Role of the instantaneous spectrum on pulse propagation in causal linear dielectrics

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A model-independent theorem demonstrates how a causal linear dielectric medium responds to the instantaneous spectrum, that is, the spectrum of the electric field pulse that is truncated at each new instant (as a given locale in the medium experiences the pulse). This process leads the medium to exchange energy with the front of a pulse differently than with the back as the instantaneous spectrum laps onto or off of nearby resonances. So-called superluminal pulse propagation in either absorbing or amplifying media as well as highly subluminal pulse propagation are understood qualitatively and quantitatively within this context.

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1. INTRODUCTION

In this paper, we present an analytical description of how pulses interact with nearby resonances in linear causal media even when the spectral content of the pulse seems insufficient to permit the interaction. This paper provides an analytical explanation for the qualitative arguments made by Chiao and co-workers[1–3] regarding superluminal pulse propagation in an amplifying medium. Their simulations and analysis of the Lorentz model showed that the leading portion of a pulse can borrow energy from an amplifying medium; this energy is then returned to the medium from the later portions of the pulse. Thus the superluminal behavior is understood to be a pulse-reshaping effect caused by an exchange of energy with the medium. Wang and co-workers[4] incorrectly dismissed this interpretation in connection with their recent superluminal observations, citing the lack of spectral overlap between the pulse and the nearby amplifying resonances.

The results presented here also provide analytical insight into the earlier research by Garrett and McCumber[5] and Chu and Wong[6] in connection with superluminal pulse propagation in absorbing linear dielectrics. Again, as was understood in qualitative and model-dependent contexts, the forward portion of an on-resonance pulse can pass through a medium relatively unattenuated, whereas the rear portion is preferentially absorbed. Likewise, highly subluminal pulse propagation[7] can be understood as a pulse-reshaping effect (i.e., the attenuation of the front of the pulse, the amplification of the back, or both). Subluminal behavior is usually discussed in the context of group velocity instead of in the present context of pulse reshaping because concerns about relativity naturally don't arise.

All these phenomena can be described in the context of the *instantaneous* power spectrum. The instantaneous spectrum is that spectrum perceived by individual points in the medium up to any given moment as the pulse sweeps past. Previously, the instantaneous power spectrum was utilized in describing the response of driven electronic circuits,[8] the acoustical response of materials to sound waves,[9] and the behavior of photon counters.[10]

We recently demonstrated[11] that the time-dependent spectrum arises naturally in Poynting's theorem when the principle of causality is invoked in the form of Kramers–Kronig.[12] This is done independently of specific models or approximations (in contrast with most discussions regarding exotic pulse-propagation phenomena). The principle of causality requires a medium experiencing a pulse to be prepared for an abrupt termination of the field at any moment, in which case further exchange of energy with the field cannot take place. Such a termination produces a truncated waveform that generally contains a wider range of spectral components than are present in the pulse taken in its entirety. This momentary spectrum can lap onto nearby absorbing or amplifying resonances. The medium accordingly attenuates or amplifies this perceived spectrum. As the medium experiences the waveform, it continually reassesses the spectrum and thereby treats the front and the rear of the pulses differently.

In presenting our arguments, we refer to Poynting's theorem and to the traditional energy-transport velocity,[13–15] which, in the context that we give it, is strictly luminal. A brief review is given in Section 2. Section 3 describes how the energy density in Poynting's theorem can be rewritten in terms of the instantaneous spectrum. Section 4 explains the role of the instantaneous spectrum in superluminal propagation in amplifying media. Section 5 explains the spectrum's role in superluminal propagation in absorbing media. Section 6 explains its role in subluminal pulse propagation. We

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discuss the results further in Section 7 and make the connection with group velocity.

2. POYNTING'S THEOREM

In this paper, we utilize Maxwell's equations in a linear, isotropic, nonmagnetic, nonconducting medium:

$$\nabla \times \mathbf{E} + \frac{\partial \mathbf{B}}{\partial t} = 0, \qquad \nabla \times \frac{\mathbf{B}}{\mu_0} - \epsilon_0 \frac{\partial \mathbf{E}}{\partial t} = \frac{\partial \mathbf{P}}{\partial t}. \quad (1)$$

We considered energy transport in anisotropic and diamagnetic media in another report.[11] By convention, we take all fields to be real in the time domain.

Poynting's theorem is a direct consequence of Eqs. (1) and can be written (under the previous assumptions) as

$$\nabla \cdot \mathbf{S} + \frac{\partial u}{\partial t} = 0, \quad (2)$$

where the Poynting vector is $\mathbf{S} \equiv \mathbf{E} \times \mathbf{B}/\mu_0$ and the total energy density is given by

$$u(t) = u_{\text{field}} + u_{\text{exchange}} + u(-\infty). \quad (3)$$

Expression (3) for the energy density includes all forms of energy, including a nonzero integration constant $u(-\infty)$ that corresponds to energy stored in the medium before the arrival of any pulse. The electromagnetic field energy is

$$u_{\text{field}} \equiv \frac{B^2}{2\mu_0} + \frac{\epsilon_0 E^2}{2}. \quad (4)$$

The time-dependent accumulation of energy transferred into the medium from the field is given by

$$u_{\text{exchange}} = \int_{-\infty}^{t} \mathbf{E} \cdot \frac{\partial \mathbf{P}}{\partial t'} \mathrm{d}t'. \quad (5)$$

As u_{exchange} increases, the energy in the medium increases; conversely, as u_{exchange} decreases, the medium surrenders energy to the electromagnetic field. Although it is possible for u_{exchange} to become negative, the combination $u_{\text{exchange}} + u(-\infty)$ should never be considered to become negative because a material cannot surrender more energy than it has to begin with.

The energy-transport velocity[13–15] is defined as

$$\mathbf{v}_E \equiv \mathbf{S}/u. \quad (6)$$

If only u_{field} is used in evaluating Eq. (6) the Cauchy–Schwartz inequality (i.e., $\alpha^2 + \beta^2 \geqslant 2\alpha\beta$) ensures that v_E is strictly bounded by c. We insist that the total energy density u never be considered to be less than u_{field}. In this we differ from previous usage of the energy-transport velocity in connection with amplifying media[1,3] in which the constant of integration $u(-\infty)$ was left at zero (apparently by default), resulting in the viewpoint of superluminal and negative (opposite to the direction of $\mathbf{S}$) energy-transport velocities.

3. INSTANTANEOUS SPECTRUM

We now turn our attention to an examination of u_{exchange} in a frequency-dependent context. We utilize the frequency-domain representation of the electric field:

$$\mathbf{E}(\omega) \equiv \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} \exp(i\omega t)\mathbf{E}(t)\mathrm{d}t. \quad (7)$$

In accord with the linear and the isotropic assumptions the temporally nonlocal constitutive relation between the electric field and the polarization can be written in terms of the linear susceptibility as

$$\mathbf{P}(\omega) = \epsilon_0 \chi(\omega)\mathbf{E}(\omega). \quad (8)$$

The polarization as a function of time is then obtained from

$$\mathbf{P}(t) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} \mathbf{P}(\omega)\exp[-(i\omega t)]\mathrm{d}\omega. \quad (9)$$

We developed the following exact representation[11] for the exchange energy density defined in Eq. (5):

$$u_{\text{exchange}} = \epsilon_0 \int_{-\infty}^{\infty} |E_t(\omega)|^2 \omega \,\mathrm{Im}\,\chi(\omega)\mathrm{d}\omega, \quad (10)$$

where

$$\mathbf{E}_t(\omega) \equiv \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{t} \mathrm{d}t'\mathbf{E}(t')\exp(i\omega t'). \quad (11)$$

Theorem (11) is obtained by the imposition of the requirements of causality on the susceptibility $\chi(\omega)$. An outline of the derivation is provided in Appendix A. As is explained below, this general form of u_{exchange} reveals physical insights into the manner in which causal dielectric materials exchange energy with different parts of an electromagnetic pulse. The description here is model independent. It is applicable to any function $\chi(\omega)$ that obeys the Kramers–Kronig relations.[12] Only the imaginary part of $\chi(\omega)$ is needed because it directly controls absorption and is therefore responsible for the energy exchange. Although the real part of $\chi(\omega)$ is not formally present in Eq. (10), it is not independent of the imaginary part and can be obtained if desired through the Kramers–Kronig relations.

u_{exchange} does not increase (or decrease) monotonically. Its value depends on the past history of the pulse up until the current time t. This time dependence enters through only the square magnitude of $E_t(\omega)$, called the *instantaneous* power spectrum. $E_t(\omega)$ can be interpreted as the Fourier transform of the pulse that is truncated at the current time t and set to zero thereafter. Obviously, a truncated pulse can include many frequency components that are not present in the pulse taken in its entirety. This relation explains why the medium responds differently to the front of a pulse versus the back. Even though absorption or amplification resonances may lie outside of the spectral envelope of the entire pulse the instantaneous spectrum perceived by the medium may momentarily overlap the resonances. The medium cannot anticipate the future of the pulse, so it must amplify or attenuate in accord with the spectrum of the pulse experienced up until the current time t. In doing this the material is always prepared for the possibility of an abrupt termination of the field, in which case there can be no further exchange of energy. This means that the amplification or attenuation must be correct at each instant.

Because the exchange energy density u_{exchange} handles the transfer of energy from the field to the medium, it is controlled by the imaginary part of $\chi(\omega)$. For a strictly absorptive medium [i.e., $\omega \operatorname{Im} \chi(\omega) \geqslant 0$, because fields are taken to be real in the time domain] u_{exchange} is seen to be positive definite so that the total energy density is always greater than u_{field} [with $u(-\infty)$ appropriately set to zero]. In an active medium [i.e., $\omega \operatorname{Im} \chi(\omega) \leqslant 0$] the integrand becomes negative because energy is pulled from the medium and given to the field. In this case $u(-\infty)$ is chosen to be sufficiently large to avoid removing (even if momentarily) energy from the medium that is not there to begin with. In either case strict luminality of the energy-transport velocity is maintained with $u \geqslant u_{\text{field}}$.

4. SUPERLUMINAL EFFECTS IN AMPLIFYING MEDIA

Equations (10) and (11) elucidate what happens when a narrow-band pulse in an amplifying medium undergoes the Chiao effect. The instantaneous spectrum dictates how the front of the pulse borrows energy from neighboring amplifying resonances even if the spectrum of the complete pulse is well outside of any gain peak. The instantaneous spectrum laps onto nearby amplifying resonances during early potions of the pulse, and the medium accordingly amplifies this perceived spectrum. As the medium experiences the waveform, it continually reassesses the spectrum in accord with Eq. (11). During the trailing portion of the pulse the instantaneous spectrum narrows as the perceived pulse spectrum withdraws from the nearby resonance and previously borrowed energy is returned to the medium.

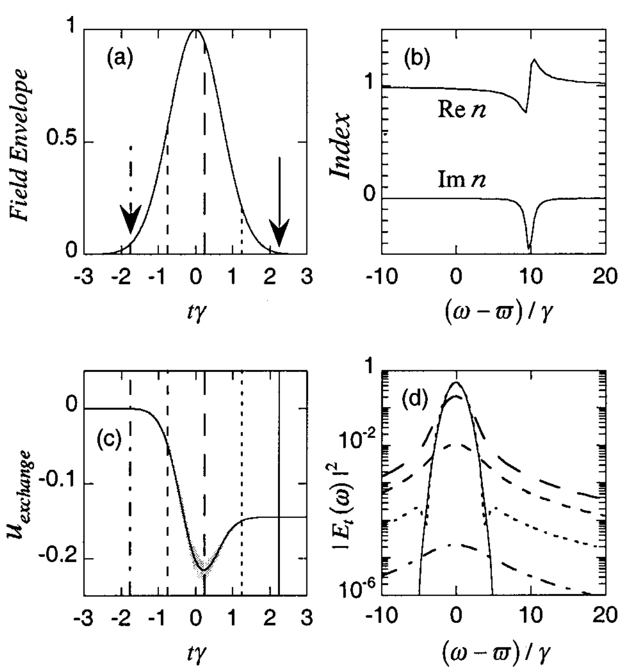

Fig. 1. (a) Electric field envelope in units of E_0. The vertical lines indicate times for the assessment of the instantaneous spectrum. (b) Refractive index associated with an *amplifying* resonance. (c) Exchange energy density in units of $\epsilon_0 E_0^2/2$. (d) Instantaneous spectra of the field pulse in units of E_0^2/γ^2. The spectra are assessed at the times indicated by the vertical lines in (a) and (c).

Figure 1(a) shows the field envelope of a Gaussian pulse given by $E(t) = E_0 \exp(-t^2/\tau^2)\cos(\bar{\omega}t)$. We consider this waveform to be passing through a point in a medium with an amplifying resonance that is centered on the frequency $\omega_0 = \bar{\omega} + 10\gamma$, where γ is the width of the resonance. For illustration purposes, we employ the Lorentz model, $\chi(\omega) = f\omega_p^2/[\omega_0^2 - \omega^2 - (i\gamma\omega)]$, where ω_p is the plasma frequency and f is the oscillator strength [note that Eqs. (10) and (11) are model independent]. The parameters of the active medium are chosen to be $\omega_0 = 100\gamma$ and $f\omega_p^2 = -100\gamma^2$. Figure 1(b) shows the real and the imaginary parts of the refractive index in the neighborhood of the resonance. As usual, the connection to the susceptibility is given by $(\operatorname{Re} n + i \operatorname{Im} n)^2 = 1 + \chi$. The duration of the pulse is chosen to be $\tau = 1/\gamma$, which results in a pulse spectral width (centered on $\bar{\omega}$) that is similar to the width of the amplifying resonance. Figure 1(c) depicts the exchange energy density u_{exchange} for a point experiencing the pulse as a function of time. The dip in the curve indicates the well-known effect of the pulse's borrowing excess energy from the medium that it returns (in part) during the later portion. The gray curve depicts the rapid oscillations (approximately 100—not resolved in the figure), whereas the black curve is time averaged.

Figure 1(d) displays the instantaneous power spectrum (used in computing u_{exchange}) evaluated at various times during the pulse. The corresponding times are indicated with vertical lines in both Figs. 1(a) and 1(c). The format of each vertical line matches a corresponding spectral curve. The instantaneous spectrum exhibits wide wings that vary in strength, depending on when the integral in Eq. (11) truncates the pulse. The spectral wings appear early during the pulse, grow stronger, and then diminish as the pulse passes. A comparison of Figs. 1(b) and 1(d) shows that the wings lap onto the amplifying resonance during the passage of the pulse. As the wings grow and access the neighboring resonance, the pulse extracts excess energy from the medium; as the wings diminish, the pulse surrenders that energy back to the medium. A similar result can also be seen when the pulse spectrum is centered above the amplifying resonance (or between two resonances, as in the experiment by Wang *et al.*[4]).

5. SUPERLUMINAL EFFECTS IN ABSORBING MEDIA

The effect reported by Garret and McCumber[5] and by Chu and Wong[6] is the converse of the description in Section 4 of the amplifying case. This effect occurs when a narrow-band pulse is centered on an absorption resonance. The instantaneous spectrum during early portions of the pulse extends *away* from the resonance. This effect is seen in Figs. 2(a)–2(d), which are similar to Figs. 1(a)–1(d). However, as is characteristic of an absorption resonance, the oscillator strength f is taken to be positive

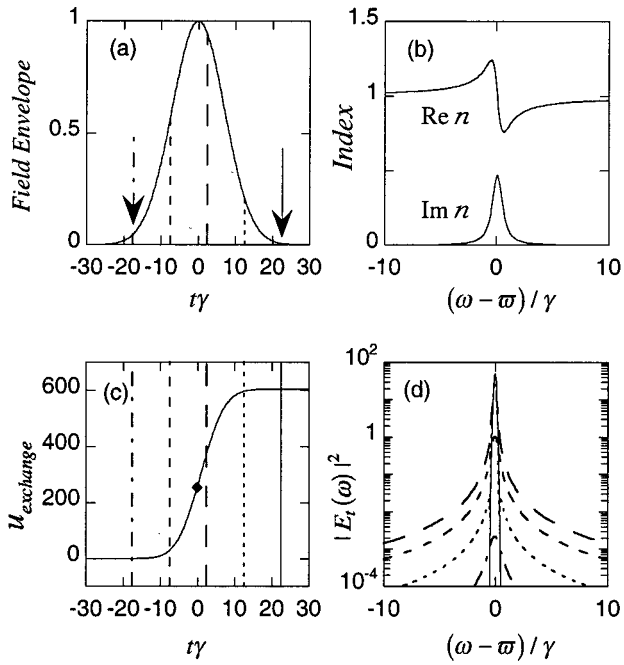

Fig. 2. (a) Electric field envelope in units of E_0. The vertical lines indicate times for the assessment of the instantaneous spectrum. (b) Refractive index associated with an *absorbing* resonance. (c) Exchange energy density in units of $\epsilon_0 E_0^2/2$. (d) Instantaneous spectra of the field pulse in units of E_0^2/γ^2. The spectra are assessed at the times indicated by the vertical lines in (a) and (c). Note that the pulse duration is longer than that shown in Fig. 1.

instead of negative. Also, the pulse duration is taken to be $\tau = 10/\gamma$ (10 times longer than that shown in Fig. 1), making the spectral content of the pulse narrower than the absorption resonance. All other parameters are the same as in Section 4 except that the center frequency of the pulse is chosen to lie on resonance (i.e., $\omega_0 = \bar{\omega}$), as can be seen from Fig. 2(d).

A comparison of Figs. 2(b) and 2(d) shows that the wings of the instantaneous spectrum extend well away from the absorbing resonance during the early part of the pulse. This position is consistent with the fact that the exchange energy seen in Fig. 2(c) is delayed in transferring energy from the field to the material. The rapid oscillations are on such a tiny scale that they are not seen in the figures. The dark diamond in the center of Fig. 2(c) corresponds to the exchange energy at time $t = 0$ (i.e., the midpoint in time of the Gaussian field profile). As is apparent, significantly less than half of the final exchange energy has transferred by this time. This condition corresponds to the fact that the early part of the field envelope is less attenuated than the rear portion. Note that, in this situation, the slope of the exchange energy is always positive. This slope indicates that the resultant electric field envelope after the exchange lies within the original pulse envelope.

6. SUBLUMINAL EFFECTS

As was described in Sections 4 and 5, superluminal behavior is manifest when a pulse propagates off resonance in an amplifying medium or on resonance in an absorbing medium. Subluminal behavior occurs in the reverse of either situation. This behavior is illustrated in Figs. 3(a)–3(d), which are identical to Figs. 1(a)–1(d) except that the oscillator strength f is positive so that the off-resonance pulse propagates in an absorbing medium. As can be seen from Fig. 3(c), the material absorbs excess energy from the front of the pulse; this energy is surrendered to the later portion. As always, this enhancement of one part of the field envelope at the expense of another is controlled directly by the instantaneous spectrum. In fact, the instantaneous spectra seen in Figs. 1(d) and 3(d) are identical. Because the instantaneous spectra access a susceptibility that is inverted from the amplifying case, the exchange energy seen in Fig. 3(c) is inverted from that in Fig. 1(c). This inversion leads to the commonly observed slowing of light in materials. In the circumstance just presented the extent of the slowing is modest.

The remaining situation to address is the case of a pulse propagating on resonance in an amplifying medium. The slowing of light results from the early instantaneous spectrum moving off of the amplifying resonance, which is followed by a narrowing of the spectrum so that the back of the pulse is preferentially amplified. The on-resonance propagation of a pulse in an amplifying medium can be highly subluminal. However, a pulse propagating on resonance in an amplifying medium can grow dramatically, just as the pulse in Figs. 2(a)–2(d) is strongly attenuated as it propagates on resonance in an absorbing medium. Nevertheless, it is naturally more fashionable to create a situation for highly subluminal propagation in which the pulse is neither amplified nor attenuated. To do this, one can employ two superimposed resonances of differing widths: one amplifying and one attenuating.

Figures 4(a)–4(d) are similar to Figs. 2(a)–2(d) except that a negative oscillator strength f is employed, representing an amplifying resonance of width γ_1. In addi-

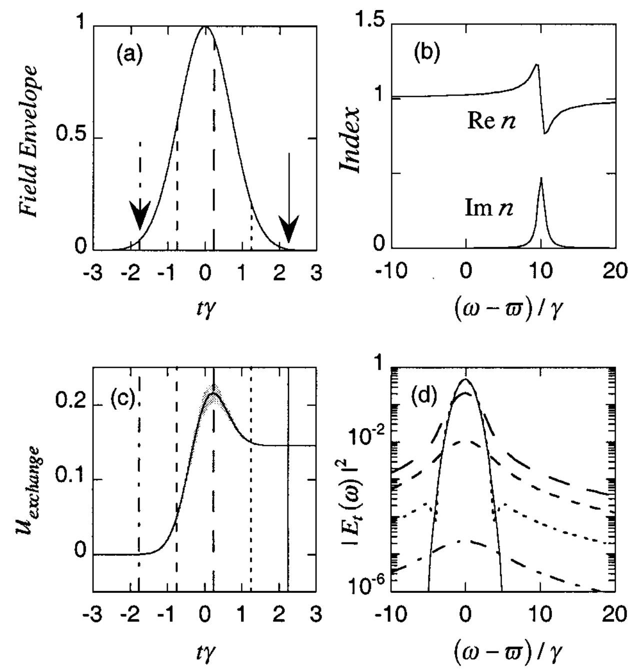

Fig. 3. Same as is shown in Fig. 1 with an *absorbing* resonance.

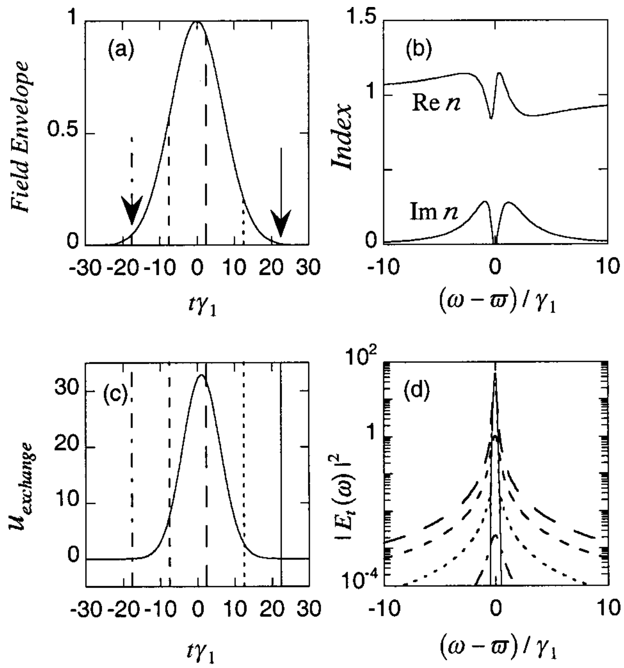

Fig. 4. Similar to Fig. 2 but with an *amplifying* resonance. In addition, a wider absorptive resonance is superimposed on the amplifying resonance.

tion, a second absorbing resonance with a spectral width of $\gamma_2 = 4.14\gamma_1$ is included in the material. The positive oscillator strength for this absorbing resonance is also 4 times greater than that of the amplifying resonance. With the exception of this second resonance the magnitude of $f\omega_p^2$ throughout this paper is consistently set to $100\gamma_1^2$.

Both resonances are centered at $\omega_0 = 100\gamma_1$, as usual. The structure of the real and the imaginary parts of the refractive index are seen in Fig. 4(b). Figures 4(a) and 4(c) show the field envelope and the exchange energy, respectively, for a pulse with a duration of $\tau = 10/\gamma_1$ that has a narrow spectrum centered at $\bar{\omega} = \omega_0$. The combined effect of the amplifying and the absorbing resonances is such that there is no net energy exchange after the passage of the entire pulse. Nevertheless, as is evident from Fig. 4(c), there is a strong transfer of energy from the early portion of the pulse to the medium that is offset nearly completely as the energy returns to the rear of the pulse. The effect is pronounced because the instantaneous spectrum accesses the absorptive spectral regions on both sides of center and finally narrows to the region of transmission[7] in which the absorption and the amplifying resonances offset each other. Although this example corresponds to strongly subluminal propagation, highly subluminal results depend on much narrower resonance structures than those used in this example.

7. DISCUSSION

In the above examples, we have considered the energy density at a single point within the medium. As the pulse passes through this point, the medium exchanges energy with the electromagnetic field, which is the mobile form of energy associated with the Poynting flux. This specific point in the medium can thereafter interact with only future portions of the pulse, the opportunity for interaction with the earlier portions having permanently passed. As the pulse continues to propagate, it is further modified at each subsequent point in the medium. In the case of superluminal behavior in an amplifying medium, what was once the far leading wing of, say, a Gaussian can grow into a hump resembling the original Gaussian, whereas the old body of the pulse eventually diminishes into the trailing edge.

To find the temporal profile (including the arrival time, as specified by some criterion) of a pulse emerging from a medium of finite thickness, one must integrate Maxwell's equations. This integration can be accomplished exactly[12] through the standard Fourier decomposition of the electromagnetic temporal waveform into its spectral components at a point at which the waveform is known. Each spectral component is then propagated (phase shift and attenuation) to any other point (assuming a homogeneous medium) at which the waveform can be reconstructed exactly. We emphasize that the results presented here [i.e., Eqs. (10) and (11)] are in every way compatible with this picture. However, the result in no way substitutes for the full description of wave propagation. The instantaneous spectrum describes how a point in the medium responds to a given waveform's passing through it. In this paper, we have selected as examples waveforms under exotic conditions in which the pulse propagation is known to exhibit superluminal or highly subluminal behaviors over finite propagation distances (avoiding the mature-dispersion regime[16–18]).

As was described in our recent paper[19] and in exact agreement with Maxwell's equations, the time interval between the *temporal* center of mass of the Poynting flux as it arrives at two distinct points is given by a spectral average over group delay (the inverse of the group velocity) that is modified by a reshaping term if the spectral amplitude becomes altered during propagation. The emergence of the pulse can, indeed, be superluminal when reckoning is by the arrival of the Poynting flux that tracks only the field energy. For example, if the (superluminal) pulse described in Fig. 1 propagates an additional distance $\Delta r = 0.1c/\gamma$ along a particular direction the temporal center of mass of the Poynting flux downstream occurs after a delay of only $\Delta t = 0.07/\gamma$ (note that $\Delta r/\Delta t = 1.4c$). If the (superluminal) pulse described in Fig. 2 propagates a similar distance the temporal center of mass of the Poynting flux downstream occurs after a delay of $\Delta t = -7/\gamma$ (occurring at the second point before the first). These results are well known and have been described and experimentally authenticated by many authors in the narrow-band limit.[1–6,12]

As the field and the medium exchange energy, the tracking of the presence of the field energy can move dramatically even though the energy-transport velocity is modest (strictly luminal). The rapid appearance of a pulse downstream is merely an artifact of not recognizing the energy already present in the medium until it converts to the form of field energy, as governed by the instantaneous spectrum. The traditional group velocity is connected to this partial accounting of the energy, which is why it can become superluminal. Group velocity (or rather, a spectral average of its inverse) is intimately linked to the Poynting flux (even for wide bandwidths) and tracks in an average sense in which the *field* energy is manifest.

We note in passing that Eqs. (10) and (11) manifestly contain the well-known Sommerfeld–Brillouin result[13,3] that a sharp signal edge cannot transmit faster than c. If a pulse begins abruptly at time t_0 the instantaneous spectrum $E_t(\omega)$ clearly remains identically zero until that time. In other words, no energy can be extracted from the medium until the field energy from the pulse arrives. Because, as was pointed out in Section 2, the Cauchy–Schwartz inequality prevents the field energy from traveling faster than c, at no point in the medium can a signal front exceed c.

In summary, we have elucidated the role of the instantaneous spectrum in situations in which linear, causal dielectric media exchange energy with the front of a pulse differently than with the back. This perspective provides an intuitive understanding of superluminal and highly subluminal phenomena in the vicinity of absorbing and amplifying resonances. This intuition is distinct from but complementary to the perspective gained through an understanding of group velocity.

APPENDIX A

We provide here a derivation of Eqs. (10) and (11) (as developed by S. A. Glasgow). The derivation is for an isotropic linear causal dielectric with the susceptibility $\chi(\omega)$, independent of any particular model. A more general proof including the possibility of anisotropy and diamagnetic effects is given in Ref. 11. From Eqs. (7)–(9) the polarization in the medium can be written as

$$\mathbf{P}(t) = \int_{-\infty}^{\infty} \mathrm{d}t' \mathbf{E}(t') G(t - t'),$$

$$G(t) \equiv \frac{\epsilon_0}{2\pi} \int_{-\infty}^{\infty} \chi(\omega) \exp[-(i\omega t)] \mathrm{d}\omega. \quad \text{(A1)}$$

The Green's function $G(t)$ can be written as the sum of two parts, the first being associated with the real part of the susceptibility and the second being associated with the imaginary part, as

$$G(t) = G_{\mathrm{Re}}(t) + G_{\mathrm{Im}}(t), \quad \text{(A2)}$$

where

$$G_{\mathrm{Re}}(t) \equiv \frac{\epsilon_0}{2\pi} \int_{-\infty}^{\infty} \mathrm{Re}[\chi(\omega)] \exp[-(i\omega t)] \mathrm{d}\omega,$$

$$G_{\mathrm{Im}}(t) \equiv i \frac{\epsilon_0}{2\pi} \int_{-\infty}^{\infty} \mathrm{Im}[\chi(\omega)] \exp[-(i\omega t)] \mathrm{d}\omega. \quad \text{(A3)}$$

We now show that $G_{\mathrm{Re}}(t)$ and $G_{\mathrm{Im}}(t)$ are equal for $t > 0$ and equal but opposite for $t < 0$. To do this, we invoke the Kramers–Kronig relation[12]

$$\mathrm{Re}\, \chi(\omega) = \frac{1}{\pi} P \int_{-\infty}^{\infty} \frac{\mathrm{Im}\, \chi(\omega')}{\omega' - \omega} \mathrm{d}\omega'. \quad \text{(A4)}$$

The letter P in front of the integral indicates the principle part. Substitution of Eq. (A4), which embodies the principle of causality, into the expression for $G_{\mathrm{Re}}(t)$ yields

$$G_{\mathrm{Re}}(t) = \frac{\epsilon_0}{2\pi^2} \int_{-\infty}^{\infty} \mathrm{d}\omega' \, \mathrm{Im}\, \chi(\omega') P \int_{-\infty}^{\infty} \frac{\exp[-(i\omega t)] \mathrm{d}\omega}{\omega' - \omega}, \quad \text{(A5)}$$

where the ordering of the integration was reversed. The final integral in Eq. (A5) can be performed analytically,[20] and it yields

$$P \int_{-\infty}^{\infty} \frac{\exp[-(i\omega t)]}{\omega' - \omega} \mathrm{d}\omega = \begin{cases} i\pi \exp[-(i\omega')] & t > 0 \\ -i\pi \exp[-(i\omega' t)] & t < 0 \end{cases}. \quad \text{(A6)}$$

Equation (A5) then becomes

$$G_{\mathrm{Re}}(t) = \begin{cases} G_{\mathrm{Im}}(t) & t > 0 \\ -G_{\mathrm{Im}}(t) & t < 0. \end{cases} \quad \text{(A7)}$$

We are now able to express the polarization $\mathbf{P}(t)$ in terms of the electric field and only the imaginary part of $\chi(\omega)$. Then Eqs. (A1) become

$$\begin{aligned} \mathbf{P}(t) &= \int_{-\infty}^{t} \mathrm{d}t' \mathbf{E}(t') 2 G_{\mathrm{Im}}(t - t') \\ &= \frac{i\epsilon_0}{\pi} \int_{-\infty}^{\infty} \mathrm{d}\omega \, \mathrm{Im}[\chi(\omega)] \exp[-(i\omega t)] \\ &\quad \times \int_{-\infty}^{t} \mathrm{d}t' \mathbf{E}(t') \exp(i\omega t'), \end{aligned} \quad \text{(A8)}$$

where again we changed the order of integration. Note that the upper limit of integration was also changed to t because Eq. (A2) is zero for negative time arguments.

To evaluate Eq. (5), we require the time derivative of Eq. (A8):

$$\begin{aligned} \frac{\partial \mathbf{P}(t)}{\partial t} &= \frac{\epsilon_0}{\pi} \int_{-\infty}^{\infty} \mathrm{d}\omega \omega \, \mathrm{Im}[\chi(\omega)] \exp[-(i\omega t)] \\ &\quad \times \int_{-\infty}^{t} \mathrm{d}t' \mathbf{E}(t') \exp(i\omega t') \\ &\quad + \frac{\epsilon_0 \mathbf{E}(t)}{\pi} \int_{-\infty}^{\infty} \mathrm{d}\omega \, \mathrm{Im}\, \chi(\omega). \end{aligned} \quad \text{(A9)}$$

The final term in Eq. (A9) vanishes because $\mathrm{Im}\, \chi(\omega)$ is an odd function of frequency [given that $\mathbf{E}(t)$ and $\mathbf{P}(t)$ are both real]. In addition, the truncated Fourier transform in Eq. (A9) can be replaced with the definition equation (11). Then the exchange energy density given by Eq. (5) becomes

$$\begin{aligned} u_{\mathrm{exchange}}(t) &= 2\epsilon_0 \int_{-\infty}^{\infty} \mathrm{d}\omega \omega \, \mathrm{Im}\, \chi(\omega) \int_{-\infty}^{t} \frac{1}{\sqrt{2\pi}} \exp[-(i\omega t')] \\ &\quad \times \mathbf{E}(t') \cdot \mathbf{E}_{t'}(\omega) \mathrm{d}t', \end{aligned} \quad \text{(A10)}$$

where the order of integration was again changed.

If we note that $\exp[-(i\omega t)]\mathbf{E}(t)/\sqrt{2\pi}$ is the time derivative of the complex conjugate of Eq. (11) the exchange energy density can be rewritten as

$$u_{\text{exchange}}(t) = 2\epsilon_0 \int_{-\infty}^{\infty} \mathrm{d}\omega\omega \,\mathrm{Im}\,\chi(\omega) \times \int_{-\infty}^{\infty} \frac{\partial \mathbf{E}_{t'}^{*}(\omega)}{\partial t'} \cdot \mathbf{E}_{t'}(\omega)\mathrm{d}t'. \quad \text{(A11)}$$

Because u_{exchange} is a real quantity, it costs nothing to add its complex conjugate and divide by 2. The exchange energy density then becomes

$$\begin{aligned} u_{\text{exchange}}(t) &= \epsilon_0 \int_{-\infty}^{\infty} \mathrm{d}\omega\omega \,\mathrm{Im}\,\chi(\omega) \int_{-\infty}^{t} \left[\frac{\partial \mathbf{E}_{t'}^{*}(\omega)}{\partial t'} \cdot \mathbf{E}_{t'}(\omega) + \mathbf{E}_{t'}^{*}(\omega) \cdot \frac{\partial \mathbf{E}_{t'}(\omega)}{\partial t'} \right] \mathrm{d}t' \\ &= \epsilon_0 \int_{-\infty}^{t} \mathrm{d}\omega\omega \,\mathrm{Im}\,\chi(\omega) \int_{-\infty}^{\infty} \frac{\partial |E_{t'}(\omega)|^2}{\partial t'} \mathrm{d}t' \\ &= \epsilon_0 \int_{-\infty}^{\infty} \mathrm{d}\omega\omega \,\mathrm{Im}\,\chi(\omega) |E_t(\omega)|^2, \quad \text{(A12)} \end{aligned}$$

and Eq. (10) is verified. This formula is well known[21] for $t = +\infty$, in which case it applies regardless of whether the medium behaves casually. The injection of causality can be made directly to the formula by the truncation of the Fourier transform at the current time. We have shown that this is consistent with the Kramers–Kronig relations.

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