Can precursors improve the transmission of energy at optical frequencies?

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The recent interest in precursors has been fuelled by the possibility of using them for the efficient transmission of information through absorbing media at radio or optical frequencies. Here we demonstrate that the low attenuation experienced by the Brillouin precursor is attributed to the inherently low absorption of dispersive media near DC, a characteristic already exploited with communications systems using the extremely low frequency (ELF) band. Pulses, regardless of their temporal width and carrier frequency, always obey Beer’s law as long as they propagate in the linear time invariant regime. We conclude with an FDTD simulation of the Maxwell–Bloch equations that shows how optical coherent bleaching effects, which take place in the linear time variant regime of the Lorentz oscillator model, can cause sustained deviations from Beer’s law over relatively long distances of water.

Keywords: anomalous absorption; Brillouin precursors; transients; coherent optics

1. Introduction

Absorption and scattering are two major hurdles impeding the transmission of signals through media. Discovering a method of sending signals through realistic media with less absorption has obvious implications for a plethora of applications, ranging from improved medical imaging to efficient communication through water.

The inherently low absorption of dispersive media near the DC frequency has justified, some years ago, the development of communication systems using the extremely low frequency (ELF) band. Unlike typical radio frequencies employed for communication purposes, ELF waves have been shown to propagate with little absorption over large distances in highly absorptive media, such as water and the Earth’s crust [1]. However, building and operating the large scale antennas required for ELF waves is a significant challenge that can be addressed for a limited range of applications only.

As a result, effort has been dedicated to determining whether the temporal profile of pulses could affect absorption, an idea rooted in Brillouin’s work on precursors [2]. The hypothesis was that a pulse that has a rise time that is temporally shorter than the medium’s response time could propagate without being affected by the absorption process. This was suggested initially because all particles have a finite mass, which necessarily means that the absorption process should not occur instantaneously [3,4]. Hence, the advent of the ultra wide band (UWB) impulse radar was the impetus to significant work in the RF regime to determine if precursors could efficiently penetrate foliage [3,5–10]. The enthusiasm on precursors has reached the optical regime, with recent experiments investigating whether femtosecond pulses in the near infrared region could be used for an ultra-efficient communications link under water [11–18]. Simulations and observations for other media and frequency regimes are also available [19–21].

Yet, despite the available literature on the topic, opinions differ on whether precursors really do improve energy transfer beyond Beer’s law [3,4,6–9,11–13,15–18,22]. This paper offers a clear argument to demonstrate that pulses, regardless of their temporal width, always obey Beer’s linear absorption law as long as they propagate in the linear time invariant regime. However, the law can be violated when pulses propagate in the time variant regime of the Lorentz oscillator model, which includes coherent and incoherent bleaching effects.

To demonstrate how to improve the energy transmission at optical frequencies, this paper first revisits Brillouin’s work on precursors using well-known physical principles [2]. We highlight how signal energy always decays according to a linear Beer’s law if pulse propagation occurs in the linear time invariant regime. This will illustrate how dispersive media’s inherent lack of absorption at DC provides convenient medium conditions for precursors.

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(and multi-exponential decay), a characteristic already exploited with communication in the ELF band [1]. Finally, we demonstrate how sub-Beer law attenuation is theoretically possible in practical media using bleaching effects.

2. Definition of Beer’s law

To insure consistency among different reader’s understanding of Beer’s law, we recall that monochromatic Beer’s law describes the rate at which energy of a monochromatic and continuous signal attenuates according to distance. The intensity of the signal attenuates exponentially according to:

\[ I(z) = I_0 \exp(-\alpha z), \]

where \( I_0 \) is the initial intensity of the wave, \( z \) is the distance, \( \alpha \) is the attenuation coefficient and \( I(z) \) is the intensity of the wave at distance \( z \).

Of course, most signals are not purely monochromatic and so monochromatic Beer’s law is no longer appropriate to describe energy attenuation. Instead, one should use a broadband Beer’s law which is effectively the sum of many exponential decays corresponding to each frequency component in the input waveform [23]:

\[ I(z) = \int I_0(\omega) \exp(-\alpha_\omega(z)\omega) \, d\omega. \]

It is clear that both of these Beer’s laws are effectively the same – the latter simply takes into account attenuation of a broadband signal, and turns into monochromatic Beer’s law in the limit of a single frequency signal. Therefore, when we speak of deviations from Beer’s law, we speak of deviations from both of these laws, which is when the absorption coefficient \( \alpha \) actually changes.

3. A qualitative description of Brillouin’s 1914 analysis

This section demonstrates that precursors are the result of pulse break-up that occurs because of the different group velocities associated with the distinct frequencies present in the pulse. We demonstrate this point by using a method similar to Jackson’s [24], but where we explicitly highlight the need for off-resonance energy to form precursors.

Let us begin by recalling the importance of group delay for precursor generation by considering the usual linear time-invariant propagation integral:

\[ E_{\text{out}}(z, t) = \int E_{\text{in}}(0, \omega) \exp[-i(\omega)\omega z/c] \exp(i\omega t) \, d\omega, \]

where \( E_{\text{out}}(z, t) \) is the output waveform after a distance \( z \), \( E_{\text{in}}(0, \omega) \) is the input spectrum, \( n(\omega) \) is the complex refractive index and \( \omega \) is the angular frequency. The equation shows that the temporal waveform at the output is the inverse Fourier transform of the input field spectrum \( E_{\text{in}}(0, \omega) \) modulated by a transfer function \( H(\omega) = \exp[-i(n(\omega)\omega z/c)]. \)

The magnitude and phase terms of this modulation function dictate the temporal evolution of the pulse as it propagates through the medium. The magnitude term \( |H(\omega)| \) determines how quickly the energy at the different frequencies of \( E_{\text{in}}(0, \omega) \) is attenuated. In other words, it modulates the input spectrum \( E_{\text{in}}(0, \omega) \) into the spectrum \( E_{\text{out}}(z, \omega) \) of the temporal output. On the other hand, the time required for groups of frequencies in the initial spectrum to propagate a distance \( z \) is given by the group delay \( r_g = -\partial H(\omega)/\partial \omega \). Pulse break-up leading to precursors happen if there is significant group delay dispersion (GDD), where \( \text{GDD} = \partial^2 r_g/\partial \omega^2 \).

For instance, Figure 1 plots the magnitude and group delay at \( z = 100 \text{ nm} \) of the single-Lorentzian

![Figure 1. Absorption coefficient (blue dashed) and group delay at \( z = 100 \text{ nm} \) of the single-Lorentzian](https://example.com/figure1.png)
medium originally considered by Brillouin, whose refractive index $n(\omega)$ is such that:

$$n(\omega) = \left(1 + \frac{\omega_p^2}{\omega_0^2 - \omega^2 - i\gamma}\right)^{1/2},$$  \hspace{1cm} (4)

where $\omega_0 = 4.47 \times 10^{16}$ rad s$^{-1}$, $\omega_p = 0.9 \times 10^{16}$ rad s$^{-1}$, $\gamma = 0.28 \times 10^{16}$ rad s$^{-1}$. The low positive group delay at high frequencies suggests that these frequencies in a spectrally broad signal are first to arrive at the output. The slightly larger group delay in the low-frequency region implies that a group of low-frequencies follow. As pointed out by Jackson [24], the resonant portion of a broadband signal is the last to arrive. Pulse break-up occurs if the frequency-dependent group delay varies sufficiently rapidly with frequency.

For the signal originally considered by Brillouin in 1914 (the truncated sinusoidal tuned to the medium resonance), the spectral distribution gives rise to pulse break-up characterized by three distinct features in the temporal output. In order of arrival, the features are the high-frequency precursor (the Sommerfeld), the low-frequency precursor (the Brillouin) and the main signal.

Pulse break-up for a sinusoidal signal could seem counter-intuitive because pulse break-up requires a broad spectrum. Yet, a truncated sinusoidal, such as the one considered by Brillouin in his original analysis is in fact, technically speaking, broadband. Indeed, the presence of a discontinuity implies that some energy must necessarily exist at many frequencies. A succinct method of demonstrating this point mathematically is to compute the Fourier transform of Brillouin’s signal, $E_{in}(t) = \delta(t) \cos(\omega_0 t)$, where $\delta(t)$ is the unit step function and $\omega_0$ is the resonant frequency of the medium. Energy is naturally present at frequencies far from $\omega_0$, as can be inferred because of the $\omega$ dependence of the Fourier transform’s last term:

$$E_{in}(\omega) = \frac{\pi}{2} \left[\delta(\omega - \omega_0) - \delta(\omega + \omega_0)\right] + \frac{i\omega}{\omega_0^2 - \omega^2}.$$  \hspace{1cm} (5)

It is this on-resonance energy that eventually dominates the heavily absorbed on-resonance energy to form precursors. Indeed, pulse break-up leading to precursors arises when (1) the frequency-dependent group delay varies so that frequency groups travel at different speeds, (2) when absorptions burns a hole in the spectrum (otherwise, only pulse smearing occurs), and (3) when the signal has an energy spectrum that is wide enough to span the region of GDD and frequency dependent absorption. When these conditions are met, the different frequency groups eventually acquire contrasting enough delay that distinct waveforms emerge in the temporal output. Combined with the fact that the energy associated with frequencies off-resonance are exposed to lesser absorption, these emerging waveforms (the precursors) eventually begin to dominate the temporal output. Therefore, this section highlights the fact that precursors are a consequence of linear dispersion theory, and thus obey linear Beer’s law of absorption.

4. Precursors generated with a Gaussian spectrum near DC

Describing any type of on-resonance propagation in physical terms is complicated by the inability to use the well-known metric of group velocity. Additionally, the spectral content of the truncated signal (its Fourier transform, Equation (5)) is expressed mathematically with the delta function, a generalized function that has little to no physical meaning.

These hurdles can be circumvented by demonstrating that precursors are easily generated with a temporal waveform whose spectrum is well defined and whose carrier frequency is not equal to the medium resonance. Note that we consider the main signal in this situation to be the one whose energy consists of spectral energy located around the carrier frequency. This choice of parameters conveniently allows us to speak of a main signal exposed to positive group delay. The portion of the spectrum which is indeed exposed to negative group velocities can conveniently be ignored, since its contribution to the temporal output is eliminated by the large absorption of the resonant region. The only consequence of note in arranging this situation is that the main signal has now a smaller group delay than the spectrum’s low-frequency tail. The resulting low-frequency ‘precursor’ therefore lags the main portion of the signal, an observation that is consistent with work by Oughstun and Balictsis [25]. For the sake of historical convention and consistency with previous literature, we continue to use the word precursor to describe any waveform that detaches itself from the main portion of the signal to eventually dominate it because of less absorption. This is despite the fact that the word ‘post-cursor’ might be more descriptive [26,27].

For example, consider a signal with a Gaussian spectrum centered at $\omega_c = 2$ rad s$^{-1}$ (a unitary speed of light $c = 1$ m s$^{-1}$ is used to scale the frequencies to single digits for clarity). The medium under consideration is a single Lorentzian with parameters $\omega_p = 5$ rad s$^{-1}$, $\mu = 0.25$ rad s$^{-1}$ and $\omega_0 = 0.9$ rad s$^{-1}$. We normalize to a unitary speed of light ($c = 1$ m s$^{-1}$) for simplicity. Figure 2 shows the spectral content and frequency-dependent group delay for different propagation distances $z$. It is clear that absorption in the
medium causes two frequency structures to emerge from the spectrum, where the low-frequency peak dominates for larger $z$. In addition, the increasing contrast in group delay associated with each frequency structure implies that their temporal separation becomes greater. Figure 3 shows the temporal signal associated with different propagation distances $z$, where it is evident that a low-frequency waveform emerges from the main signal. For reference purposes, the dot represents the delay given by $\tau_p = z \times \epsilon/(n(0))$.

Since the attenuation property of precursors has been their appeal for practical purposes, we consider the decay of different quantities describing the pulse of this section by plotting (1) the pulse area of the output field (red), (2) the maximum amplitude of the output field (blue), (3) the overall intensity of the output field (black), and (4) the normalized $1/z$ and $1/z^{1/2}$ algebraic decays (purple and green). The pulse area is included here because it is a useful variable in the semi-classical description of absorption, a quantity often used to describe the energy threshold of coherent bleaching [28]. It is given by $\theta = (\mu/h) \int |E(t,z)| \, dt$ where $\mu$ is the dipole moment, $h$ is Planck’s constant and $|E(t,z)|$ is the field envelope.

The sharp discontinuity present in the maximum field amplitude plot at $z \approx 2$ m corresponds to the point at which the precursor amplitude begins to dominate the temporal output. What is apparent is that the maximum field amplitude follows a $1/z$ decay. This agrees with the simulations of precursor propagation in a semiconductor with a Gaussian input by Ni and Alfano [19]. Although this rate differs from the historical $1/z^{1/2}$ decay calculated by Brillouin, it should be noted that Brillouin’s decay rate has since been revised with more advanced mathematical methods, as discussed by Oughstun and Sherman [29].

The salient feature of Figure 4 is that while the signal’s overall intensity (as would be measured in a laboratory with a square-law detector [13]) decays more rapidly than either of the $1/z$ or $1/z^{1/2}$ algebraic decays, it decays non-exponentially nonetheless. This is certainly not a Beer’s law violation. Rather, it is the result of the individual frequencies present in the pulse being exposed to the medium’s frequency-dependent absorption coefficient. This is precisely the linear broadband Beer’s law alluded to by Alfano et al. and discussed in more detail in Gibson and Österberg [18,23].

Figure 2. The field spectrum and frequency-dependent group delay for propagation distances $z$. The low absorption near DC causes the temporal output to be dominated by a low-frequency waveform, whose origins are identical to the ones of the Brillouin precursor. (The color version of this figure is included in the online version of the journal.)
5. Discussion

The linear time invariant assumption inherent to the propagation integral used to describe precursors, Equation (3), means that any waveform that emerges from a dispersive medium consists of frequencies that were present in the original pulse. This is regardless of their initial position with respect to each other (phase/chirp). Thus, for a given spectrum $E(\omega)$ propagating through a dispersive medium, absorption is always the same regardless of whether the phases of the different frequency components in the spectrum are constant with respect to each other (e.g. a femtosecond pulse from an ultrafast laser) or randomly varying (e.g. a cw incoherent flashlight). While the temporal envelopes at the output will necessarily be different for two light sources with identical spectrum but different phase, it is a physical and mathematical certainty that the amount of absorbed energy will be identical. This observation agrees with a comment made by Roberts where the author demonstrated the importance of linear time invariance by making use of Parseval’s theorem for a bandwidth limited pulse, Equation (6), [30].

$$\int |E_{\text{out}}(z, t)|^2 \, dt \leq \int |E_{\text{in}}(0, \omega)|^2 \exp \left[ -\alpha_{\text{min}} z \right] \, d\omega.$$  \hspace{1cm} (6)
Therefore, the best possible transmission possible in the linear time-invariant regime follows from intuition: it is achieved by using a light source composed of frequencies that are least susceptible to absorption.

As we have shown, precursors do exist under certain conditions, but special care should be given to their potential for increased depth penetration in a practical application. Indeed, the slow decay rate of the Brillouin precursor waveform is attributable to the zero absorption at DC frequency inherent to dispersive media. This is precisely the characteristic exploited by the US military for communication with underwater vehicles using the extremely low frequency (ELF) and ultra low frequency (ULF) band [1,31]. From our analysis, it is apparent that both the slowly attenuating Brillouin precursor and ELF/ULF waveforms rely on the natural property of dispersive media being less absorptive near DC. Therefore, the answer to this paper’s title is ‘no’.

(Note that while this paper uses the Lorentz oscillator model for absorption because of previous work performed in the optical regime, the argument could be translated to the Drude and Debye absorption models for other frequency regimes.)

Nevertheless, deviations from Beer’s law in the optical regime are possible if pulse propagation occurs in the time-variant regime of the Lorentz oscillator model. This regime is modelled with the coupled Maxwell–Bloch equations

\[
\frac{\partial \rho_1}{\partial t} = -\frac{1}{T_2^1} \rho_1 + \omega_0 \rho_2,
\]

\[
\frac{\partial \rho_2}{\partial t} = -\frac{1}{T_2^2} \rho_2 - \omega_0 \rho_1 + \frac{2\mu}{\hbar} E \rho_3,
\]

\[
\frac{\partial \rho_3}{\partial t} = -\frac{1}{T_1} (\rho_3 - \rho_{30}) - \frac{2\mu}{\hbar} E \rho_2,
\]

where \(\mu\) represents the dipole moment of the medium, \(E\) is the incident electric field, \(\rho_{30}\) is the initial state of inversion for the population, and \(\rho_1\), \(\rho_2\), and \(\rho_3\) represent the dispersive (in-phase) polarization, the absorptive (in-quadrature) polarization, and the inversion parameter of the two-level system approximation of the medium, respectively. A value \(\rho_3 = -1\) represents an atomic system where all atoms are in their ground state, and \(\rho_3 = 0\) one that is completely transparent. This can be seen from the quantum mechanical description of the absorption coefficient for an inhomogeneously broadened medium, \(\omega(t, z; \rho_3) \approx \alpha_0 \rho_3(t, z)\), [28].

A pulse with enough energy to cause atomic inversion can sustain Beer’s law deviations over relatively large distances if its temporal width is shorter or comparable than the medium dephasing time \(T'\) [39]. This process, known as coherent bleaching, occurs when the medium could store energy momentarily before returning it to the pulse via stimulated emission. The effect of Self-Induced Transparency, whereby a pulse induces complete transparency in the medium, occurs in the limit that the pulse is much shorter than the dephasing time \(T'\).

To demonstrate the potential of coherent bleaching, in general, for transmitting energy more efficiently through water (e.g. for communication and imaging), a computer program was written to solve the Maxwell–Bloch equations numerically with an FDTD technique combined with a predictor-corrector iterative method, [32]. The input parameters of the simulation were chosen to simulate a 52 fs sech^2 pulse (intensity profile) propagating on the 2.94 \(\mu\)m resonance of water. This resonance was chosen because it is a strong and well-defined absorption peak in the water spectrum, and because its dephasing lifetimes are well documented in the literature, \(T_1 = 1\) ps and \(T' = 10\) fs, [33,34]. We assume that the experimentally measured transition dipole \(\mu = 6.2 \times 10^{-30}\) C m obtained by Callegari et al. [35] for the (4, 0) transition (739 nm) is representative of the transition dipole for the 2.94 \(\mu\)m resonance.

Figure 5 shows the evolution of the pulse when its peak intensity is 2.31 \(\text{W cm}^{-2}\), which is low enough that the pulse only induces negligible inversion (\(\rho_3 \approx 1\)). The pulse decays according to linear \((\text{broadband})\) Beer’s law. Note that an absorption length is approximately 3.4 \(\mu\)m at this wavelength [36]. Since the absorption line is narrower than the pulse spectrum, absorption leads to a hole in the spectrum and therefore temporal oscillations in the pulse. Note that this is one of the three conditions listed in Section 3 that must be satisfied to give rise to precursors. In addition, it should be highlighted that the pulse that evolves from this scenario is precisely the \(0\)–\(\pi\) pulse described in thorough detail by Crisp [37].

The simulation is then repeated for identical conditions except that the input intensity is increased to 760 \(\text{GW cm}^{-2}\). This intensity level is still below water’s breakdown threshold, which is on the order of 10 \(\text{TW cm}^{-2}\) for 100 fs pulses [38]. As shown in Figure 6, coherent bleaching causes the pulse to propagate with negligible absorption thus illustrating how linear Beer’s law, monochromatic or broadband, could be violated over relatively long distances in the linear time invariant regime of the Lorentz oscillator model.

While the intensity used in this simulation is below water’s breakdown threshold, current typical amplified ultrafast lasers could only provide such elevated intensities by focusing the beam to small spot sizes and therefore for short distances. For instance,
a Spectra Physics OPA-800C can provide femtosecond pulses of 50 μJ energy at the 2.94 μm wavelength when fed with a Spectra Physics Pro F1K system with femtosecond pulses of 1 mJ energy, 1 kHz repetition rate at 800 nm. Assuming a pulse width of 50 fs, the peak power of 1 GW can yield the required intensity of 760 GW cm\(^{-2}\) by focusing the beam to a spot with a radius of 205 μm. This corresponds to a Rayleigh length of 4.5 cm.

Given this limitation, exploitation of coherent bleaching for decreased attenuation in long distance communications channels is contingent upon the development of sufficiently intense ultrafast pulsed laser technology.

6. Conclusion
This paper highlighted how precursors are simply the result of a pulse break-up, and hence do not violate Beer’s law since they take place in a regime where the output field is both linear and time-invariant with respect to the input field. Therefore, the answer to the title of this paper is ‘no’. However, sustained deviations from Beer’s law of absorption can occur through coherent bleaching effects using sufficiently short and intense laser pulses interacting with a medium resonance.

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