Fresnel Formulas for the Forced Electromagnetic Pulses and Their Application for Optical-to-Terahertz Conversion in Nonlinear Crystals

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We show that the usual Fresnel formulas for a free-propagating pulse are not applicable for a forced terahertz electromagnetic pulse supported by an optical pulse at the end of a nonlinear crystal. The correct linear reflection and transmission coefficients that we derive show that such pulses can experience a gain or loss at the boundary. This energy change depends on linear dielectric constants only. We also predict a regime where a complete disappearance of the forced pulse under oblique incidence occurs, an effect that has no counterpart for free-propagating pulses.

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Almost 200 years ago, Fresnel presented formulas for the transmission and reflection of electromagnetic pulses at the interface between two media [1]. The Fresnel coefficients depend only on two material parameters—the refractive indices of the media, which determine completely the dynamics of free-propagating pulses.

However, linear Maxwell's equations allow for another important class of pulses—forced pulses. These pulses propagate with velocities that are not determined by the refractive index of the medium. A textbook example of a forced solution is the field of a moving charge. Another practical example, on which we focus in this Letter, is the terahertz pulse created by a femtosecond optical pulse in nonlinear crystal, such as $LiNbO_3$. The optical pulse induces nonlinear polarization that mimics the optical intensity envelope. This polarization gives rise to the terahertz electric and magnetic fields through *linear* Maxwell's equations. The velocity of this new pulse is not determined by its field components but is fixed by the velocity of the optical pulse, unlike the case for a free pulse. Because of the presence of an external source, the optical pulse in our example, these pulses are referred to as forced pulses. Similar to free pulses, forced pulses experience reflection and transmission at interfaces. However, one cannot directly apply the usual Fresnel formulas to the forced pulses as their velocity fixed by their source does not appear in the Fresnel formulas.

The purpose of this Letter is twofold: first, to extend the concept of Fresnel reflection and transmission to forced pulses; second, to apply the derived formulas to study the generation of free-space terahertz radiation from forced pulses at the end of nonlinear crystal.

With respect to the first purpose, we revisit the fundamental electromagnetic problem of pulse reflection and transmission and derive corresponding coefficients for the forced pulses. We find that these coefficients depend only on the linear properties of the media and, thus, play the same role as the usual Fresnel formulas for free pulses. However, unlike free pulses, the energy of forced pulses is not conserved upon reflection and transmission. This change depends on refractive indices only, i.e., on linear material properties. Thus, although nonlinearity is required to create such pulses, it does not play any role in pulse reflection and transmission.

With respect to the second purpose, there exist two views on the mechanism of the optical-to-terahertz conversion in the literature. According to the first one [2–4], the conversion occurs in the bulk of a slab made of an electro-optic material: the terahertz field is assumed to have vanishing value at the entrance boundary of the crystal and gradually grows with distance within the coherence length. The transmission of the formed terahertz signal to vacuum at the exit boundary of the crystal is described by the ordinary Fresnel transmission coefficient. According to the other view, proposed in Ref. [5], two terahertz pulses emitted from the slab are interpreted as transition radiation from the slab boundaries produced by the moving nonlinear polarization. In a recent paper [6] the two viewpoints were opposed to each other in favor of the second. In this Letter, we focus on the processes that occur at the end of the crystal and exploit our results to discuss the existing viewpoints. We clearly show that one cannot use the usual Fresnel formulas and present the new Fresnel formulas for the forced pulses. These new formulas, on the other hand, allow one to avoid using the concept of transition radiation at the exit boundary.

We now describe our model. We consider a slab ($0 < z < L$) of nonlinear material characterized by the refractive index in the terahertz range n_t and optical group refractive index n_g (see Fig. 1). The slab is surrounded on both sides by linear material with the refractive index in the terahertz range n_0. To obtain an analytical result for terahertz fields, we neglect terahertz absorption and dispersion; i.e., n_t and n_0 are real constants.

Assuming that a femtosecond laser pulse is incident normally on the slab and its transverse size is much larger

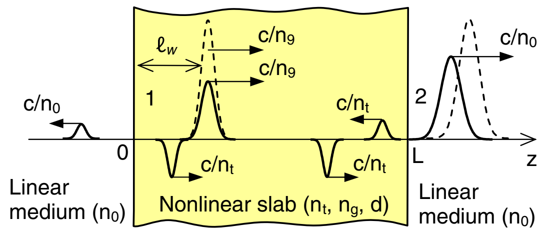

FIG. 1 (color online). Generation of terahertz pulses (solid lines) by an optical pulse (dashed lines) propagating through a nonlinear slab. The snapshots 1 and 2 refer to the moments when the optical pulse is inside the slab and after transmission, respectively.

than the terahertz wavelength, we use the one-dimensional model. We neglect the pulse depletion due to linear absorption (typically weak in such crystals as $LiNbO_3$ and GaAs below the band gap) and nonlinear processes, such as two-photon absorption and second-harmonic generation. The latter is a reasonable approximation at the not very high pump intensities we are interested in here. We do not account for the optical pulse reflection from the interface $z = L$, which can be prevented, for example, by putting an antireflective coating on the surface. Within the described model, the nonlinear polarization induced in the slab via optical rectification is

$$\mathbf{P}^{\rm NL} = \mathbf{p}F(\xi)\Pi(z), \qquad \xi = t - zn_g/c, \qquad (1)$$

where $\Pi(z) = 1$ inside the slab ($0 < z < L$) and $\Pi(z) = 0$ elsewhere. The function $F(\xi)$ is the time-dependent envelope of the optical intensity. We will use the Gaussian function $F(\xi) = e^{-\xi^2/\tau^2}$, where τ is the pulse duration [the standard full width at half maximum (FWHM) is $\tau_{\rm FWHM} = 2\sqrt{\ln 2}\tau$]. The orientation of the amplitude vector $\mathbf{p}$ is determined by the polarization of the optical beam and orientation of the crystallographic axes of the sample. We assume $p_y = p_z = 0$ and $p_x \propto dI_0$ with I_0 the peak laser intensity and d the nonlinear coefficient.

To find the terahertz radiation generated by the moving polarization (1), we use Maxwell's equations. After eliminating the magnetic field and applying the Fourier transformation with respect to time (ω is the Fourier variable and ˜ denotes quantities in the Fourier domain), the equation for the electric field transform $\tilde{E}_x(\omega)$ becomes

$$\frac{\partial^2 \tilde{E}_x}{\partial z^2} + \frac{\omega^2}{c^2} n^2(z)\tilde{E}_x = -\frac{4\pi\omega^2 p_x}{c^2}\tilde{F}(\omega)\Pi(z)e^{-i\omega z n_g/c}, \qquad (2)$$

where $n(z)$ is n_t inside the slab and n_0 outside; $\tilde{F}(\omega) = (\tau/2\sqrt{\pi})e^{-\omega^2\tau^2/4}$ is the Fourier transform of $F(\xi)$.

To proceed, we solve Eq. (2) in the homogeneous regions ($z < 0$, $0 < z < L$, and $z > L$) and match the solutions by the boundary conditions of continuity $\tilde{E}_x$ and $\tilde{B}_y = (ic/\omega)\partial\tilde{E}_x/\partial z$. Since in practice (for $L > 100\ \mu$m) the duration of the generated terahertz pulse is typically smaller than the round-trip time of the pulse in the slab, it is convenient to consider successively the processes at the entrance and exit boundaries of the crystal. To do that, we consider, at first, the crystal as semi-infinite ($0 < z < \infty$) and match the solutions of Eq. (2) at $z = 0$. This gives

$$\tilde{E}_x = \tilde{F}(\omega)\begin{cases} C_1 e^{i\omega z n_0/c}, & z < 0, \\ C_2 e^{-i\omega z n_t/c} - A e^{-i\omega z n_g/c}, & z > 0, \end{cases} \qquad (3)$$

with $A = 4\pi p_x/(n_t^2 - n_g^2)$, $C_1 = -A(n_t - n_g)/(n_t + n_0)$, and $C_2 = A(n_g + n_0)/(n_t + n_0)$. Transforming Eq. (3) into the time domain we obtain for $z < 0$:

$$E_x(z,t) = -\frac{n_t - n_g}{n_t + n_0} A F\left(t + \frac{zn_0}{c}\right), \qquad (4)$$

and for $z > 0$:

$$E_x(z,t) = A\left[\frac{n_g + n_0}{n_t + n_0} F\left(t - \frac{zn_t}{c}\right) - F\left(t - \frac{zn_g}{c}\right)\right]. \qquad (5)$$

Equation (4) predicts the formation of a backward propagating terahertz pulse in the linear material ($z < 0$) after the optical pulse enters the slab (Fig. 1, snapshot 1). In the slab, Eq. (5) predicts the generation of two terahertz pulses of the same shape, which mimics the envelope of optical intensity, but with different amplitudes and opposite signs (Fig. 1, snapshot 1). The forced wave response [the second term in Eq. (5)] is near field, and the free wave response (the first term) is free-space radiation. Although the presence of two pulses agrees with Ref. [2,4], the amplitudes in (5) are different due to our use of rigorous continuity of the field at $z = 0$ instead of setting it to zero. Expressions similar to (4) and (5) were obtained in Ref. [7]; however, the subsequent interaction of the generated pulses with the exit boundary was treated incorrectly.

The pulses in the slab propagate with different velocities defined by n_t and n_0. Near the entrance ($z = 0$), the pulses overlap and partially compensate each other; in the course of propagation they become separated and, therefore, the total terahertz field increases. The position inside the crystal at which the total terahertz field shows two separate pulses as a function of time, as sketched on Fig. 1, snapshot 1, can be called a walk-off length $\ell_w = c\tau_{\rm FWHM}/|n_t - n_g|$.

The free pulse can travel faster or slower than the forced pulse, depending on specific values of n_t and n_g. We focus on the superluminal case when $n_t > n_g$, a common situation in electro-optic crystals. In practice, this case can be subdivided further: (i) strongly superluminal case, when $n_g \ll n_t$ like in $LiNbO_3$, and (ii) weakly superluminal case, when n_g is only slightly less than n_t ($n_g \approx n_t$) like in GaAs (see parameters below). In the strongly superluminal case ($n_g \ll n_t$), the forced pulse, according to Eq. (5), significantly exceeds the free pulse in amplitude and, therefore, a finite total terahertz field appears just after the optical pulse enters the crystal. The forced and free pulses split up at a small distance, smaller than the laser pulse length ($\ell_w \approx c\tau_{\rm FWHM}/n_t \ll c\tau_{\rm FWHM}/n_g$), due to the large difference in their velocities, and propagate further separately. For example, in the case of $LiNbO_3$ excited with Ti:sapphire laser pulses (~800 nm wavelength) with $\tau_{\rm FWHM} = 150$ fs, $n_g = 2.23$, $n_t = 5.1$, we have $\ell_w \approx 16\ \mu$m. Thus, for typical slabs with $L > 100\ \mu$m, the forced and free pulses arrive at the exit boundary separately and interact with it independently (Fig. 1, snapshot 2).

In the weakly superluminal case ($n_g \approx n_t$), the pulses have almost equal amplitudes and practically cancel each other near $z = 0$. The total terahertz field

$$E_x \approx -4\pi p_x z c^{-1}(n_t + n_g)^{-1}F'(\xi), \qquad 0 < z \lesssim \ell_w, \tag{6}$$

grows linearly with z until the pulses become separated at $z \approx \ell_w \gg c\tau_{\rm FWHM}/n_g$.

Equation (5) also allows us to calculate the time-dependent terahertz energy in the slab

$$W_s = A^2 \frac{c\tau a}{8(2\pi)^{1/2}}\left[\frac{n_t^2 + n_g^2}{2n_g} + n_t\left(\frac{n_g + n_0}{n_t + n_0}\right)^2 - \frac{2\eta b}{a} \times \frac{n_t(n_g + n_0)}{(n_t + n_0)} \exp\left(-\frac{t^2c^2}{\ell_w^2}\frac{4\ln 2}{n_t^2 + n_g^2}\right)\right], \tag{7}$$

where $a = 1 + \mathrm{erf}(\sqrt{2}t/\tau)$, $\eta = (n_t + n_g)/\sqrt{2(n_t^2 + n_g^2)}$, $b = 1 + \mathrm{erf}(\sqrt{2}\eta t/\tau)$. When $tc \gg \ell_w$, the first term in (7) gives the energy W_A of the forced pulse, the second term gives the energy W_B of the free pulse, and the third (interference) term vanishes.

Now let us consider the incidence of the terahertz field (5) on the boundary $z = L$. Using the same Fourier transform approach, we obtain a backward propagating (reflected) terahertz field in the slab ($z < L$):

$$E_x^{(r)}(z, t) = AR_F \frac{n_g + n_0}{n_t + n_0} F\left(t - \frac{Ln_t}{c} + \frac{(z - L)n_t}{c}\right) - AR_N F\left(t - \frac{Ln_g}{c} + \frac{(z - L)n_t}{c}\right), \tag{8}$$

and a forward propagating (transmitted) terahertz field in the linear material ($z > L$):

$$E_x^{(t)}(z, t) = AT_F \frac{n_g + n_0}{n_t + n_0} F\left(t - \frac{Ln_t}{c} - \frac{(z - L)n_0}{c}\right) - AT_N F\left(t - \frac{Ln_g}{c} - \frac{(z - L)n_0}{c}\right). \tag{9}$$

In Eqs. (8) and (9), we defined the usual Fresnel reflection and transmission coefficients

$$R_F = \frac{n_t - n_0}{n_t + n_0}, \qquad T_F = \frac{2n_t}{n_t + n_0}. \tag{10}$$

We also introduced two new coefficients R_N and T_N:

$$R_N = \frac{n_g - n_0}{n_t + n_0}, \qquad T_N = \frac{n_t + n_g}{n_t + n_0}. \tag{11}$$

Equations (8) and (9) show that the free pulses formed in the slab and linear material after the incidence of the free pulse are related to its amplitude by the usual Fresnel formulas for R_F and T_F. However, the free pulses formed in the slab and linear material after the incidence of the forced pulse are related to its amplitude by the new Fresnel formulas for R_N and T_N.

The generation of free pulses by a forced pulse can also be viewed as transition radiation produced by a moving polarization source that suddenly disappears as suggested in [5,6]. This follows from the general concept of transition radiation [8]. The use of the derived Fresnel formulas presents a different view on this problem and, just like the usual Fresnel formulas, helps to calculate the reflection and transmission coefficients based on the results that follow from Maxwell's equations.

To probe the analogy between free and forced pulses further, we focus now on the energy conversion coefficients. Integrating the Poynting vector $S_z = (c/4\pi)E_x B_y$ for the reflected and transmitted terahertz pulses over an infinite time interval, we obtain the energy reflection and transmission coefficients for the forced pulse:

$$\frac{W_r}{W_A} = R_N^2 \frac{2n_t n_g}{n_t^2 + n_g^2}, \qquad \frac{W_t}{W_A} = T_N^2 \frac{2n_0 n_g}{n_t^2 + n_g^2}, \tag{12}$$

and the relative change of the total terahertz energy:

$$\frac{\Delta W}{W_A} = \frac{W_r + W_t - W_A}{W_A} = \frac{-(n_t - n_g)^2}{(n_t^2 + n_g^2)(n_t + n_0)}\left[n_0 + \frac{2n_g^2 + n_g n_t + n_t^2}{n_t - n_g}\right]. \tag{13}$$

Depending on the relation between n_0, n_t, and n_g, the energy change ΔW can be either positive or negative.

The nonconservation of energy can be explained by calculating the work $W_{\rm NL}$ of the moving nonlinear source on the incident and reflected terahertz pulses

$$W_{\rm NL} = -\int_{-\infty}^{\infty} dt \int_{-\infty}^{L} dz j_x^{\rm NL}(E_x^{(i)} + E_x^{(r)}), \tag{14}$$

where $j_x^{\rm NL} = p_x F'(\xi)$, and $E_x^{(i)}$ and $E_x^{(r)}$ are given by the

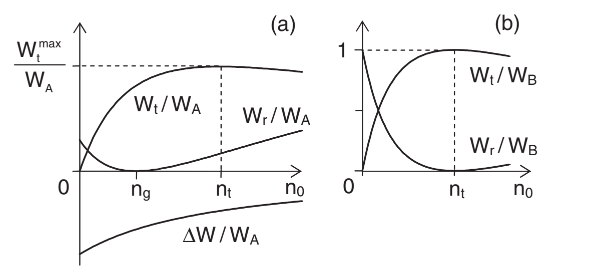

FIG. 2. (a) Energies of the transmitted (W_t/W_A) and reflected (W_r/W_A) free pulses formed by the incidence of the forced pulse and the total energy change $\Delta W/W_A$ as functions of n_0. The curves are plotted for $n_t > n_g$. (b) Same as (a) but for the incident free pulse. The energy change is zero.

second terms in Eqs. (5) and (8), respectively. Evaluating (14) yields $W_{\rm NL} = \Delta W$.

Figure 2(a) shows these energies as functions of the refractive index n_0. The transmitted energy is maximal at $n_0 = n_t$: $W_t^{\max}/W_A < 1$ for $n_g < n_t$ and $W_t^{\max}/W_A > 1$ for $n_g > n_t$. For $LiNbO_3$, $W_t^{\max}/W_A = 0.38$. The minimum of reflection $W_r = 0$ occurs at $n_0 = n_g$. A negative ΔW means partial energy absorption by the nonlinear polarization through work (14). The results for the forced pulses [Fig. 2(a)] differ significantly from those for the free pulses described by the usual Fresnel equations [Fig. 2(b)]. For free pulses the maximum of transmission is unity and is achieved when the reflection is zero.

Focusing on the experiments with $LiNbO_3$ in vacuum ($n_0 = 1$), we obtain the ratio of the amplitudes of the free and forced pulses in the crystal 0.53 [see Eq. (5)]. The transmission and reflection coefficients [$T_F = 1.67$, $R_F = 0.67$, $T_N = 1.20$, and $R_N = 0.20$ obtained using Eqs. (10) and (11)] depend quite significantly on whether the pulse is free or forced. In vacuum, $z > L$, the ratio of the amplitudes of the pulse produced by the forced pulse and the one produced by the free pulse is $1.20/(1.67 \times 0.53) \approx 1.36$. Moreover, absorption in the crystal attenuates the free pulse: its amplitude decreases approximately by a factor of 2 in a 1 mm slab, according to Fig. 2 of Ref. [6]. This changes the ratio to $1.36 \times 2 \approx 2.7$, which is close to the experimental value ~ 2.5 of Ref. [6].

In the weakly superluminal case of GaAs excited with fiber laser pulses ($\sim$1.5–2 μm wavelength) we have $n_g = 3.55$, $n_t = 3.59$, $\ell_w \approx 1.1$ mm. For the slabs with $L \leq \ell_w$, typically used in experiments, there is no sufficient distance for the forced and free pulses to become separated, and the total terahertz field arriving at the boundary $z = L$ has the form (6). Since $n_g \approx n_t$, we have $R_N \approx R_F$ and $T_N \approx T_F$. Thus, the reflection and transmission of the total terahertz field at $z = L$ obey the ordinary Fresnel law without any change of energy.

The presence of terahertz dispersion that we neglected does not change our conclusions. The dispersion affects only the free pulse during its propagation in the slab and only when $n_t \approx n_g$ (GaAs), as was confirmed by our numerical Fourier transform of Eq. (2) using frequency dependent n_t.

The concept of Fresnel reflection and transmission for forced pulses that we discussed for normal incidence can also be easily adopted for oblique incidence. Here we predict an interesting feature of oblique incidence: a complete disappearance of the forced pulse at the boundary. Let us take a forced pulse incident under an angle α from nonlinear medium ($z < 0$) to linear one ($z > 0$). Using equality of the tangential components of the wave vectors for the Fourier transforms of the incident, reflected, and transmitted fields, we obtain the laws of reflection and refraction (β and γ are the angles at which the reflected and transmitted free pulses propagate, respectively):

$$n_g \sin\alpha = n_t \sin\beta = n_0 \sin\gamma \qquad (15)$$

that generalize the classical law of specular reflection and Snell's law established for free pulses [1]. If $n_0 < n_g$ and the forced pulse is incident at $\alpha > \arcsin(n_0/n_g)$, there will be no propagating transmitted wave, an effect similar to the total internal reflection for incident free pulses. Surprisingly, there will be no reflected propagating pulse either if at the same time $n_t < n_g$ and $\alpha > \arcsin(n_t/n_g)$. Thus, the incident forced pulse completely disappears after hitting the boundary. Its energy is absorbed by the work of the nonlinear polarization. The condition $n_t < n_g$ occurs in, for example, GaP excited by a Ti:sapphire laser [9]. This disappearance is independent from the pulse polarization as it follows from kinematic relations (15). The effect of disappearance can be verified in practice by measuring the transmitted terahertz signal as a function of the angle at which the optical pulse propagates inside the crystal.

To conclude, we showed that the commonly used Fresnel formulas for the free pulses are inapplicable for calculating the transmission and reflection of the forced terahertz pulses at the end of a nonlinear crystal, and we introduced new Fresnel formulas for the forced pulses. The obtained values for the generated terahertz pulses agree with the measurements in $LiNbO_3$ crystals [6]. To maximize the transmitted terahertz signal, the refractive index of the surrounding medium should be equal to that inside the nonlinear crystal at terahertz frequencies. We also predicted that forced pulses can completely disappear at the boundary under oblique incidence.

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