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II.
Pulse-Mode Expansions and Refractive Indices in Plane-Wave Propagation

B. W. Shore, R. Sacks, T. Karr, J. Morris and J. A. Paisner

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Abstract

This memo presents basic background theory for treating simultaneous propagation of electromagnetic pulses of various colors, directed along a common ray, through a molecular vapor. The memo discusses some techniques for expanding the positive frequency part of the transverse electric field into pulse modes, characterized by carrier frequencies within a modulated envelope. We discuss, in the approximation of plane waves with slowly varying envelopes, a set of uncoupled envelope equations in which a polarization mode-envelope acts as a source for an electric-field envelope. These equations, when taken with a prescription for the polarization field, are the basic equations of plane-wave pulse propagation through a molecular medium.

We discuss two ways of treating dispersive media, one based upon expansions in the frequency domain and the other based in the time domain. In both cases we find envelope equations that involve group velocities.

This memo represents a portion of a more extensive treatment of propagation to be presented separately. Many of the equations presented here have been described in various books and articles. They are collected and described here as a summary and review of contemporary theory.

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§II.4 Introduction and Summary

The propagation of electromagnetic waves through non-magnetic material can be described by two fields, the electric field $E(r,t)$ and the polarization field $P(r,t)$. It is the solenoidal (or transverse) portion of the electric field that constitutes the propagating radiation, and the solenoidal part of the polarization field acts as a source for this field. The solenoidal parts of these fields obey the wave equation

$$\left[\nabla^2 - \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \right] E(r,t) = \frac{4\pi}{c^2} \frac{\partial^2}{\partial t^2} P(r,t). \quad (\text{II.4-1})$$

The polarization field describes the density of dipole moments, and so it expresses the behavior of the medium through which the field propagates. It is to be determined from some appropriate model of the medium, including the response of the molecules of the medium to the radiation field as it propagates. Both of these fields can be decomposed into positive and negative frequency components, in the form

$$E(r,t) = E^{(+)}(r,t) + E^{(-)}(r,t). \quad (\text{II.4-2})$$

We here examine further decomposition of the fields, with intended application to the description of pulses of radiation.

Our basic result will be a set of envelope equations,

$$\frac{\partial}{\partial z} \mathcal{E}_\lambda(z,r) = \frac{2\pi i}{c} \omega_\lambda \mathcal{P}_\lambda(z,r) \quad (\text{II.4-3})$$

in which a polarization envelope $\mathcal{P}_\lambda$ acts as a source for an electric-field envelope $\mathcal{E}_\lambda$. The pulse envelope argument r is a local time, moving with the pulse (the precise connection between r and t and z will be defined below). These two envelope functions represent functions that occur in the expansion of the positive frequency parts of the transverse electric field and the transverse polarization field into travelling pulses, as in the expression

$$E^{(+)}(r,t) = \frac{1}{2} \int_{\lambda} \mathcal{E}_\lambda(r,r) \exp[ik_\lambda \cdot r - i\omega_\lambda t]. \quad (\text{II.4-4})$$

A corresponding expansion applies for the polarization field. The symbol $\int_{\lambda}$ appear-

ing here denotes a summation over discrete variables, such as two polarization directions, and an integral over continuum variables, if any. The label λ denotes the parameters (polarization, frequency, and propagation direction) needed to identify a mode.

Fourier Expansions

The simplest and most direct approach introduces a Fourier expansion both in space and in time: we introduce a continuum of wavevectors $\mathbf{k}$ and a continuum of frequencies ω , together with unit vectors $\mathbf{e}(\mu, \mathbf{k})$, and write the positive-frequency field as a Fourier integral, in space and time, in the form

$$\mathbf{E}^{(+)}(\mathbf{r}, t) = \frac{1}{2} \sum_{\mu} \int d\mathbf{k} \int d\omega \tilde{\mathcal{E}}(\mu, \mathbf{k}, \omega) \mathbf{e}(\mu, \mathbf{k}) \exp(i\mathbf{k} \cdot \mathbf{r} - i\omega t). \quad (\Pi.4-5)$$

The function $\tilde{\mathcal{E}}(\mu, \mathbf{k}, \omega)$ is the Fourier coefficient for polarization μ . To simplify expressions we abbreviate the labels of polarization μ , propagation $\mathbf{k}$ and frequency ω by the single label λ , writing in place of Eqn. (II.4-5) the equation

$$\mathbf{E}^{(+)}(\mathbf{r}, t) = \left[\int_{\lambda} \right] \mathbf{E}^{(+)}_{\lambda}(\mathbf{r}, t) = \frac{1}{2} \left[\int_{\lambda} \right] \tilde{\mathcal{E}}(\lambda) \mathbf{e}(\lambda) \exp(i\mathbf{k}_{\lambda} \cdot \mathbf{r} - i\omega_{\lambda} t). \quad (\Pi.4-6)$$

The function $\tilde{\mathcal{E}}(\lambda)$ is the Fourier coefficient for mode λ . We make a similar decomposition of the polarization field,

$$\mathbf{P}^{(+)}(\mathbf{r}, t) = \left[\int_{\lambda} \right] \mathbf{P}^{(+)}_{\lambda}(\mathbf{r}, t) = \frac{1}{2} \left[\int_{\lambda} \right] \tilde{\mathcal{P}}(\lambda) \mathbf{e}(\lambda) \exp(i\mathbf{k}_{\lambda} \cdot \mathbf{r} - i\omega_{\lambda} t). \quad (\Pi.4-7)$$

In these expansions the vector properties of the fields are expressed by unit vectors $\mathbf{e}(\lambda)$, possibly complex:

$$|\mathbf{e}(\lambda)|^2 = \mathbf{e}(\lambda) \cdot \mathbf{e}(\lambda)^* = 1. \quad (\Pi.4-8)$$

These vectors are assumed to be perpendicular to the propagation direction $\mathbf{k}_{\lambda}$, in order that the fields be solenoidal,

$$\mathbf{k}_{\lambda} \cdot \mathbf{e}(\lambda) = 0, \quad (\Pi.4-9)$$

but they are otherwise arbitrary. They may be helicity vectors or, more generally, vectors of elliptical polarization. For each propagation direction there exist a pair of independent basis vectors. The superposition formula presents an integral over wavevectors and frequencies and a sum over polarizations.

The Fourier expansion, taken with the wave equation, produces a set of algebraic equations

$$\left[(\mathbf{k}_{\lambda})^2 - \frac{(\omega_{\lambda})^2}{c^2} \right] \tilde{\mathcal{E}}(\lambda) = 4\pi \frac{(\omega_{\lambda})^2}{c^2} \tilde{\mathcal{P}}(\lambda). \quad (\Pi.4-10)$$

In the absence of polarization, $\tilde{\mathcal{P}}(\lambda) = 0$, this equation has solutions for arbitrary $\tilde{\mathcal{E}}(\lambda)$,

so long as the magnitude ck_λ and ω_λ are related by the simple dispersion relation

$$c^2(k_\lambda) = (\omega_\lambda)^2. \quad (\text{II.4-11})$$

The two solutions to this quadratic equation for k_λ correspond to forward (or outward) travelling waves and backward (or inward) travelling waves. The second-order wave equation allows both such solutions.

Whereas the expansion of the electric field represents an analysis of a solution to Maxwell's equations, the polarization field expansion must incorporate properties of the molecules. When we deal with propagation involving single-photon transitions the polarization field may be regarded as a given field, although it must, in fact, be determined from a model of molecular behavior consistent with the presence of the electric field. However, when we deal with Raman transitions the polarization field is explicitly directly proportional to combinations of electric fields. (It is also implicitly dependent upon the fields via molecular excitation). Under these circumstances the polarization field depends both upon molecular properties and upon the fields. Consequently the two situations require slightly different treatments. The simplest case occurs when $\mathcal{P}(\lambda)$ may be considered independent of $\mathcal{E}(\lambda)$, for then the electric field may be obtained as the Fourier integral

$$E^{(+)}(\mathbf{r}, t) = 2\pi \int_{\lambda} (\omega_\lambda)^2 \tilde{\mathcal{P}}(\lambda) e(\lambda) \frac{\exp(i\mathbf{k}_\lambda \cdot \mathbf{r} - i\omega_\lambda t)}{(ck_\lambda + \omega_\lambda)(ck_\lambda - \omega_\lambda)} \quad (\text{II.4-12})$$

We can recognize the occurrence of forward and backward travelling waves in this expression, originating from the evaluation of the Fourier integral at the two poles, $ck_\lambda = \pm \omega_\lambda$. When we consider only forward travelling waves we neglect one of these poles. Such an approximation is equivalent to replacing the second-order differential equation by a first-order equation.

Pulse Mode Expansions

The Fourier expansion places all of the space and time variation into an exponential phase, the function $\xi_\lambda = \mathbf{k}_\lambda \cdot \mathbf{r} - \omega_\lambda t$ that is given *a priori*. The description of propagation then becomes a set of linear algebraic equations for the Fourier coefficients $\mathcal{E}(\lambda)$. When one deals with short pulses it often proves useful to incorporate part of the field variation into an envelope function $\mathcal{E}_\lambda(\mathbf{r}, t)$, by writing an expansion of the form

$$E^{(+)}(\mathbf{r}, t) = \sum_{\lambda} E^{(+)}_{\lambda}(\mathbf{r}, t) = \frac{1}{2} \sum_{\lambda} \mathcal{E}_{\lambda}(\mathbf{r}, t) e(\lambda) \exp(i\xi_{\lambda}) \quad (\text{II.4-13a})$$

$$P^{(+)}(\mathbf{r}, t) = \sum_{\lambda} P^{(+)}_{\lambda}(\mathbf{r}, t) = \frac{1}{2} \sum_{\lambda} \mathcal{P}_{\lambda}(\mathbf{r}, t) e(\lambda) \exp(i\xi_{\lambda}). \quad (\text{II.4-13b})$$

This expansion represents the total electric field (at point $\mathbf{r}$ and time t) as a superposition of a set of pulse fields (or *pulse modes*) indexed by the symbol λ . The factors in

this expansion have the following significance. As with the Fourier expansion, the unit vectors $\mathbf{e}(\lambda)$ express the polarization properties of the field. The dominant dependence upon position $\mathbf{r}$ and time t is presumed to be contained in the carrier-wave phase $\xi_\lambda \equiv \xi_\lambda(\mathbf{r}, t)$. The propagation vector $\mathbf{k}_\lambda$ and (instantaneous) frequency ω_λ are defined as space and time derivatives of the phase ξ_λ :

$$\mathbf{k}_\lambda = \nabla \xi_\lambda, \quad \omega_\lambda = -\frac{\partial}{\partial t} \xi_\lambda. \quad (\text{II.4-14})$$

The pulse amplitudes $\mathcal{E}_\lambda(\mathbf{r}, \tau)$ and $\mathcal{P}_\lambda(\mathbf{r}, \tau)$, possibly complex, contain the remainder of the time and space dependence. We have introduced a local time τ as an argument of these envelopes, anticipating that it will prove convenient to introduce some such change of variables. For the moment we need only assume that there exists a functional relationship between τ and the variables $\mathbf{r}, t$.

When describing atomic excitation at fixed position it sometimes proves useful to write the expansion in the form of a temporal Fourier integral,

$$\mathbf{E}^{(+)}(\mathbf{r}, t) = \frac{1}{2} \sum_{\lambda} \int d\omega \tilde{\mathcal{E}}_{\lambda}(\mathbf{r}, \omega) \mathbf{e}(\lambda) \exp(-i\omega t) \quad (\text{II.4-15a})$$

$$\mathbf{P}^{(+)}(\mathbf{r}, t) = \frac{1}{2} \sum_{\lambda} \int d\omega \tilde{\mathcal{P}}_{\lambda}(\mathbf{r}, \omega) \mathbf{e}(\lambda) \exp(-i\omega t). \quad (\text{II.4-15b})$$

The Fourier coefficient $\tilde{\mathcal{E}}_{\lambda}(\mathbf{r}, \omega)$ that appears here varies with spatial location. Because it describes a pulse, it is centered in frequency within some bandwidth $\Delta\omega_\lambda$ about some mean frequency ω_λ . By writing an expansion in discrete pulse modes we assume that the Fourier transform of the electric field at any position can be unambiguously separated into a succession of frequency distributions whose separation is much larger than the bandwidth. When this assumption fails, so that the individual pulses become confused in the frequency domain, it is not so useful to introduce pulse mode expansions.

The Magnetic Field and the Poynting Vector

We shall assume, as is customary, that the exponential phase ξ_λ incorporates the most rapidly changing dependence of the fields upon space and time. Under this assumption the positive frequency part of the magnetic field $\mathbf{B}(\mathbf{r}, t)$ is approximated by the expansion

$$\begin{aligned} \mathbf{B}^{(+)}(\mathbf{r}, t) &= \sum_{\lambda} \mathbf{B}^{(+)}_{\lambda}(\mathbf{r}, t) \\ &= \frac{1}{2} \sum_{\lambda} \frac{ck_{\lambda}}{\omega_{\lambda}} \mathcal{E}_{\lambda}(\mathbf{r}, \tau) [\hat{\mathbf{k}}_{\lambda} \times \mathbf{e}(\lambda)] \exp(i\xi_{\lambda}). \end{aligned} \quad (\text{II.4-16})$$

Here $\hat{k}_\lambda$ is a unit vector along the propagation vector k_λ . This expansion produces a solution to Maxwell's equation

$$\nabla \times \mathbf{E} + \frac{1}{c} \frac{\partial}{\partial t} \mathbf{B} = 0 \quad (\text{II.4-17})$$

when the spatial and temporal derivatives of the envelope function are much smaller than the corresponding changes of the phase ξ_λ . From this expansion we can evaluate the time averaged Poynting vector,

$$\langle \mathbf{S} \rangle = \frac{c}{4\pi} \langle \mathbf{E} \times \mathbf{B} \rangle = \frac{c}{2\pi} \langle \mathbf{E}^{(-)} \times \mathbf{B}^{(+)} \rangle. \quad (\text{II.4-18})$$

Here $\langle \dots \rangle$ denotes a time average. The absolute magnitude of this average is the radiation intensity. The time average

$$\langle \exp(i\xi_\lambda - i\xi_{\lambda'}) \rangle$$

picks out modes with common frequency. Let us consider waves that, for given carrier frequency, form a well collimated beam, so that the spatial phase difference in this exponential is negligible. Different polarization vectors, for common propagation direction, are orthogonal. We therefore obtain, for the radiation intensity, the expression

$$I(\mathbf{r}, t) = \sum_{\lambda} I_{\lambda}(\mathbf{r}, t) = \sum_{\lambda} \frac{c}{8\pi} \frac{ck_{\lambda}}{\omega_{\lambda}} |\mathcal{E}_{\lambda}(\mathbf{r}, t)|^2 \quad (\text{II.4-19})$$

Pulse Phase

There are two general ways to define the pulse phase ξ_λ . The simplest approach is to require that k_λ and ω_λ be independent of time and position, so that all second derivatives of the phase ξ_λ vanish:

$$\frac{\partial^2}{\partial t^2} \xi_\lambda = 0, \quad \nabla^2 \xi_\lambda = 0, \quad \nabla \frac{\partial}{\partial t} \xi_\lambda = 0. \quad (\text{II.4-20})$$

That is, the phase can be written, just as with Fourier decomposition, as

$$\xi_\lambda = \mathbf{k}_\lambda \cdot \mathbf{r} - \omega_\lambda t, \quad (\text{II.4-21})$$

where $\mathbf{k}_\lambda$ and ω_λ are independent of position and time but may depend on the mode. This assumption still leaves the precise relationship between k_λ and ω_λ undefined; in general this connection can be written in the form

$$ck_\lambda = \eta(\omega_\lambda) \omega_\lambda \quad (\text{II.4-22})$$

thereby defining a (frequency dependent) index of refraction $\eta(\omega)$. Although the phase ξ_λ is the phase of Fourier analysis, the envelope expansion is not a Fourier expansion. The coefficients of the exponentials (the envelope functions $\mathcal{E}_\lambda$ and $\mathcal{F}_\lambda$) are here allowed to vary with position and time, whereas in a Fourier expansion they would be constants.

A more complicated approach allows small but nonzero second derivatives of the phase. That is, both the frequency ω_λ and the wavevector k_λ defined above are allowed to vary slowly with time and position. The slow variation may be expressed through the relationship

$$\left| \nabla^2 \xi_\lambda \right| \ll (k_\lambda)^2, \quad \left| \frac{\partial^2}{\partial t^2} \xi_\lambda \right| \ll (\omega_\lambda)^2, \quad \left| \nabla \frac{\partial}{\partial t} \xi_\lambda \right| \ll |k_\lambda \omega_\lambda|. \quad (\text{II.4-23})$$

With either of these choices of phase the envelope function $\mathcal{E}_\lambda(r, t)$ refers to a particular carrier frequency ω_λ , propagation direction $\mathbf{k}_\lambda$ and polarization direction $\mathbf{e}(\lambda)$. It is assumed to vary slowly over a time comparable to a carrier period (and hence over a spatial distance of a carrier wavelength). This is the *slowly varying envelope approximation* (SVEA).

It is important to keep in mind that, although the expansion of the electric field into pulse modes can be done with arbitrary definition of phase and amplitude, the polarization field is determined by microscopic properties. We must somehow fit those properties (the atomic dipole moment) to the form assumed for the polarization field. This is easier to do with the definition of Eqn. (II.4-21), and so we shall follow that procedure.

The Nondispersive Field-Envelope Equations

The basic equation of propagation is the transverse wave equation (II.4-1). This wave equation simply expresses Maxwell's equations for transverse fields in the absence of free charges or magnetization. We now introduce a mode expansion of $\mathbf{E}$ and of $\mathbf{P}$. With any such expansion we obtain the set of coupled equations

$$\sum_{\lambda} \left[\nabla^2 - \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \right] \mathbf{E}^{(+)}_{\lambda} = \sum_{\lambda} \frac{4\pi}{c^2} \frac{\partial^2}{\partial t^2} \mathbf{P}^{(+)}_{\lambda}. \quad (\text{II.4-24})$$

The vector field $\mathbf{P}^{(+)}_{\lambda}$ that appears as a source term is the entire transverse polarization field. That is, we have not separated the polarization field into linear and nonlinear portions (as is done when one defines an index of refraction).

We now require that Eqn. (II.4-24) hold for each pulse mode separately. That is, we equate separately the coefficients of each exponential and each unit vector. We thereby require that the amplitude functions to be such that each separate mode λ satisfies the wave equation,

$$\left[\nabla^2 - \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \right] \mathbf{E}^{(+)}_{\lambda} = \frac{4\pi}{c^2} \frac{\partial^2}{\partial t^2} \mathbf{P}^{(+)}_{\lambda}. \quad (\text{II.4-25})$$

For the Fourier expansion this equality is automatic. In the pulse-mode expansion it is a condition that we impose upon the mode amplitude functions. It is not an approximation; it is a constraint on the class of solutions we seek.

The derivatives needed in the wave equation involve two functions: the exponentiated phase ξ_λ , and the envelope functions $\mathcal{E}_\lambda(r, t)$ and $\mathcal{P}_\lambda(r, t)$. We have explicitly

$$\begin{aligned} \nabla^2 \mathcal{E}_\lambda(r, r) \exp(i\xi_\lambda) \\ = \exp(i\xi_\lambda) \left[-k_\lambda^2 + i(\nabla^2 \xi_\lambda) + 2ik_\lambda \cdot \nabla + \nabla^2 \right] \mathcal{E}_\lambda(r, r) \end{aligned} \quad (\text{II.4-26})$$

$$\begin{aligned} \frac{\partial^2}{\partial t^2} \mathcal{S}_\lambda(r, r) \exp(i\xi_\lambda) \\ = \exp(i\xi_\lambda) \left[-\omega_\lambda^2 + i \left(\frac{\partial^2}{\partial t^2} \xi_\lambda \right) - 2i\omega_\lambda \frac{\partial}{\partial t} + \frac{\partial^2}{\partial t^2} \right] \mathcal{S}_\lambda(r, r). \end{aligned} \quad (\text{II.4-27})$$

We therefore obtain the equations

$$\begin{aligned} \left[-k_\lambda^2 + i(\nabla^2 \xi_\lambda) + 2ik_\lambda \cdot \nabla + \nabla^2 \right] \mathcal{E}_\lambda(r, r) \\ + \frac{1}{c^2} \left[\omega_\lambda^2 - i \left(\frac{\partial^2}{\partial t^2} \xi_\lambda \right) + 2i\omega_\lambda \frac{\partial}{\partial t} - \frac{\partial^2}{\partial t^2} \right] \mathcal{S}_\lambda(r, r) \\ = \frac{4\pi}{c^2} \left[-\omega_\lambda^2 + i \left(\frac{\partial^2}{\partial t^2} \xi_\lambda \right) - 2i\omega_\lambda \frac{\partial}{\partial t} + \frac{\partial^2}{\partial t^2} \right] \mathcal{S}_\lambda(r, r). \end{aligned} \quad (\text{II.4-28})$$

These equations hold, exactly, for any of the mode expansions. It proves useful to organize this equation as follows:

$$\begin{aligned} -\frac{i}{2ck_\lambda} \left[(\omega_\lambda)^2 - (ck_\lambda)^2 + ic^2 \left[(\nabla^2 \xi_\lambda) - \frac{1}{c^2} \left(\frac{\partial^2}{\partial t^2} \xi_\lambda \right) \right] \right] \mathcal{E}_\lambda(r, r) \\ + \left[\hat{k}_\lambda \cdot \nabla + \frac{\omega_\lambda}{c^2 k_\lambda} \frac{\partial}{\partial t} - \frac{i}{2k_\lambda} \left[\nabla^2 - \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \right] \right] \mathcal{E}_\lambda(r, r) \\ = \frac{2\pi i \omega_\lambda}{c^2 k_\lambda} \left[1 - \frac{i}{(\omega_\lambda)^2} \left(\frac{\partial^2}{\partial t^2} \xi_\lambda \right) + \frac{2i}{\omega_\lambda} \frac{\partial}{\partial t} - \frac{1}{(\omega_\lambda)^2} \frac{\partial^2}{\partial t^2} \right] \mathcal{S}_\lambda(r, r) \end{aligned} \quad (\text{II.4-29})$$

The first line of this equation involves (possibly) time and space variations of the phase ξ_λ but no derivatives of the envelope function $\mathcal{E}_\lambda$. The second line involves first and second order derivatives of the envelope function, with respect to space and time. To proceed we must specify more details of the mode expansion and the properties of the phase ξ_λ . We wish to do this in such a way that the first line of this equation will vanish, and that we retain only first derivatives in the second line.

Expansions with Constant Frequency

Consider the case where k_λ and ω_λ are independent of position and time, so that all second derivatives of the phase ξ_λ vanish. We now define the relationship between k_λ and ω_λ to be

$$ck_\lambda = \omega_\lambda. \quad (\text{II.4-30})$$

(This is the definition of k_λ for the present; below we shall introduce an alternative definition in which the phase velocity is frequency dependent.) With this definition each envelope equation becomes

$$\begin{aligned} & \left[2ik_\lambda \left(\hat{\mathbf{k}}_\lambda \cdot \nabla + \frac{1}{c} \frac{\partial}{\partial t} \right) + \nabla^2 - \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \right] \mathcal{E}_\lambda(\mathbf{r}, t) \\ &= \frac{4\pi}{c^2} \left[-\omega_\lambda^2 - 2i\omega_\lambda \frac{\partial}{\partial t} + \frac{\partial^2}{\partial t^2} \right] \mathcal{P}_\lambda(\mathbf{r}, t). \end{aligned} \quad (\text{II.4-31})$$

These equations have not yet introduced any approximations concerning the waves. In particular, they need not all propagate along a common axis. However, if the equations are to be of practical use it is desirable that the rapid variation of the field be contained within the phase ξ_λ so that the envelope functions will be slowly varying. We are here assuming that this condition will occur for the choice $ck_\lambda = \omega_\lambda$. Although this is a conventional assumption, it requires verification in each instance, and need not be true in general.

The SVEA

Let us rewrite the mode propagation equation (with $ck_\lambda = \omega_\lambda$) as

$$\begin{aligned} & \left[\hat{\mathbf{k}}_\lambda \cdot \nabla + \frac{1}{c} \frac{\partial}{\partial t} \right] \mathcal{E}_\lambda(\mathbf{r}, t) - \frac{i}{2k_\lambda} \left[\nabla^2 - \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \right] \mathcal{E}_\lambda(\mathbf{r}, t) \\ &= \frac{2\pi i(\omega_\lambda)^2}{c^2 k_\lambda} \left[1 + \frac{2i}{\omega_\lambda} \frac{\partial}{\partial t} - \frac{1}{(\omega_\lambda)^2} \frac{\partial^2}{\partial t^2} \right] \mathcal{P}_\lambda(\mathbf{r}, t). \end{aligned} \quad (\text{II.4-32})$$

We are interested in pulses whose envelopes vary slowly in space and time, compared with the variation extracted with the carrier phase $\mathbf{k}_\lambda \cdot \mathbf{r} - \omega_\lambda t$. For such pulses we make the slowly varying envelope approximation (SVEA) by neglecting second derivatives of the electric field envelope and neglecting the second derivative of the polarization-field envelope. We thereby obtain the SVAE approximation for mode λ ,

$$\left[\hat{\mathbf{k}}_\lambda \cdot \nabla + \frac{1}{c} \frac{\partial}{\partial t} \right] \mathcal{E}_\lambda(\mathbf{r}, t) = \frac{2\pi i(\omega_\lambda)^2}{c^2 k_\lambda} \left[1 + \frac{2i}{\omega_\lambda} \frac{\partial}{\partial t} \right] \mathcal{P}_\lambda(\mathbf{r}, t). \quad (\text{II.4-33})$$

Although we have omitted the second derivative of the polarization envelope we have retained the first derivative. This procedure allows us to develop approximations for the real and imaginary parts of the equation. Furthermore, when we consider two-photon excitation, such as occurs with Raman processes, the polarization field is proportional to the electric field, and we must retain the first time derivative of $\mathcal{P}_\lambda$ for consistency.

The Paraxial SVEA

We are interested in radiation that propagates primarily along a well defined direction, say the z axis. Let us write the propagation vector as

$$\mathbf{k}_\lambda = k_{z\lambda} \mathbf{e}(z) + k_{x\lambda} \mathbf{e}(x) + k_{y\lambda} \mathbf{e}(y) \quad (\text{II.4-34})$$

where $k_{z\lambda} \approx k_\lambda$ is the dominant component, and $k_{x\lambda}$ and $k_{y\lambda}$ are much smaller than k_λ . We can then write the mode equation as

$$\begin{aligned} & \left[\frac{\partial}{\partial z} + \frac{1}{c} \frac{\partial}{\partial t} \right] \mathcal{E}_\lambda(r, t) \\ & + \left[\frac{k_{x\lambda}}{k_\lambda} \frac{\partial}{\partial x} + \frac{k_{y\lambda}}{k_\lambda} \frac{\partial}{\partial y} - \frac{i}{2k_\lambda} \left[\left(\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} \right) + \frac{\partial^2}{\partial z^2} - \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \right] \right] \mathcal{E}_\lambda(r, t) \\ & = \frac{2\pi i (\omega_\lambda)^2}{c^2 k_\lambda} \left[1 + \frac{2i}{\omega_\lambda} \frac{\partial}{\partial t} - \frac{1}{(\omega_\lambda)^2} \frac{\partial^2}{\partial t^2} \right] \mathcal{E}_\lambda(r, t). \end{aligned} \quad (\text{II.4-35})$$

The second derivatives in x and y define the transverse Laplacian

$$(\nabla^T)^2 = \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2}$$

Next we assume that the longitudinal variation of the envelope is slight over a wavelength, so that we can neglect the spatial second derivative $\partial^2/\partial z^2$. However, we retain the transverse Laplacian. This term is responsible for diffraction effects. The neglect of second derivatives in z and t constitutes the slowly varying envelope approximation (SVEA). In making this approximation we assume that the envelope functions $\mathcal{E}_\lambda$ and $\mathcal{E}_\lambda^*$ vary more slowly with space and time than do the phases ξ_λ . This approximation allows us to neglect the second derivative terms of $\mathcal{E}_\lambda$ and of $\mathcal{E}_\lambda^*$. It eliminates consideration of waves flowing in the backward direction. With the neglect of second derivatives in time we obtain the equation

$$\begin{aligned} & \left[\frac{\partial}{\partial z} + \frac{1}{c} \frac{\partial}{\partial t} \right] \mathcal{E}_\lambda(r, t) \\ & + \left[\frac{k_{x\lambda}}{k_\lambda} \frac{\partial}{\partial x} + \frac{k_{y\lambda}}{k_\lambda} \frac{\partial}{\partial y} - \frac{i}{2k_\lambda} \left(\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} \right) \right] \mathcal{E}_\lambda(r, t) \\ & = \frac{2\pi i (\omega_\lambda)^2}{c^2 k_\lambda} \left[1 + \frac{2i}{\omega_\lambda} \frac{\partial}{\partial t} \right] \mathcal{E}_\lambda(r, t). \end{aligned} \quad (\text{II.4-36})$$

When we further restrict consideration to those modes whose carrier wave vectors are directed along the common z axis (the paraxial approximation) we obtain the *paraxial wave equation*

$$\left[\frac{\partial}{\partial z} + \frac{1}{c} \frac{\partial}{\partial t} - \frac{i}{2k_\lambda} \left(\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} \right) \right] \mathcal{E}_\lambda(r, r) = \frac{2\pi i (\omega_\lambda)^2}{c^2 k_\lambda} \left[1 + \frac{2i}{\omega_\lambda} \frac{\partial}{\partial t} \right] \mathcal{P}_\lambda(r, r). \quad (\text{II.4-36})$$

Apart from the transverse Laplacian, this equation describes plane waves. The second derivatives allow diffraction.

The Plane Wave SVEA

Further simplification is possible if we assume that, at least locally, the pulses are plane waves, so that we can neglect the transverse derivatives. This approximation is satisfactory if the following conditions hold: first, the initial wavefront must be nearly flat, with its normal directed along the z axis; second, the properties of the medium must not vary appreciable with the transverse coordinate (i.e. there must be no discontinuities in the material); third, the laser fields must have no transverse changes over distances comparable to a wavelength.

With the SVEA the result (with $ck_\lambda = \omega_\lambda$) is the set of one dimensional plane-wave envelope equations

$$\left[\frac{\partial}{\partial z} + \frac{1}{c} \frac{\partial}{\partial t} \right] \mathcal{E}_\lambda(z, r) = \frac{2\pi i}{c} \omega_\lambda \left[1 + \frac{2i}{\omega_\lambda} \frac{\partial}{\partial t} \right] \mathcal{P}_\lambda(z, r). \quad (\text{II.4-37a})$$

In descriptions of traditional single-photon excitation processes (as contrasted with Raman or higher-order processes) one neglects the time derivative of $\mathcal{P}_\lambda$ to obtain the equation

$$\left[\frac{\partial}{\partial z} + \frac{1}{c} \frac{\partial}{\partial t} \right] \mathcal{E}_\lambda(z, r) = \frac{2\pi i}{c} \omega_\lambda \mathcal{P}_\lambda(z, r). \quad (\text{II.4-37b})$$

This is the traditional basic equation of propagation for pulses, in the plane-wave slowly-varying-envelope approximation, when the index of refraction is unity. It shows that each pulse envelope $\mathcal{E}_\lambda$ has an individual source term, the polarization mode envelope $\mathcal{P}_\lambda$.

Although it might appear that each envelope propagates independently of any other pulse, this need not be so. The equations for $\mathcal{P}_\lambda$ (which must be derived from the Schrödinger equation of atomic excitation or an equivalent equation) couple excitation of one polarization mode to electric fields in other modes.

It is tempting to generalize this result to a dispersive medium by introducing an index of refraction. To see when this is justified we need to examine in more detail the origin of the index of refraction. This we shall do below.

Moving Reference Frame

In the approximation, made above, of unit index of refraction, each separate pulse envelope involves a common phase velocity -- the velocity of light c . It is customary (cf. Lamb or Isevgi and Lamb) to introduce a coordinate system that moves with this speed. To accomplish this we replace the two variables (z, t) by the pair of variables

(Z,T) through the definition

$$T = t - \frac{z}{c}, \quad Z = z. \quad (\text{II.4-38})$$

The new time variable T is just the time measured from a hypothetical fiducial mark, at the leading edge of the pulse, moving at the velocity c. At the entry face to the material, $z = 0$, the time $T = 0$ coincides with the start of the pulse. We shall take this variable T as the local time τ that appears as the argument of the pulse envelope function. That is, the argument T of $\mathcal{E}_\lambda(Z,T)$ refers to the local (retarded) time at position Z, starting from $T = 0$ at the arrival from the entry face of a signal at speed c. The new variables lead to derivatives

$$\left[\frac{\partial}{\partial Z} \right]_T = \left[\frac{\partial}{\partial z} \right]_t + \frac{1}{c} \left[\frac{\partial}{\partial t} \right]_z, \quad \left[\frac{\partial}{\partial T} \right]_Z = \left[\frac{\partial}{\partial t} \right]_z \quad (\text{II.4-39})$$

and thence to the envelope equations

$$\frac{\partial}{\partial Z} \mathcal{E}_\lambda(Z,T) = \frac{2\pi i}{c} \omega_\lambda \mathcal{E}_\lambda(Z,T). \quad (\text{II.4-40})$$

There is one such equation for each pulse mode, i.e. for each carrier frequency and polarization. The total electric field is

$$\mathbf{E}(\mathbf{r},t) = \mathcal{R}e \sum_{\lambda} \mathbf{e}(\lambda) \mathcal{E}_\lambda \left(z, t - \frac{z}{c} \right) \exp(i\mathbf{k}_\lambda \cdot \mathbf{r} - i\omega_\lambda t). \quad (\text{II.4-41})$$

The remainder of this chapter will discuss only plane-wave approximations. To simplify notation we shall use z in place of Z as the position variable. The variable T is what was earlier denoted by τ .

The preceding result is basically the conventional short-pulse propagation equation (cf. Lamb) extended to multiple pulses, i.e. multiple carrier frequencies and polarization directions. Conventional treatments introduce an index of refraction, and as a consequence, deal with phase velocity that is not equal to the vacuum light speed c .

The pulse envelope equations represent half of a set of radiation-matter equations. They describe the propagation of electric fields, given the polarization field. The other half of the equations describe the time variation of the polarization, given the electric field.

Phase and Intensity Equations

The envelope $\mathcal{E}_\lambda$ may be complex valued, and so changes may occur both in magnitude and in phase. Let us examine the two types of change. Define the phase Φ_λ by the equations

$$\mathcal{E}_\lambda = |\mathcal{E}_\lambda| \exp(i\Phi_\lambda). \quad (\text{II.4-42})$$

Then from the pulse mode equation multiplied by $\mathcal{E}_\lambda^*$ we obtain the result

$$|\mathcal{E}_\lambda| \frac{\partial}{\partial z} |\mathcal{E}_\lambda| + i |\mathcal{E}_\lambda|^2 \frac{\partial}{\partial z} \Phi_\lambda = \frac{i(2\pi\omega_\lambda)}{c} \mathcal{E}_\lambda^* \mathcal{P}_\lambda. \quad (\text{II.4-43})$$

Upon separating real and imaginary parts, and introducing the mode intensity I_λ by the definition (we are taking $ck_\lambda = \omega_\lambda$ in the present discussion)

$$I_\lambda = \frac{c}{8\pi} \frac{ck_\lambda}{\omega_\lambda} |\mathcal{E}_\lambda|^2 = \frac{c}{8\pi} |\mathcal{E}_\lambda|^2, \quad (\text{II.4-44})$$

we obtain the two equations

$$\frac{\partial}{\partial z} I_\lambda = -I_\lambda 4\pi k_\lambda \mathcal{I}m \left(\frac{\mathcal{P}_\lambda}{\mathcal{E}_\lambda} \right) \quad (\text{II.4-45})$$

$$\frac{\partial}{\partial z} \Phi_\lambda = 2\pi k_\lambda \mathcal{R}e \left(\frac{\mathcal{P}_\lambda}{\mathcal{E}_\lambda} \right). \quad (\text{II.4-46})$$

The first of these equations is recognizable as the equation of radiative transfer, but with a dynamical time-dependent absorption coefficient

$$\kappa(\omega_\lambda) = 4\pi k_\lambda \mathcal{I}m \left(\frac{\mathcal{P}_\lambda}{\mathcal{E}_\lambda} \right). \quad (\text{II.4-47})$$

The second of these equations expresses possible shifts in carrier frequency. In particular, it accounts for the phase change that is usually incorporated into an index of refraction. In each case the significant quantity is the ratio of instantaneous polarization field to instantaneous electric field.

To complete our description of pulse propagation we require some prescription for the polarization field. It is this field that expresses the atomic properties, and the response of the atoms to radiation.

§II.5 Pulse Propagation in a Dispersive Medium: Frequency Dependent Index

It often proves useful to assume that the (transverse) polarization field can be separated into two parts, lin P and non P :

$$\mathbf{P} = \text{lin P} + \text{non P} \quad (\text{II.5-1})$$

The nonlinear portion appears as a source term in the wave equation, whereas the linear term appears as an index of refraction η or as a susceptibility χ through the relationship

$$4\pi \text{lin P} = 4\pi\chi\mathbf{E} = (\eta^2 - 1)\mathbf{E} = (\epsilon - 1)\mathbf{E} \quad (\text{II.5-2})$$

Elementary theory takes χ and η to be scalar constants or, more generally, as time-independent tensors. What happens if we relax this restriction and allow χ to depend upon frequency or time? This question has been examined by Ackerhalt, Drummond and Eberly (1986). Following is an examination and extension of their results, applied to pulse-mode expansions.

The Plane wave Equation

Let us consider propagation of plane waves through a medium in which there is a linear and nonlinear polarization field. We assume that there are no sharp discontinuities in the material properties that would produce reflections. We are interested in solving the transverse plane-wave equation

$$\left[\frac{\partial^2}{\partial z^2} - \frac{1}{c^2} \frac{\partial}{\partial t^2} \right] \mathbf{E}(z,t) - \frac{4\pi}{c^2} \frac{\partial}{\partial t^2} \text{lin P}(z,t) = \frac{4\pi}{c^2} \frac{\partial^2}{\partial t^2} \text{non P}(z,t) \quad (\text{II.5-3})$$

for pulses of radiation travelling in the positive z direction. (We will neglect the backward travelling waves.) We would like to account for the linear portion of the polarization field as accurately as possible by means of a frequency dependent index of refraction, so that the nonlinear portion of the polarization can be regarded as a disturbance of that propagation.

We shall refer to the contribution the nonlinear polarization, non P , as the inhomogeneous *source* term in the wave equation, and shall refer to propagation in the absence of this nonlinear polarization as being source-free propagation.

The wave equation deals with variations of fields in space and time. There are two general procedures for treating the time dependence. First, we may deal explicitly with functions of time (envelope functions) and with attendant derivatives with respect to time. Alternatively we may introduce Fourier transforms of the time dependence (transform functions) and deal with algebraic equations involving frequencies. We shall describe both approaches.

Mode Expansions

We have previously expanded the positive frequency part of the (transverse) fields in the form

$$E^{(+)}(z,t) = \frac{1}{2} \sum_{\lambda} e(\lambda) \exp(ik_{\lambda} z - i\omega_{\lambda} t) \mathcal{E}_{\lambda}(z,t) \quad (\text{II.5-4})$$

$$\text{lin } P^{(+)}(z,t) = \frac{1}{2} \sum_{\lambda} e(\lambda) \exp(ik_{\lambda} z - i\omega_{\lambda} t) \text{lin } \mathcal{P}_{\lambda}(z,t). \quad (\text{II.5-5})$$

The mode frequencies ω_{λ} and wavenumbers k_{λ} that appear in this expansion define plane wave carriers that are modified by the envelope function $\mathcal{E}_{\lambda}(z,t)$. By extracting these explicit phases we should be left with envelope functions that vary slowly with space and time. That is, the Fourier transform of the envelope function $\mathcal{E}_{\lambda}(z,t)$ should contain only spatial wavevectors whose magnitude is much less than k_{λ} and frequencies that are much smaller than ω_{λ} .

Fourier Analysis

To obtain a relationship between the functions $\mathcal{E}$ and $\text{lin } \mathcal{P}$ we consider a Fourier expansion of the spatial and temporal portions of the mode envelopes, in the form of double fourier integrals

$$E^{(+)}(z,t) = \frac{1}{2} \sum_{\lambda} e(\lambda) \int dK \int d\omega \exp(iKz - i\omega t) \tilde{\mathcal{E}}_{\lambda}(K,\omega) \quad (\text{II.5-6})$$

$$\text{lin } P^{(+)}(z,t) = \frac{1}{2} \sum_{\lambda} e(\lambda) \int dK \int d\omega \exp(iKz - i\omega t) \text{lin } \tilde{\mathcal{P}}_{\lambda}(K,\omega). \quad (\text{II.5-7})$$

A similar expression applies to the nonlinear portion of the polarization field. Our interest will be with fields in which the Fourier coefficients $\tilde{\mathcal{E}}_{\lambda}(K,\omega)$ cluster about wavevectors k_{λ} and frequencies ω_{λ} . That is, we assume that the radiation comprises well separated carrier waves such that the separation between any two carrier frequencies is much larger than the bandwidth around either carrier. By this hypothesis the integrals over dK and $d\omega$ can therefore be separated into distinct disjoint segments, each of which is identifiable as a particular mode λ .

The Susceptibility

We now assume that the Fourier components $\text{lin } \tilde{\mathcal{P}}$ are linearly related to the Fourier components $\tilde{\mathcal{E}}$. In general this will be a tensor relationship. For simplicity, let us assume a scalar relationship

$$\text{lin } \tilde{\mathcal{P}}_{\lambda}(\mathbf{K}, \omega) = 4\pi\chi(\omega) \tilde{\mathcal{E}}_{\lambda}(\mathbf{K}, \omega). \quad (\text{II.5-8})$$

We assume implicitly with this relationship that any spatial changes in the susceptibility occur only over large distances, so that locally the material may be considered homogeneous.^[*] We assume that the susceptibility $\chi(\omega)$ defined by this equation is known, as a function of frequency ω . This means that we assume some model (such as the Lorentz model) of linear response of molecules to fields. In general $\chi(\omega)$ will be a complex-valued function of frequency. The complex-valued frequency-dependent index of refraction $n(\omega)$ is defined, for the scalar susceptibility being considered, as

$$n(\omega) \equiv \sqrt{1 + 4\pi\chi(\omega)}. \quad (\text{II.5-9})$$

Using the frequency dependent susceptibility we write the linear portion of the polarization field as the double Fourier integral

$$\text{lin } \mathbf{P}^{(+)}(\mathbf{z}, t) = \frac{1}{2} \sum_{\lambda} \mathbf{e}(\lambda) \int d\mathbf{K} \int d\omega \exp(i\mathbf{K} \cdot \mathbf{z} - i\omega t) \chi(\omega) \tilde{\mathcal{E}}_{\lambda}(\mathbf{K}, \omega). \quad (\text{II.5-10})$$

The Transformed Source-Free Wave Equation

Let us apply this Fourier analysis to the wave equation for the field ${}^0\mathbf{E}$ in the absence of nonlinear polarization. With the definitions above we convert the source-free wave equation for plane-waves

$$0 = \left[\frac{\partial^2}{\partial z^2} - \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \right] {}^0\mathbf{E}(\mathbf{z}, t) - \frac{4\pi}{c^2} \frac{\partial^2}{\partial t^2} \text{lin } \mathbf{P}(\mathbf{z}, t) \quad (\text{II.5-11})$$

into an equation for mode λ :

$$0 = \frac{\partial^2}{\partial z^2} \int d\mathbf{K} \int d\omega \exp(i\mathbf{K} \cdot \mathbf{z} - i\omega t) {}^0\tilde{\mathcal{E}}_{\lambda}(\mathbf{K}, \omega) - \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \int d\mathbf{K} \int d\omega \exp(i\mathbf{K} \cdot \mathbf{z} - i\omega t) [1 + 4\pi\chi(\omega)] {}^0\tilde{\mathcal{E}}_{\lambda}(\mathbf{K}, \omega) \quad (\text{II.5-12})$$

^[*] footnote Were we to consider changes in the medium over distances comparable to a wavelength we would need to express the relationship between linear polarization and electric field as a convolution. We would also need to consider backward travelling waves generated by the discontinuity.

The differentiation, taken with the definition of $\eta(\omega)$, produces the equation

$$0 = [c^2 K^2 - \omega^2 \eta(\omega)^2] {}^0\tilde{\mathcal{E}}_\lambda(K, \omega). \quad (\text{II.5-13})$$

We can see that if we require that $K \equiv K(\omega)$ and ω be related by the dispersion relation

$$c^2 K(\omega)^2 = \omega^2 \eta(\omega)^2 \equiv \omega^2 [1 + 4\pi\chi(\omega)] \quad (\text{II.5-14})$$

then the wave equation will be satisfied, for *any distribution* of Fourier coefficients ${}^0\tilde{\mathcal{E}}_\lambda(K, \omega)$, so long as there is no non-linear polarization. The two solutions to this quadratic equation correspond to forward travelling waves and backward travelling waves. The electric field in this source-free case (no nonlinear polarization) may be written as a single Fourier integral

$${}^0\mathbf{E}^{(+)}(z, t) = \frac{1}{2} \sum_\lambda \mathbf{e}(\lambda) \int d\omega \exp[iK(\omega)z - i\omega t] {}^0\tilde{\mathcal{E}}_\lambda(\omega) \quad (\text{II.5-15})$$

The Fourier coefficient ${}^0\tilde{\mathcal{E}}_\lambda(\omega)$ is independent of space and time and may be determined from the time dependence of the pulse at $z = 0$:

$$\sum_\lambda \mathbf{e}(\lambda) {}^0\tilde{\mathcal{E}}_\lambda(\omega) = \frac{1}{\pi} \int_{-\infty}^{\infty} dt \exp(+i\omega t) {}^0\mathbf{E}^{(+)}(0, t) \quad (\text{II.5-16})$$

The individual modes are, by hypothesis, identifiable as distinct clusters of Fourier amplitudes about the discrete frequencies ω_λ . That is, ${}^0\tilde{\mathcal{E}}_\lambda(\omega)$ is peaked within a frequency band of width $2\Delta\omega_\lambda$ around the frequency $\omega = \omega_\lambda$. The electric field at other positions is obtained from Fourier synthesis with these same coefficients.

Other Choices for $K(\omega)$

Let us regard the preceding results from a slightly different viewpoint. Suppose we look for a solution to the source-free propagation equation in the form of a single Fourier integral

$${}^0\mathbf{E}^{(+)}(z, t) = \frac{1}{2} \sum_\lambda \mathbf{e}(\lambda) \int d\omega \exp[iK(\omega)z - i\omega t] {}^0\tilde{\mathcal{E}}_\lambda(z, \omega) \quad (\text{II.5-17})$$

where $K(\omega)$ is an arbitrary function of frequency and the Fourier amplitude ${}^0\tilde{\mathcal{E}}_\lambda(z, \omega)$ may depend upon position in a way that remains to be determined. The wave equation then leads to the temporal Fourier transform equation

$$0 = \frac{\partial^2}{\partial z^2} \int d\omega \exp[iK(\omega)z - i\omega t] {}^0\tilde{\mathcal{E}}_\lambda(z, \omega)$$

$$-\frac{1}{c^2} \frac{\partial^2}{\partial t^2} \int d\omega \exp[iK(\omega)z - i\omega t] \eta(\omega)^2 {}^0\tilde{\mathcal{E}}_\lambda(z, \omega) \quad (\text{II.5-18})$$

From this we obtain the equation for the Fourier function ${}^0\tilde{\mathcal{E}}_\lambda(z, \omega)$

$$0 = \left[\frac{\partial^2}{\partial z^2} + 2iK(\omega) \frac{\partial}{\partial z} - K(\omega)^2 + \frac{\omega^2}{c^2} \eta(\omega)^2 \right] {}^0\tilde{\mathcal{E}}_\lambda(z, \omega) \quad (\text{II.5-19})$$

This equation may also be written as

$$0 = \left[\frac{\partial}{\partial z} + iK(\omega) + \frac{i\omega\eta(\omega)}{c} \right] \left[\frac{\partial}{\partial z} + iK(\omega) - \frac{i\omega\eta(\omega)}{c} \right] {}^0\tilde{\mathcal{E}}_\lambda(z, \omega). \quad (\text{II.5-20})$$

This equation is exactly equivalent to the wave equation for plane waves in homogeneous medium in the absence of nonlinear polarization. The function $\eta(\omega) \equiv \sqrt{1 + 4\pi\chi(\omega)}$ that appears here is, for present purposes, taken to be a known function of frequency, while the function $K(\omega)$, like the z dependence of ${}^0\tilde{\mathcal{E}}_\lambda(z, \omega)$, remains as yet to be determined. The vanishing of this latter expression may be produced by either factor. We are interested in forward travelling waves, which we may require to satisfy the equation

$$0 = \left[\frac{\partial}{\partial z} + iK(\omega) - \frac{i\omega\eta(\omega)}{c} \right] {}^0\tilde{\mathcal{E}}_\lambda(z, \omega). \quad (\text{II.5-21})$$

We have previously chosen $K(\omega)$ to satisfy the dispersion relation $cK(\omega) = \omega\eta(\omega)$. Suppose instead that we choose $K(\omega)$ to differ from this definition by some frequency dependent term $g(\omega)$,

$$K(\omega) = \frac{\omega\eta(\omega)}{c} + g(\omega). \quad (\text{II.5-22})$$

Then we have the equation

$$0 = \left[\frac{\partial}{\partial z} + ig(\omega) \right] {}^0\tilde{\mathcal{E}}_\lambda(z, \omega). \quad (\text{II.5-23})$$

This equation has the solution, for any ω ,

$${}^0\tilde{\mathcal{E}}_\lambda(z, \omega) = \exp[-ig(\omega)z] {}^0\tilde{\mathcal{E}}_\lambda(0, \omega). \quad (\text{II.5-24})$$

From this we deduce that any nonzero choice of $g(\omega)$ will produce a spatial modulation of the transform function. The choice of $K(\omega)$ that produces the slowest spatial variation in the transform function (namely no variation) is the choice $cK(\omega) = \omega\eta(\omega)$. Only for this choice can we obtain a transform function that is independent of z . That is, we can obtain the simplest solution, consisting of an explicitly z -dependent phase and a constant amplitude, by choosing $cK(\omega) = \omega\eta(\omega)$. With any other choice of $g(\omega)$ we must construct an amplitude function whose z dependence cancels the z dependence introduced by $g(\omega)$, in order that the final solution produce the result

$$eE^{(+)}(z,t) = \frac{1}{2} \sum_{\lambda} e(\lambda) \int d\omega \exp \left[\frac{i\omega}{c} \{ \eta(\omega)z - ct \} \right] e\tilde{E}_{\lambda}(z,\omega). \quad (\text{II.5-25})$$

Including Spatial Variation of the Susceptibility

Consider the effect of spatial variation in the susceptibility $\chi(z,\omega)$ and hence in the index of refraction $\eta(z,\omega)$. Let us consider a construction of the form

$$eE^{(+)}(z,t) = \frac{1}{2} \sum_{\lambda} e(\lambda) \int d\omega \exp[i\theta(z,\omega) - i\omega t] e\tilde{E}_{\lambda}(z,\omega). \quad (\text{II.5-26})$$

where $\theta(z,\omega)$ is a phase to be determined. In place of Eqn. (II.5-26) we obtain the source-free propagation equation

$$\left[\frac{\partial^2}{\partial z^2} + 2iK(z,\omega) \frac{\partial}{\partial z} - K(z,\omega)^2 + i \left(\frac{\partial K}{\partial z} \right) + \frac{\omega^2}{c^2} \eta(z,\omega)^2 \right] e\tilde{E}_{\lambda}(z,\omega) = 0 \quad (\text{II.5-27})$$

or, equivalently,

$$\begin{aligned} & \left[\frac{\partial}{\partial z} + iK(z,\omega) + \frac{i\omega\eta(z,\omega)}{c} \right] \left[\frac{\partial}{\partial z} + iK(z,\omega) - \frac{i\omega\eta(z,\omega)}{c} \right] e\tilde{E}_{\lambda}(z,\omega) \\ &= -i e\tilde{E}_{\lambda}(z,\omega) \left[\frac{\partial}{\partial z} \frac{\omega\eta(z,\omega)}{c} \right] \end{aligned} \quad (\text{II.5-28})$$

where $K(z,\omega)$ is the spatial derivative of the phase $\theta(z,\omega)$:

$$K(z,\omega) = \frac{\partial}{\partial z} \theta(z,\omega). \quad (\text{II.5-29})$$

The spatial variation of the refractive index produces a source term on the right-hand side of the equation. Nevertheless we can take $cK(z,\omega) = \omega\eta(z,\omega)$ as before, so that the phase is

$$\theta(z,\omega) = \int_0^z dz' K(z',\omega) = \frac{\omega}{c} \int_0^z dz' \sqrt{1 + 4\pi\chi(z',\omega)} = \frac{\omega}{c} \int_0^z dz' \eta(z',\omega) \quad (\text{II.5-30})$$

This choice produces the equation

$$\left[\frac{\partial}{\partial z} + 2iK(z,\omega) \right] \frac{\partial}{\partial z} e\tilde{E}_{\lambda}(z,\omega) = -i \left[\frac{\partial K}{\partial z} \right] e\tilde{E}_{\lambda}(z,\omega) \quad (\text{II.5-31})$$

If the spatial variation of $\chi(z,\omega)$ or $\eta(z,\omega)$ is slow compared with the local wavelength, then we can neglect the derivative $\partial K/\partial z$ and we recover the previous equations, but

with parametric dependence of $K(\omega)$, $\eta(\omega)$ and $\chi(\omega)$ upon position. The equation becomes

$$0 = \left[\frac{\partial}{\partial z} + 2iK(z, \omega) \right] \frac{\partial}{\partial z} \tilde{\mathcal{E}}_{\lambda}(z, \omega). \quad (\text{II.5-32})$$

This equation has solutions $\tilde{\mathcal{E}}_{\lambda}(\omega)$ that are independent of z (these are the forward-travelling waves). We conclude that when we neglect the derivative $\partial K/\partial z$ we have the construction

$$\mathcal{E}^{(+)}(z, t) = \frac{1}{2} \sum_{\lambda} \mathbf{e}(\lambda) \int d\omega \exp[i\theta(z, \omega) - i\omega t] \tilde{\mathcal{E}}_{\lambda}(\omega). \quad (\text{II.5-33})$$

When we include the effect of the derivative $\partial K/\partial z$ but assume that the transform function varies slowly with z , so that $\partial/\partial z$ is negligible compared with $K(z, \omega)$, we have the equation

$$2iK(z, \omega) \frac{\partial}{\partial z} \tilde{\mathcal{E}}_{\lambda}(z, \omega) = -i \left[\frac{\partial K}{\partial z} \right] \tilde{\mathcal{E}}_{\lambda}(z, \omega) \quad (\text{II.5-35})$$

This equation has solutions

$$\tilde{\mathcal{E}}_{\lambda}(z, \omega) = \tilde{\mathcal{E}}_{\lambda}(0, \omega) \frac{\sqrt{K(0, \omega)}}{\sqrt{K(z, \omega)}}. \quad (\text{II.5-36})$$

With this refinement the field construction becomes

$$\mathcal{E}^{(+)}(z, t) = \frac{1}{2} \sum_{\lambda} \mathbf{e}(\lambda) \int d\omega \exp[i\theta(z, \omega) - i\omega t] \tilde{\mathcal{E}}_{\lambda}(0, \omega) \frac{\sqrt{K(0, \omega)}}{\sqrt{K(z, \omega)}} \quad (\text{II.5-37})$$

Monochromatic Wave

We may note that, in the approximation of a monochromatic wave at frequency ω_{λ} , the wavevector is

$$ck_{\lambda} = \omega_{\lambda} \eta(\omega_{\lambda}) \quad (\text{II.5-38})$$

and the field is simply

$$\mathcal{E}^{(+)}(z, t) = \frac{1}{2} \sum_{\lambda} \mathbf{e}(\lambda) \exp(ik_{\lambda} z - i\omega_{\lambda} t) \tilde{\mathcal{E}}_{\lambda}(\omega_{\lambda}) \quad (\text{II.5-39})$$

with summation over polarization. This monochromatic forward-travelling wave obeys

the first-order equation

$$\left[\frac{\partial}{\partial z} + \frac{\eta_\lambda}{c} \frac{\partial}{\partial t} \right] {}^0E^{(+)}(z,t) = 0 \quad (\text{II.5-40})$$

in addition to obeying the second-order equation

$$\left[\frac{\partial^2}{\partial z^2} - \frac{(\eta_\lambda)^2}{c^2} \frac{\partial^2}{\partial t^2} \right] {}^0E^{(+)}(z,t) = 0. \quad (\text{II.5-41})$$

The first order equation selects forward travelling waves from the set of solutions to the second-order equation. Because there is no inhomogeneous (or source) term on the right hand side of these equations we can make the replacement

$$\left[\frac{\partial}{\partial z} - \frac{\eta_\lambda}{c} \frac{\partial}{\partial t} \right] \left[\frac{\partial}{\partial z} + \frac{\eta_\lambda}{c} \frac{\partial}{\partial t} \right] {}^0E^{(+)}(z,t) = \left[\frac{\partial}{\partial z} + \frac{\eta_\lambda}{c} \frac{\partial}{\partial t} \right] {}^0E^{(+)}(z,t) \quad (\text{II.5-42})$$

thereby obtaining a first-order equation for forward-directed waves. When a source term is present we no longer have this equality.

§II.6 Source-Free Envelope Equations

The Fourier transform equation is not the equation that is customarily used in treating pulse propagation. The customary equation deals with both space and time variables, not just space variables. In general (i.e. in the presence of nonlinear polarization) we introduce the needed envelope functions $\mathcal{E}_\lambda(z,t)$ by the definition

$$\exp[ik_\lambda z - i\omega_\lambda t] \mathcal{E}_\lambda(z,t) = \int d\omega \exp[iK(\omega)z - i\omega t] \tilde{\mathcal{E}}_\lambda(z,\omega) \quad (\text{II.6-1})$$

That is, the envelope function $\mathcal{E}_\lambda(z,t)$ is expressible as a temporal Fourier transform taken about a mean frequency ω_λ and a mean wavenumber k_λ . These explicit phases are introduced in order that the envelope function $\mathcal{E}_\lambda(z,t)$ be slowly varying for forward-travelling waves. (With these phases present the envelopes describing backward waves contain a rapidly varying phase factor $\exp[-2K(\omega)z]$.)

This section continues the discussion of source-free propagation, i.e. propagation in the absence of nonlinear polarization. We continue to denote the envelope function for that special case as ${}^0\mathcal{E}_\lambda$.

We have already discussed the choice of $K(\omega)$. For source-free propagation (through a medium with negligible gradients of refractive index) we choose $cK(\omega) = \omega n(\omega)$ and obtain a Fourier amplitude that is independent of distance. We have then the connection between the time-varying envelope function ${}^0\mathcal{E}_\lambda(z,t)$ and the Fourier amplitude ${}^0\tilde{\mathcal{E}}_\lambda(\omega)$ the formula

$${}^0\mathcal{E}_\lambda(z,t) = \int d\omega \exp[i(K(\omega) - k_\lambda)z - i(\omega - \omega_\lambda)t] {}^0\tilde{\mathcal{E}}_\lambda(\omega) \quad (\text{II.6-2})$$

Although $K(\omega)$ has been determined, to produce the most slowly varying transform function, the relationship between ω_λ and k_λ remains to be defined. We have assumed that the pulses have well defined carrier frequencies, so that the amplitude ${}^0\tilde{\mathcal{E}}_\lambda(\omega)$ is appreciable only within a band of frequencies of width $\Delta\omega_\lambda$ around $\omega = \omega_\lambda$. Therefore the spatial phase will be least when we choose k_λ within the range of values $K(\omega_\lambda - \Delta\omega_\lambda)$ to $K(\omega_\lambda + \Delta\omega_\lambda)$. The center of this distribution is the value $K_\lambda \equiv K(\omega_\lambda)$, and this value would appear to be one possible choice. We shall show below the significance of this choice for k_λ .

The Envelope Equation

Using these envelope functions we have the mode expansions

$$E^{(+)}(z,t) = \frac{1}{2} \sum_\lambda e(\lambda) \exp(ik_\lambda z - i\omega_\lambda t) \mathcal{E}_\lambda(z,t) \quad (\text{II.6-3})$$

$$\text{non P}^{(+)}(z,t) = \frac{1}{2} \sum_{\lambda} \alpha(\lambda) \exp(ik_{\lambda} z - i\omega_{\lambda} t) \text{non } \mathcal{P}_{\lambda}(z,t). \quad (\text{II.6-4})$$

We now require a similar expansion for the linear part of the polarization field. This field has the property

$$\exp[ik_{\lambda} z - i\omega_{\lambda} t] \text{lin } \mathcal{P}_{\lambda}(z,t) = \int d\omega \exp[iK(\omega)z - i\omega t] \chi(\omega) \tilde{\mathcal{E}}_{\lambda}(z,\omega) \quad (\text{II.6-5})$$

Our intention is to treat forward-travelling pulses whose Fourier components cluster about discrete frequencies. To treat these envelope functions we introduce, around each of the discrete frequencies ω_{λ} , a series expansion of the susceptibility

$$\chi(\omega) = \sum_{n=0}^{\infty} \frac{\chi_{\lambda}^{(n)}}{n!} (\omega - \omega_{\lambda})^n \quad (\text{II.6-6})$$

The coefficient $\chi_{\lambda}^{(n)}$ is defined as the n th derivative of $\chi(\omega)$ with respect to ω , evaluated at the frequency ω_{λ} :

$$\chi_{\lambda}^{(n)} \equiv \left[\frac{d^n}{d\omega^n} \chi(\omega) \right]_{\lambda} \quad (\text{II.6-7})$$

When used in the Fourier integral for the linear portion of the polarization, Eqn. (II.6-4), this expansion gives the result

$$\begin{aligned} & \int d\omega \exp[iK(\omega)z - i\omega t] 4\pi \chi(\omega) \tilde{\mathcal{E}}_{\lambda}(z,\omega) \\ &= \sum_{n=0}^{\infty} \frac{4\pi \chi_{\lambda}^{(n)}}{n!} \int d\omega \exp[iK(\omega)z - i\omega t] (\omega - \omega_{\lambda})^n \tilde{\mathcal{E}}_{\lambda}(z,\omega) \\ &= \exp(-i\omega_{\lambda} t) \sum_{n=0}^{\infty} \frac{4\pi \chi_{\lambda}^{(n)}}{n!} \left(i \frac{\partial}{\partial t} \right)^n \int d\omega \exp[iK(\omega)z - i\omega t + i\omega_{\lambda} t] \tilde{\mathcal{E}}_{\lambda}(z,\omega) \\ &= \exp(-i\omega_{\lambda} t) \sum_{n=0}^{\infty} \frac{4\pi \chi_{\lambda}^{(n)}}{n!} \left(i \frac{\partial}{\partial t} \right)^n \exp(ik_{\lambda} z) \mathcal{E}_{\lambda}(z,t) \end{aligned} \quad (\text{II.6-8})$$

Using this result, and the definition of the pulse envelope, we obtain the source-free

wave equation

$$0 = \left[\frac{\partial^2}{\partial z^2} - \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \right] \exp(ik_\lambda z - i\omega_\lambda t) {}^0\mathcal{E}_\lambda(z, t) - \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \exp(ik_\lambda z - i\omega_\lambda t) \sum_{n=0}^{\infty} \frac{4\pi\chi_\lambda^{(n)}}{n!} \left(i \frac{\partial}{\partial t} \right)^n {}^0\mathcal{E}_\lambda(z, t) \quad (\text{II.6-9})$$

Upon carrying out the differentiation we obtain the equation

$$0 = \left[\frac{\partial^2}{\partial z^2} + 2ik_\lambda \frac{\partial}{\partial z} - (k_\lambda)^2 \right] {}^0\mathcal{E}_\lambda(z, t) - \frac{1}{c^2} \left[\frac{\partial^2}{\partial t^2} - 2i\omega_\lambda \frac{\partial}{\partial t} - (\omega_\lambda)^2 \right] {}^0\mathcal{E}_\lambda(z, t) - \frac{1}{c^2} \left[\frac{\partial^2}{\partial t^2} - 2i\omega_\lambda \frac{\partial}{\partial t} - (\omega_\lambda)^2 \right] \sum_{n=0}^{\infty} \frac{4\pi\chi_\lambda^{(n)}}{n!} \left(i \frac{\partial}{\partial t} \right)^n {}^0\mathcal{E}_\lambda(z, t) \quad (\text{II.6-10})$$

These terms can be rearranged in the following form:

$$0 = - \frac{1}{c^2} \left[(ck_\lambda)^2 - (\omega_\lambda)^2 \right] \{ 1 + 4\pi\chi_\lambda^{(0)} \} {}^0\mathcal{E}_\lambda(z, t) + \left[2ik_\lambda \frac{\partial}{\partial z} + \frac{2i(\omega_\lambda)}{c} \left[1 + 4\pi\chi_\lambda^{(0)} + \frac{1}{2} \omega_\lambda 4\pi\chi_\lambda^{(1)} \right] \frac{1}{c} \frac{\partial}{\partial t} \right] {}^0\mathcal{E}_\lambda(z, t) + \left[\frac{\partial^2}{\partial z^2} - \left[1 + 4\pi\chi_\lambda^{(0)} + 2\omega_\lambda 4\pi\chi_\lambda^{(1)} + \frac{1}{2} (\omega_\lambda)^2 4\pi\chi_\lambda^{(2)} \right] \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \right] {}^0\mathcal{E}_\lambda(z, t) + \frac{1}{c^2} \sum_{n=3}^{\infty} \left[\frac{(\omega_\lambda)^2 4\pi\chi_\lambda^{(n)}}{n!} + 2\omega_\lambda \frac{4\pi\chi_\lambda^{(n-1)}}{(n-1)!} + \frac{4\pi\chi_\lambda^{(n-2)}}{(n-2)!} \right] \left(i \frac{\partial}{\partial t} \right)^n {}^0\mathcal{E}_\lambda(z, t)$$

Note that third and higher-order time derivatives originate entirely with the frequency variation of the susceptibility. A portion of the first and second derivative terms also expresses this frequency variation.

Phase and Group Velocity

The appearance of this equation can be simplified by introducing a few definitions. Let us define the function $K(\omega)$ by the relationship

$$cK(\omega) = \omega\eta(\omega) \equiv \omega\sqrt{1 + 4\pi\chi(\omega)}. \quad (\text{II.6-12})$$

Let $K_\lambda = K(\omega_\lambda)$ be the value of this function at $\omega = \omega_\lambda$. The following properties hold for this function and various derivatives evaluated at $\omega = \omega_\lambda$:

$$cK_\lambda \equiv cK(\omega_\lambda) = \omega_\lambda \sqrt{1 + 4\pi\chi_\lambda^{(0)}} = \omega_\lambda \eta_\lambda \quad (\text{II.6-13})$$

$$c^2 \left[\frac{dK^2}{d\omega} \right]_\lambda = \left[\frac{d}{d\omega} \omega^2 \eta(\omega)^2 \right]_\lambda = 2\omega_\lambda \left[1 + 4\pi\chi_\lambda^{(0)} + \frac{1}{2} \omega_\lambda 4\pi\chi_\lambda^{(1)} \right]$$

$$c^2 \left[\frac{d^2 K^2}{d\omega^2} \right]_\lambda = \left[\frac{d^2}{d\omega^2} \omega^2 \eta(\omega)^2 \right]_\lambda = 2 \left[1 + 4\pi\chi_\lambda^{(0)} + 2\omega_\lambda 4\pi\chi_\lambda^{(1)} + \frac{1}{2} (\omega_\lambda)^2 4\pi\chi_\lambda^{(2)} \right]$$

and, for $n > 2$,

$$\frac{c^2}{n!} \left[\frac{d^n K^2}{d\omega^n} \right]_\lambda = \left[\frac{4\pi\chi_\lambda^{(n-2)}}{(n-2)!} + 2\omega_\lambda \frac{4\pi\chi_\lambda^{(n-1)}}{(n-1)!} + (\omega_\lambda)^2 \frac{4\pi\chi_\lambda^{(n)}}{n!} \right]. \quad (\text{II.6-14})$$

These expressions can also be written in terms of a phase velocity $v_{ph}(\lambda)$ and a group velocity $v_{gr}(\lambda)$ defined as

$$\frac{1}{v_{ph}(\lambda)} \equiv \frac{K_\lambda}{\omega_\lambda} = \frac{\eta_\lambda}{c} \quad (\text{II.6-15})$$

$$\frac{1}{v_{gr}(\lambda)} \equiv \left[\frac{dK}{d\omega} \right]_\lambda = \frac{1}{c\eta_\lambda} [1 + 4\pi\chi_\lambda^{(0)} + 2\pi\omega_\lambda \chi_\lambda^{(1)}]. \quad (\text{II.6-16})$$

When we can neglect $\chi_\lambda^{(1)}$ the group velocity and the phase velocity coincide. When the two velocities do not coincide, it is the group velocity that is needed in the pulse propagation equation (the phase velocity occurs when the wave is monochromatic). We can write higher derivatives of $K(\omega)$ as derivatives of the inverse group velocity, as in the expression

$$\frac{1}{2} \left[\frac{d^2 K^2}{d\omega^2} \right]_\lambda = \left[\frac{d}{d\omega} \frac{K(\omega)}{v_{gr}(\omega)} \right]_\lambda. \quad (\text{II.6-17})$$

Using these definitions we can rewrite Eqn. (II.6-11), for source-free propagation, as

$$0 = [(K_\lambda)^2 - (k_\lambda)^2] {}^0\mathcal{E}_\lambda(z,t) + \left[\frac{\partial^2}{\partial z^2} + 2ik_\lambda \frac{\partial}{\partial z} \right] {}^0\mathcal{E}_\lambda(z,t)$$

$$+ \left[i \left[\frac{\partial K^2}{\partial \omega} \right]_{\lambda} \frac{\partial}{\partial t} + \sum_{n=2}^{\infty} \frac{1}{n!} \left[\frac{d^n K^2}{d\omega^n} \right]_{\lambda} \left(i \frac{\partial}{\partial t} \right)^n \right] {}^0\mathcal{E}_{\lambda}(z,t) \quad (\text{II.6-18})$$

or else as

$$0 = \left[\frac{\partial^2}{\partial z^2} + 2ik_{\lambda} \frac{\partial}{\partial z} - (k_{\lambda})^2 + \sum_{n=0}^{\infty} \frac{1}{n!} \left[\frac{d^n K^2}{d\omega^n} \right]_{\lambda} \left(i \frac{\partial}{\partial t} \right)^n \right] {}^0\mathcal{E}_{\lambda}(z,t) \quad (\text{II.6-19})$$

These elaborate equations for the envelope function are equivalent to the simple Eqn. (II.5-32) for the transform function, and to the wave equation for a homogeneous medium in the absence of nonlinear polarization. Although the original wave equation was second order in time, the additional time derivatives are needed, in part, to account for the frequency dependence of the index of refraction.

Frequency Dependence

Typically the susceptibility has the form

$$4\pi\chi(\omega) = \sum_m \frac{F_m}{\omega - \omega_m - i\Gamma_m} \quad (\text{II.6-20})$$

where the summation goes over the set of resonance frequencies ω_m and widths Γ_m of the molecule that are accessible from the initial state. The required derivatives are, for this frequency dependence,

$$4\pi\chi_{\lambda}^{(n)} = (-1)^n n! \sum_m \frac{F_m}{(\omega_{\lambda_0} - \omega_m - i\Gamma_m)^{n+1}} \quad (\text{II.6-21})$$

This expression provides the formula

$$\frac{c^2}{n!} \left[\frac{d^n K^2}{d\omega^n} \right]_{\lambda} = (-1)^n \sum_m \frac{(F_m [(\omega_{\lambda} - \omega_m - i\Gamma_m)^2 - 2\omega_{\lambda}(\omega_{\lambda_0} - \omega_m - i\Gamma_m)] + (\omega_{\lambda})^2)}{(\omega_{\lambda} - \omega_m - i\Gamma_m)^{n+1}} \quad (\text{II.6-22})$$

In some cases it is useful to deal with near-resonant susceptibility. Under these conditions one need only consider that term in the summation for which $\omega_m \approx \omega_{\lambda}$, and one retains only the term that is proportional to $(\omega_{\lambda})^2$. This procedure yields the approximation

$$\frac{c^2}{n!} \left[\frac{d^n K^2}{d\omega^n} \right]_{\lambda} \simeq (-1)^n (\omega_{\lambda})^2 \frac{F_m}{(\omega_{\lambda} - \omega_m - i\Gamma_m)^{n+1}} \equiv (\omega_{\lambda})^2 \frac{4\pi\chi_{\lambda}^{(n)}}{n!}. \quad (\text{II.6-23})$$

This is not the approximation of interest to us. Our treatment assumes that such resonant terms are dealt with explicitly in the molecular dynamics. The cases of interest to us are those for which ω_{λ} is far from resonance, and is in fact much smaller than any of the resonance frequencies, $\omega_{\lambda} \ll \omega_m$. Under these conditions we can neglect the term $(\omega_{\lambda})^2$ and the term proportional to ω_{λ} and obtain the approximation

$$\frac{c^2}{n!} \left[\frac{d^n K^2}{d\omega^n} \right]_{\lambda} \simeq (-1)^n \sum_m \frac{F_m}{(\omega_{\lambda} - \omega_m - i\Gamma_m)^{n+1}} \equiv \frac{4\pi\chi_{\lambda}^{n-2}}{(n-2)!}. \quad (\text{II.6-24})$$

Choosing the Wavevector

We have yet to specify the connection between the wavevector k_{λ} and the frequency ω_{λ} . We wish to choose this so that the envelope function ${}^0\mathcal{E}_{\lambda}(z,t)$ has the least variation with z . Suppose that we have made such a choice, and that the envelope does vary only slowly. Let us also assume that the higher time derivatives are negligible. With these assumptions we may approximate the equation for the envelope function by the first-order equation

$$0 = \left[2ik_{\lambda} \frac{\partial}{\partial z} - (k_{\lambda})^2 + (K_{\lambda})^2 + i2K_{\lambda} \left[\frac{dK}{d\omega} \right]_{\lambda} \frac{\partial}{\partial t} \right] {}^0\mathcal{E}_{\lambda}(z,t) \quad (\text{II.6-25})$$

The simplest choice for k_{λ} is the value $K_{\lambda} \equiv K(\omega_{\lambda})$, because this provides the equation

$$0 = \left[\frac{\partial}{\partial z} + \frac{1}{v_g(\lambda)} \frac{\partial}{\partial t} \right] {}^0\mathcal{E}_{\lambda}(z,t) \quad (\text{II.6-26})$$

This is the equation for an envelope function that moves, without distortion, at the group velocity $v_g(\lambda)$. That is, the envelope function for mode λ , under the approximations embodied in Eqn. (II.6-26), is a function of z and t only through the combination $z - v_g(\lambda)t$. For fixed z , the envelope depends on time only through the retarded-time variable

$$T_{\lambda} = t - \frac{z}{v_g(\lambda)} \equiv t - \frac{z}{v_{\lambda}}$$

Having chosen $k_{\lambda} = K_{\lambda}$ by this criterion, we obtain the more general (but still source-free) wave equation as

$$0 = \left[\frac{\partial^2}{\partial z^2} + 2ik_{\lambda} \frac{\partial}{\partial z} \right] {}^0\mathcal{E}_{\lambda}(z,t)$$

$$+ \left[i \left[\frac{dK^2}{d\omega} \right]_{\lambda} \frac{\partial}{\partial t} + \sum_{n=2}^{\infty} \frac{1}{n!} \left[\frac{d^n K^2}{d\omega^n} \right]_{\lambda} \left(i \frac{\partial}{\partial t} \right)^n \right] {}^0\mathcal{E}_{\lambda}(z, t) \quad (\text{II.6-27})$$

These are the equations that one typically uses, with the neglect of various derivatives, for propagation of envelope functions. (They are used, with the addition of a source term, to treat propagation in the presence of nonlinear polarization.) These envelope functions are used in the field construction

$${}^0E^{(+)}(z, t) = \frac{1}{2} \sum_{\lambda} e(\lambda) \exp[ik_{\lambda} z - i\omega_{\lambda} t] {}^0\mathcal{E}_{\lambda}(z, t) \quad (\text{II.6-28})$$

$$ck_{\lambda} \equiv cK(\omega_{\lambda}) = \omega_{\lambda} \sqrt{1 + 4\pi\chi(\omega_{\lambda})} \equiv \omega_{\lambda} \eta_{\lambda}$$

They are the pulse envelope functions that correspond to the rather simple Fourier transform equation

$$0 = \left[\frac{\partial}{\partial z} + 2iK(\omega) \right] \frac{\partial}{\partial z} {}^0\tilde{\mathcal{E}}_{\lambda}(z, \omega). \quad (\text{II.6-29})$$

We recognize that our choice of $cK(\omega) = \omega\eta(\omega)$ and the selection of forward waves gives a transform equation whose solution is independent of position.

Retarded Times

In a source-free homogeneous medium the transform function can be used to produce the envelope function by means of the formula

$${}^0\mathcal{E}_{\lambda}(z, t) = \int d\omega \exp[i(K(\omega) - k_{\lambda})z - (\omega - \omega_{\lambda})t] {}^0\tilde{\mathcal{E}}_{\lambda}(\omega) \quad (\text{II.6-30})$$

This connection can be expressed in terms of the local mode-time T_{λ} at position z in the form

$${}^0\mathcal{E}_{\lambda} \left[z, T_{\lambda} + \frac{z}{v_{\lambda}} \right] = \int d\omega \exp[i(K_{\lambda}(\omega)z - (\omega - \omega_{\lambda})T_{\lambda})] {}^0\tilde{\mathcal{E}}_{\lambda}(\omega) \quad (\text{II.6-31})$$

where

$$\frac{1}{v_{\lambda}} \equiv \left[\frac{dK}{d\omega} \right]_{\lambda}, \quad K_{\lambda}(\omega) \equiv K(\omega) - K(\omega_{\lambda}) - \left[\frac{dK}{d\omega} \right]_{\lambda} (\omega - \omega_{\lambda}) \quad (\text{II.6-32})$$

When the pulse travels without changing form (i.e. when one can neglect derivatives

higher than first order) then the envelope function is independent of z . In this approximation $K_\lambda(\omega)$ is negligible. In more general cases (when $K_\lambda(\omega)$ is not negligible) we can still use this construction, given a Fourier transform function, to produce an envelope function.

We recognize that the introduction of a local time $T_v = t - z/v$, retarded with the aid of some velocity v , is equivalent to the relationship

$$\mathcal{E}_\lambda(z, T_v + z/v) = \int d\omega \exp[i(K_v(\omega)z - (\omega - \omega_\lambda)T_v)] \tilde{\mathcal{E}}_\lambda(z, \omega) \quad (\text{II.6-33})$$

in which the frequency dependent function $K_v(\omega)$ is defined by

$$cK_v(\omega) = \omega\eta(\omega) - \omega_\lambda\eta(\omega_\lambda) - \frac{c}{v}(\omega - \omega_\lambda) \quad (\text{II.6-34})$$

This function vanishes at $\omega = \omega_\lambda$. If the range of frequencies present in $\tilde{\mathcal{E}}_\lambda(z, \omega)$ is small compared with the variation of refractive index, then we can neglect $K_v(\omega)$ and can approximate the envelope function by

$$\mathcal{E}_\lambda(z, T_v + z/v) \simeq \int d\omega \exp[-i(\omega - \omega_\lambda)T_v] \tilde{\mathcal{E}}_\lambda(z, \omega) \quad (\text{II.6-35})$$

When this approximation holds we can write the source-free field as

$${}^0\mathcal{E}_\lambda(z, T_v + z/v) \simeq \int d\omega \exp[-i(\omega - \omega_\lambda)T_v] {}^0\tilde{\mathcal{E}}_\lambda(\omega) \equiv {}^0\mathcal{E}_\lambda(T_v) \quad (\text{II.6-36})$$

That is, the envelope function depends only on the time interval in a retarded reference frame. This approximation will hold so long as there is negligible value for the phase,

$$\begin{aligned} 2\pi \gg |K_v(\omega_\lambda + \Delta)z| &= \left| \frac{z\omega_\lambda}{c} \left[\eta(\omega_\lambda + \Delta) - \eta(\omega_\lambda) + \frac{\Delta}{\omega_\lambda} \left[\eta(\omega_\lambda + \Delta) - \frac{c}{v} \right] \right] \right| \\ &\simeq \left| \frac{z\Delta}{c} \left[\eta_\lambda - \frac{c}{v} + \frac{1}{\omega_\lambda} \left[\frac{\partial \eta}{\partial \omega} \right]_\lambda \right] \right| \end{aligned} \quad (\text{II.6-37})$$

where Δ is a measure of the frequency spread, away from $\omega = \omega_\lambda$, present in the Fourier amplitude. As distances become sufficiently long, or contributing bandwidths Δ become sufficiently large, this approximation fails (even in the absence of nonlinear polarization), and then one must use the more accurate formula (II.6-33) to construct the envelope function from the Fourier transform.

As long as we deal only with the Fourier transform we need not be concerned with the choice of a retarded time T_v or with the choice of a velocity v with which the retarded reference frame moves. This choice is only needed when we wish to economize our description of time variations at a fixed point, by selecting a time window that includes the dominant portions of the field. When we deal with fixed reference time t , rather than with retarded time, there is no need for a group velocity.

When the medium is very dispersive, or when nonlinear polarization terms significantly affect the propagation, pulse shapes become very distorted. One may find that a pulse breaks into separate solitons, each moving with a different group velocity. The result is a waveform (or succession of pulses) that fill a large time interval. The precise choice of a velocity v for the center of this window is less important than the width of the window.

§II.7 The Nonlinear Polarization

When nonlinear polarization is present we can use a double Fourier transform, in space and time, to provide the following formal expression for the electric field:

$$\mathbf{E}^{(+)}(\mathbf{z}, t) = {}^0\mathbf{E}^{(+)}(\mathbf{z}, t) \quad (\text{II.7-1})$$

$$+ \frac{1}{2} \sum_{\lambda} \mathbf{e}(\lambda) \int d\mathbf{K} \int d\omega \frac{\exp[i\mathbf{K}\mathbf{z} - i\omega t]}{[c\mathbf{K} - \eta(\omega)\omega][c\mathbf{K} + \eta(\omega)\omega]} (\omega)^2 4\pi^{\text{non}} \tilde{\mathcal{P}}_{\lambda}(\mathbf{K}, \omega)$$

Here ${}^0\mathbf{E}$ is a solution to the homogeneous wave equation, in the absence of ${}^{\text{non}}\mathbf{P}$. This expression is only formal because the dependence of the nonlinear polarization field upon the electric field has been left implicit. Nevertheless it shows the presence of forward and backward scattering waves, obtained by evaluating the integral at the two poles. By omitting one of these poles (or by dealing with a first-order equation) we eliminate the backward-wave contribution to the field.

Including Nonlinear Polarization

Now let us consider the effect of the nonlinear polarization. We cannot use the source-free construction (II.5-15) as it stands for this field, because that construction produces a solution in the absence of nonlinear polarization. We can modify that construction in several ways, including

I.) Alter the dispersion relation on $\mathbf{K}(\omega)$

II.) Remove the requirement that the Fourier envelope function be independent of position.

To permit these approaches we introduce the construction

$$\mathbf{E}^{(+)}(\mathbf{z}, t) = \frac{1}{2} \sum_{\lambda} \mathbf{e}(\lambda) \int d\omega \exp[i\mathbf{K}(\omega)\mathbf{z} - i\omega t] \tilde{\mathcal{E}}_{\lambda}(\mathbf{z}, \omega). \quad (\text{II.7-2})$$

That is, we now have $\tilde{\mathcal{E}}_{\lambda}(\mathbf{z}, \omega)$ in place of ${}^0\tilde{\mathcal{E}}_{\lambda}(\omega)$. The phase $\mathbf{K}(\omega)\mathbf{z}$ is introduced with the intention of removing the dominant part of the spatial variation for forward-travelling waves, leaving a function that varies, on average, more slowly with distance. (Note, however, that any backward-travelling waves will contribute a rapidly varying phase $\exp[-2i\mathbf{K}(\omega)\mathbf{z}]$ to the envelope.) The wave equation becomes

$$\frac{\partial^2}{\partial z^2} \int d\omega \exp[i\mathbf{K}(\omega)\mathbf{z} - i\omega t] \tilde{\mathcal{E}}_{\lambda}(\mathbf{z}, \omega)$$

$$\begin{aligned}
& -\frac{1}{c^2} \frac{\partial^2}{\partial t^2} \int d\omega \exp[iK(\omega)z - i\omega t] \eta(\omega)^2 \tilde{\mathcal{E}}_\lambda(z, \omega) \\
& = \frac{4\pi}{c^2} \frac{\partial^2}{\partial t^2} \int d\omega \exp[iK(\omega)z - i\omega t] \omega^2 \tilde{\mathcal{P}}_\lambda(z, \omega)
\end{aligned} \quad (\text{II.7-3})$$

We can obtain from this equation the equation for the temporal Fourier transform:

$$\left[\frac{\partial^2}{\partial z^2} + 2iK(\omega) \frac{\partial}{\partial z} - K(\omega)^2 + \frac{\omega^2}{c^2} \eta(\omega)^2 \right] \tilde{\mathcal{E}}_\lambda(z, \omega) = -\frac{4\pi}{c^2} \omega^2 \text{non} \tilde{\mathcal{P}}_\lambda(z, \omega). \quad (\text{II.7-4})$$

This equation may also be written as

$$\left[\frac{\partial}{\partial z} + iK(\omega) + \frac{i\omega\eta(\omega)}{c} \right] \left[\frac{\partial}{\partial z} + iK(\omega) - \frac{i\omega\eta(\omega)}{c} \right] \tilde{\mathcal{E}}_\lambda(z, \omega) = -\frac{4\pi}{c^2} \omega^2 \text{non} \tilde{\mathcal{P}}_\lambda(z, \omega). \quad (\text{II.7-5})$$

The function $\eta(\omega) \equiv \sqrt{1 + 4\pi\chi(\omega)}$ that appears here is, for present purposes, taken to be a known function of frequency. In particular, we assume that we can evaluate this function, as well as all derivatives with respect to ω , at any desired frequency.

Let us assume that the function $K(\omega)$ remains, as in the source-free case for forward-travelling waves,

$$cK(\omega) = \omega\eta(\omega) \equiv \sqrt{1 + 4\pi\chi(\omega)}. \quad (\text{II.7-6})$$

This dispersion relation provides a connection between time derivatives and space derivatives of the field E . With this relationship the propagation equation becomes the transform equation

$$\left[\frac{\partial^2}{\partial z^2} + 2iK(\omega) \frac{\partial}{\partial z} \right] \tilde{\mathcal{E}}_\lambda(z, \omega) = -\frac{4\pi}{c^2} \omega^2 \text{non} \tilde{\mathcal{P}}_\lambda(z, \omega). \quad (\text{II.7-7})$$

When there is no nonlinear polarization the solution is any arbitrary space-independent function of ω . That source-free equation may be written

$$\left[\frac{\partial}{\partial z} + 2iK(\omega) \right] \frac{\partial}{\partial z} \tilde{\mathcal{E}}_\lambda(\omega) = 0. \quad (\text{II.7-8})$$

or as

$$\left[\frac{\partial}{\partial z} + iK(\omega) - \frac{i\omega\eta(\omega)}{c} \right] \tilde{\mathcal{E}}_\lambda(\omega) = 0. \quad (\text{II.7-10a})$$

or as

$$\left[K(\omega) \frac{\partial}{\partial z} + iK(\omega)^2 - \frac{i\omega^2 \eta(\omega)^2}{c^2} \right] {}^0\tilde{\mathcal{E}}_\lambda(\omega) = 0. \quad (\text{II.7-10b})$$

These equations are satisfied because the spatial derivatives vanish and because $cK(\omega) = \omega\eta(\omega)$ for forward waves. When nonlinear polarization is present one must consider the spatial variation of the transform $\tilde{\mathcal{E}}_\lambda(z, \omega)$. (One could also alter the dispersion relationship.)

If the source term (the nonlinear polarization) produces slow spatial variations of the envelope we can neglect the second derivative $\partial^2/\partial z^2$. What is relevant to such an approximation is the distance scale $1/K(\omega)$. We ask, is the variation slow over this distance? If $K(\omega)$ changes appreciably with ω over the frequency interval that contributes to the pulse (e.g. there is a resonance) then different frequencies will have different scale lengths. This is not the case we are interested in. If there are such resonance effects they should be placed into the nonlinear polarization, not the linear polarization.

§II.8 Spatially First Order Equations

We usually assume that backward travelling waves may be neglected. The lack of backward waves (traceable to the absence of spatial gradients in material properties on a distance scale comparable to a wavelength) allows us to drop the second derivative $\partial^2/\partial z^2$. One may also argue for the neglect of this derivative on the basis that spatial changes in the envelope function occur over a distance that is much larger than $1/k_\lambda$. We therefore wish an equation that is first order in z .

We also assume that the time scale for temporal change in the envelope is long compared with various derivatives of the susceptibility. The relevant scale for the n th derivative is $d^n K^2/d\omega^n$.

The propagation equation may be viewed either in the time domain, where it becomes an equation for an envelope function $\mathcal{E}_\lambda(z, t)$, or in the frequency domain, where it becomes an equation for a transform function $\tilde{\mathcal{E}}_\lambda(z, \omega)$. Consider the wave equation for the temporal Fourier transform

$$\left[\frac{\partial^2}{\partial z^2} + 2iK(\omega) \frac{\partial}{\partial z} - K(\omega)^2 + \frac{\omega^2}{c^2} \eta(\omega)^2 \right] \tilde{\mathcal{E}}_\lambda(z, \omega) = - \frac{4\pi}{c^2} \omega^2 \text{non} \tilde{\mathcal{P}}_\lambda(z, \omega). \quad (\text{II.8-1})$$

The factor $K(\omega)^2$ originates with spatial derivatives whereas the factor $\omega^2 \eta(\omega)^2/c^2$ originates with time derivatives; the two terms are equal. We introduced above an expansion for the variable $c^2 K^2(\omega) = \omega^2 \eta(\omega)^2$ about the value $\omega = \omega_\lambda$ and wrote

$$\left[\frac{\partial^2}{\partial z^2} + 2iK(\omega) \frac{\partial}{\partial z} - K(\omega)^2 + \sum_{n=0}^{\infty} \frac{1}{n!} \left[\frac{d^n K^2}{d\omega^n} \right]_\lambda (\omega - \omega_\lambda)^n \right] \tilde{\mathcal{E}}_\lambda(z, \omega) = - \frac{4\pi}{c^2} \omega^2 \text{non} \tilde{\mathcal{P}}_\lambda(z, \omega). \quad (\text{II.8-2})$$

This Fourier transform equation produces an envelope equation that is second order in the spatial derivative,

$$\left[\frac{\partial^2}{\partial z^2} + 2ik_\lambda \frac{\partial}{\partial z} - (k_\lambda)^2 + \sum_{n=0}^{\infty} \frac{1}{n!} \left[\frac{d^n K^2}{d\omega^n} \right]_\lambda \left[i \frac{\partial}{\partial t} \right]^n \right] \mathcal{E}_\lambda(z, t) = - \frac{4\pi}{c^2} (\omega_\lambda)^2 \left[1 + \frac{2i}{\omega_\lambda} \frac{\partial}{\partial t} - \frac{1}{(\omega_\lambda)^2} \frac{\partial^2}{\partial t^2} \right] \text{non} \mathcal{P}_\lambda(z, t) \quad (\text{II.8-3})$$

We desire an equation that is first order in $\partial/\partial z$. Two possibilities may be considered: either drop the $\partial^2/\partial z^2$ from the envelope equation, or else drop the $\partial^2/\partial z^2$ from the

transform equation and then determine the envelope equation corresponding to this transform equation.

Truncating the Envelope Equation

Let us first consider the envelope equation, and neglect the second derivative $\partial^2/\partial z^2$ in this equation. We obtain the approximate first-order envelope equation

$$\left[\frac{\partial}{\partial z} + \frac{1}{2ik_\lambda} \sum_{n=1}^{\infty} \frac{1}{n!} \left[\frac{d^n K^2}{d\omega^n} \right]_\lambda \left(i \frac{\partial}{\partial t} \right)^n \right] \mathcal{E}_\lambda(z, t) \\ = \frac{2\pi i}{c^2} \frac{(\omega_\lambda)^2}{k_\lambda} \left[1 + 2i\omega_\lambda \frac{\partial}{\partial t} - \frac{\partial^2}{\partial t^2} \right] \text{non } \mathcal{E}_\lambda(z, t) \quad (\text{II.8-4})$$

We may note that, as a result of dropping the derivative $\partial^2/\partial z^2$, our pulse equation is equivalent to the Fourier transform equation

$$\left[2ik_\lambda \frac{\partial}{\partial z} - 2k_\lambda K(\omega) + (k_\lambda)^2 + \sum_{n=0}^{\infty} \frac{1}{n!} \left[\frac{d^n K^2}{d\omega^n} \right]_\lambda (\omega - \omega_\lambda)^n \right] \tilde{\mathcal{E}}_\lambda(z, \omega) \\ = - \frac{4\pi}{c^2} \omega^2 \text{non } \tilde{\mathcal{E}}_\lambda(z, \omega). \quad (\text{II.8-5})$$

Upon replacing the infinite sum by $K(\omega)^2$ we obtain the transform equation

$$\left[2ik_\lambda \frac{\partial}{\partial z} + [K(\omega) - k_\lambda]^2 \right] \tilde{\mathcal{E}}_\lambda(z, \omega) = - \frac{4\pi}{c^2} \omega^2 \text{non } \tilde{\mathcal{E}}_\lambda(z, \omega). \quad (\text{II.8-6})$$

That is, without truncating the series of time derivatives, the approximate envelope equation differs from what we would obtain by neglecting the second derivative of the transform equation.

Error I

When we apply this equation in the absence of nonlinear polarization,

$$0 = \left[2ik_\lambda \frac{\partial}{\partial z} + [K(\omega) - k_\lambda]^2 \right] {}^0\tilde{\mathcal{E}}_\lambda(z, \omega) \quad (\text{II.8-6})$$

it does not have, as solutions, transform functions that are independent of z . Instead the solutions contain a z -dependent phase,

$${}^0\tilde{\mathcal{E}}_\lambda(z, \omega) = \exp[iQ_\lambda(\omega)z] {}^0\tilde{\mathcal{E}}_\lambda(0, \omega) \quad (\text{II.8-7})$$

where

$$Q_\lambda(\omega) = \frac{1}{k_\lambda} (K(\omega) - k_\lambda)^2 \quad (\text{II.8-8})$$

This phase represents an error in the solution. The size of this error will be proportional to the distance z and will depend on the bandwidth Δ of the frequency distribution of the transform function $\tilde{\mathcal{E}}_\lambda(\omega)$ about $\omega = \omega_\lambda$. We may estimate the effect of this error in a frequency bandwidth Δ by the formula

$$Q_\lambda(L_\lambda + \Delta)z \approx \frac{z}{2k_\lambda} \left[\left[\frac{dK}{d\omega} \right]_\lambda \right] z \Delta^2 \quad (\text{II.8-9})$$

The First Order Equation

When we retain only the lowest derivatives the envelope equation is

$$\left[\frac{\partial}{\partial z} + \left[\frac{dK}{d\omega} \right]_\lambda \frac{\partial}{\partial t} \right] \mathcal{E}_\lambda(z, t) = \frac{2\pi i}{c^2} \frac{(\omega_\lambda)^2}{k_\lambda} \left[1 + \frac{2i}{\omega_\lambda} \frac{\partial}{\partial t} \right] \text{non } \mathcal{E}_\lambda(z, t) \quad (\text{II.8-10})$$

The coefficient of the time derivative is the inverse of the group velocity $v_{gr}(\lambda)$ for mode λ . This definition provides the conventional plane-wave pulse propagation equation

$$\left[\frac{\partial}{\partial z} + \frac{1}{v_{gr}(\lambda)} \frac{\partial}{\partial t} \right] \mathcal{E}_\lambda(z, t) = \frac{2\pi i \omega_\lambda}{c n_\lambda} \left[1 + \frac{2i}{\omega_\lambda} \frac{\partial}{\partial t} \right] \text{non } \mathcal{E}_\lambda(z, t) \quad (\text{II.8-11})$$

This equation differs from that of nondispersive propagation by the presence of the group velocity rather than the velocity of light c (or the phase velocity) as the multiplier of the time derivative, and by the presence of the index of refraction on the right-hand side.

In deriving this equation we have fixed the pulse carrier frequency ω_λ and the wavevector k_λ , and we have assumed that this choice makes the envelope $\mathcal{E}_\lambda(z, t)$ slowly varying in space and time. This assumption requires verification for particular cases.

It is possible to introduce moving coordinate systems, as one does with constant-index propagation. However, it will be necessary to introduce a separate time scale for each mode when each mode has a different propagation velocity.

This envelope equation is equivalent to the Fourier transform equation

$$\begin{aligned} 2ik_\lambda \left[\frac{\partial}{\partial z} + i \left[K(\omega) - k_\lambda - \left[\frac{dK}{d\omega} \right]_\lambda (\omega - \omega_\lambda) \right] \right] \tilde{\mathcal{E}}_\lambda(z, \omega) \\ = - \frac{4\pi}{c^2} [\omega^2 - (\omega - \omega_\lambda)^2] \text{non } \tilde{\mathcal{E}}_\lambda(z, \omega). \end{aligned} \quad (\text{II.8-12})$$

These equations offer the simplest approximate equations that are first order in spatial derivative.

Error 1a

When we apply the first-order equation to propagation in the absence of nonlinear polarization we obtain a solution of the form of Eqn. (II.8-7), in which there is a z-dependent phase error. With the first order equation the error is

$$Q_\lambda(\omega)z = \frac{z}{2k_\lambda} \left[[K(\omega) - k_\lambda]^2 - K(\omega)^2 + \sum_{n=0}^1 \frac{1}{n!} \left[\frac{d^n K^2}{d\omega^n} \right]_\lambda (\omega - \omega_\lambda)^2 \right] \quad (\text{II.8-13})$$

We may estimate the effect of this error in a frequency bandwidth Δ by the formula

$$Q_\lambda(\omega_\lambda + \Delta)z \approx \frac{z}{2k_\lambda} \left[\left[\left[\frac{dK}{d\omega} \right]_\lambda \right]^2 + \frac{1}{2} \left[\frac{d^2 K^2}{d\omega^2} \right]_\lambda \right] \Delta^2$$

Second-Order Equation

Let us truncate the series of time derivatives at second order. The envelope equation becomes

$$\begin{aligned} & \left[\frac{\partial}{\partial z} + \left[\frac{dK}{d\omega} \right]_\lambda \frac{\partial}{\partial t} + \frac{i}{4k_\lambda} \left[\frac{d^2 K^2}{d\omega^2} \right]_\lambda \frac{\partial^2}{\partial t^2} \right] \mathcal{E}_\lambda(z, t) \\ &= \frac{2\pi i}{c^2} \frac{(\omega_\lambda)^3}{k_\lambda} \left[1 + \frac{2i}{\omega_\lambda} \frac{\partial}{\partial t} - \frac{1}{(\omega_\lambda)^2} \frac{\partial^2}{\partial t^2} \right] \text{non } \mathcal{P}_\lambda(z, t) \end{aligned} \quad (\text{II.8-14})$$

This equation represents one possibility for an equation that is first order in z and second order in t. It has been derived from the second order wave equation by assuming a simple susceptibility expression for the relationship between Fourier transforms of electric field and linear polarization field, by then making series expansions of that frequency dependent susceptibility about the carrier frequencies, and finally by neglecting $\partial^2/\partial z^2$ compared with $k_\lambda \partial/\partial z$. It may be written in terms of group and phase velocities as

$$\begin{aligned} & \left[\frac{\partial}{\partial z} + \frac{1}{v_{gr}(\lambda)} \frac{\partial}{\partial t} + \frac{i}{4k_\lambda} \left[\frac{d^2 K^2}{d\omega^2} \right]_\lambda \frac{\partial^2}{\partial t^2} \right] \mathcal{E}_\lambda(z, t) \\ &= \frac{2\pi i}{c^2} v_{ph}(\lambda) \omega_\lambda \left[1 + \frac{2i}{\omega_\lambda} \frac{\partial}{\partial t} - \frac{1}{(\omega_\lambda)^2} \frac{\partial^2}{\partial t^2} \right] \text{non } \mathcal{P}_\lambda(z, t) \end{aligned} \quad (\text{II.8-15})$$

This equation is equivalent to the equation for the Fourier transform,

$$\left[2ik_{\lambda} \frac{\partial}{\partial z} + [K(\omega) - k_{\lambda}]^2 - K(\omega)^2 + \sum_{n=0}^2 \frac{1}{n!} \left[\frac{d^n K^2}{d\omega^n} \right]_{\lambda} (\omega - \omega_{\lambda})^n \right] \tilde{\mathcal{E}}_{\lambda}(z, \omega)$$

$$= - \frac{4\pi}{c^2} \omega^2 \text{non} \tilde{\mathcal{P}}_{\lambda}(z, \omega).$$

(II.8-16)

Error 1b

The envelope equation, applied to propagation in the absence of nonlinear polarization, produces an envelope function of the form of Eqn. (II.8-7). The phase error is

$$Q_{\lambda}(\omega)z = \frac{z}{2k_{\lambda}} \left[[K(\omega) - k_{\lambda}]^2 - K(\omega)^2 + \sum_{n=0}^2 \frac{1}{n!} \left[\frac{d^n K^2}{d\omega^n} \right]_{\lambda} (\omega - \omega_{\lambda})^2 \right] \quad (\text{II.8-13})$$

We may estimate the magnitude of this error for a frequency bandwidth of Δ by the formula

$$Q_{\lambda}(\omega_{\lambda} + \Delta)z \approx \frac{z}{2k_{\lambda}} \left[\left[\frac{dK}{d\omega} \right]_{\lambda} \right]^2 \Delta^2$$

Truncating the Transform equation

This second-order equation may be approached in a different manner. Let us consider instead the transform equation written in factored form as

$$\left[\frac{\partial}{\partial z} + iK(\omega) + \frac{i\omega}{c} \eta(\omega) \right] \left[\frac{\partial}{\partial z} + iK(\omega) - \frac{i\omega}{c} \eta(\omega) \right] \tilde{\mathcal{E}}_{\lambda}(z, \omega) = - \frac{4\pi}{c^2} \omega^2 \text{non} \tilde{\mathcal{P}}_{\lambda}(z, \omega)$$

(II.8-13)

The terms $iK(\omega)$ come from spatial derivatives of the envelope, whereas the terms $i\omega\eta(\omega)/c$ come from the time derivatives. When we concentrate on forward travelling waves we use the definition $cK(\omega) = \omega\eta(\omega)$ to produce the equation

$$\left[\frac{\partial}{\partial z} + 2iK(\omega) \right] \frac{\partial}{\partial z} \tilde{\mathcal{E}}_{\lambda}(z, \omega) = - \frac{4\pi}{c^2} \omega^2 \text{non} \tilde{\mathcal{P}}_{\lambda}(z, \omega)$$

(II.8-14)

When there is no source term on the right-hand side we can omit the first bracketed factor because $\partial/\partial z$ alone, acting on $\tilde{\mathcal{E}}_{\lambda}(\omega)$. When nonlinear polarization is present we neglect the spatial derivative in the first bracketed factor (as is appropriate for forward travelling waves) and obtain the approximate equation

$$2i\eta(\omega) \left[\frac{\partial}{\partial z} + iK(\omega) - \frac{i\omega}{c} \eta(\omega) \right] \tilde{\mathcal{E}}_\lambda(z, \omega) = - \frac{4\pi}{c} \omega \text{ non } \tilde{\mathcal{P}}_\lambda(z, \omega) \quad (\text{II.8-15})$$

We could, of course, add or subtract from the left hand side of this equation any vanishing terms such as multiples of $cK(\omega) - \omega\eta(\omega)$. The particular combination $\partial/\partial z + iK(\omega)$ becomes $\partial/\partial z + ik_\lambda$ acting on the envelope function, so it is useful to express transform equations in terms of this combination. Whereas we have previously dealt with the frequency dependence of $K(\omega)^2$ or $\eta(\omega)^2$, here we have factored those squares, $\eta(\omega)^2 = \eta(\omega)\eta(\omega)$. We see that now we need to introduce an expansion of $cK(\omega) = \omega\eta(\omega)$ rather than the previous expansion of $c^2K(\omega)^2 = \omega^2\eta(\omega)^2$. To do this we can express the frequency variation of $K(\omega)$ as a series in $(\omega - \omega_\lambda)$,

$$K(\omega) = \sum_{n=0}^{\infty} \frac{1}{n!} \left[\frac{d^n K}{d\omega^n} \right]_\lambda (\omega - \omega_\lambda)^n \quad (\text{II.8-16})$$

Upon taking the Fourier transform this becomes a series of time derivatives. We will thereby obtain mixed space-time derivatives. To avoid these, and obtain the desired form of an equation that involves $\partial/\partial z$ alone, we may use the lowest order approximation $K(\omega) \simeq k_\lambda$ and deal with the operator $k_\lambda \partial/\partial z$. This is a second approximation, beyond our neglect of $\partial^2/\partial z^2$. Our approximate transform equation becomes

$$2ik_\lambda \frac{\partial}{\partial z} \tilde{\mathcal{E}}_\lambda(z, \omega) = - \frac{4\pi}{c^2} \omega^2 \text{ non } \tilde{\mathcal{P}}_\lambda(z, \omega). \quad (\text{II.8-17})$$

This equation can be written as

$$\begin{aligned} 2ik_\lambda \left[\frac{\partial}{\partial z} + iK(\omega) - i \sum_{n=0}^{\infty} \frac{1}{n!} \left[\frac{d^n K}{d\omega^n} \right]_\lambda (\omega - \omega_\lambda)^n \right] \tilde{\mathcal{E}}_\lambda(z, \omega) \\ = - \frac{4\pi}{c^2} \omega^2 \text{ non } \tilde{\mathcal{P}}_\lambda(z, \omega) \end{aligned} \quad (\text{II.8-18})$$

This transform equation is equivalent to the spatially first-order envelope equation

$$\begin{aligned} \left[\frac{\partial}{\partial z} - i \sum_{n=1}^{\infty} \frac{1}{n!} \left[\frac{d^n K}{d\omega^n} \right]_\lambda \left(i \frac{\partial}{\partial t} \right)^n \right] \mathcal{E}_\lambda(z, t) \\ = + \frac{2\pi i}{c^2} \frac{(\omega_\lambda)^2}{k_\lambda} \left[1 + \frac{2i}{\omega_\lambda} \frac{\partial}{\partial t} - \frac{1}{(\omega_\lambda)^2} \frac{\partial^2}{\partial t^2} \right] \text{ non } \mathcal{P}_\lambda(z, t) \end{aligned} \quad (\text{II.8-19})$$

Retaining only first and second order time derivatives we obtain the envelope equation

$$\left[\frac{\partial}{\partial z} + \frac{1}{v_{gr}(\lambda)} \frac{\partial}{\partial t} + \frac{i}{2} \left[\frac{d^2 K}{d\omega^2} \right]_{\lambda} \frac{\partial^2}{\partial t^2} \right] \mathcal{E}_{\lambda}(z, t) \\ = + \frac{2\pi i}{c^2} v_{ph}(\lambda) \omega_{\lambda} \left[1 + \frac{2i}{\omega_{\lambda}} \frac{\partial}{\partial t} - \frac{1}{(\omega_{\lambda})^2} \frac{\partial^2}{\partial t^2} \right] \text{non } \mathcal{P}_{\lambda}(z, t). \quad (\text{II.8-20})$$

This equation is not the equation that we obtain by dropping the second derivative term from the second-order envelope equation. It differs in the coefficient of $\partial^2/\partial t^2$. However, we should recognize that this approximation has replaced $\eta(\omega)^2$ by $\eta(\omega)\eta_{\lambda}$. When transformed to the time domain this approximation alters the second time derivative. The difference in the coefficients of $\partial^2/\partial t^2$ in the two cases is

$$\frac{1}{4k_{\lambda}} \left[\frac{d^2 K^2}{d\omega^2} \right]_{\lambda} - \frac{1}{2} \left[\frac{d^2 K}{d\omega^2} \right]_{\lambda} = \frac{1}{2k_{\lambda}} \frac{1}{(v_{gr}(\lambda))^2} \quad (\text{II.8-21})$$

The present envelope equation is equivalent to the transform equation

$$2ik_{\lambda} \left[\frac{\partial}{\partial z} + iK(\omega) - i \sum_{n=0}^2 \frac{1}{n!} \left[\frac{d^n K}{d\omega^n} \right]_{\lambda} (\omega - \omega_{\lambda})^n \right] \tilde{\mathcal{E}}_{\lambda}(z, \omega) \\ = - \frac{4\pi}{c^2} \omega^2 \text{non } \tilde{\mathcal{P}}_{\lambda}(z, \omega). \quad (\text{II.8-22})$$

Error II

When applied to source-free propagation these approximate equations produce an envelope function of the form of Eqn. (II.8-7) that contains a phase error. This error is

$$Q_{\lambda}(\omega)z = K(\omega) - \sum_{n=0}^2 \frac{1}{n!} \left[\frac{d^n K}{d\omega^n} \right]_{\lambda} (\omega - \omega_{\lambda})^n. \quad (\text{II.8-23})$$

We may estimate the effect of this error by considering a frequency bandwidth Δ . We obtain the estimate

$$Q_{\lambda}(\omega_{\lambda} + \Delta)z \simeq \frac{z}{6} \left[\frac{d^3 K}{d\omega^3} \right]_{\lambda} \Delta^3 \quad (\text{II.8-24})$$

Improved Approximation

We can improve the approximation by starting from the transform equation

$$\frac{\partial}{\partial z} \tilde{\mathcal{E}}_{\lambda}(z, \omega) = \frac{2\pi i}{c^2} \frac{\omega^2}{K(\omega)} \text{non } \tilde{\mathcal{P}}_{\lambda}(z, \omega) \quad (\text{II.8-25})$$

and making an expansion for $K(\omega)^{-1}$:

$$\frac{1}{K(\omega)} = \frac{1}{(k_\lambda)^2} \left[k_\lambda - \left[\frac{dK}{d\omega} \right]_\lambda (\omega - \omega_\lambda) + \left[\frac{d^2K}{d\omega^2} \right]_\lambda - \frac{1}{k_\lambda} \frac{1}{(v_{gr}(\lambda))^2} \right] (\omega - \omega_\lambda)^2 + \dots \quad (\text{II.8-26})$$

Upon truncating the series expansions at second order we obtain the transform equation

$$\begin{aligned} & \left[\frac{\partial}{\partial z} + iK(\omega) - ik_\lambda - \frac{i(\omega - \omega_\lambda)}{v_{gr}(\lambda)} - \frac{i}{2} \left[\frac{d^2K}{d\omega^2} \right]_\lambda (\omega - \omega_\lambda)^2 \right] \tilde{\mathcal{E}}_\lambda(z, \omega) \\ &= \frac{2\pi i \omega^2}{(ck_\lambda)^2} \left[k_\lambda - \frac{\omega - \omega_\lambda}{v_{gr}(\lambda)} + \left[\frac{d^2K}{d\omega^2} \right]_\lambda - \frac{1}{k_\lambda} \frac{1}{(v_{gr}(\lambda))^2} \right] (\omega - \omega_\lambda)^2 \text{non } \tilde{\mathcal{E}}_\lambda(z, \omega) \end{aligned} \quad (\text{II.8-27})$$

This equation can be converted into an envelope equation. For simplicity we omit the second derivatives of the polarization field. The result is the envelope equation

$$\begin{aligned} & \left[\frac{\partial}{\partial z} + \frac{1}{v_{gr}(\lambda)} \frac{\partial}{\partial t} - \frac{i}{2} \left[\frac{d^2K}{d\omega^2} \right]_\lambda \frac{\partial^2}{\partial t^2} \right] \mathcal{E}_\lambda(z, t) \\ &= \frac{2\pi i (\omega_\lambda)^2}{c^2 k_\lambda} \left[1 + \frac{2i}{\omega_\lambda} \frac{\partial}{\partial t} \right] \text{non } \mathcal{E}_\lambda(z, t) + \frac{1}{v_{gr}(\lambda)} \frac{\partial}{\partial t} 2\pi (\eta_\lambda)^2 \text{non } \mathcal{E}_\lambda(z, t) \end{aligned} \quad (\text{II.8-28})$$

Summary

We have the choice of two pulse-envelope equations that are first order in space and second-order in time. The first of these equations,

$$\begin{aligned} & \left[\frac{\partial}{\partial z} + \frac{1}{v_{gr}(\lambda)} \frac{\partial}{\partial t} + \frac{i}{4k_\lambda} \left[\frac{d^2K^2}{d\omega^2} \right]_\lambda \frac{\partial^2}{\partial t^2} \right] \mathcal{E}_\lambda(z, t) \\ &= \frac{2\pi i}{c^2} v_{ph}(\lambda) \omega_\lambda \left[1 + \frac{2i}{\omega_\lambda} \frac{\partial}{\partial t} - \frac{1}{(\omega_\lambda)^2} \frac{\partial^2}{\partial t^2} \right] \text{non } \mathcal{E}_\lambda(z, t) \end{aligned} \quad (\text{II.8-29})$$

is derived by dropping the second derivative $\partial^2/\partial z^2$ from the envelope equation. When applied to source-free propagation (the absence of nonlinear polarization) it produces an envelope equation whose transform,

$${}^0\tilde{E}_\lambda(z, \omega) = \exp[iQ_\lambda(\omega)z] {}^0\tilde{E}_\lambda(0, \omega) \quad (\text{II.8-30})$$

contains a phase error. For frequency bandwidth Δ the phase error may be estimated as

$$Q_\lambda(\omega_\lambda + \Delta)z \simeq \frac{z}{2k_\lambda} \left[\left[\frac{dK}{d\omega} \right]_\lambda \right] z \Delta^2 \quad (\text{II.8-31})$$

The second-order envelope equation may be regarded as derived from the transform equation

$$\left[2ik_\lambda \frac{\partial}{\partial z} + [K(\omega) - k_\lambda]^2 \right] \tilde{\mathcal{E}}_\lambda(z, \omega) = - \frac{4\pi}{c^2} \omega^2 \text{ non } \tilde{\mathcal{P}}_\lambda(z, \omega). \quad (\text{II.8-32})$$

by truncating the series of time derivatives needed to describe frequency variation of the refractive index.

The second of the equations,

$$\begin{aligned} & \left[\frac{\partial}{\partial z} + \frac{1}{v_g(\lambda)} \frac{\partial}{\partial t} + \frac{i}{2} \left[\frac{d^2 K}{d\omega^2} \right]_\lambda \frac{\partial^2}{\partial t^2} \right] \mathcal{E}_\lambda(z, t) \\ & = + \frac{2\pi i}{c^2} v_{ph}(\lambda) \omega_\lambda \left[1 + \frac{2i}{\omega_\lambda} \frac{\partial}{\partial t} - \frac{1}{(\omega_\lambda)^2} \frac{\partial^2}{\partial t^2} \right] \text{non } \mathcal{P}_\lambda(z, t). \end{aligned} \quad (\text{II.8-33})$$

may be derived from the transform equation

$$2ik_\lambda \frac{\partial}{\partial z} \tilde{\mathcal{E}}_\lambda(z, \omega) = - \frac{4\pi}{c^2} \omega^2 \text{ non } \tilde{\mathcal{P}}_\lambda(z, \omega). \quad (\text{II.8-34})$$

by truncating the series of time derivatives. When there is no nonlinear polarization this equation produces a solution whose transform contains the phase error

$$Q_\lambda(\omega)z = K(\omega) - \sum_{n=0}^2 \frac{1}{n!} \left[\frac{d^n K}{d\omega^n} \right]_\lambda (\omega - \omega_\lambda)^n. \quad (\text{II.8-35})$$

We may estimate the effect of this error by considering a frequency bandwidth Δ . We obtain the estimate

$$Q_\lambda(\omega_\lambda + \Delta)z \simeq \frac{z}{6} \left[\frac{d^3 K}{d\omega^3} \right]_\lambda \Delta^3 \quad (\text{II.8-36})$$

This error may be expected to be smaller than the error of Eqn. (II.8-31).

A third equation

$$\left[\frac{\partial}{\partial z} + \frac{1}{v_g(\lambda)} \frac{\partial}{\partial t} + \frac{i}{2} \left[\frac{d^2 K}{d\omega^2} \right]_\lambda \frac{\partial^2}{\partial t^2} \right] \mathcal{E}_\lambda(z, t)$$

$$\begin{aligned}
&= \frac{2\pi i \omega_\lambda}{c \eta_\lambda} \left[1 + \frac{2i}{\omega_\lambda} \frac{\partial}{\partial t} \right] \text{non } \mathcal{G}_\lambda(z, t) \\
&+ \frac{2\pi i \omega_\lambda}{c \eta_\lambda} \left[1 - \frac{2c}{v_{gr}(\lambda) \eta_\lambda} \frac{\partial^2}{\partial t^2} \right] \text{non } \mathcal{G}_\lambda(z, t).
\end{aligned}
\tag{II.8-29}$$

Each of these transform equations is an approximation to the second-order transform equation

$$\left[\frac{\partial^2}{\partial z^2} + 2iK(\omega) \frac{\partial}{\partial z} \right] \tilde{\mathcal{G}}_\lambda(z, \omega) = - \frac{4\pi}{c^2} \omega^2 \text{non } \tilde{\mathcal{G}}_\lambda(z, \omega).
\tag{II.8-37}$$

The difference in the coefficients of $\partial^2/\partial t^2$ in the two cases is

$$\frac{1}{4k_\lambda} \left[\frac{d^2 K}{d\omega^2} \right]_\lambda - \frac{1}{2} \left[\frac{d^2 K}{d\omega^2} \right]_\lambda = \frac{1}{2k_\lambda} \frac{1}{(v_{gr}(\lambda))^2}
\tag{II.8-38}$$

Comment

The treatment of propagation through dispersive medium by means of time-dependent equations for envelope functions inevitably introduces a phase error. This error accompanies the truncation of an infinite series of time derivatives, needed to treat frequency dependence of the susceptibility. When one works in the frequency domain, with transform functions, this error does not occur. With this Fourier approach one solves a set of uncoupled equations for separate frequency components, and then constructs the required time-dependent envelope functions by a Fourier transform.