions to the optical properties as following [5, 11, 14, 15]

$$\varepsilon_{\rm tot}(\omega_{\rm T}) = \varepsilon_{\rm osc}(\omega_{\rm T}) - \frac{\omega_{\rm pl}^2}{\omega_{\rm T}^2 + i\omega_{\rm T}/\tau_{\rm sc}},\tag{8}$$

The contribution of the free carries encoded the second term of  $\varepsilon_{tot}(\omega_T)$  are implemented along the line of a Drude model with electron scattering length  $\tau_{sc}$  [10] and with the plasma frequency  $\omega_{pl}^2$  proportional to the density of free charge carries  $\rho_{\rm FC}$ . In our previous works [5, 11, 14, 15]

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we developed a phenomenological description for  $\rho_{\rm FC}$  in a PPLN.

$$\rho_{\rm FC}(F_{\rm L}) = \begin{cases} \rho_{\rm 3PA}(F_{\rm L}) & F_{\rm L} \le F_{\rm 0} \\ \\ \rho_{\rm s} - A \; {\rm e}^{-a(F_{\rm L} - F_{\rm 0})} & F_{\rm L} > F_{\rm 0}, \end{cases} \tag{9}$$

For fluences smaller than a transition fluence  $F_0$  we describe the FC density by the three-photon-adsorption process (3PA) of the pump beam in the medium. For larger fluences we model in this way a saturation of the FC density in agreement with experimental observations [10].

#### (b) Decreasing of the Pump Intensity

As second approach (b) in order to include the effects of the free carries have to be considered directly the decreasing of the pump intensity [7] induced by the three photon adsorption and given in frequency domain by [11, 14, 15]

$$I_{\rm L}(\omega_{\rm T},z) = {\rm e}^{-\iota q z} \sum_{n=0}^{\infty} \tilde{I}_n \, z^n, \text{ with } \tilde{I}_n = \frac{u_n}{2\sigma_n \sqrt{\pi}} {\rm e}^{-\frac{\omega_{\rm T}^2}{4\sigma_n^2}}$$
(10)

where we use the following notation  $u_n \begin{pmatrix} -1/2 \\ n \end{pmatrix} I_0 \left(2\gamma_3 I_0^2\right)^n$ ,  $\sigma_n^2 = \frac{2}{\tau^2} (2n+1)$  and  $q = \omega_{\rm T} n_{\rm opt}^{\rm gr}/c$ .

The nonlinear polarization and the intensity are proportional, if  $P_{\rm T}$  includes quadratic terms of the electrical field only. Therefore, within the linear approximation, see. Eq. (7), the inhomogeneous terms of the coupled system in SVA given by Eq. (3) read

$$G_{\rm T}(\omega_{\rm T},z) = -\iota \frac{\omega_{\rm T}^2}{2k(\omega_{\rm T})c^3 n_0 \varepsilon_0} I_{\rm L}(\omega_{\rm T},z) {\rm e}^{\iota k(\omega_{\rm T})z}$$
(11)

$$G_{\rm L}\omega_{\rm L}, z) = {\rm FT}_{\rm t} \left[ -\frac{\gamma_3}{4} n(\omega_0 c \varepsilon_0)^2 E_{\rm L}^3(t) \left( E_{\rm L}^*(t) \right)^2 \right], (12)$$

where  $FT_{t\rightarrow}$  indicate the Fourier transform from time to frequency domain. Here we neglect the linear adsorption in the optical regime and we use  $\alpha_{\rm T}$  calculated from the oscillator model dielectric function given in [13].

For a Gaussian pulse an analytic expression for the amplitude  $A_{\rm T}$  can be derived as  $A_{\rm T}(\omega_{\rm T}, z) = \sum_{n=0}^{\infty} a_n z^n$ , where the coefficient  $a_n$  are given by [14, 15]

$$a_n = \begin{cases} e^{-iqz} \sum_{m=1}^{\infty} \frac{p_{n+m}}{b} \frac{(n+m)!}{m!} & \text{if } n \ge 1; \\ e^{-\alpha z/2} \sum_{m=1}^{\infty} \frac{p_{n+m}}{b} \frac{(n+m)!}{m!} & \text{if } n = 0. \end{cases}, \quad (13)$$

with  $b = \alpha/2 - \iota \Delta k_0$  and  $p_m = -\iota \frac{\Omega \chi_{\text{eff}}^{(2)}}{2kc^3 n_0 \varepsilon_0} \tilde{I}_n$ .

#### RESULTS

In this manuscript we investigate a periodic polarized congruent lithium niobate crystal with  $\chi^{(2)}(z) = \chi^{(2)}_{\text{eff}} e^{-i2\pi z/\Lambda}$ , where the parameter  $\chi^{(2)}_{\text{eff}} = 336 \,\text{pm V}^{-1}$ is the effective second order nonlinear susceptibility and  $\Lambda = 237.74 \,\mu\text{m}$  is the quasi-phase-matching orientationreversal period. We consider a Gaussian laser beam pulse with central wave length  $\lambda_0 = 1030$  nm and a pulse duration at full width of half-maximum  $\tau_{\rm FWHM}$  = 25 fs [3–5, 8, 10, 11, 14, 15].

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# Conversion Efficiency

Because for the optimization of the synchronization an efficient and stable generation of THz pulses is a fundamental task, the conversion efficiency of the THz generation, defined as [3,4]

$$\eta = \frac{\pi \epsilon_0 c \int_0^\infty d\omega_{\rm T} n(\omega_{\rm T}) |E_{\rm T}(\omega_{\rm T}, z)|^2}{F_{\rm L}},\qquad(14)$$

where  $F_{\rm L}$  indicates the pump fluence, has to be maximized. In order to understand the dependence of  $\eta$  on the optical properties we calculate the efficiency by solving directly the equation of motion as differential equation of the second order, i.e. Eq. (1) as well as using the SVA, i.e. Eq. 3. In Fig. 1 we compare the results for the conversion efficiency  $\eta$ for a fixed pump fluence  $F_{\rm L} = 5$  mJ cm<sup>-2</sup> as function of the crystal length *L* between the second order calculation (solid lines) and the SVA (dashed lines). All results in this figure are obtained using the approach (a) for the description of FC, because at this fluence the results with the strategy (b) does not present significative deviations.



Figure 1: The conversion efficiency  $\eta$  for a fixed pump fluence  $F_p = 5 \text{ mJ cm}^{-2}$  as function of the crystal length *L* for the second order calculation (solid lines) and the SVA (dashed lines) labeled by different colors as indicated.

In the linear approximation is used, see. Eq. (7), we note a saturation of  $\eta$  for large crystal lengths as well as deviations between the second order and SVA calculations.

Using the Sellmeier equation, the conversion efficiency shows a local maximum about around  $L \approx 10$  mm and is a decreasing function of the larger crystal length. We note effects of the FC contributions as well as larger deviations between the results coming between from second order calculation and SVA. Therefore, at small fluences the second order dynamics effects are stronger than the contribution of the free charge carries. However, at large laser pump intensities this finding changes and the contribution of FC plays a key role, whereas the effects of the second order can be neglected [5, 15].

Therefore, for the investigation of the  $\eta$  at higher pump fluence we can restrict ourselves to the SVA calculation and we consider the efficiency as function of  $F_{\rm L}$  at fixed crystal length using both approaches for the inclusion of the FC contributions. In Fig. 2 we compare  $\eta$  calculated obtained within a (minimal) depleted calculation (b) for a crystal length of



Figure 2: Comparison of  $\eta$  obtained within a (minimal) depleted calculation (b) for a crystal length of L = 40 mm as function of  $F_L \eta$  calculated in SVA (undepleted) using the linear approximation within the approach (a) for the different parameterizations of the FC density as well as for vanishing FC density as indicated.

L = 40 mm as function of  $F_{\rm L}$  as well as the efficiency  $\eta$  calculated in SVA within the linear approximation within the approach (a) for the different parameterizations of the FC density. For comparison, the results for the efficiency calculated at vanishing FC are plotted, which shows a linear behavior.

In the approach (a), for unsaturated density of FC, the  $\eta$  holds a non-monotonic behavior with a local maximum around 10 mJ cm<sup>-2</sup> and then it decreases to an asymptotic value for large fluences, whereas for saturated  $\rho_{\rm FC}$  with  $F_0 = 9$  mJ cm<sup>-2</sup> exhibits a monotonic increasing behavior. Nevertheless, a change of the slope of this behavior occurs at the value of the fluence, where the local maximum of the efficiency for the unsaturated FC density is located.

Using the strategy (b) the depleted calculation shows a deviation from the linear dependence on  $F_{\rm L}$ . Furthermore, the asymptotic behavior for large fluences follows a power law  $\eta \propto F_{\rm L}^{\delta}$  with  $\delta = 0.79$ , whereas the efficiency calculated including the FC contribution parameterized by  $F_0 = 9$  mJ cm<sup>-2</sup> shows a linear asymptotic functional dependence.

# **Optimum Crystal Length**

For the development of the planned feed-back system a estimation of the optimum crystal length  $l_0$  for the generation is a fundamental task. For this purpose, we restrict ourselves on the SVA and we firstly consider the approach (a). Under these assumptions, the absolute value of the field squared can be factorized as [5,7,15]

$$|A_{\rm T}(\omega_{\rm T}, z)|^2 = |F(\omega_{\rm T})|^2 \ |L_{\rm Gen}(\omega_{\rm T}, z)|^2, \qquad (15)$$

where we define

$$L_{\text{Gen}}(\omega_{\text{T}}, z) = \int_{0}^{\infty} d\omega E_{\text{L}}(\omega_{\text{T}} + \omega, z) E_{\text{L}}^{*}(\omega, z) D(\omega, \omega_{\text{T}}),$$
(16)

with

$$D(\omega, \omega_{\rm T}) = \frac{\epsilon_0 \chi_{\rm eff}^{(2)} \left( e^{-\iota \Delta kz} - e^{\alpha_{\rm T} z/2} \right)}{P_{\rm NL}(\omega_{\rm T}, z)} \frac{1}{\iota \Delta k - \alpha_{\rm T}/2}$$
(17)  
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publisher, and with the mismatching  $\Delta k = k(\omega_{\rm T} + \omega_{\rm L}) - k(\omega_{\rm L}) - k(\omega_{\rm T}) + k(\omega_{\rm L}) - k(\omega_{\rm L}) - k(\omega_{\rm L}) + k(\omega_{\rm L}) - k(\omega_{\rm L}) - k(\omega_{\rm L}) - k(\omega_{\rm L}) + k(\omega_{\rm L}) - k(\omega_{$  $\frac{2\pi}{\Lambda}$ . The quantity has the unit of a length and can be considered as an effective generation length of the THz generation. It includes the whole spatial dependence of the electric field work,  $A_{\rm T}$  and depends strongly on the optical properties of the material. We can generalize  $|L_{\text{Gen}}|^2$  for the approach (b) in

> $|L_{\text{Gen}}(\omega_{\text{T}}, z)|^2 = \frac{|A_{\text{T}}(\omega_{\text{T}}, z)|^2}{|F_{(\text{b})}(\omega_{\text{T}})|^2}$ (18)

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$$F_{\rm (b)}(\omega_{\rm T}) = \omega_{\rm T}^2 P_{\rm NL}(\omega_{\rm T}, z=0) / \left(2k(\omega_{\rm T})c^2\epsilon_0\right). \tag{19}$$

Therefore, at the optimum crystal length  $l_0$  for the THz generation, the generation length, i.e. the quantity

$$L_{\max}(\omega_{\rm T}) = |L_{\rm Gen}(\omega_{\rm T}, l_0)|, \qquad (20)$$

is a frequency dependent metric conversion efficiency, where  $E_{\rm max}(\omega_{\rm T}) = \omega_{\rm T}^2 P_{\rm NL}(\omega_{\rm T})$ is a frequency dependent measure for the highest possible conversion efficiency, whereas  $\eta$  is an inclusive measure of must 1 the efficiency over the full spectrum.

work In general  $L_{\max}(\omega_{\rm T})$  and  $l_0(\omega_{\rm T})$  as functions of the frequency exhibit a maximum at the critical frequency  $\omega_{cr}$ , this where the mismatching vanishes, i.e.  $\Delta k(\omega_{cr}) = 0$ . Accordof ing on the using of the linear approximation or the complete from this work may be used under the terms of the CC BY 3.0 licence (© 2019). Any distribution Sellmeier equation and on the choice for treatment of the influence of the free carries, i.e. approach (a) or (b), following features can be summarized:

- *i*) In the approach (a) without the FC contribution the  $L_{\rm max}(\omega_{\rm T})$  is independent from the laser fluence.
- ii) In the approach (a) with non-vanishing FC contribution this scaling behavior is broken and we observe a shift of the critical frequency and a dropping of the corresponding maximum  $L_{max}(\omega_{cr})$  by increasing of the fluence. As example we show in the panel (A) of the Fig. 3  $L_{max}(\omega)$  for vanishing and unsaturated FC density using the complete Sellmeier equation at different fluencies.
- iii) In the minimal depleted calculation (b) only the decreasing of  $L_{\text{max}}(\omega_{\text{cr}})$  as function of  $F_{\text{L}}$  occurs.
- *iv*) The optimum crystal length  $l_0$  in the approach (a) using the linear approximation diverges for  $\omega_{\rm T} \rightarrow \omega_{\rm cr}$ , whereas the maximum od the generation length is finite and given by  $L_{\text{max}}(\omega_{\text{cr}}) = 2/\alpha_{\text{T}}(\omega_{\text{cr}})$ .
- v) Using the complete Sellmeier equation within the approach (a) as well using the depleted calculation (b) the optimum crystal length  $l_0(\omega_{cr})$  becomes finite. Additionally, it becomes larger by increasing of the fluence.

The dependence of  $l_0(\omega_{\rm cr})$  on the fluence  $F_{\rm L}$  is shown in the panel (B) of the Fig. 3 for the depleted calculation (b) as well as for different FC density within the approach (a). The comparison suggests that the linear approximation

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for the polarization in the depleted calculation (b) leads to an overestimation of the optimum crystal length. In general we note the importance of the details of the dynamics, i.e. the choice the FC density within the approach (a), for the determination of  $l_0$ .



Figure 3: (A):  $L_{\max}(\omega)$  for vanishing and for unsaturated FC density using the complete Sellmeier equation at different fluencies as indicated. (B):  $l_0(\omega_{cr})$  as function of  $F_L$  for the depleted calculation (b) and different choices for the FC density within the approach (a) as indicated.

# CONCLUSION

The influence of the optical properties of the lithium niobate crystal on the conversion efficiency as well as on the optimum crystal length of the generation of THz pulses has been investigated. We compare different approximation for the modeling of the generation dynamic (SVA vs. second order calculation) and of the influence of the free carries and we present a (minimal) depleted calculation.

We clearly show the importance of a consistent description of the optical properties as well as a detailed generation dynamics in order to develop the planned shot to shot feedback system, which have to perform the synchronization between electron bunch and the ultrashort laser with a time resolution of less than 1 fs.

## ACKNOWLEDGEMENT

The work of S. Mattiello is supported by the German Federal Ministry of Education and Research (BMBF) under contract no. 05K16ROA.

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