Generation of propagating quasistatic fields using ultrashort laser pulses in electro-optic crystals

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Abstract. We explore the effect of quasistatic field generation in a nonstationary medium. The nonstationarity is provided by time-dependent density of free carriers created by an ultrashort laser pulse, a situation which is common in electro-optic schemes of THz generation. The quasistatic fields can serve as indicators of ultrafast carrier dynamics and can be used for particle acceleration, control of magnetic materials, streaking techniques, and spectroscopy.

1. Introduction
Optical rectification of ultrashort laser pulses in electro-optic crystals is an established way of terahertz (THz) generation. The laser pulse produces nonlinear polarization, which moves with its group velocity and acts as a source of the radiation. Specific generation schemes include collinear phase matching [1, 2, 3] as well as non-collinear regimes based on using tilt-front pulses [4] or Cherenkov effect [5].

A key challenge for the THz generation schemes is to increase the strength of the output. Strong THz fields are of interest for various applications, such as table-top particle acceleration [6], terahertz-assisted attosecond generation [7], manipulation of magnetic order in matter [8], nonlinear terahertz spectroscopy [9]. The demand for stronger THz pulses requires increasing the laser pump intensity. This, however, gives rise to multi-photon absorption and therefore, depletion of the pump. Also, the created carriers absorb the generated THz radiation. Thus, free-carrier generation (FCG) is commonly considered as a detrimental effect.

Recently, it was shown that FCG can give rise to an unexpected physical effect, namely, the creation of quasistatic electromagnetic fields which have both electric and magnetic components [10]. This effect cannot be accounted for by simply including the free-carrier contribution to the complex dielectric permittivity of the crystal, a model which is routinely used to analyze free-carrier absorption [11, 12]. The physical mechanism of the effect is related to the nonstationarity of the medium in which carriers are created. The newly born carriers are accelerated by the electric field that copropagates the nonlinear polarization. The acceleration produces a burst of an electric current, which in turn generates the quasistatic fields that propagate ahead of the laser pulse. The predictions of Ref. [10] were based on a one-dimensional (1D) model. However, in practice the laser beams are usually focused to enhance the nonlinear polarization. Here we first outline the main features of the 1D model and then explore the role of laser pulse focusing on the formation of quasistatic fields using a more practical two-dimensional (2D) model.
2. 1D case

In the 1D case [10], a laser pulse propagating along the x direction with velocity \( V = c/n_{\text{opt}} \), where \( n_{\text{opt}} \) is the group refractive index in the optical range, in an electro-optic crystal, creates intensity distribution \( I(x, t) = I_0 f(x, t) \) and induces nonlinear polarization

\[
P(x, t) = P_0 f(x, t), \quad f(x, t) = \exp[-(t - x/V)^2/\tau^2],
\]

where \( I_0 \) is the peak optical intensity, \( f(x, t) \) is the pulse shape. The amplitude \( P_0 = d\mathcal{E}^2_0 \) is determined by the nonlinear coefficient \( d \) and maximum of the pulse envelope \( \mathcal{E}_0 \). The orientation of \( P_0 \) is determined by the polarization of the optical pulse and orientation of the crystallographic axes of the crystal. We assume that \( P_0 \) is along the y axis; therefore, only \( \{E_y, H_z\} \) field components exist. At the same time, the pulse creates free carriers with density \( N(x, t) \) which then form current \( J_{Ec} \):

\[
\frac{\partial N}{\partial t} = \alpha I^2, \quad \frac{\partial J_{Ec}}{\partial t} = \frac{\omega^2(x, t)}{4\pi \varepsilon_b} \varepsilon_b,
\]

where \( \alpha = \beta_2/(2\hbar\omega) \), \( \beta_2 \) is the two-photon absorption coefficient, \( \hbar \omega \) is the quantum energy of the laser radiation, \( \omega^2(x, t) = 4\pi\varepsilon^2 N(x, t)/(m \varepsilon_b) \), \( \varepsilon_b \) is the background permittivity. To find the field, which is generated by polarization (1) in the presence of free carrier current (2), we solve the Maxwell equations using an in-house developed FDTD code.

We model the incidence of a laser pulse on an electro-optic slab located in vacuum. The width of the slab is 3 mm, its dielectric permittivity is \( \varepsilon_b = 10.9 \), which corresponds to GaP at low frequencies, and \( n_{\text{opt}} = 3.66 \). The presence of phonons in GaP does not play any crucial role in the formation of quasistatic fields. The results are shown in Fig. 1 for two cases: without \( (f_p = 0) \) and with \( (f_p = 0.5 \text{ THz}) \) FCG. Without FCG, the field inside the slab consists of two pulses. The first pulse describes the transient solution formed when the laser pulse enters the slab, see Fig. 1(a). It moves with velocity \( c/\sqrt{\varepsilon_p} \). The second pulse is the forced solution which moves together with the laser pulse with velocity \( c/n_{\text{opt}} \). Since the pulses move with different velocities, the distance between them increases with time. Eventually, they encounter the right boundary and form transmitted and reflected pulses, Fig. 1(b). The transmitted pulses exceed in amplitude the pulses inside the crystal; moreover, the transmission coefficients for the transient and forced pulses differ [13]. The case in which carriers are created is more interesting. Fig. 1(a) shows that FCG leads to the formation of a plateau between the pulses. The plateau is the quasistatic field. The size of the plateau also increases with distance from the left boundary. The plateau also remains upon the transmission of the fields through the right boundary \( (x = 3 \text{ mm}) \) of the crystal, see Fig 1(b).

![Figure 1](image-url)

**Figure 1.** Generation of the THz field by an optical pulse incident on a 3 mm wide electro-optic crystal \((0 < x < 3 \text{ mm})\) in 1D geometry. Field distribution \( E_y(x, t) \) (a) inside the crystal and (b) after the field transmits through the right boundary at \( x = 3 \text{ mm} \). The density of the created carriers is parameterized through their plasma frequency \( f_p = 0 \) (without FCG) and \( f_p = \omega_p/(2\pi) = 0.5 \text{ THz} \) (with FCG). The pulse has \( \tau = 0.2 \text{ ps} \).
3. 2D case

We now turn to the 2D case which is much closer to experiments with focused laser pulses. We assume that the fields are independent of the \( z \) coordinate and the laser pulse shape is

\[
    f(x, y, t) = \exp\left[-(t - x/V)^2/\tau^2\right] \cdot \exp\left(-y^2/a_0^2\right),
\]

where \( a_0 \) is the focusing parameter. To find the generated field, a 2D FDTD code was developed that included equation (2) for carrier dynamics.

The fields created by a focused pulse interacting with an electro-optic slab are shown in Fig. 2. Without FCG, one observes the formation of two distinct waveforms inside the crystal, see Fig. 2(a). In contrast to the 1D case, they have different shapes: the first waveform represents the transient field that spreads in the transverse direction (like a cylindrical wave) with distance. The second waveform does not spread since it follows the envelope of the pump pulse. Once the waveforms exit the crystal, both transmitted waveforms start to spread with distance. With FCG, one can clearly observe the formation of quasistatic fields in the region between the two waveforms, see Fig. 2(c,d). Figure 3 shows slices at \( y = 0 \) and \( y = 0.8 \) mm through the 2D field distributions shown in Fig. 2. While the pulse is inside the crystal, the quasistatic field is predominantly in the region limited by the width of the pulse in the transverse direction. At \( y = 0.8 \) mm the quasistatic field is negligible. Once the waveforms exit the crystal, the fields spread in the transverse direction and we can observe that the quasistatic component is basically the same at \( y = 0 \) and at \( y = 0.8 \) mm. The presence of this quasistatic component clearly differentiate the case with FCG from that without FCG.

![Figure 2](image1.png)

**Figure 2.** Generation of the THz field by an optical pulse incident on a 3 mm wide electro-optic crystal \((0 < x < 3 \) mm) in 2D geometry. (a1, a2) Field distribution \( E_y(x, y, t)/P_0 \) inside the crystal and (b1, b2) after the field transmits through the right boundary. The focusing parameter is \( a_0 = 0.2 \) mm.

![Figure 3](image2.png)

**Figure 3.** Slices through 2D distributions shown in Fig. 2.
4. Conclusion
To conclude, we studied the generation of quasistatic electromagnetic fields by ultrashort optical pulses interacting with electro-optic crystals in the regime where free carriers are generated. The nonstationarity of the problem gives rise to the formation of quasistatic fields in the spatial region between the transient waveform, which originates from the entrance boundary, and the forced waveform, which accompanies the pump pulse. It is shown that focusing of the laser pulse does not destroy the formation of the quasistatic field but rather leads to its additional localization in the transverse direction. Upon exiting the crystal, the field spreads and its intensity decreases.

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