Generalized time delay, velocity, and absorption in dispersive and absorbing media

Guilhem Gallot*
Laboratoire d'Optique et Biosciences, Ecole Polytechnique, CNRS, INSERM, 91128 Palaiseau, France

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The time delay and absorption of ultrashort electromagnetic pulses are defined from the local maximum of the wave's electric field and envelope. A time-domain framework is derived to obtain, in dispersive and absorbing linear media, the expressions of the phase and group time delays, and of the absorption undergone by the wave. These expressions are related to the dielectric properties of the medium but also to the local curvature of the wave's electric field. The result provides a unified and rational picture of the propagation, in dispersive and absorbing media, of ultrashort electromagnetic pulses in transmission and attenuated total reflection (ATR) geometries.

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I. INTRODUCTION

The concept of phase and group velocities plays a major role in optics, quantum mechanics, plasma physics, and geophysics [1–3]. The concept of group velocity goes back to Hamilton in 1839 [4]. The definition of the group velocity of a wave-packet was then given by Lord Rayleigh [5]. The propagation of short pulses in a dispersive medium was developed by Sommerfeld and Brillouin [6], analyzing the group velocity for complex wave numbers. In 1970, Garrett and McCumber [7] theoretically studied the propagation of a Gaussian pulse through a slab of absorbing medium and demonstrated that the envelope's maximum could propagate with abnormal group velocity. In particular, the peak of the pulse can follow the group velocity, and be greater than c or even negative. Due to the attenuation of various parts of the pulse, the reshaping of the envelope affects the location of the pulse's peak. This result, experimentally confirmed by Chu and Wong [8], triggered many theoretical and experimental studies [9–20].

The generalization of the group velocity to absorbing media requires one to deal with the complex dispersion relation $\omega(\mathbf{k}, \mathbf{r})$. Many attempts were made to give meaning to a complex wave number, frequency, or even time [9–14]. Among them, two theories can be highlighted. The first is based on the saddle point expansion [9–12]. To obtain the asymptotic expansion of the propagated field, for large values of the propagating distance, the saddle points of the complex phase function are investigated. One finds that the central wave number is not conserved in absorbing media, but undergoes a drift proportional to the imaginary part of the group velocity calculated at a shifted frequency. The second was developed by Peatross *et al.* [13], and is based on the centroid of power flow of the electromagnetic wave using the Poynting vector. In their analysis, the propagation velocity is defined in terms of net group and reshaping delays, it is always significant even in case of strong attenuation or dispersion, and it requires knowledge of the signal over the frequency domain. However, the pulse reshaping not only affects the definition of the time delays, it also modifies the attenuation of the pulse maximum, requiring an effective absorption coefficient taking into account dispersion. But this aspect received much less attention, as in the case of reflection geometries. For instance, the question of the time delay after attenuated total reflection (ATR) in absorbing media still remains open.

Here, we present a time domain framework that extends the definitions of time delays, velocities, and absorptions for the propagation of electromagnetic pulses through slabs of absorbing media, but also in reflection geometries such as ATR. This framework is based on the perturbation analysis of the transfer functions.

The following notations and conventions will be used relative to an electric field $E(t)$: the Fourier transform in the frequency domain is $\tilde{E}(\omega) = \mathcal{F}[E](\omega) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} E(t)\, e^{-i\,\omega t} dt$, the analytical signal is $\hat{E}(t) = E(t) + i(\mathcal{H}E)(t)$, where $(\mathcal{H}E)(t)$ is the Hilbert transform, and the field envelope is $\mathcal{E}(t) = |\hat{E}(t)|$ [21]. We consider a general transfer function $H(t)$ which provides the output field $E(t)$ from an input field. After a change in the system (thickness, dielectric constant, incident angle, etc.), the transfer function is changed into $H(t) + \Delta H(t)$ and the new output is $\tilde{\hat{E}}_+(\omega) = \tilde{\hat{E}}(\omega)[1 + \tilde{\rho}(\omega)]$, where $\tilde{\rho}(\omega) = \Delta\tilde{H}(\omega)/\tilde{H}(\omega)$ is the relative perturbation.

II. GENERALIZED TIME DELAYS AND ATTENUATIONS

We consider the shift in time and amplitude of the maximum of the electric field $E(t)$, given by (i) $E^{(1)}(t_M) = \mathrm{Re}\,\mathcal{F}^{-1}[i\,\omega\,\tilde{\hat{E}}(\omega)](t_M) = 0$, where t_M is the time of the local maximum. After perturbation, we similarly obtain (ii) $\mathrm{Re}\,\mathcal{F}^{-1}[i\,\omega\,\tilde{\hat{E}}(\omega)[1 + \tilde{\rho}(\omega)]](t_M + \Delta t_\phi) = 0$, where Δt_ϕ is the maximum delay shift. Assuming small perturbations so that $\Delta t_\phi \ll 1/\omega$, taking the difference between (i) and (ii), and introducing

$$\Delta_E(t) = \mathcal{F}^{-1}[\tilde{\hat{E}}(\omega)\,\tilde{\rho}(\omega)](t), \tag{1}$$

*Corresponding author: Guilhem.Gallot@polytechnique.edu

one obtains the delay shift of the maximum,

$$\Delta t_\phi = -\frac{\mathrm{Re}\,\Delta_E^{(1)}(t_M)}{E^{(2)}(t_M)}, \tag{2}$$

related to the phase delay time. The variation of the maximum amplitude $\Delta E = E_+(t_M + \Delta t_\phi) - E(t_M)$ is then obtained using Eq. (2) and (i). The relative variation of the amplitude maximum is

$$\frac{\Delta E}{E} = \frac{\mathrm{Re}\,\Delta_E(t_M)}{E(t_M)}. \tag{3}$$

The peak evolution of the field's envelope $\mathcal{E}(t)$, related to the group velocity, is provided by $\mathcal{E}^{(1)} = 0 \Leftrightarrow \mathrm{Re}\,[\hat{E}(t)\,\hat{E}^{(1)*}(t)] = 0$. This condition is applied to $\hat{E}$(t) at the envelope's peak time t_0 and to $\hat{E}_+$(t) at time $t_0 + \Delta t_g$, where Δt_g is the group delay time of the envelope's peak. Assuming small perturbations, one obtains

$$\Delta t_g = -\frac{\mathrm{Re}\left[\left(\hat{E}\,\Delta_E^{(1)*}\right)(t_0) + \left(\Delta_E\,\hat{E}^{(1)*}\right)(t_0)\right]}{\mathrm{Re}[(\hat{E}\,\hat{E}^{(2)*})(t_0) + (\hat{E}^{(1)}\,\hat{E}^{(1)*})(t_0)]}. \tag{4}$$

Last, the relative variation of the field intensity $I = \mathcal{E}^2(t)$ is

$$\frac{\Delta I}{I} = \frac{2\,\mathrm{Re}[\hat{E}\,\Delta_E^*](t_0)}{[\hat{E}\,\hat{E}^*](t_0)}. \tag{5}$$

The framework for the calculation of the time delays and attenuations in dispersive and absorbing media is provided by Eqs. (1) to (5). The complex function $\Delta_E(t)$ plays a central role in the transfer of the perturbation to the electric field. These definitions rely on the local properties of $E(t)$ and $\mathcal{E}(t)$ around their maximum in the time domain through the derivatives. We then expect the influence of the shape of the pulse on the propagation constants, as shown by Peatross *et al.* [13].

III. GENERALIZED VELOCITIES AND ABSORPTIONS

The transfer function for the propagation through a homogeneous slab of thickness z and complex refractive index $\bar{n}(\omega) = n(\omega) - i\kappa(\omega)$ is given by $\tilde{H}(\omega) = e^{-i\,\phi(\omega)}$, where $\phi(\omega) = \omega\,\bar{n}(\omega)\,z/c$ [1]. For a small change Δz, the relative perturbation is $\tilde{\rho}(\omega) = -i\,\omega\,\bar{n}(\omega)\Delta z/c$. Since $\tilde{\rho}(\omega)$ is proportional to Δz, the same is true for Eqs. (2) to (5). Defining $\Delta_{E0}(t) = \mathcal{F}^{-1}[\tilde{\hat{E}}(\omega)\bar{n}(\omega)](t)$, so that $\Delta_E = -\Delta z\,\Delta_{E0}^{(1)}(t)/c$, we then obtain consistent definitions of the phase and group velocities v_ϕ and v_g, associated with the field and envelope velocities, respectively:

$$v_\phi = \frac{\Delta z}{\Delta t_\phi} = c\,\frac{E^{(2)}(t_M)}{\mathrm{Re}\,\Delta_{E0}^{(2)}(t_M)} \tag{6}$$

$$v_g = \frac{\Delta z}{\Delta t_g} = c\,\frac{\mathrm{Re}[(\hat{E}\,\hat{E}^{(2)*})(t_0) + (\hat{E}^{(1)}\,\hat{E}^{(1)*})(t_0)]}{\mathrm{Re}\left[\left(\hat{E}\,\Delta_{E0}^{(2)*}\right)(t_0) + \left(\Delta_{E0}^{(1)}\,\hat{E}^{(1)*}\right)(t_0)\right]}. \tag{7}$$

The field and envelope absorptions α_ϕ and α_g are defined by $E(z) = e^{-\alpha_\phi z}$ and $\mathcal{E}(z) = e^{-\alpha_g z}$, and are derived from Eqs. (3) and (5) as

$$\alpha_\phi = \frac{1}{c}\frac{\mathrm{Re}\,\Delta_{E0}^{(1)}(t_M)}{E(t_M)} \quad \text{and} \quad \alpha_g = \frac{2}{c}\frac{\mathrm{Re}\left[\hat{E}\,\Delta_{E0}^{(1)*}\right](t_0)}{[\hat{E}\,\hat{E}^*](t_0)}. \tag{8}$$

Considering now the instructive case of a pulse duration converging toward 0, so that $E(t)$ is a Dirac function $\delta(t)$, the spectrum uniformly spreads over all frequencies, and the physically realistic refractive index has to converge toward unity at high frequencies. One then obtains $\Delta_{E0}(t) = \hat{\delta}(t)$ and finds that Eqs. (6) and (7) converge toward c. Therefore, v_ϕ and v_g both reduce to c as a response to a discontinuity, consistently with the Brillouin and Sommerfeld principle of non-superliminality of transients [6].

We now apply this framework to practical examples, and show that we can obtain simple and efficient expressions for the velocities and absorptions in dispersive and absorbing media. For that purpose, we expand $\tilde{\rho}(\omega)$ in series of powers of ω. The relative perturbation is $\tilde{\rho}(\omega) = \sum_{p=0}^{\infty}(A_p - i\,B_p)\omega^p$ and then Eq. (1) is

$$\Delta_E(t) = \sum_{p=0}^{\infty}(-i)^p(A_p - i\,B_p)\left[E^{(p)} + i(\mathcal{H}E)^{(p)}\right](t). \tag{9}$$

We expand $\bar{n}(\omega)$ in formal Puisieux series around frequency ω_0, so $n(\omega) = n_{-1}/\omega + \sum_{p=0}^{\infty}\frac{n_p}{p!}(\omega-\omega_0)^p$ and $\kappa(\omega) = \kappa_{-1}/\omega + \sum_{p=0}^{\infty}\frac{\kappa_p}{p!}(\omega-\omega_0)^p$, with $n_p = n^{(p)}(\omega_0)$ and $\kappa_p = \kappa^{(p)}(\omega_0)$ for $p \geqslant 0$. The term κ_{-1} takes into account the special case of constant absorption α_0 for which $\kappa_{-1} = \alpha_0 c/2$.

For a chirped Gaussian pulse $E_{CG}(t) = e^{-\frac{1}{2}a^2t^2}\cos(\omega_0 t + \frac{1}{2}bt^2)$, the Hilbert transform is $(\mathcal{H}E_{CG})(t) = e^{-\frac{1}{2}a^2t^2}\sin(\omega_0 t + \frac{1}{2}bt^2)$ [21]. Introducing the developments of $n(\omega)$ and $\kappa(\omega)$ into Eq. (9) at peak positions $t_0 = t_M = 0$, one obtains the following expressions for the velocities and absorptions at order 2:

$$\begin{aligned}\Delta t_\phi = \Bigg[&\frac{\omega_0}{a^2+\omega_0^2}\,n_{-1} + n_0 + \frac{2a^2\omega_0}{a^2+\omega_0^2}\,n_1 \\ &+ \frac{3a^4 + a^2\omega_0^2 - 3b^2}{2\left(a^2+\omega_0^2\right)}\,n_2 - \frac{b}{a^2+\omega_0^2}\,\kappa_0 \\ &- \frac{2b\,\omega_0}{a^2+\omega_0^2}\,\kappa_1 - \frac{b}{2}\frac{6a^2+\omega_0^2}{a^2+\omega_0^2}\,\kappa_2\Bigg]\frac{\Delta z}{c},\end{aligned} \tag{10}$$

$$\begin{aligned}\Delta t_g = \Bigg[&n_0 + \omega_0\,n_1 + \frac{3(a^4-b^2)}{2a^2}\,n_2 - \frac{b}{a^2}\,\kappa_0 \\ &- \frac{b\,\omega_0}{a^2}\,\kappa_1 - 3b\,\kappa_2\Bigg]\frac{\Delta z}{c},\end{aligned} \tag{11}$$

$$\alpha_\phi = \frac{1}{c}\left[\kappa_{-1} + \omega_0\kappa_0 + a^2\kappa_1 - \frac{1}{2}a^2\omega_0\kappa_2 + b\,n_1 + \frac{1}{2}b\,\omega_0 n_2\right] \quad \text{and} \quad \alpha_g = 2\alpha_\phi. \tag{12}$$

The propagation constants exhibit a clear dependence on the shape parameters of the pulse, a and b, as well as on the power series expansions of $n(\omega)$ and $\kappa(\omega)$. Both time delays and absorptions depend on dispersive n_p and attenuative κ_p parameters to take into account the complicated reshaping during propagation. For a quasi-monochromatic ($a \to 0$) and unchirped ($b = 0$) pulse, we retrieve back the classical expressions $v_\phi = c/n_0$, $v_g = c/(n_0 + n_1\omega_0)$, $\alpha_\phi = \alpha_0/2$, and $\alpha_g = \alpha_0$ as expected. A comparison with numerical simulation of the propagation of the chirped Gaussian pulses through a dispersive and absorbing medium, using the transfer function

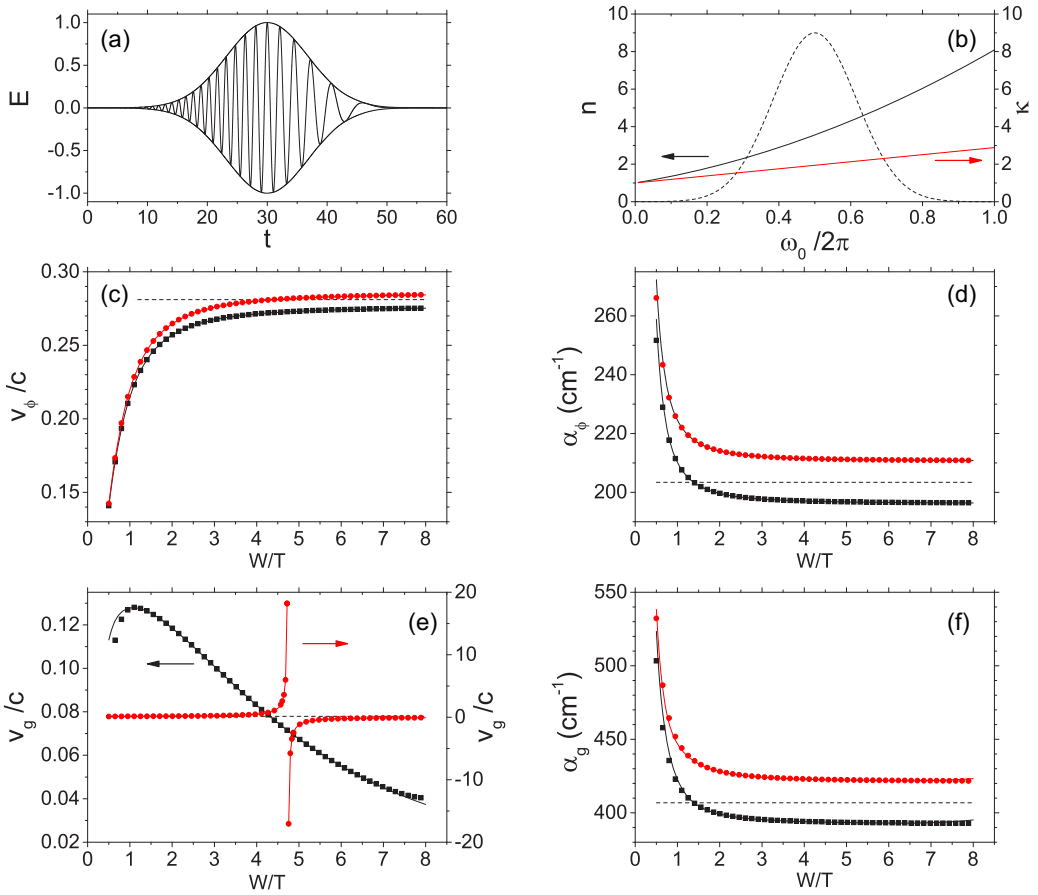

FIG. 1. Numerical calculation of the propagation constants for a chirped Gaussian pulse propagating in a dispersive and absorbing medium with $n_0 = 3.5$, $n_1 = 7$, $n_2 = 8$, $\kappa_0 = 2$, $\kappa_1 = 2$, and $\kappa_2 = 0$. (a) Electric field of the pulse. (b) Normalized spectrum of the pulse (dotted line), refractive index n (black line), and extinction coefficient κ (red line) of the slab medium. (c)–(f) Propagation constants v_ϕ (c), α_ϕ (d), v_g (e), and α_g (f) for $b = -0.15$ (black) and $b = +0.15$ (red), from numerical simulation (dots) and Eqs. (10) (solid line). The dashed lines are classical theory.

in the Fourier space $\tilde{H}(\omega) = e^{-i\phi(\omega)}$, is presented Fig. 1, introducing the pulse envelope width $W = \sqrt{8\ln 2}/a$ and the central period $T = 2\pi/\omega_0$. The agreement is very good with Eqs. (10)–(12) even for very short pulses, and we observe in Figs. 1(c)–1(f) a clear difference for opposite chirp b. Most interestingly, we also observe superluminal and negative group velocities as outlined by Garrett and McCumber for Gaussian pulses [7]. The divergence in the group velocity in Fig. 1(e) originates from the zeroing of Δt_g due to the counterbalance between the positive terms from dispersion and the negative terms from absorption, when $b > 0$. Furthermore, the parameters converge toward constant values for broad pulses, even though these values are different from classical ones due to the presence of chirp. A residual deviation may also appear for very short pulses, because a higher order development would be necessary.

We now consider the processing of signals known only locally, either because the analytic signal cannot be calculated from elementary functions or because the signal is obtained from experimental data. The calculation is based on the $\mathrm{sinc}(x) = \sin(\pi x)/\pi x$ sampling approximation, and on the property that the Hilbert transform of $\mathrm{sinc}(x)$ is $(\mathcal{H}\,\mathrm{sinc})(x) = [1 - \cos(\pi x)]/\pi x$ [22]. The approximations of $E^{(p)}(t)$ and $(\mathcal{H}E)^{(p)}(t)$ are found in Eqs. (A1) in the Appendix. Inserting these equations into Eqs. (2)–(5) extends the calculation of the delays, velocities, and absorptions related to real experimental pulses. For most cases, a small-order development is sufficient to achieve a very rapid convergence of the approximation. Therefore, very few sampling points are required around the point of interest to analyze the pulse propagation. This is consistent with the picture that local knowledge of the electric field is enough to fully characterize the propagation.

IV. WIGNER TIME DELAY IN ATR GEOMETRIES

Finally, we consider the attenuated total reflection (ATR) geometry. Here, the incident electric field impinges on an interface between two dispersive and absorbing media, with an incident angle θ, and generates an evanescent wave which extends into the second medium. The first and second media have a complex refractive index $\bar{n}_q(\omega) = n_q(\omega) - i\kappa_q(\omega)$ with $q = 1$ to 2, respectively. From the Snell-Descartes law [1], the complex refraction angle ψ is given by $\cos\psi = \sqrt{1 - [(\bar{n}_1/\bar{n}_2)\sin\theta]^2}$, and the reflection transfer functions $\tilde{H}^{\mathrm{s}}(\omega)$ and $\tilde{H}^{\mathrm{p}}(\omega)$ for s and p polarizations are found in Appendix Eq. (A2). In the following, $\tilde{H}(\omega)$ will refer either to s or p polarizations.

For small variations Δn_q and $\Delta\kappa_q$ of n_q and κ_q, respectively, the relative perturbation for ATR is

$$\tilde{\rho}(\omega) = \tilde{\rho}_1(\omega) + \tilde{\rho}_2(\omega) = \sum_{q=1}^{2} C_q(\omega)[\Delta n_q(\omega) - i\,\Delta\kappa_q(\omega)], \tag{13}$$

where $C_q(\omega) = (\frac{1}{\tilde{H}}\frac{\partial \tilde{H}}{\partial n_q})(\omega)$ stands for the complex constants C_q^{s} and C_q^{p} detailed in Appendix Eq. (A3). Equation (13) is inserted into Eqs. (1)–(5) to obtain the time delays and absorptions for ATR geometry. We will consider two cases of particular interest.

The first is ATR with small variations of $\bar{n}_2$ assuming $\bar{n}_1$ constant, as in time-domain ATR imaging [23]. At first order in ω, and for a Gaussian pulse $E(t) = e^{-\frac{1}{2}a^2t^2}\cos\omega_0 t$, the phase delay time is given by

$$\Delta t^{\mathrm{ATR}} = -\frac{\omega_0}{a^2+\omega_0^2}\,\mathrm{Im}\left[\tilde{\rho}_2(\omega_0) + \frac{a^2+\omega_0^2}{\omega_0}\tilde{\rho}_2^{(1)}(\omega_0)\right], \tag{14}$$

and the group delay time, also called the Wigner time delay, is

$$\Delta t_g^{\mathrm{ATR}} = -\,\mathrm{Im}\,\tilde{\rho}_2^{(1)}(\omega_0), \tag{15}$$

which provides a consistent description of the time delays in total reflection with dispersive and absorbing media. Since Eq. (15) only depends on the derivative of $\tilde{\rho}_2(\omega)$, we retrieve the result that $\Delta t_g^{\mathrm{ATR}} = 0$ for nondispersive, nonabsorbing media [24]. In contrast, $\Delta t_g^{\mathrm{ATR}} \neq 0$ in dispersive media, and it also exhibits a divergence at critical angle. Absorptions can similarly be obtained from Eqs. (3) and (5).

The second case is for $\bar{n}_2(\omega)$ constant, and for the electric field impinging from an absorbing medium $\bar{n}_1(\omega)$. Under a small change $\Delta\theta$ of angle of incidence, $\tilde{\rho}_1(\omega_0,\theta)$ is

$$\tilde{\rho}_1^{(1)}(\omega_0,\theta) = \frac{\partial}{\partial n_1}\left(\frac{1}{\tilde{H}}\frac{\partial\tilde{H}}{\partial\theta}\right)(\omega_0,\theta)[n_1 - i\,\kappa_1]\Delta\theta \tag{16}$$

and then $\Delta t_g^{\mathrm{ATR}} = -\,\mathrm{Im}\,\tilde{\rho}_1^{(1)}(\omega_0,\theta)$. Since $\frac{1}{\tilde{H}}\frac{\partial\tilde{H}}{\partial\theta}$ diverges at critical angle, the same goes for $\tilde{\rho}^{(1)}$ and for $\Delta t_g^{\mathrm{ATR}}$, as soon as absorption, even a very low one, exists in the first medium. Therefore, and contrary to the result without absorption [24], one expects a diverging Wigner time delay at critical angle for any realistic experiment since absorption is never zero. This divergence at critical angle could be experimentally tested by measuring the delay difference from an absorbing prism with air or metal as the external medium.

V. CONCLUSION

In summary, we developed a framework that consistently describes the time delay and absorption of short electromagnetic pulses in dispersive and absorbing media. It provides accurate definitions of phase and group velocities as well as corresponding absorptions for pulses propagating through dispersive and absorbing media, even for very short pulses or when the group velocity is negative or superluminal. Both time delays and absorptions depend on the balance between dispersive and attenuation. The framework also provides a rigorous processing of time delays in ATR geometry when absorption is taken into account. These results can be used in a variety of domains, such as frequency comb metrology, carrier-envelope phase control, nonlinear optics, ultrafast pulse propagation, or time-domain ATR imaging.

APPENDIX

Approximations of $E^{(p)}(t)$ and $(\mathcal{H}E)^{(p)}(t)$ using the Sinc expansion [22] at order N are

$$\begin{aligned} E^{(p)}(t) &\approx \sum_{m=-N}^{+N} E(m\tau)\,s_m(t), \\ (\mathcal{H}E)^{(p)}(t) &\approx \sum_{m=-N}^{+N} E(m\tau)\,c_m(t), \end{aligned} \tag{A1}$$

with $s_m(t) = \mathrm{sinc}[(t-p\tau)/\tau]$ and $c_m(t) = (\mathcal{H}\,\mathrm{sinc})[(t-p\tau)/\tau]$, where $(\mathcal{H}\,\mathrm{sinc})(t) = [1-\cos(\pi t)]/\pi t$, assuming that the spectrum of E is null outside the interval $[-\pi/\tau, \pi/\tau]$.

In ATR geometry, the reflection transfer functions $H^{\mathrm{s}}(\omega)$ and $H^{\mathrm{p}}(\omega)$ for s and p polarizations, from a first medium with complex refractive index $\bar{n}_1$ to a second one with $\bar{n}_2$, are given by [1]

$$\begin{aligned} \tilde{H}^s(\omega) &= \frac{\bar{n}_1\cos\theta - \bar{n}_2\cos\psi}{\bar{n}_1\cos\theta + \bar{n}_2\cos\psi}, \\ \tilde{H}^p(\omega) &= \frac{\bar{n}_2\cos\theta - \bar{n}_1\cos\psi}{\bar{n}_2\cos\theta + \bar{n}_1\cos\psi}, \end{aligned} \tag{A2}$$

with $\cos\psi = \sqrt{1-[(\bar{n}_1/\bar{n}_2)\sin\theta]^2}$, and the expressions of $C_q(\omega) = \frac{1}{\tilde{H}}\frac{\partial\tilde{H}}{\partial n_q}$ for s and p polarizations are

$$\begin{aligned} \frac{C_1^s}{\bar{n}_2}(\omega) &= \frac{C_2^s}{\bar{n}_1}(\omega) = \frac{2\cos\theta\sec\psi}{\bar{n}_2^2-\bar{n}_1^2}, \\ \frac{C_1^p}{\bar{n}_2}(\omega) &= -\frac{C_2^p}{\bar{n}_1}(\omega) = \frac{2\cos\theta\sec\psi\left[1-2\left(\frac{\bar{n}_1}{\bar{n}_2}\sin\theta\right)^2\right]}{\bar{n}_1^2\cos^2\psi - \bar{n}_2^2\cos^2\theta}. \end{aligned} \tag{A3}$$

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