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THE STUDY OF THE INTERACTION OF INTENSE
PICOSECOND LIGHT PULSE WITH MATERIALS:
A THEORY OF THIRD HARMONIC GENERATION
IN AN ISOTROPIC NONLINEAR MEDIUM WITH
ULTRASHORT LIGHT PULSES

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WITH MATERIALS

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CHI H. LEE

and

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13. ABSTRACT This report contains the development of a theory for third harmonic generation (THG) due to a broad band optical pulses such as existing in the picosecond time region. The theory is than applied to several examples. Specifically, Maxwell's equations are used as the starting point and a wave equation for the THG is developed for a uniform plane wave in a dielectric, isotropic medium. The material parameters considered are the nonlinear susceptibility giving rise to the third harmonic, the respective phase and group velocities, and the interaction length. Three examples consisting of a sine wave, a Gaussian pulse and a linearly chirped pulse are evaluated.		

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IN



A THEORY OF THIRD HARMONIC GENERATION
IN AN ISOTROPIC NONLINEAR MEDIUM WITH
ULTRASHORT LIGHT PULSES

by

David W. Coffey*

and

Chi H. Lee

*Submitted to the Faculty of the Graduate School of the University of Maryland in partial fulfillment of the requirements for the degree of the Master of Science, 1972.

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SECTION I

INTRODUCTION

This thesis contains a theoretical investigation into non-linear electromagnetic-field theory as applied to third harmonic generation in bulk materials. Such a topic is of interest since the invention of the field known as non-linear optics. With the high power levels available from lasers, practical experiments in non-linear optics have become a reality. Of course, the generation of harmonics and the heterodyning of different frequencies or wavelengths is desirable for various reasons. The most compelling reason for investigating third harmonic generation (THG), the topic of this thesis, is for use as a tool in investigating the structure of ultra-short laser pulses known as pico-second pulses.¹ These pulses are on the order of pico-seconds, a time regime so short as to be unresolvable in real time by present techniques. As a result, non-linear processes such as harmonic generation are used to infer the characteristics of these pico-second pulses. It is for this purpose that the investigation was undertaken.

In format, this thesis consists of introduction, the development of the THG theory and several examples of the application of the theory to elementary fundamental functions such as a sine wave, a gaussian pulse and a linearly chirped pulse. The theory begins with

1. DeMaria, A. J., D. A. Stetser, and W. H. Glenn, Jr., "Ultrashort Light Pulses," Science, Vol. 156, No. 3782, pp 1557-1568, 23 June (1967).

Maxwell's Equation and concludes with an expression for THG under conditions of a uniform plane wave in an isotropic non-linear medium. As a result of the developed theory, the influence of excitation and material parameters can be evaluated. Appendices are included to elaborate on portions of the various developments that would be unwieldy in the body of the thesis as well as to elaborate on interesting issues of insufficient importance to be included as part of the principal effort.

SECTION II

A THEORY OF THG

A. Definitions. This section of the paper presents a description of THG as developed for a uniform plane wave in an isotropic non-linear medium. The equations that are obtained are sufficiently general to take any real temporal function, whether casual or not, and obtain the resulting third harmonic conversion in both the time and frequency domains. For a point at which to begin, the definition of a Fourier transform pair is given and also symbology that will be employed in the mathematical developments. Thus, the Fourier transform,

$$F(\omega) = \int_{-\infty}^{\infty} \mathcal{F}(t) e^{-j\omega t} dt \quad ,$$

and its inverse transform,

$$\mathcal{F}(t) = \frac{1}{2\pi} \int_{-\infty}^{\infty} F(\omega) e^{j\omega t} d\omega \quad ,$$

are given. In the functional notation to be used, functions designated by script capital letters belong in the time domain and those designated by block capital letters are in the frequency domain. Also, the Fourier transform relationship may be symbolized as

$$\mathcal{F}(t) \leftrightarrow F(\omega) \quad .$$

The functions of interest are defined as the fundamental function or exciting function, $\mathcal{E}_f(z, t)$, as

$$\mathcal{E}_f(z, t) = \frac{1}{2\pi} \int_{-\infty}^{\infty} E_f(z, \omega) e^{-jk(\omega)z} e^{j\omega t} d\omega \quad ,$$

the generated function or response function, $E_g(z, t)$, as

$$E_g(z, t) = \frac{i}{2\pi} \int_{-\infty}^{\infty} E_g(z, \omega) e^{-jk(\omega)z} e^{j\omega t} d\omega ,$$

and the polarizability, $P(z, t)$, as

$$P(z, t) = \frac{1}{2\pi} \int_{-\infty}^{\infty} P(z, \omega) e^{j\omega t} d\omega .$$

$E_f(z, \omega)$ and $E_g(z, \omega)$ represent their respective temporal functions less phase factors associated with propagation while $P(z, \omega)$ is a complete description at all points.

B. Relating $E_g(z, \omega)$ to $P(z, \omega)$. Beginning with Maxwell's

Equations, a mathematical development will be pursued that results in finding $E_g(z, \omega)$ as a function of $P(z, \omega)$. There will be approximations made as the development progresses and these will be explicitly noted.

Maxwell's Equations:

$$\vec{\nabla} \times \vec{E} = - \frac{\partial}{\partial t} \vec{B}$$

$$\vec{\nabla} \cdot \vec{D} = \rho$$

$$\vec{\nabla} \times \vec{H} = \vec{J} + \frac{\partial}{\partial t} \vec{D}$$

$$\vec{\nabla} \cdot \vec{B} = 0$$

and the additional relationships:

$$\vec{B} = \mu \vec{H}$$

$$\vec{D} = \epsilon_0 \vec{E} + \vec{P}$$

Using Maxwell's Equations, a wave equation is derived. It is assumed that the medium is charge free ($\rho=0$) and of zero conductivity ($\sigma=0$). The following sequence outlines the derivation of the wave equations.

$$\begin{aligned}
\vec{\nabla} \times \vec{E} &= -\frac{\partial}{\partial t} \vec{B} \\
&= -\mu \frac{\partial}{\partial t} \vec{H} \\
\vec{\nabla} \times \vec{H} &= \vec{J} + \frac{\partial}{\partial t} \vec{D} \\
&= \epsilon_0 \frac{\partial}{\partial t} \vec{E} + \frac{\partial}{\partial t} \vec{P} \\
\frac{\partial}{\partial t} (\vec{\nabla} \times \vec{H}) &= \vec{\nabla} \times \left(\frac{\partial}{\partial t} \vec{H} \right) \\
&= \epsilon_0 \frac{\partial^2}{\partial t^2} \vec{E} + \frac{\partial^2}{\partial t^2} \vec{P} \\
&= \frac{1}{\mu} \vec{\nabla} \times \vec{\nabla} \times \vec{E} \\
&= -\frac{1}{\mu} (\vec{\nabla} \cdot \vec{E} - \nabla^2 \vec{E}) .
\end{aligned}$$

$$\text{Thus, } \nabla^2 \vec{E} = \mu \epsilon \frac{\partial^2}{\partial t^2} \vec{E} + \mu \frac{\partial^2}{\partial t^2} \vec{P} .$$

Because the medium is assumed to be isotropic, propagation can be restricted to any axis without loss of generality. Consequently, let a uniform plane wave propagate down the z-axis. The wave equation has now become:

$$\frac{\partial^2}{\partial z^2} \vec{E} = \mu \epsilon_0 \frac{\partial^2}{\partial t^2} \vec{E} + \mu \frac{\partial^2}{\partial t^2} \vec{P} .$$

The wave equation as given above possesses no coupling between coordinates due to vector operations. Therefore, the expression of the wave equation for the components in each coordinate axis is independent of the other two. It is possible to examine either the x-axis or y-axis component and not lose any accuracy. Consequently, no distinction will be made and only one component will be used with the result that the wave equation is now a scalar equation of the form:

$$\frac{\partial^2}{\partial z^2} \mathcal{E}(z,t) = \mu \epsilon_0 \frac{\partial^2}{\partial t^2} \mathcal{E}(z,t) + \mu \frac{\partial^2}{\partial t^2} \rho(z,t) .$$

Now that a satisfactory wave equation has been obtained, the relationship between the exciting field and the generated field must be obtained. To do this, let

$$\mathcal{E}(z,t) = \mathcal{E}_f(z,t) + \mathcal{E}_g(z,t) .$$

This relates the instantaneous field, $\mathcal{E}(z,t)$, to the fundamental or exciting field, $\mathcal{E}_f(z,t)$, and the generated field, $\mathcal{E}_g(z,t)$. The wave equation now becomes:

$$\frac{\partial^2}{\partial z^2} [\mathcal{E}_f(z,t) + \mathcal{E}_g(z,t)] = \mu \epsilon_0 \frac{\partial^2}{\partial t^2} [\mathcal{E}_f(z,t) + \mathcal{E}_g(z,t)] + \mu \frac{\partial^2}{\partial t^2} \rho(z,t) .$$

It is desirable to relate these functions to their frequency domain counterparts. For this reason, the following definitions, previously presented, will be applied to the latest form of the wave equation.

$$\mathcal{E}_f(z,t) = \frac{1}{2\pi} \int_{-\infty}^{\infty} E_f(z,\omega) e^{-jk(\omega)z} e^{j\omega t} d\omega$$

$$\mathcal{E}_g(z,t) = \frac{1}{2\pi} \int_{-\infty}^{\infty} E_g(z,\omega) e^{-jk(\omega)z} e^{j\omega t} d\omega$$

$$\rho(z,t) = \frac{1}{2\pi} \int_{-\infty}^{\infty} P(z,\omega) e^{j\omega t} d\omega .$$

Thus,

$$\begin{aligned} & \frac{\partial^2}{\partial z^2} \left[\frac{1}{2\pi} \int_{-\infty}^{\infty} E_f(z, \omega) e^{-jk(\omega)z} e^{j\omega t} d\omega + \frac{1}{2\pi} \int_{-\infty}^{\infty} E_g(z, \omega) e^{-jk(\omega)z} e^{j\omega t} d\omega \right] \\ & + \epsilon_0 \frac{\partial^2}{\partial t^2} \left[\frac{1}{2\pi} \int_{-\infty}^{\infty} E_f(z, \omega) e^{-jk(\omega)z} e^{j\omega t} d\omega + \frac{1}{2\pi} \int_{-\infty}^{\infty} E_g(z, \omega) e^{-jk(\omega)z} e^{j\omega t} d\omega \right] \\ & + \mu \frac{\partial^2}{\partial t^2} \left[\frac{1}{2\pi} \int_{-\infty}^{\infty} P(z, \omega) e^{j\omega t} d\omega \right] . \end{aligned}$$

Performing the indicated differentiation results in

$$\begin{aligned} & \frac{1}{2\pi} \int_{-\infty}^{\infty} \left[\frac{\partial^2}{\partial z^2} E_f(z, \omega) - j2k(\omega) \frac{\partial}{\partial z} E_f(z, \omega) - k(\omega)^2 E_f(z, \omega) \right] e^{-jk(\omega)z} e^{j\omega t} d\omega \\ & + \frac{1}{2\pi} \int_{-\infty}^{\infty} \left[\frac{\partial^2}{\partial z^2} E_g(z, \omega) - j2k(\omega) \frac{\partial}{\partial z} E_g(z, \omega) - k(\omega)^2 E_g(z, \omega) \right] e^{-jk(\omega)z} e^{j\omega t} d\omega \\ & + \epsilon_0 \left[\frac{1}{2\pi} \int_{-\infty}^{\infty} (-\omega^2) E_f(z, \omega) e^{-jk(\omega)z} e^{j\omega t} d\omega + \frac{1}{2\pi} \int_{-\infty}^{\infty} (-\omega^2) E_g(z, \omega) e^{-jk(\omega)z} e^{j\omega t} d\omega \right] \\ & + \mu \left[\frac{1}{2\pi} \int_{-\infty}^{\infty} (-\omega^2) P(z, \omega) e^{j\omega t} d\omega \right] . \end{aligned}$$

Assume that the conversion of the fundamental to the third harmonic is sufficiently small that the amplitude of $E_f(z, \omega)$ is essentially unchanged as the wave passes through the medium. Then the derivatives with respect to z of $E_f(z, \omega)$ may be set equal to zero. Using the above assumption and dropping the common factor of $\frac{1}{2\pi}$, the wave equation is

$$\begin{aligned}
& - \int_{-\infty}^{\infty} k(\omega)^2 E_f(z, \omega) e^{-jk(\omega)z} e^{j\omega t} d\omega + \int_{-\infty}^{\infty} \left[\frac{\partial^2 E_g}{\partial z^2}(z, \omega) - j2k(\omega) \frac{\partial E_g}{\partial z}(z, \omega) - \right. \\
& \left. k(\omega)^2 E_g(z, \omega) \right] e^{-jk(\omega)z} e^{j\omega t} d\omega = -\mu \epsilon_0 \int_{-\infty}^{\infty} \omega^2 E_f(z, \omega) e^{-jk(\omega)z} e^{j\omega t} d\omega \\
& - \mu \epsilon_0 \int_{-\infty}^{\infty} \omega^2 E_g(z, \omega) e^{-jk(\omega)z} e^{j\omega t} d\omega - \mu \int_{-\infty}^{\infty} \omega^2 P(z, \omega) e^{j\omega t} d\omega .
\end{aligned}$$

The propagation factor, $k(\omega)$, is equal to $(\mu \epsilon)^{\frac{1}{2}} \omega$, where $\epsilon = \epsilon(\omega)$ for most materials and $\mu = \mu_0$ (assuming dielectric materials). Using the relationship, the wave equation simplifies to

$$\begin{aligned}
& \int_{-\infty}^{\infty} \left[\frac{\partial^2 E_g}{\partial z^2}(z, \omega) - j2k(\omega) \frac{\partial E_g}{\partial z}(z, \omega) \right] e^{-jk(\omega)z} e^{j\omega t} d\omega \\
& = -\mu \int_{-\infty}^{\infty} \omega^2 P(z, \omega) e^{j\omega t} d\omega .
\end{aligned}$$

This may be rewritten to take advantage of the common variable of integration and an equation consisting of one integral can be obtained.

$$\int_{-\infty}^{\infty} \left[\frac{\partial^2 E_g}{\partial z^2}(z, \omega) e^{-jk(\omega)z} - j2k(\omega) \frac{\partial E_g}{\partial z}(z, \omega) e^{-jk(\omega)z} + \mu \omega^2 P(z, \omega) \right] e^{j\omega t} d\omega = 0 .$$

It is felt that the factor $\frac{\partial^2 E_g}{\partial z^2}(z, \omega)$ may be equated to zero by assuming that the envelope of the wave is slowly varying.

This results in the equation,

$$\int_{-\infty}^{\infty} \left[\mu \omega^2 P(z, \omega) - j2k(\omega) \frac{\partial E_g}{\partial z}(z, \omega) e^{-jk(\omega)z} \right] e^{j\omega t} d\omega = 0 .$$

For this relationship to be true over the nearly unlimited conditions placed upon it, the integrand must equal zero. Thus, the following equation is obtained

$$\frac{\partial E_g(z, \omega)}{\partial z} = -\frac{\mu \omega^2}{j2k(\omega)} P(z, \omega) e^{jk(\omega)z}$$

Recall that $k(\omega) = (\mu \epsilon)^{\frac{1}{2}} \omega$ and the above equation becomes

$$\frac{\partial E_g(z, \omega)}{\partial z} = -j \frac{k(\omega)}{2\epsilon} P(z, \omega) e^{jk(\omega)z}$$

At this point, an equation relating $E_g(z, \omega)$ to a generating function, $P(z, \omega)$ has been obtained.

C. $P(z, \omega)$ as a function of $E_f(z, \omega)$. In this section, a relationship is developed between $P(z, \omega)$ and $E_f(z, \omega)$. In IIA, the following definition was given:

$$\mathcal{P}(z, t) = \frac{1}{2\pi} \int_{-\infty}^{\infty} P(z, \omega) e^{j\omega t} d\omega$$

It follows that

$$P(z, \omega) = \int_{-\infty}^{\infty} \mathcal{P}(z, t) e^{-j\omega t} dt$$

Let $\mathcal{P}(z, t)$ be defined in such a way that it possesses no linear susceptibility term. This may be achieved by allowing the permittivity, ϵ_0 , of the preceding section to be replaced with ϵ which contains the linear susceptibility term. Because the interest of this paper is THG, only the non-vanishing fourth rank component of the electric susceptibility tensor will be considered. Thus,

$$\mathcal{P}(z, t) = \epsilon_0 \chi \mathcal{E}_f(z, t)^3$$

where χ is the non-vanishing susceptibility component.

$$P(z,t) = \epsilon_0 \chi \left[\frac{1}{2\pi} \int_{-\infty}^{\infty} E_f(z,\omega) e^{-jk(\omega)z} e^{j\omega t} d\omega \right]^3$$

$$= \epsilon_0 \chi \left(\frac{1}{2\pi} \right)^3 \left[\int_{-\infty}^{\infty} E_f(z,\omega') e^{-jk(\omega')z} e^{j\omega' t} d\omega' \right]$$

$$\left[\int_{-\infty}^{\infty} E_f(z,\omega'') e^{-jk(\omega'')z} e^{j\omega'' t} d\omega'' \right] \left[\int_{-\infty}^{\infty} E_f(z,\omega''') e^{-jk(\omega''')z} e^{j\omega''' t} d\omega''' \right]$$

$P(z,\omega)$ is merely the Fourier transform of this equation.

$$P(z,\omega) = \int_{-\infty}^{\infty} \left\{ \epsilon_0 \chi \left(\frac{1}{2\pi} \right)^3 \int_{-\infty}^{\infty} E_f(z,\omega') e^{-jk(\omega')z} e^{j\omega' t} d\omega' \right.$$

$$\left. \int_{-\infty}^{\infty} E_f(z,\omega'') e^{-jk(\omega'')z} e^{j\omega'' t} d\omega'' \int_{-\infty}^{\infty} E_f(z,\omega''') e^{-jk(\omega''')z} e^{j\omega''' t} d\omega'''\right\} e^{-j\omega t} dt$$

This may be rewritten as

$$P(z,\omega) = \epsilon_0 \chi \left(\frac{1}{2\pi} \right)^3 \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} E_f(z,\omega') E_f(z,\omega'') E_f(z,\omega''')$$

$$e^{-j[k(\omega') + k(\omega'') + k(\omega''')]z} e^{-j[\omega - \omega' - \omega'' - \omega''']t} d\omega' d\omega'' d\omega''' dt$$

The relationship

$$\delta(\omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} e^{-j\omega t} dt$$

may be used to obtain the equation

$$P(z, \omega) = \epsilon_0 \chi \left(\frac{1}{2\pi} \right)^2 \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} E_f(z, \omega') E_f(z, \omega'') E_f(z, \omega''') \delta(\omega - \omega' - \omega'' - \omega''') \cdot$$

$$e^{-j[k(\omega') + k(\omega'') + k(\omega''')] z} d\omega' d\omega'' d\omega''' .$$

$P(z, \omega) = 0$ except when $\omega - \omega' - \omega'' - \omega''' = 0$.

This condition is used in the form $\omega' = \omega - \omega'' - \omega'''$ to get

$$P(z, \omega) = \epsilon_0 \chi \left(\frac{1}{2\pi} \right)^2 \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} E_f(z, \omega - \omega'' - \omega''') E_f(z, \omega'') E_f(z, \omega''') \cdot$$

$$e^{-j[k(\omega - \omega'' - \omega''') + k(\omega'') + k(\omega''')] z} d\omega'' d\omega''' .$$

The expression derived in the preceding paragraph is quite general. At this point, let it be assumed that the spectrum of $E_f(z, \omega)$ is highly concentrated about ω_0 , the fundamental frequency. The degree of concentration that is desired is enough so $k(\omega)$ may be represented by the first two terms of a Taylor's Series Expansion. Under these conditions, the following expression is valid.

$$k(\omega) = k(\omega_0) + (\omega - \omega_0) \left. \frac{\partial k}{\partial \omega} \right|_{\omega = \omega_0} .$$

$$\text{Let } k(\omega_0) = k_0$$

$$\text{and } \left. \frac{\partial k}{\partial \omega} \right|_{\omega = \omega_0} = \alpha .$$

Then

$$k(\omega - \omega'' - \omega''') = k_0 + \alpha (\omega - \omega'' - \omega''' - \omega_0)$$

$$k(\omega'') = k_0 + \alpha (\omega'' - \omega_0)$$

$$k(\omega''') = k_0 + \alpha (\omega''' - \omega_0) ,$$

and the sum,

$$k(\omega - \omega'' - \omega''') + k(\omega'') + k(\omega''') = 3k_0 + \alpha(\omega - 3\omega_0) .$$

Thus,

$$P(z, \omega) = \epsilon_0 \chi \left(\frac{1}{2\pi} \right)^2 \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} E_f(z, \omega - \omega'' - \omega''') E_f(z, \omega'') E_f(z, \omega''') e^{-j[3k_0 + \alpha(\omega - 3\omega_0)]z} d\omega'' d\omega''' .$$

Because the phase factor in the integrand has no dependence upon either ω'' or ω''' , it can be removed from the integral and placed with the constant factor. If

$$G(z, \omega) \equiv \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} E_f(z, \omega - \omega'' - \omega''') E_f(z, \omega'') E_f(z, \omega''') d\omega'' d\omega''' ,$$

then

$$P(z, \omega) = \epsilon_0 \chi \left(\frac{1}{2\pi} \right)^2 e^{-j[3k_0 + \alpha(\omega - 3\omega_0)]z} G(z, \omega) .$$

It is interesting to note that $G(z, \omega)$ is the Fourier transform of $E_f(z, t)^3$ without its associated phase factor. Because $E_f(z, t)^3$ is essentially unaltered by its passage through the non-linear medium, then $E_f(0, t)^3 \sim E_f(z, t)^3$ and $G(0, \omega) \sim G(z, \omega)$. It can also be seen that $G(0, \omega)$ has as its inverse Fourier transform $E_f(0, t)^3$. Thus, $G(0, \omega) \leftrightarrow E_f(0, t)^3$. At this stage in the derivations, the relationship between the fundamental field in the time domain, $E_f(z, t)$, and the rate of growth of the third harmonic in the frequency domain, $\frac{\partial}{\partial z} E_g(z, \omega)$, is emerging.

D. Determination of $E_g(z, \omega)$. The actual spectral density of the third harmonic can be calculated by assuming it to be zero at $z=0$ and integrating to z .

$$E_g(z, \omega) = \int_0^z \frac{\partial}{\partial z} E_g(z, \omega) dz .$$

The result of IIB was that

$$\frac{\partial}{\partial z} E_g(z, \omega) = -j \frac{k(\omega)}{2\epsilon} P(z, \omega) e^{jk(\omega)z}$$

Then

$$E_g(z, \omega) = -j \frac{k(\omega)}{2\epsilon} \int_0^z P(z, \omega) e^{jk(\omega)z} dz$$

Using the result of IIC,

$$E_g(z, \omega) = -j \frac{k(\omega)}{2\epsilon} \int_0^z \left\{ \epsilon_0 \chi \left(\frac{1}{2\pi} \right)^2 e^{-j[3k_0 + \alpha(\omega - 3\omega_0)]z} G(o, \omega) \right\} e^{jk(\omega)z} dz$$

Because of the origin of $P(z, \omega)$, it is sharply peaked about $3\omega_0$, and the expansion of $k(\omega)$ about $3\omega_0$ is valid.

$$k(\omega) = k(3\omega_0) + (\omega - 3\omega_0) \left. \frac{\partial k}{\partial \omega} \right|_{\omega=3\omega_0}$$

Let

$$\gamma = \left. \frac{\partial k}{\partial \omega} \right|_{\omega=3\omega_0}$$

$$\text{then } k(\omega) = k_3 + \gamma (\omega - 3\omega_0)$$

and then

$$\begin{aligned} E_g(z, \omega) &= -j \frac{k(\omega) \epsilon_0 \chi}{2\epsilon (2\pi)^2} \int_0^z G(o, \omega) e^{-j[3k_0 + \alpha(\omega - 3\omega_0)]z} e^{j[k_3 + \gamma(\omega - 3\omega_0)]z} dz \\ &= -j \frac{k(\omega) \epsilon_0 \chi}{2\epsilon (2\pi)^2} G(o, \omega) \int_0^z e^{-j[3k_0 - k_3 + (\alpha - \gamma)(\omega - 3\omega_0)]z} dz \\ &= -j \frac{k(\omega) \epsilon_0 \chi}{2\epsilon (2\pi)^2} G(o, \omega) \frac{e^{-j[3k_0 - k_3 + (\alpha - \gamma)(\omega - 3\omega_0)]z}}{-j[3k_0 - k_3 + (\alpha - \gamma)(\omega - 3\omega_0)]} \Bigg|_0^z \end{aligned}$$

Evaluating this and recognizing that $\epsilon_r = \epsilon/\epsilon_0$,

$$E_g(z, \omega) = -j \frac{k(\omega) x}{2\epsilon_r (2\pi)^2} G(o, \omega) e^{-j[3k_0 - k_3 + (\alpha - \gamma)(\omega - 3\omega_0)]z} \frac{-1}{-j[3k_0 - k_3 + (\alpha - \gamma)(\omega - 3\omega_0)]}$$

The last factor can be converted into the form $e^{-j\frac{\theta}{2}} \sin \frac{\theta}{2}$. Also, since $E_g(z, \omega)$ is highly concentrated about $3\omega_0$, let the amplitude factor, $k(\omega)$, be replaced with k_3 . The final equation for this section

is then

$$E_g(z, \omega) = j \frac{k_3 x z}{2\epsilon_r (2\pi)^2} G(o, \omega) e^{-j[3k_0 - k_3 + (\alpha - \gamma)(\omega - 3\omega_0)]z/2} \cdot \frac{\sin\{[3k_0 - k_3 + (\alpha - \gamma)(\omega - 3\omega_0)]z/2\}}{[3k_0 - k_3 + (\alpha - \gamma)(\omega - 3\omega_0)]z/2}$$

E. Determination of $\mathcal{E}_g(z, t)$. The final step is to convert

$E_g(z, \omega)$ into its time domain counterpart. This is done by taking the inverse Fourier transform which, due to the complexity of $E_g(z, \omega)$, is not a simple task. The equation which gives $\mathcal{E}_g(z, t)$ is:

$$\mathcal{E}_g(z, t) = \frac{1}{2\pi} \int_{-\infty}^{\infty} E_g(z, \omega) e^{-jk(\omega)z} e^{j\omega t} d\omega$$

This, upon insertion of the developed form of $E_g(z, \omega)$, becomes:

$$\mathcal{E}_g(z, t) = \frac{1}{2\pi} \int_{-\infty}^{\infty} j \frac{k_3 x z}{2\epsilon_r (2\pi)^2} G(o, \omega) e^{-j[3k_0 - k_3 + (\alpha - \gamma)(\omega - 3\omega_0)]z/2} \cdot \frac{\sin\{[3k_0 - k_3 + (\alpha - \gamma)(\omega - 3\omega_0)]z/2\}}{[3k_0 - k_3 + (\alpha - \gamma)(\omega - 3\omega_0)]z/2} e^{-jk(\omega)z} e^{j\omega t} d\omega$$

The integrand contains three principal factors which are listed here:

1) $G(o, \omega)$,

$$2) e^{-j[3k_0 - k_3 + (\alpha - \gamma)(\omega - 3\omega_0)]z/2} \frac{\sin\{[3k_0 - k_3 + (\alpha - \gamma)(\omega - 3\omega_0)]z/2\}}{[3k_0 - k_3 + (\alpha - \gamma)(\omega - 3\omega_0)]z/2}$$

3) $e^{-jk(\omega)z}$, assuming that k_3 , χ and r_r are constant. These will be treated in order.

Earlier it was stated without proof that $G(o, \omega) \leftrightarrow \mathcal{E}_f(o, t)^3$. This is quite true except for the scale factor associated with $\mathcal{E}_f(o, t)^3$.

The derivation of this relationship follows,

$$P(z, \omega) = \epsilon_0 \chi \left(\frac{1}{2\pi}\right)^2 e^{-j[3k_0 + \alpha(\omega - 3\omega_0)]z} G(z, \omega)$$

from IIC. At $z=0$

$$P(o, \omega) = \epsilon_0 \chi \left(\frac{1}{2\pi}\right)^2 G(o, \omega)$$

It will be recalled that

$$\rho(z, t) = \epsilon_0 \chi \mathcal{E}_f(z, t)^3$$

Therefore, if $\mathcal{H}(o, t) \leftrightarrow G(o, \omega)$, then

$$\epsilon_0 \chi \left(\frac{1}{2\pi}\right)^2 \mathcal{H}(o, t) = \epsilon_0 \chi \mathcal{E}_f(o, t)^3$$

and then, by simply rewriting,

$$\mathcal{H}(o, t) = (2\pi)^2 \mathcal{E}_f(o, t)^3$$

The second of the three principal factors is the most difficult to evaluate. For this reason, the actual development has been placed in Appendix A. The result, however, is

$$f(t) = \begin{cases} \frac{1}{|\alpha - \gamma|z} e^{j[3\omega_0 - \frac{3k_0 - k_3}{\alpha - \gamma}]t} & , \alpha t \leq (\alpha - \gamma)z \\ 0, & \text{all other } t \end{cases}$$

when it is assumed that $\alpha - \gamma \geq 0$. A change in the sense of $\alpha - \gamma$ merely

results in a change in the sense of the inequality.

The last factor is $e^{-jk(\omega)z}$. Using $k(\omega) = k_3 + \gamma(\omega - 3\omega_0)$, the factor becomes

$$e^{-j[k_3 + \gamma(\omega - 3\omega_0)]z}$$

which is equivalent to

$$e^{-j[k_3 - 3\omega_0\gamma]z} e^{-j\gamma zw}$$

The last form consists of a phase factor for propagation of

$$e^{-j[k_3 - 3\omega_0\gamma]z}$$

and a time shift of γz seconds.

The final bit of work consists of using the information gathered about the three integrand factors to obtain $\mathcal{E}_g(z, t)$. This hinges on the fact that

$$F(\omega)G(\omega) \leftrightarrow \int f(\tau)g(t-\tau)d\tau$$

Identifying $F(\omega)$ with the $\sin \omega/\omega$ function,

$$G(\omega) \text{ with } \mathcal{E}_f(0, t)^3$$

and the time delay with the dependent variable, the following expression can be obtained

$$\mathcal{E}_g(z, t + \gamma z) = \frac{jk_3 \chi z}{2\epsilon_r (2\pi)^2} \int_0^{(\alpha - \gamma)z} \frac{1}{|\alpha - \gamma|z} e^{j\left\{ [3\omega_0 - \frac{3k_0 - k_3}{\alpha - \gamma}] \tau - (k_0 - 3\omega_0 \gamma) z \right\}}$$

$$\cdot (2\pi)^2 \mathcal{E}_f(0, t - \tau)^3 d\tau$$

Define a change of variables

$$\tau' = \tau + \gamma z$$

and $t' = t + \gamma z$

Then, using these changes, and cleaning up some simple algebra,

$$\xi_g(z, t') = \frac{jk_3 X}{2\epsilon_r |\alpha - \gamma|} \int_{\gamma z}^{\alpha z} e^{j\left\{ \left[3\omega_0 - \frac{3k_0 - k_3}{\alpha - \gamma} \right] (\tau' - \gamma z) - (k_3 - 3\omega_0 \gamma) z \right\}} \xi_f(0, t' - \tau')^3 d\tau'.$$

Dropping all of the primes gives the third harmonic

$$\xi_g(z, t) = \frac{jk_3 X}{2\epsilon_r |\alpha - \gamma|} \int_{\gamma z}^{\alpha z} e^{j\left\{ \left[3\omega_0 - \frac{3k_0 - k_3}{\alpha - \gamma} \right] \tau + \left[\frac{3k_0 - k_3}{\alpha - \gamma} \gamma - k_3 \right] z \right\}} \xi_f(0, t - \tau)^3 d\tau.$$

In the time domain, the privilege of examining both the real and the imaginary components of a function does not exist. Consequently, a choice must be made. If the fundamental function, $\xi_f(0, t)$, is a real function, then the generated function, $\xi_g(z, t)$, must also be real in order to be observed. Applying the restriction that only $\text{Re} \{ \xi_g(z, t) \}$ is meaningful, then $\xi_g(z, t)$ for this paper becomes

$$\xi_g(z, t) = \text{Re} \left\{ \frac{jk_3 X}{2\epsilon_r |\alpha - \gamma|} \int_{\gamma z}^{\alpha z} e^{j\left\{ \left[3\omega_0 - \frac{3k_0 - k_3}{\alpha - \gamma} \right] \tau + \left[\frac{3k_0 - k_3}{\alpha - \gamma} \gamma - k_3 \right] z \right\}} \xi_f(0, t - \tau)^3 d\tau \right\}.$$

This becomes the final equation of section II.

$$\xi_g(z, t) = \frac{-k_3 X}{2\epsilon_r |\alpha - \gamma|} \int_{\gamma z}^{\alpha z} \sin\left[\left[3\omega_0 - \frac{3k_0 - k_3}{\alpha - \gamma} \right] \tau + \left[\frac{3k_0 - k_3}{\alpha - \gamma} \gamma - k_3 \right] z \right] \xi_f(0, t - \tau)^3 d\tau.$$

$\xi_g(z, t)$ has been found as a function of the fundamental field,

$\xi_f(0, t)$ and parameters of the medium such as $k(\omega)$, $\frac{\partial k}{\partial \omega}(\omega)$, z , ϵ_r and X .

SECTION III

EXAMPLES

A. Eternal Sinusoid. To illustrate the use of the previously obtained equation and as a basic check on the validity of the theory, the elementary case of the eternal sinusoid is considered. Let

$$\mathcal{E}_f(o,t) = \mathcal{E}_o \sin \omega_o t.$$

Then,

$$\mathcal{E}_g(z,t) = \frac{-k_3 X}{2\epsilon_r |\alpha - \gamma|} \int_{\gamma z}^{\alpha z} \sin \left\{ \left[3\omega_o \frac{3k_o - k_3}{\alpha - \gamma} \right] \tau + \left[\frac{3k_o - k_3}{\alpha - \gamma} \gamma - k_3 \right] z \right\}$$

$$\cdot \mathcal{E}_o^3 \sin^3 \{ \omega_o (t - \tau) \} d\tau .$$

Using the identity $\sin^3 X = 3/4 \sin X - 1/4 \sin 3X$ and retaining only the third harmonic term results in

$$\mathcal{E}_g(z,t) = \frac{k_3 X \mathcal{E}_o^3}{8\epsilon_r |\alpha - \gamma|} \int_{\gamma z}^{\alpha z} \sin \left[\left(3\omega_o \frac{3k_o - k_3}{\alpha - \gamma} \right) \tau - \left[\frac{3k_o - k_3}{\alpha - \gamma} \gamma - k_3 \right] z \right] \sin \{ 3\omega_o (t - \tau) \} d\tau .$$

When the group velocities are matched,² $\alpha = \gamma$, and the group velocities equal the average phase velocity,³ $(3k_o + k_3)/2$, then

$$\mathcal{E}_g(z,t) = \frac{-k_3 X \mathcal{E}_o^3 z}{8\epsilon_r} \cdot \frac{\sin \frac{3k_o - k_3}{2} z}{\frac{3k_o - k_3}{2}} \cos \{ 3\omega_o t - k_3 z \} .$$

2. See Appendix B.

3. See Appendix C.

This is in excellent agreement with previously established results⁴ which have the intensity at $3\omega_0$ proportional to

$$x^2 \frac{\sin^2(\frac{l}{2}\Delta k)}{(\frac{l}{2}\Delta k)^2}$$

Thus, it is reasonable to assume this theoretical development to be valid.

B. Gaussian Pulse. This portion of Section III consists of an idealized pulse in the form of a sine wave which is amplitude modulated with a gaussian. The third harmonic that is generated is examined in both the time and frequency domains under conditions of both matched phase velocity and matched group velocity.

The input or fundamental pulse, $\xi_f(o,t)$, is a gaussian modulated sine wave. The temporal characteristics are completely described by the equation

$$\xi_f(o,t) = \xi_0 e^{-t^2/T^2} \sin(\omega_0 t)$$

where T establishes the pulse width. The spectrum, $E_f(o,\omega)$, is defined by the Fourier transform of $\xi_f(o,t)$. This is given by

$$E_f(o,\omega) = \mathcal{F}\{\xi_0 e^{-t^2/T^2} \sin(\omega_0 t)\}$$

$$= \frac{\xi_0}{2\pi} \mathcal{F}\{e^{-t^2/T^2}\} * \mathcal{F}\{\sin \omega_0 t\}$$

where, as usual, $\mathcal{F}\{ \}$ denotes the Fourier transform and the astrisk

4. Bey, Giuliani and Rabin, "Phase-Matched Optical Harmonic Generation in Liquid Media Employing Anomalous Dispersion", IEEE Journal of Quantum Electronics, Vol. QE-4, number 11, pp 932-939, November 1968.

(*) denotes convolution. Appendix E is a proof of the convolution relationship.

$$E_f(o, \omega) = \frac{\xi_o}{2\pi} \{ \sqrt{\pi} T e^{-T^2 \omega^2 / 4} \} * \{ j\pi [\delta(\omega + \omega_o) - \delta(\omega - \omega_o)] \}$$

$$= j \frac{\sqrt{\pi}}{2} \xi_o T \left[e^{-T^2 (\omega - \omega_o)^2 / 4} - e^{-T^2 (\omega + \omega_o)^2 / 4} \right]$$

The power spectrum, $S_f(o, \omega)$, is given by

$$S_f(o, \omega) = E_f(o, \omega) E_f(o, \omega)^* \sqrt{\frac{\epsilon}{\mu}}$$

where the asterisk (*) as a superscript denotes the complex conjugate.

Thus,

$$S_f(o, \omega) = \frac{\pi}{4} T^2 \sqrt{\frac{\epsilon}{\mu}} \left[e^{-T^2 (\omega - \omega_o)^2 / 2} + e^{-T^2 (\omega + \omega_o)^2 / 2} \right] \xi_o^2$$

if it is assumed that the positive and negative frequency components do not overlap. This assumption causes a component about zero frequency to be dropped, but in the case of a laser, this causes no difficulty.

Next, the generated field, $\xi_g(z, t)$, will be calculated. From Section II.E, the following equation is obtained:

$$\xi_g(z, t) = \frac{k_3 x}{2|\alpha - \gamma| \epsilon_r} \int_{yz}^{\alpha z} \sin \left[\left[3\omega_o \frac{3k_o - k_3}{\alpha - \gamma} \right] \tau + \left[\frac{3k_o - k_3}{\alpha - \gamma} \gamma - k_3 \right] z \right] \xi_f(o, t - \tau)^3 d\tau$$

$$\text{For } \xi_f(o, t) = \xi_o e^{-t^2 / T^2} \sin(\omega_o t),$$

$$\xi_g(z, t) = \frac{-k_3 x \xi_o^3}{2|\alpha - \gamma| \epsilon_r} \int_{yz}^{\alpha z} \sin \left[\left[3\omega_o \frac{3k_o - k_3}{\alpha - \gamma} \right] \tau + \left[\frac{3k_o - k_3}{\alpha - \gamma} \gamma - k_3 \right] z \right] e^{-3(t - \tau)^2 / T^2} \cdot \sin^3[\omega_o(t - \tau)] d\tau$$

By use of the trigonometric identity, $\sin^3 x = 3/4 \sin x - 1/4 \sin 3x$, in the above expression and retaining only the term containing $3x$ because the only interest is in the third harmonic, the following equation can be obtained.

$$\begin{aligned} \mathcal{E}_g(z,t) = & \frac{k_3 \epsilon_0^3}{8|\alpha-\gamma| \epsilon_r} \int_{\gamma z}^{\alpha z} \sin\left\{ \left[3\omega_0 \frac{3k_0-k_3}{\alpha-\gamma} \right] \tau + \left[\frac{3k_0-k_3}{\alpha-\gamma} \gamma - k_3 \right] z \right\} e^{-3(t-\tau)^2/T^2} \\ & \cdot \sin[3\omega_0(t-\tau)] d\tau . \end{aligned}$$

The identity, $\sin x \sin y = [\cos(x-y) - \cos(x+y)]/2$, may be used to rid the integrand of the product of trigonometric functions. Thus,

$$\begin{aligned} \mathcal{E}_g(z,t) = & \frac{k_3 \epsilon_0^3}{16|\alpha-\gamma| \epsilon_r} \left[\int_{\gamma z}^{\alpha z} e^{-3(t-\tau)^2/T^2} \right. \\ & \cdot \cos\left\{ \left[6\omega_0 \frac{3k_0-k_3}{\alpha-\gamma} \right] \tau + \left[\frac{3k_0-k_3}{\alpha-\gamma} \gamma - k_3 \right] z - 3\omega_0 t \right\} d\tau \\ & \left. - \int_{\gamma z}^{\alpha z} e^{-3(t-\tau)^2/T^2} \cos\left\{ \left[\frac{3k_0-k_3}{\alpha-\gamma} \right] \tau + \left[\frac{3k_0-k_3}{\alpha-\gamma} \gamma - k_3 \right] z + 3\omega_0 t \right\} d\tau \right] . \end{aligned}$$

For the case where the phase velocities are matched, e.g. $3k_0 = k_3$ (see Appendix C), the expression for $\mathcal{E}_g(z,t)$ becomes

$$\begin{aligned} \mathcal{E}_g(z,t) = & \frac{k_3 \epsilon_0^3}{16|\alpha-\gamma| \epsilon_r} \left[\int_{\gamma z}^{\alpha z} e^{-3(t-\tau)^2/T^2} \right. \\ & \left. \cdot \cos\{6\omega_0 \tau - k_3 z - 3\omega_0 t\} d\tau - \cos\{3\omega_0 t - k_3 z\} \int_{\gamma z}^{\alpha z} e^{-3(t-\tau)^2/T^2} d\tau \right] . \end{aligned}$$

For the case in which the gaussian modulation varies slowly when compared to $6\omega_0$ and the interval $(\alpha-\gamma)z \gg 6\omega_0$, the first integral

can be neglected and then

$$\mathcal{E}_g(z,t) = \frac{-k_3^2 \chi \epsilon_0^3}{16 |\alpha - \gamma| \epsilon_r} \cos[3\omega_0 t - k_3 z] \int_{yz}^{\alpha z} e^{-3(t-\tau)^2/T^2} d\tau.$$

The spectrum for the above expression is given by

$$\mathcal{E}_g(z,\omega) = \mathcal{F}\{\mathcal{E}_g(z,t)\}$$

$$\frac{-jk_3^2 \chi \epsilon_0^3 T \sqrt{\pi}}{32 \sqrt{3} |\alpha - \gamma| \epsilon_r} \left\{ e^{-jk_3 z} e^{-T^2(\omega - 3\omega_0)^2/12} \frac{e^{-j(\omega - 3\omega_0)\alpha z} e^{-j(\omega - 3\omega_0)\gamma z}}{\omega - 3\omega_0} + e^{jk_3 z} e^{-T(\omega + 3\omega_0)^2/12} \frac{e^{-j(\omega + 3\omega_0)\alpha z} e^{-j(\omega + 3\omega_0)\gamma z}}{\omega + 3\omega_0} \right\}.$$

The power spectrum is found as before. Neglecting the positive and negative frequency overlap,

$$S_g(z,\omega) = \frac{k_3^2 \chi^2 \epsilon_0^6 T^2 \pi z^2}{3072 \epsilon_r^2} \sqrt{\frac{\epsilon}{\mu}} \left\{ \left[\frac{\sin[(\omega - 3\omega_0)(\alpha - \gamma)\frac{z}{2}]}{(\omega - 3\omega_0)(\alpha - \gamma)\frac{z}{2}} \right]^2 e^{-T^2(\omega - 3\omega_0)^2/6} + \left[\frac{\sin[(\omega + 3\omega_0)(\alpha - \gamma)\frac{z}{2}]}{(\omega + 3\omega_0)(\alpha - \gamma)\frac{z}{2}} \right]^2 e^{-T^2(\omega + 3\omega_0)^2/6} \right\}.$$

The above calculations are similar in principle to those performed earlier for the fundamental wave. Due to the more complicated functions, these calculations are more involved and not particularly enlightening. Consequently, only the results are shown here.

The third harmonic, $\mathcal{E}_g(z,t)$, can also be found under conditions of matched group velocities between the fundamental, $\mathcal{E}_f(o,t)$, and the third harmonic. The output under matched group velocities, e.g. $\alpha = \gamma$ (see Appendix B), is obtained by taking the limit of $\mathcal{E}_g(z,t)$ as α approaches γ . The treatment is as follows.

$$\begin{aligned} \mathcal{E}_g(z,t) = \lim_{\alpha \rightarrow \gamma} \left\{ \frac{k_3 \chi \mathcal{E}_0^3}{16|\alpha - \gamma| \epsilon_r} \left[\int_{\gamma z}^{\alpha z} e^{-3(t-\tau)^2/T^2} \right. \right. \\ \left. \left. \cdot \cos\left\{ \left[6\omega_0 \frac{3k_0 - k_3}{\alpha - \gamma} \right] \tau + \left[\frac{3k_0 - k_3}{\alpha - \gamma} \gamma - k_3 \right] z - 3\omega_0 t \right\} d\tau \right. \right. \\ \left. \left. - \int_{\gamma z}^{\alpha z} e^{-3(t-\tau)^2/T^2} \cos\left\{ \left[-\frac{3k_0 - k_3}{\alpha - \gamma} \right] \tau + \left[\frac{3k_0 - k_3}{\alpha - \gamma} \gamma - k_3 \right] z + 3\omega_0 t \right\} d\tau \right] \right\}. \end{aligned}$$

Because the limit is being taken, it is reasonable to extract the gaussian function from the integrand by substituting γz for r . The equation then becomes

$$\begin{aligned} \mathcal{E}_g(z,t) = \frac{k_3 \chi \mathcal{E}_0^3}{16\epsilon_r} e^{-3(t-\gamma z)^2/T^2} \lim_{\alpha \rightarrow \gamma} \left\{ \frac{1}{|\alpha - \gamma|} \left[\int_{\gamma z}^{\alpha z} \cos\left\{ \left[6\omega_0 \frac{3k_0 - k_3}{\alpha - \gamma} \right] \tau \right. \right. \right. \\ \left. \left. + \left[\frac{3k_0 - k_3}{\alpha - \gamma} \gamma - k_3 \right] z - 3\omega_0 t \right\} d\tau \right. \right. \\ \left. \left. - \int_{\gamma z}^{\alpha z} \cos\left\{ \left[-\frac{3k_0 - k_3}{\alpha - \gamma} \right] \tau + \left[\frac{3k_0 - k_3}{\alpha - \gamma} \gamma - k_3 \right] z + 3\omega_0 t \right\} d\tau \right] \right\}. \end{aligned}$$

This can be reduced to

$$\begin{aligned} \mathcal{E}_g(z,t) = \frac{-k_3 \chi \mathcal{E}_0^3 z}{8\epsilon_r} \frac{\sin\left\{ \frac{3k_0 - k_3}{2} z \right\}}{\frac{3k_0 - k_3}{2} z} \cos\left\{ \frac{3k_0 + k_3}{2} z - 3\omega_0 \gamma z \right\} \\ \cdot \cos\{3\omega_0 t - 3\omega_0 \gamma z\} e^{-3(t-\gamma z)^2/T^2} \lim_{\alpha \rightarrow \gamma} \frac{\alpha - \gamma}{\alpha - \gamma}. \end{aligned}$$

The limit in the above expression merely establishes the sign of $\mathcal{E}_g(z,t)$. A $\sin(x)/x$ relationship with respect to the phase velocity match is obtained while an oscillating relationship is obtained between the average of the phase velocities and the group velocity which is then weighted by z .

The various spectra are found to be

$$E_g(z, \omega) = \frac{-k_3 \chi \epsilon_0^3 z \sqrt{\pi} T}{16\sqrt{3} \epsilon_r} \frac{\sin\left\{\frac{3k_0 - k_3}{2} z\right\}}{\frac{3k_0 - k_3}{2} z} \\ \cdot \cos\left\{\frac{3k_0 - k_3}{2} z - 3\omega_0 \gamma z\right\} e^{-j\omega \gamma z} \left\{ e^{-T^2(\omega - 3\omega_0)^2/12} \right. \\ \left. + e^{-T^2(\omega + 3\omega_0)^2/12} \right\} \lim_{\alpha \rightarrow \gamma} \frac{\alpha - \gamma}{\alpha - \gamma}$$

and

$$S_g(z, \omega) = \frac{k_3^2 \chi^2 \epsilon_0^6 z^2 T^2 \pi}{768 \epsilon_r^2} \sqrt{\frac{\epsilon}{\mu}} \frac{\sin^2\left\{\frac{3k_0 - k_3}{2} z\right\}}{\left(\frac{3k_0 - k_3}{2} z\right)^2} \\ \cdot \cos^2\left\{\frac{3k_0 - k_3}{2} z - 3\omega_0 \gamma z\right\} \left\{ e^{-T^2(\omega - 3\omega_0)^2/6} + e^{-T^2(\omega + 3\omega_0)^2/6} \right\}$$

A concluding comment and calculation give some insight as to the effects of the phase velocities and group velocities on THG. If the peak intensity of the THG pulse is calculated and then normalized to the peak intensity obtained when the independent variable is zero, then a plot of this ratio can be made and a qualitative feeling for the process developed. In the case of matched phase velocity, the ratio is given by the equation

$$\frac{\hat{I}}{\hat{I}_m} = \left(\frac{2T}{(\alpha - \gamma)z} \right)^2 \left(\int_0^{\frac{2T}{(\alpha - \gamma)z}} e^{-3x^2} dx \right)^2,$$

where $\hat{I} \equiv$ peak intensity as a function of the independent variable,

$\hat{I}_m \equiv$ peak intensity under the condition of matched group velocity

($\alpha=\gamma$) and

$T \equiv$ a parameter directly related to pulsewidth.

In the case of matched group velocities, the expression is

$$\frac{\hat{I}}{\hat{I}_m} = \frac{\sin^2\left\{\frac{3k_0 - k_3}{2}z\right\}}{\left\{\frac{3k_0 - k_3}{2}z\right\}^2}$$

when the group velocities equal the average phase velocity. These functions are plotted and presented in the accompanying Figures I and II.

C. Linearly Chirped Rectangular Pulse. This portion of Section III presents the types of calculations made and the results obtained from these calculations for the third harmonic generated by a linearly chirped rectangular pulse. The calculations will be only briefly described because of the complexity of the expressions and the difficulty and bulkiness involved with executing the required operations.

Assume a fundamental pulse

$$\mathcal{E}_f(o,t) = \begin{cases} \mathcal{E}_o \cos\left\{\omega_o t + \frac{1}{2} \mu t^2\right\}, & -\frac{T}{2} \leq t \leq \frac{T}{2} \\ 0, & \text{otherwise} \end{cases}$$

The spectrum of this pulse is given by

$$\begin{aligned} E_f(o,\omega) &= \mathcal{F}\{\mathcal{E}_f(o,t)\} \\ &= \int_{-T/2}^{T/2} \mathcal{E}_o \cos\left\{\omega_o t + \frac{1}{2} \mu t^2\right\} e^{-j\omega t} dt \end{aligned}$$

$$\begin{aligned}
&= \frac{1}{2} e^{-j \frac{(\omega - \omega_0)^2}{2\mu}} \xi_0 \int_{-T/2}^{T/2} e^{j \frac{\mu}{2} \left[t - \frac{\omega - \omega_0}{\mu} \right]^2} \\
&\quad \cdot dt + \frac{1}{2} e^{-j \frac{(\omega + \omega_0)^2}{2\mu}} \xi_0 \int_{-T/2}^{T/2} e^{j \frac{\mu}{2} \left[t - \frac{\omega + \omega_0}{\mu} \right]^2} dt .
\end{aligned}$$

By the use of the Fresnel Integrals,

$$C(X) = \int_0^X \cos\left(\frac{\pi}{2} x^2\right) dx \text{ and}$$

$$S(X) = \int_0^X \sin\left(\frac{\pi}{2} x^2\right) dx ,$$

it is found that

$$\begin{aligned}
E_f(\omega) = & \frac{1}{2} \sqrt{\frac{\pi}{\mu}} e^{-j \frac{(\omega - \omega_0)^2}{2\mu}} \xi_0 \left[C\left(\sqrt{\frac{\mu}{\pi}} \left[\frac{T}{2} + \frac{\omega - \omega_0}{\mu} \right]\right) \right. \\
& \left. + C\left(\sqrt{\frac{\mu}{\pi}} \left[\frac{T}{2} - \frac{\omega - \omega_0}{\mu} \right]\right) + j \left\{ S\left(\sqrt{\frac{\mu}{\pi}} \left[\frac{T}{2} + \frac{\omega - \omega_0}{\mu} \right]\right) \right. \right. \\
& \left. \left. + S\left(\sqrt{\frac{\mu}{\pi}} \left[\frac{T}{2} - \frac{\omega - \omega_0}{\mu} \right]\right) \right\} \right]
\end{aligned}$$

$$\begin{aligned}
& + S\left(\sqrt{\frac{L}{\pi}} \left[\frac{T}{2} - \frac{\omega - \omega_0}{\mu}\right]\right) \left\} + \frac{1}{2} \sqrt{\frac{L}{\pi}} e^{-j \frac{(\omega + \omega_0)^2}{2\mu}} \left[C\left(\sqrt{\frac{L}{\pi}} \left[\frac{T}{2} + \frac{\omega + \omega_0}{\mu}\right]\right) \right. \right. \\
& + C\left(\sqrt{\frac{L}{\pi}} \left[\frac{T}{2} - \frac{\omega + \omega_0}{\mu}\right]\right) \\
& \left. \left. - j \left\{ S\left(\sqrt{\frac{L}{\pi}} \left[\frac{T}{2} + \frac{\omega + \omega_0}{\mu}\right]\right) + S\left(\sqrt{\frac{L}{\pi}} \left[\frac{T}{2} - \frac{\omega + \omega_0}{\mu}\right]\right) \right\} \right].
\end{aligned}$$

Following the previous examples, it is seen that the spectral power density, $S_f(o, \omega)$, is equal to

$$\begin{aligned}
S_f(o, \omega) = \frac{\pi}{4L} \sqrt{\frac{L}{\pi}} \left\{ \left[C\left(\sqrt{\frac{L}{\pi}} \left[\frac{T}{2} + \frac{\omega - \omega_0}{\mu}\right]\right) \right. \right. \\
\left. \left. + C\left(\sqrt{\frac{L}{\pi}} \left[\frac{T}{2} - \frac{\omega - \omega_0}{\mu}\right]\right) \right]^2 + \left[S\left(\sqrt{\frac{L}{\pi}} \left[\frac{T}{2} + \frac{\omega - \omega_0}{\mu}\right]\right) \right. \right.
\end{aligned}$$

$$\begin{aligned}
& + S\left(\sqrt{\frac{\mu}{\pi}}\left[\frac{T}{2} - \frac{\omega - \omega_0}{\mu}\right]\right)^2 + \left[C\left(\sqrt{\frac{\mu}{\pi}}\left[\frac{T}{2} + \frac{\omega + \omega_0}{\mu}\right]\right) + C\left(\sqrt{\frac{\mu}{\pi}}\left[\frac{T}{2} - \frac{\omega + \omega_0}{\mu}\right]\right) \right]^2 \\
& + \left[S\left(\sqrt{\frac{\mu}{\pi}}\left[\frac{T}{2} + \frac{\omega + \omega_0}{\mu}\right]\right) + S\left(\sqrt{\frac{\mu}{\pi}}\left[\frac{T}{2} - \frac{\omega + \omega_0}{\mu}\right]\right) \right]^2 \left. \right\} \epsilon_0^2.
\end{aligned}$$

The equations presented in the preceding paragraph are fairly large and do not leave the reader with a reasonable feel for the shape of the function. The equations developed for the IHG as a result of this fundamental excitation become much more cumbersome and unwieldy. For these reasons, information on the spectra will not be presented.

The third harmonic, $\epsilon_g(z, t)$, that is generated by the chirped fundamental will be calculated for the case of matched phase velocities between the fundamental and the third harmonic. From Section II, part E,

$$\epsilon_g(z, t) = \frac{-k_3 x}{2|\alpha - \gamma| \epsilon_r} \int_{yz}^{\alpha z} \sin \left\{ \left[3\omega_0 - \frac{3k_0 - k_3}{\alpha - \gamma} \right] \tau + \left[\frac{3k_0 - k_3}{\alpha - \gamma} \gamma - k_3 \right] z \right\}$$

$$\bullet \epsilon_f(0, t - \tau)^3 d\tau.$$

Under the condition of matched phase velocities, $3k_0 = k_3$, then

$$\xi_g(z,t) = \frac{-k_3 x}{2|\alpha-\gamma|} \int_{\gamma z}^{\alpha z} \sin\{3\omega_0 \tau - k_3 z\} \xi_f(o,t-\tau)^3 d\tau .$$

Because $\xi_f(o,t)$ is expressed in a piece-wise fashion, $\xi_g(z,t)$ is also obtained in a piece-wise fashion. The limits of the integral in $\xi_g(z,t)$ are affected and the expression of $\xi_g(z,t)$ is rather cumbersome. An explanation of the effect that $\xi_f(o,t)$ has on $\xi_g(z,t)$ with respect to the integration, and an example establishing the integral limits for the various regions of $\xi_g(z,t)$ is presented in Appendix F.

There is a general form for $\xi_g(z,t)$, under various conditions of α , γ , z , T and t . The following equation presents this form while the accompanying table, Table I, presents the conditions, regions, and integral limits for the four cases considered.

In general,

$$\xi_g(z,t) = \frac{-k_3 x \xi_o^3}{8|\alpha-\gamma| \epsilon_r} \int_{\tau_1}^{\tau_2} \sin\{3\omega_0 \tau - k_3 z\} \cos\{3\omega_0(t-\tau) + \frac{3}{2} \bar{\nu} (t-\tau)^2\} d\tau .$$

$\xi_g(z,t)$ is necessarily zero except in regions I, II and III as given in Table I. The actual solution for the first two cases will be given. This is considered to be sufficient, because these consider the situations in which the pulse is smaller than $|\alpha-\gamma|z$ and also larger than $|\alpha-\gamma|z$. Interchanging the group velocities does not cause a drastic change in the character of the solution. The solutions are:

$$1) \alpha > \gamma, T \geq |\alpha - \gamma| z$$

$$\begin{aligned} \varepsilon_g(z, t) = & \left. \begin{aligned} & \frac{k_3 \chi \varepsilon_0^3}{16 |\alpha - \gamma| \varepsilon_r} \sqrt{\frac{\pi}{3\mu}} \left\{ \left[-C \left[\sqrt{\frac{3\mu}{\pi}} \frac{T}{2} \right] - C \left[\sqrt{\frac{3\mu}{\pi}} (t - \gamma z) \right] \right] \sin (3\omega_0 t - k_3 z) \right. \\ & \left. + \left[-S \left[\sqrt{\frac{3\mu}{\pi}} \frac{T}{2} \right] - S \left[\sqrt{\frac{3\mu}{\pi}} (t - \gamma z) \right] \right] \cos (3\omega_0 t - k_3 z) \right\}, \gamma z - \frac{T}{2} \leq t \leq \alpha z - \frac{T}{2} \\ & \frac{k_3 \chi \varepsilon_0^3}{16 |\alpha - \gamma| \varepsilon_r} \sqrt{\frac{\pi}{3\mu}} \left\{ \left[C \left[\sqrt{\frac{3\mu}{\pi}} (t - \alpha z) \right] - C \left[\sqrt{\frac{3\mu}{\pi}} (t - \gamma z) \right] \right] \sin (3\omega_0 t - \right. \\ & \left. k_3 z) + \left[S \left[\sqrt{\frac{3\mu}{\pi}} (t - \alpha z) \right] - S \left[\sqrt{\frac{3\mu}{\pi}} (t - \gamma z) \right] \right] \cos (3\omega_0 t - k_3 z) \right\}, \\ & \alpha z - \frac{T}{2} \leq t \leq \gamma z + \frac{T}{2} \\ & \frac{k_3 \chi \varepsilon_0^3}{16 |\alpha - \gamma| \varepsilon_r} \sqrt{\frac{\pi}{3\mu}} \left\{ \left[C \left[\sqrt{\frac{3\mu}{\pi}} (t - \alpha z) \right] - C \left[\sqrt{\frac{3\mu}{\pi}} \frac{T}{2} \right] \right] \sin (3\omega_0 t - k_3 z) \right. \\ & \left. + \left[S \left[\sqrt{\frac{3\mu}{\pi}} (t - \alpha z) \right] - S \left[\sqrt{\frac{3\mu}{\pi}} \frac{T}{2} \right] \right] \cos (3\omega_0 t - k_3 z) \right\}, \gamma z + \frac{T}{2} \leq t \leq \alpha z + \frac{T}{2} \end{aligned} \right. \end{aligned}$$

and

$$2) \alpha > \gamma, T \leq |\alpha - \gamma| z$$

$$\begin{aligned}
 \varepsilon_g(z,t) = & \left\{ \frac{k_3 x \varepsilon_0^3}{16|\alpha-\gamma|\varepsilon_r} \sqrt{\frac{\pi}{3\mu}} \left[-C \left[\sqrt{\frac{3\mu}{\pi}} \frac{T}{2} \right] - C \left[\sqrt{\frac{3\mu}{\pi}} (t-\gamma z) \right] \right] \sin(3\omega_0 t - k_3 z) \right. \\
 & \left. + \left[-S \left[\sqrt{\frac{3\mu}{\pi}} \frac{T}{2} \right] - S \left[\sqrt{\frac{3\mu}{\pi}} (t-\gamma z) \right] \right] \cos(3\omega_0 t - k_3 z) \right\}, \quad \gamma z - \frac{T}{2} \leq t \leq \gamma z + \frac{T}{2} \\
 & \frac{-k_3 x \varepsilon_0^3}{16|\alpha-\gamma|\varepsilon_r} \sqrt{\frac{\pi}{3\mu}} \left\{ -C \left[\sqrt{\frac{3\mu}{\pi}} \frac{T}{2} \right] \sin(3\omega_0 t - k_3 z) \right. \\
 & \left. - S \left[\sqrt{\frac{3\mu}{\pi}} \frac{T}{2} \right] \cos(3\omega_0 t - k_3 z) \right\}, \quad \gamma z + \frac{T}{2} \leq t \leq \alpha z - \frac{T}{2} \\
 & \frac{k_3 x \varepsilon_0^3}{16|\alpha-\gamma|\varepsilon_r} \sqrt{\frac{\pi}{3\mu}} \left\{ \left[C \left[\sqrt{\frac{3\mu}{\pi}} (t-\alpha z) \right] - C \left[\sqrt{\frac{3\mu}{\pi}} \frac{T}{2} \right] \right] \sin(3\omega_0 t - k_3 z) + \right. \\
 & \left. \left[S \left[\sqrt{\frac{3\mu}{\pi}} (t-\alpha z) \right] - S \left[\sqrt{\frac{3\mu}{\pi}} (t-\gamma z) \right] \right] \cos(3\omega_0 t - k_3 z) \right\}, \quad \alpha z - \frac{T}{2} \leq t \leq \alpha z + \frac{T}{2}
 \end{aligned}$$

For all t other than specified above, $\varepsilon_g(z,t)$ is necessarily zero.

The third harmonic, $\xi_g(z, t)$, that is generated under matched group velocity conditions will now be calculated. This is obtained by evaluating $\xi_g(z, t)$ in the limit as $\alpha \rightarrow \gamma$. Thus,

$$\xi_g(z, t) = \lim_{\alpha \rightarrow \gamma} \frac{-k_3 X}{2|\alpha - \gamma| \epsilon_r} \int_{\gamma z}^{\alpha z} \sin\left[\left(3\omega_0 - \frac{3k_0 - k_3}{\alpha - \gamma}\right)\tau + \left(\frac{3k_0 - k_3}{\alpha - \gamma} \gamma - k_3\right)z\right] \\ \cdot \xi_p(0, t - \tau)^3 d\tau.$$

For the chirped pulse case, this can be seen to reduce to

$$\xi_g(z, t) = \frac{-k_3 X \xi_0^3}{8\epsilon_r} \cos\left[3\omega_0(t - \gamma z) + \frac{3}{2} \bar{\mu} (t - \gamma z)^2\right] \lim_{\alpha \rightarrow \gamma} \frac{1}{\alpha - \gamma} \\ \int_{\gamma z}^{\alpha z} \sin\left[\left(3\omega_0 - \frac{3k_0 - k_3}{\alpha - \gamma}\right)\tau + \left(\frac{3k_0 - k_3}{\alpha - \gamma} \gamma - k_3\right)z\right] d\tau.$$

Upon evaluation, $\xi_g(z, t)$ becomes

$$\xi_g(z, t) = \begin{cases} \frac{-k_3 X \xi_0^3}{8\epsilon_r} \frac{\sin\left[\frac{3k_0 - k_3}{2} z\right]}{\frac{3k_0 - k_3}{2} z} \\ \cdot \lim_{\alpha \rightarrow \gamma} \left(\frac{\alpha - \gamma}{\alpha - \gamma}\right) \sin\left[3\omega_0 \gamma z - \frac{3k_0 + k_3}{2} z\right] \cos\left[3\omega_0(t - \gamma z) + \frac{3}{2} \bar{\mu} (t - \gamma z)^2\right] \\ \text{for } \gamma z - \frac{T}{2} \leq t \leq \gamma z + \frac{T}{2} \\ 0, \text{ otherwise} \end{cases}$$

The effects of several parameters on peak intensity will be shown now. As before, the information will be shown graphically and in a normalized manner. The development used is quite similar to that of the gaussian case, although this time it is not possible to present as much information as would be desired due to the number of variables involved and the complexity of the function. For the phase velocity matched case, the particular case of $\alpha > \gamma$, $T \leq |\alpha - \gamma|z$ will be considered. In addition, only Region II of the solution will be evaluated under the assumption that it dominates the peak intensity. After normalizing, the peak intensity is given by:

$$\left(C(X)^2 + S(X)^2 \right) / X^2, \text{ where } X = \sqrt{\frac{3\mu}{\pi}} \frac{T}{2},$$

thus containing both $\bar{\mu}$ and T . A plot of this function is shown in Figure III, where the pulse width, T , is held constant.

Another case, when $\bar{\mu}$ is held constant and T is the variable parameter, is also presented. The peak intensity is proportional to $C(X)^2 + S(X)^2$ and a plot of this function is shown in Figure IV.

Finally, the case of matched group velocity is shown in Figure V. This is simply the $\sin^2(x)/x^2$ function that appears to be the characteristic function associated with mismatched phase velocities.

SECTION IV

CONCLUSIONS

A theory for THG has been developed from Maxwell's Equations for a uniform plane wave in an isotropic medium. This theory is fairly general with the only outstanding assumptions being that the generation process is assumed to be relatively inefficient and that the spectral bandwidth of the pulse about the center frequency is sufficiently small that the propagation coefficients of the fundamental and third harmonic can be represented by a truncated Taylor's expansion. For the present state-of-the-art in optics, these assumptions are quite practical.

A series of three examples were then examined. The first of these, an eternal sinusoid, was examined to determine whether the results of this theory fit previously obtained results. A comparison was made with the results of a recognized group of workers in the field and a favorable outcome was obtained.

The second example was a sine wave that was amplitude modulated with a gaussian wave form. This fundamental pulse was then examined for THG under conditions of matched group velocities and of matched phase velocities. For matched group velocities, the variation of peak intensity followed a $\sin^2(x)/x^2$ function while for matched phase velocities, the peak intensity followed a $\text{erf}(x)^2/x^2$ function.

The third example was a linearly chirped rectangular pulse. This example was quite difficult from a mathematical point of view and only the most interesting results and calculations were presented. The THG pulse was examined under the conditions of matched group

velocities and of matched phase velocities. For the condition of matched group velocities, the peak intensity followed a $\sin^2(x)/x^2$ function. For the condition of matched phase velocities, the THG pulse was examined as a function of frequency modulation and as a function of pulse width. In both cases oscillatory behavior was observed. When the frequency modulated case was examined, the peak intensity followed a $(C(x)^2 + S(x)^2)/x^2$ function. When the pulse width modulated case was examined, the peak intensity followed a $C(x)^2 + S(x)^2$ function. This oscillatory behavior is quite interesting and could possibly be seen in an experiment if, for example, a train of pico-second laser pulses were to change pulse width as the pulse train progressed in time.

In conclusion, this work has developed a seemingly valid theory of THG and has, within limits, begun to identify THG results as a function of material and excitation parameters. It is sincerely hoped that this effort will benefit others in this field of endeavor.

<u>Case</u>	<u>Region</u>	<u>Interval</u>	<u>T_1</u>	<u>T_2</u>
$\alpha > \gamma$	I	$\gamma z - \frac{T}{2} \leq t \leq \alpha z - \frac{T}{2}$	γz	$t + \frac{T}{2}$
$T \geq \alpha - \gamma z$	II	$\alpha z - \frac{T}{2} \leq t \leq \gamma z + \frac{T}{2}$	γz	αz
	III	$\gamma z + \frac{T}{2} \leq t \leq \alpha z + \frac{T}{2}$	$t - \frac{T}{2}$	αz
	I	$\gamma z - \frac{T}{2} \leq t \leq \gamma z + \frac{T}{2}$	γz	$t + \frac{T}{2}$
$T \leq \alpha - \gamma z$	II	$\gamma z + \frac{T}{2} \leq t \leq \alpha z - \frac{T}{2}$	$t - \frac{T}{2}$	$t + \frac{T}{2}$
	III	$\alpha z - \frac{T}{2} \leq t \leq \alpha z + \frac{T}{2}$	$t - \frac{T}{2}$	αz
	I	$\alpha z - \frac{T}{2} \leq t \leq \gamma z - \frac{T}{2}$	$t + \frac{T}{2}$	αz
$T \geq \alpha - \gamma z$	II	$\gamma z - \frac{T}{2} \leq t \leq \alpha z + \frac{T}{2}$	γz	αz
	III	$\alpha z + \frac{T}{2} \leq t \leq \gamma z + \frac{T}{2}$	γz	$t - \frac{T}{2}$
	I	$\alpha z - \frac{T}{2} \leq t \leq \alpha z + \frac{T}{2}$	$t + \frac{T}{2}$	αz
$T \leq \alpha - \gamma z$	II	$\alpha z + \frac{T}{2} \leq t \leq \gamma z - \frac{T}{2}$	$t + \frac{T}{2}$	$t - \frac{T}{2}$
	III	$\gamma z - \frac{T}{2} \leq t \leq \gamma z + \frac{T}{2}$	γz	$t - \frac{T}{2}$

TABLE I. Integral Limits for THG of Linearly Chirped Pulse.

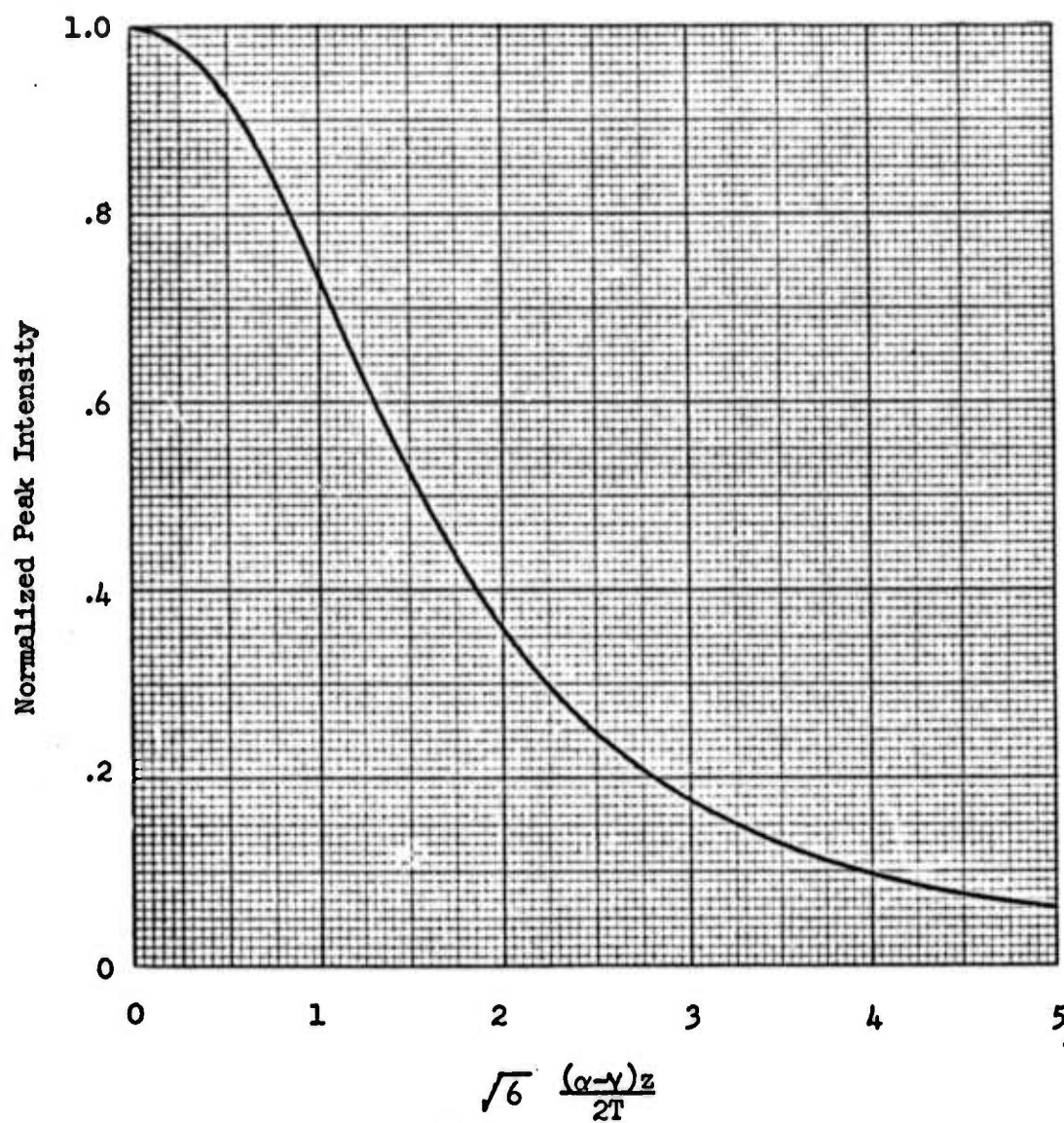


FIGURE I. Peak Intensity of THG of a Gaussian Pulse as a Function of Group Velocity Mismatch.

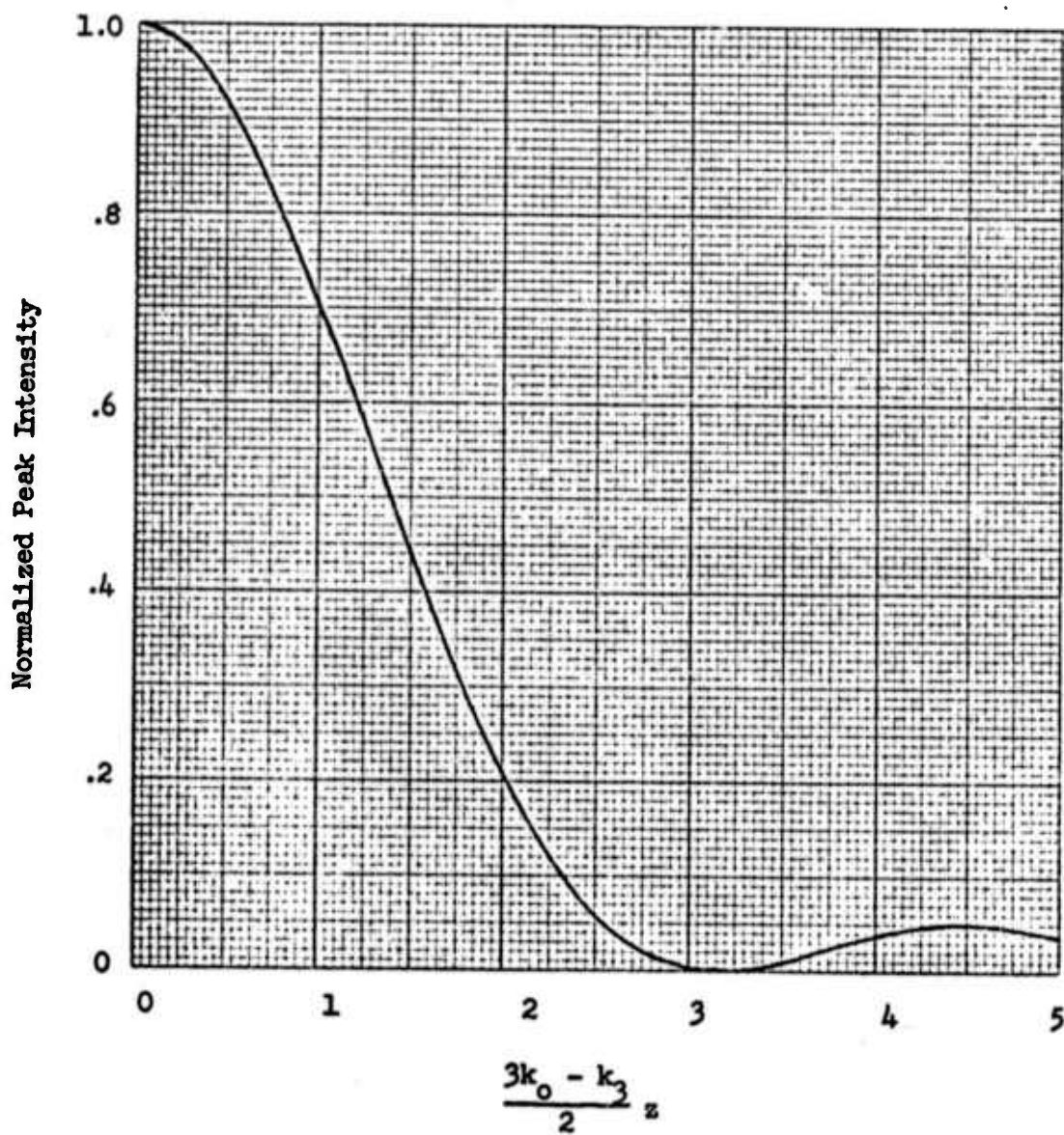


FIGURE II. Peak Intensity of THG of a Gaussian Pulse as a Function of Phase Velocity Mismatch.

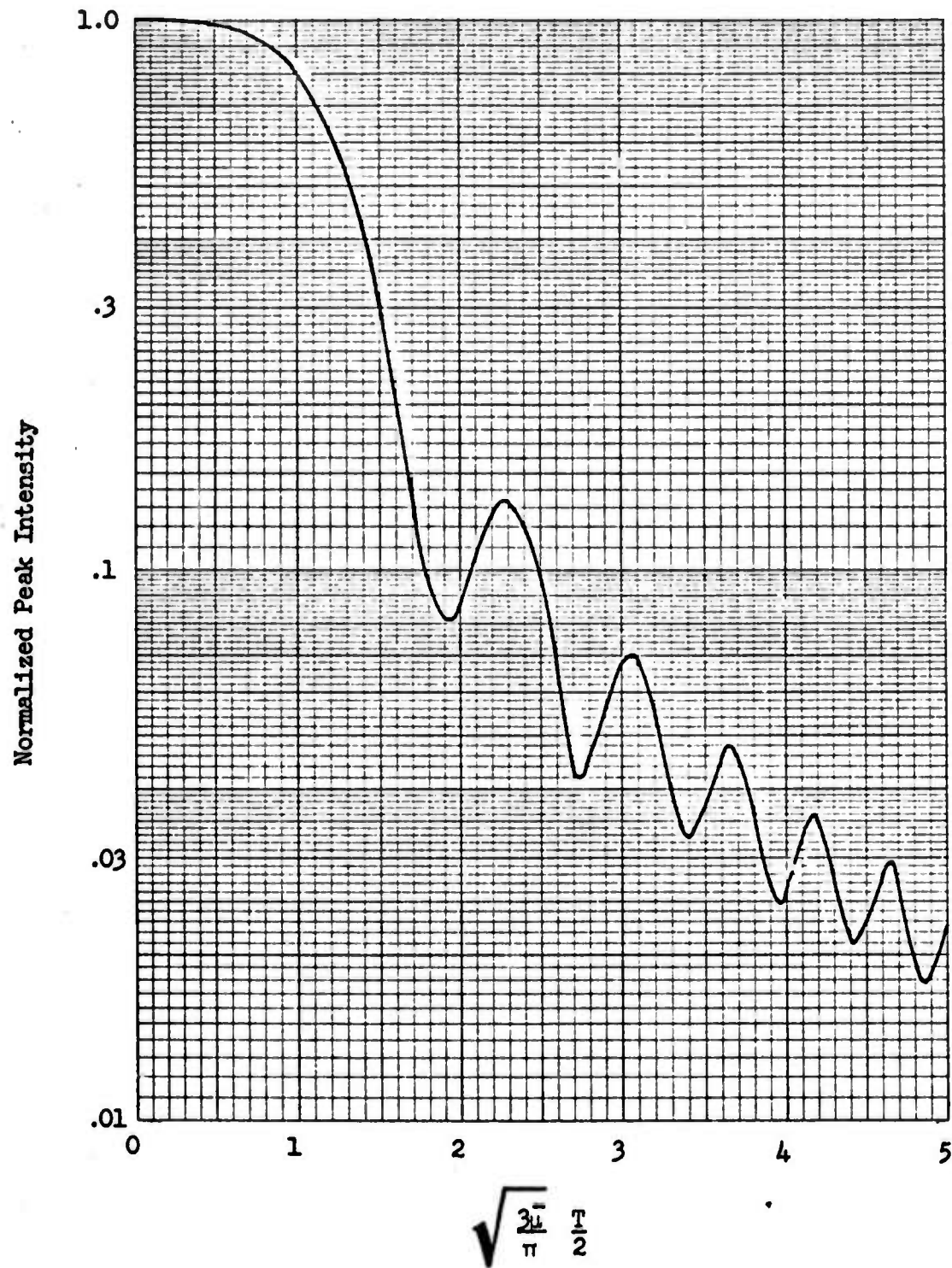


FIGURE III. Peak Intensity of THG of a Linearly Chirped Pulse as a Function of Variable Chirp.

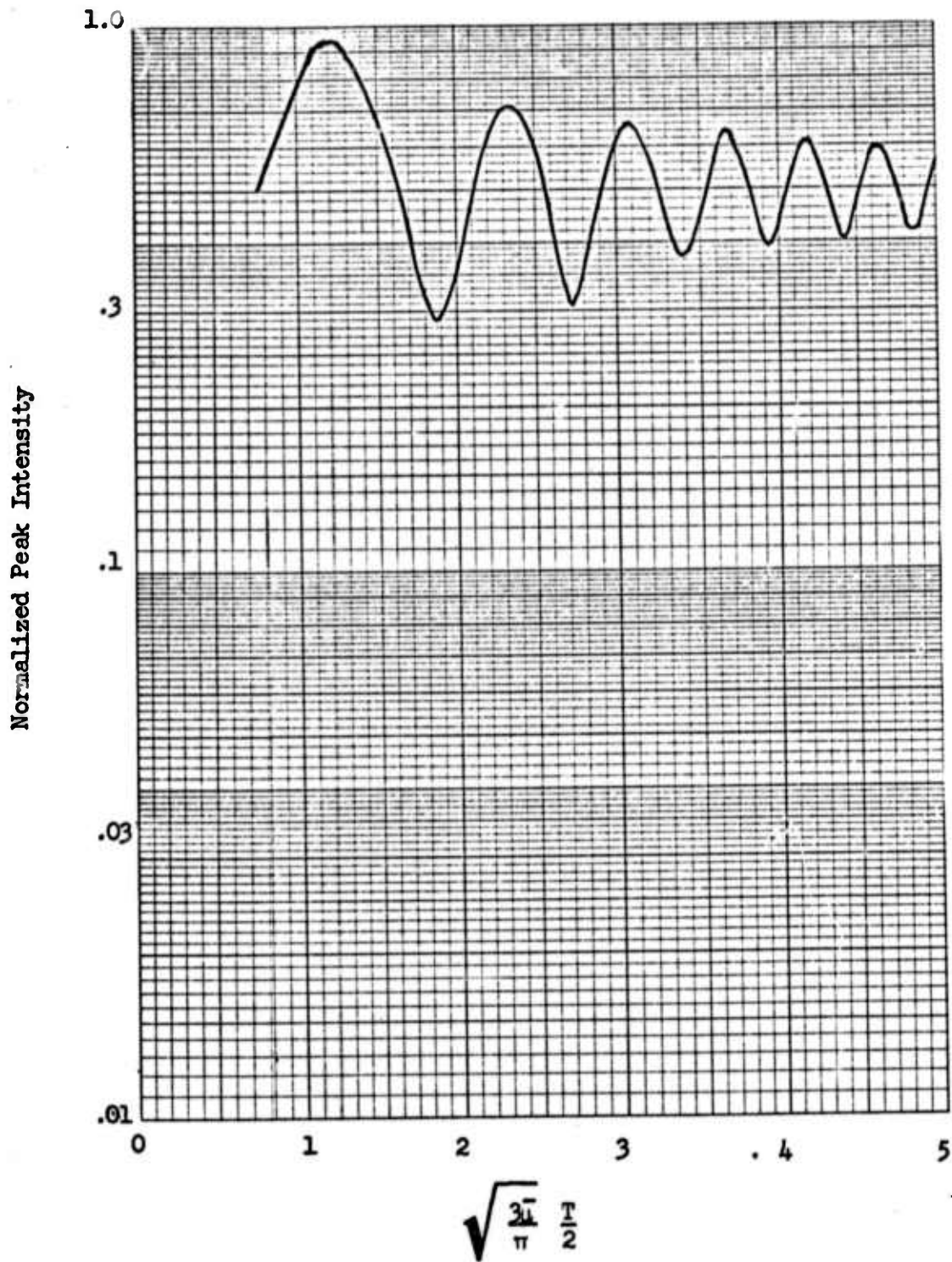


FIGURE IV. Peak Intensity of THG of a Linearly Chirped Pulse as a Function of Pulse Duration.

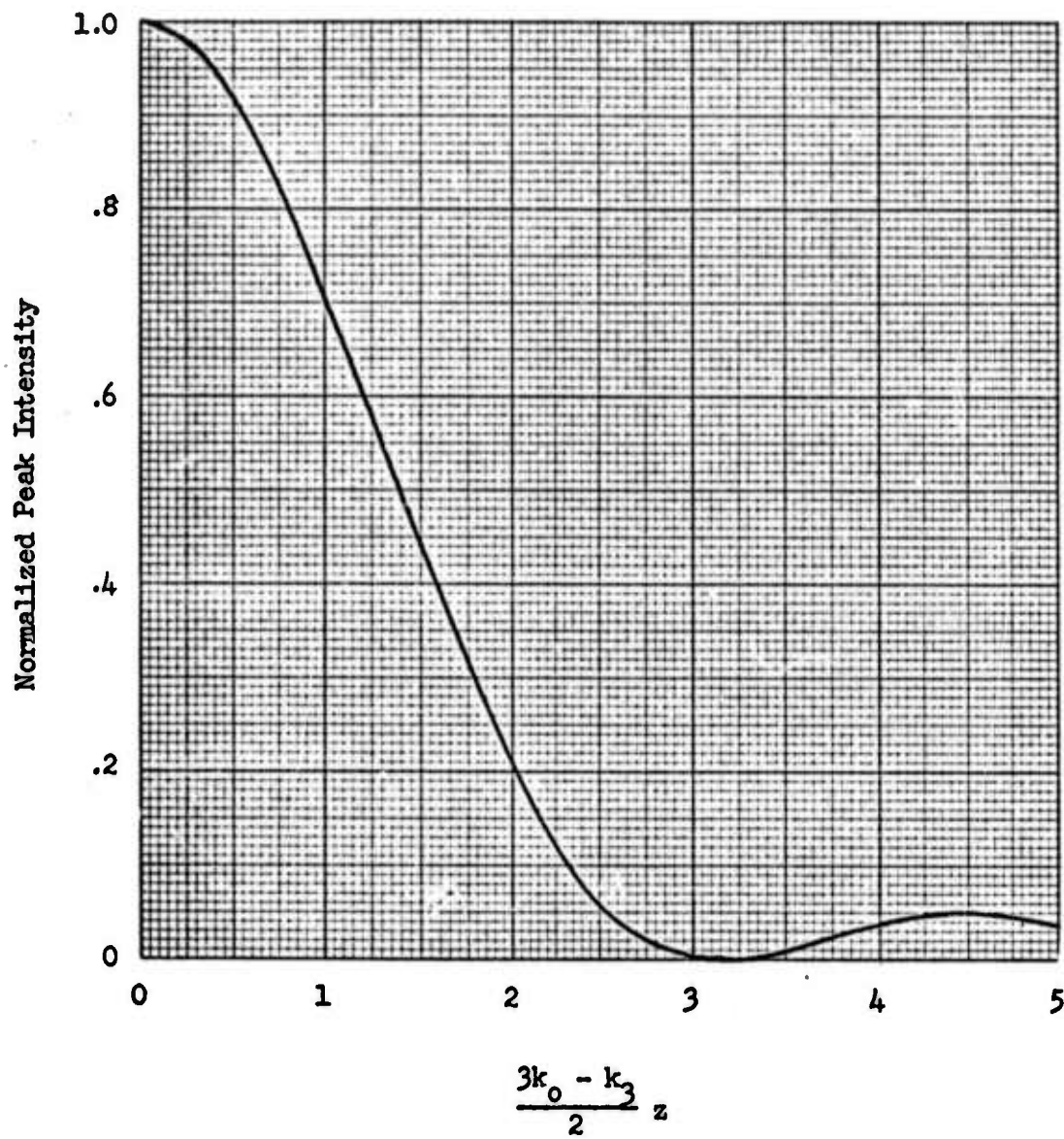


FIGURE V. Peak Intensity of THG of a Linearly Chirped Pulse as a Function of Phase Velocity Mismatch.

APPENDIX A

EVALUATION OF THE INVERSE FOURIER TRANSFORM OF:

$$e^{-j[3k_0 - k_3 + (\alpha - \gamma)(\omega - 3\omega_0)]z/2} \frac{\sin\{[3k_0 - k_3 + (\alpha - \gamma)(\omega - 3\omega_0)]z/2\}}{[3k_0 - k_3 + (\alpha - \gamma)(\omega - 3\omega_0)]z/2}$$

Postulate a function $f(t)$ and knowledge of its Fourier transform, $F(\omega)$. Denote the transform pair relationship by the symbology

$$f(t) \leftrightarrow F(\omega)$$

$$\text{Let } F(\omega) = e^{-jA(\omega - n)} \frac{\sin[A(\omega - n)]}{A(\omega - n)}$$

From the expression in the title, it is seen that

$$A \equiv (\alpha - \gamma)z/2$$

$$n \equiv 3\omega_0 - \frac{3k_0 - k_3}{\alpha - \gamma}$$

$$\text{Thus } f(t) \leftrightarrow e^{-jA(\omega - n)} \frac{\sin[A(\omega - n)]}{A(\omega - n)}$$

The scaling relationship,

$$f(at) \leftrightarrow \frac{1}{|a|} F\left(\frac{\omega}{a}\right),$$

is applied yielding

$$f(At) = \frac{1}{|A|} e^{j(\omega - n)} \frac{\sin[\omega - n]}{\omega - n}$$

The frequency shifting property,

$$f(t) e^{j\omega_0 t} \leftrightarrow F(\omega - \omega_0),$$

is applied yielding

$$f(At) e^{-jnAt} \leftrightarrow \frac{1}{|A|} e^{-j\omega} \frac{\sin \omega}{\omega}$$

Finally, the time shifting property,

$$f(t - t_0) \leftrightarrow F(\omega) e^{-j\omega t_0}$$

is applied and the scale factor $|A|$ is transposed. Thus,

$$|A| f(A[t + 1]) e^{-j\Omega A(t + 1)} \leftrightarrow \frac{\sin \omega}{\omega}$$

If $d(t) \leftrightarrow \frac{\sin \omega t}{\omega}$, then

$$d(t) = \begin{cases} \frac{1}{2}, & -1 \leq t \leq 1 \\ 0, & \text{otherwise} \end{cases} .$$

There now exists the equivalence

$$d(t) = |A| f(A[t+1]) e^{-j\Omega A(t+1)} .$$

Allow a change of variable,

$$t' = A[t+1] .$$

$$\text{Thus } f(t') = \frac{1}{|A|} d\left(\frac{t'}{A} - 1\right) e^{j\omega t'} .$$

Deleting the primes and using the definition for $d(t)$,

$$f(t) = \begin{cases} \frac{1}{2|A|} e^{j\Omega t}, & 0 \leq t \leq 2A \\ 0, & \text{otherwise} \end{cases} .$$

If $A < 0$, the inequality is reversed. Using the definitions of A and Ω originally stated,

$$f(t) = \begin{cases} \frac{1}{|a-\gamma|z} e^{j\left[3\omega_0 - \frac{3k_0 - k_3}{a-\gamma}\right] t}, & 0 \leq t \leq (a-\gamma)z \\ 0, & \text{otherwise} \end{cases} .$$

APPENDIX B

EVALUATING $\xi_g(z, t)$ WHEN $\alpha \rightarrow \gamma$

$$\text{Let } \xi_f(0, t) = \xi_0 \sin \omega_0 t$$

$$\xi_g(z, t) = \frac{-k_3 \chi}{2|\alpha - \gamma| \epsilon_r} \int_{\gamma t}^{\alpha z} \sin\left[\left(3\omega_0 - \frac{3k_0 - k_3}{\alpha - \gamma}\right)\tau + \left[\frac{3k_0 - k_3}{\alpha - \gamma} \gamma - k_3\right]z\right] \\ \cdot \xi_0^3 \sin^3\{\omega_0(t - \tau)\} d\tau .$$

Considering only the third harmonic, $\xi_g(z, t)$ becomes

$$\xi_g(z, t) = \frac{k_3 \chi \xi_0^3}{8|\alpha - \gamma| \epsilon_r} \int_{\gamma z}^{\alpha z} \sin\left[\left(3\omega_0 - \frac{3k_0 - k_3}{\alpha - \gamma}\right)\tau + \left[\frac{3k_0 - k_3}{\alpha - \gamma} \gamma - k_3\right]z\right] \\ \cdot \sin\{3\omega_0(t - \tau)\} d\tau .$$

Using $\sin x \sin y = \frac{1}{2}[\cos(x - y) - \cos(x + y)]$,

$$\xi_g(z, t) = \frac{k_3 \chi \xi_0^3}{16|\alpha - \gamma| \epsilon_r} \left[\int_{\gamma z}^{\alpha z} \cos\left[\left(6\omega_0 - \frac{3k_0 - k_3}{\alpha - \gamma}\right)\tau + \left[\frac{3k_0 - k_3}{\alpha - \gamma} \gamma - k_3\right]z - 3\omega_0 t\right] d\tau \right. \\ \left. - \int_{\gamma z}^{\alpha z} \cos\left[\left(-\frac{3k_0 - k_3}{\alpha - \gamma}\right)\tau + \left[\frac{3k_0 - k_3}{\alpha - \gamma} \gamma - k_3\right]z + 3\omega_0 t\right] d\tau \right] .$$

Performing the indicated integration and simplifying,

$$\begin{aligned} \xi_g(z,t) = \frac{k_3 \chi \xi_0^3}{16|\alpha-\gamma| \epsilon_r} & \left[\frac{1}{6\omega_0 - \frac{3k_0-k_3}{\alpha-\gamma}} \left(\sin[6\omega_0 \alpha z - 3k_0 z - 3\omega_0 t] \right. \right. \\ & \left. \left. - \sin[6\omega_0 \gamma z - k_3 z - 3\omega_0 t] \right) \right. \\ & \left. + \frac{1}{\frac{3k_0-k_3}{\alpha-\gamma}} \left(\sin[3\omega_0 t - 3k_0 z] - \sin[3\omega_0 t - k_3 z] \right) \right]. \end{aligned}$$

Taking the limit as $\alpha \rightarrow \gamma$,

$$\begin{aligned} \xi_g(z,t) = \frac{k_3 \chi \xi_0^3}{16\epsilon_r} & \frac{1}{3k_0-k_3} \left[\sin[6\omega_0 \gamma z - k_3 z - 3\omega_0 t] \right. \\ & \left. - \sin[6\omega_0 \gamma z - 3k_0 z - 3\omega_0 t] + \sin[3\omega_0 t - 3k_0 z] - \sin[3\omega_0 t - k_3 z] \right]. \end{aligned}$$

Using $\sin x - \sin y = 2 \sin \frac{1}{2}(x-y) \cos \frac{1}{2}(x+y)$,

$$\begin{aligned} \xi_g(z,t) = \frac{k_3 \chi \xi_0^3 z}{16\epsilon_r} & \frac{\sin\left[\frac{3k_0-k_3}{2} z\right]}{\frac{3k_0-k_3}{2} z} \left[\cos\left[3\omega_0 t + \frac{3k_0-k_3}{2} z - 6\omega_0 \gamma z\right] \right. \\ & \left. + \cos\left[3\omega_0 t - \frac{3k_0-k_3}{2} z\right] \right]. \end{aligned}$$

Finally, using $\cos x + \cos y = 2 \cos \frac{1}{2}(x+y) \cos \frac{1}{2}(x-y)$,

$$\xi_g(z,t) = \frac{k_3 \chi \xi_0^3 z}{16\epsilon_r} \frac{\sin\left[\frac{3k_0-k_3}{2} z\right]}{\frac{3k_0-k_3}{2} z} \cos\left[3\omega_0 \gamma z - \frac{3k_0-k_3}{2} z\right] \cos[3\omega_0 t - 3\omega_0 \gamma z].$$

APPENDIX C

RELATING k TO PHASE AND GROUP VELOCITIES

The accepted definition of phase velocity is the velocity at which a fixed point in phase travels. For a simple traveling sine wave, $\sin(\omega t - kz)$, the phase velocity is given by

$$v_p = \frac{\omega}{k}$$

$$\text{Thus, } k = \frac{\omega}{v_p} .$$

$$\text{If } 3k_0 = k_3, \text{ then } \frac{3\omega_0}{v_0} = \frac{\omega_3}{v_3}$$

and since $\omega_3 = 3\omega_0$, the conclusion is that $v_{p_0} = v_{p_3}$.

Historically, the phase velocity has been considered to be the speed of light in the medium under consideration. Thus,

$$v_p = c/n$$

where c = speed of light

n = index of refraction.

If $v_{p_0} = v_{p_3}$, then necessarily $n_0 = n_3$.

Packets of electromagnetic energy are usually considered to travel at a velocity called group velocity rather than the phase velocity. Using a spectrum closely grouped about ω_0 , the group velocity is

$$v_g = \frac{\partial \omega}{\partial k} .$$

Thus α and γ as used in the paper are reciprocal group velocities.

APPENDIX D

RELATING $\frac{\partial k}{\partial \omega}$ TO n AND $\frac{\partial n}{\partial \lambda}$

Because most information relating propagation to materials used in optics is given in the form of $n(\lambda)$, it is appropriate to relate the group velocity to $n(\lambda)$.

$$\frac{\partial k}{\partial \omega} = \frac{\partial}{\partial \omega} \left(\frac{2\pi}{\lambda} \right)$$

but $\lambda = \lambda_0/n$, where λ_0 is in vacuo, then

$$\frac{\partial k}{\partial \omega} = \frac{\partial}{\partial \omega} \left(\frac{2\pi n}{\lambda_0} \right)$$

Because

$$\frac{2\pi}{\lambda_0} = \frac{\omega}{c}$$

$$\begin{aligned} \frac{\partial k}{\partial \omega} &= \frac{\partial}{\partial \omega} \left(\frac{\omega n}{c} \right) \\ &= \frac{\omega}{c} \frac{\partial n}{\partial \omega} + \frac{n}{c} \end{aligned}$$

The more common optical term is dispersion which is $\frac{\partial n}{\partial \lambda_0}$. This will be related to $\frac{\partial n}{\partial \omega}$.

$$\frac{\partial n}{\partial \lambda_0} = \frac{\partial n}{\partial \omega} \frac{\partial \omega}{\partial \lambda_0}$$

$$\therefore = \frac{2\pi c}{\lambda_0^2}$$

$$\frac{\partial \omega}{\partial \lambda_0} = - \frac{2\pi c}{\lambda_0^2}$$

$$\therefore \frac{\partial n}{\partial \omega} = - \frac{\lambda_0^2}{2\pi c} \frac{\partial n}{\partial \lambda_0}$$

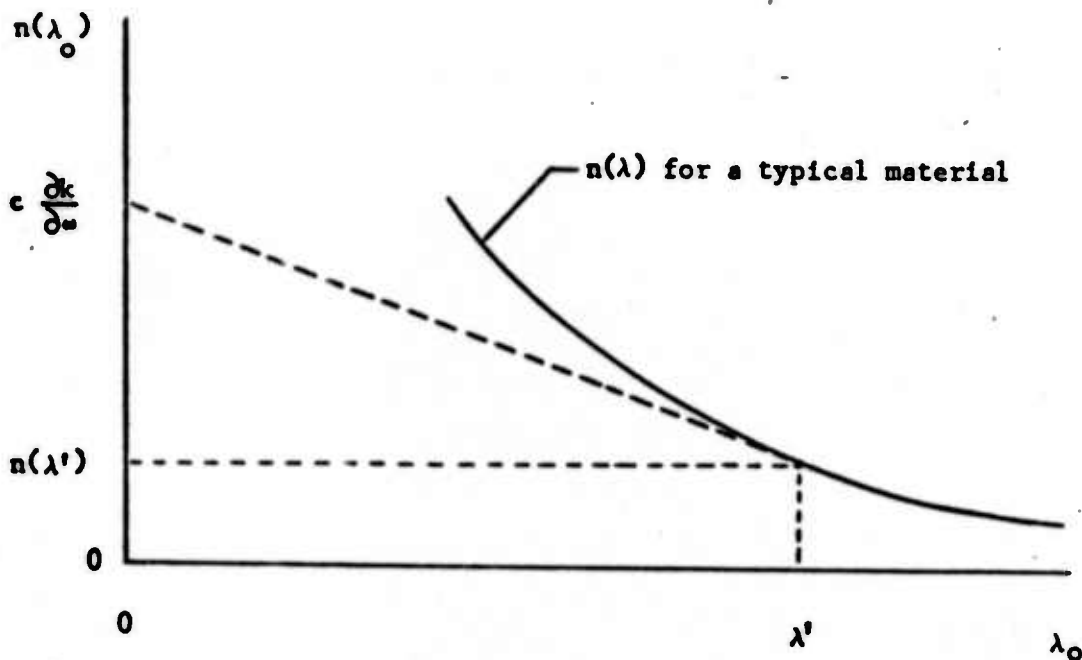
$$\begin{aligned}
 \text{and now, } \frac{\partial k}{\partial \omega} &= \frac{n}{c} - \frac{\omega}{c} \frac{\lambda_0^2}{2\pi c} \frac{\partial n}{\partial \lambda_0} \\
 &= \frac{n}{c} - \frac{2\pi}{\lambda_0} \frac{\lambda_0^2}{2\pi c} \frac{\partial n}{\partial \lambda_0} \\
 &= \frac{1}{c} \left(n - \lambda_0 \frac{\partial n}{\partial \lambda_0} \right) .
 \end{aligned}$$

Because α and γ are each expressed as $\frac{\partial k}{\partial \omega}$,

$$\alpha = \frac{1}{c} \left(n_f - \lambda_f \frac{\partial n}{\partial \lambda_0} \Big|_{\lambda_0 = \lambda_f} \right) \quad \text{and} \quad \gamma = \frac{1}{c} \left(n_g - \lambda_g \frac{\partial n}{\partial \lambda_0} \Big|_{\lambda_0 = \lambda_g} \right) .$$

A graphical interpretation of this is possible using normally available graphical material. The graph below is an example which shows λ_0 ,

$\frac{\partial n}{\partial \lambda_0}$ and $c \frac{\partial k}{\partial \omega}$.



The ordinate intercept gives directly the value $c \frac{\partial k}{\partial \omega}$. From a graphical construction such as this, a match of phase velocity and group velocity can easily be made.

APPENDIX E

A CONVOLUTION RELATIONSHIP

Prove:

$$\frac{1}{2\pi} F(\omega) * G(\omega) \leftrightarrow f(t)g(t)$$

Proof:

$$\begin{aligned} \frac{1}{2\pi} F(\omega) * G(\omega) &\leftrightarrow \mathcal{F}^{-1} \left\{ \frac{1}{2\pi} \int_{-\infty}^{\infty} F(\omega-u)G(u)du \right\} \\ &\leftrightarrow \frac{1}{2\pi} \int_{-\infty}^{\infty} \left\{ \frac{1}{2\pi} \int_{-\infty}^{\infty} F(\omega-u)G(u)du \right\} e^{j\omega t} d\omega \\ &\leftrightarrow \frac{1}{2\pi} \int_{-\infty}^{\infty} G(u) \left\{ \frac{1}{2\pi} \int_{-\infty}^{\infty} F(\omega-u) e^{j\omega t} d\omega \right\} du \end{aligned}$$

Due to the frequency shifting property,

$$\frac{1}{2\pi} \int_{-\infty}^{\infty} F(\omega-u) e^{j\omega t} d\omega = f(t) e^{jut}$$

Thus

$$\begin{aligned} \frac{1}{2\pi} F(\omega) * G(\omega) &\leftrightarrow \frac{1}{2\pi} \int_{-\infty}^{\infty} G(u) f(t) e^{jut} du \\ &\leftrightarrow f(t) \left\{ \frac{1}{2\pi} \int_{-\infty}^{\infty} G(u) e^{jut} du \right\} \end{aligned}$$

$$\therefore \frac{1}{2} F(\omega) * G(\omega) \leftrightarrow f(t)g(t)$$

APPENDIX F

ESTABLISHING THE LIMITS OF INTEGRATION OF $\xi_g(z,t)$ FOR A
RECTANGULARLY PULSED $\xi_f(o,t)$

For expressions of $\xi_f(o,t)$ that are in a piecewise manner rather than a single analytically expressed function, establishing the limits of integration for $\xi_g(z,t)$ is not always simple. It is the purpose of this appendix to illustrate a useful graphical technique for determining these limits.

$$\text{Let } \xi_f(o,t) = \begin{cases} 1, & -\frac{T}{2} \leq t \leq \frac{T}{2} \\ 0, & \text{otherwise} \end{cases}$$

and let

$$\xi_g(z,t) = \int_{\gamma z}^{\alpha z} \xi_f(o,t-\tau)^3 d\tau.$$

Such a function is sufficiently close to the actual function of $\xi_f(o,t)$ and $\xi_g(z,t)$ for this illustration to be valid but simple enough to convey easily the intent of the illustration. Assume $\alpha z > \gamma z$ and that $T \leq \alpha - \gamma z$. For $t < \gamma z - \frac{T}{2}$, Figure A-IV(a) represents the region where the integrand is non-zero and the region of integration do not overlap. The integral is necessarily zero for this condition. As t increases, $\xi_f(o,t-\tau)$ moves to the right and finally enters the region of integration. This occurs at $t = \gamma z - \frac{T}{2}$ and the limits of integration become γz and $t + \frac{T}{2}$ until $t + \frac{T}{2} = \alpha z$. This condition is shown in Figure A-IV(b). As $\xi_f(o,t-\tau)$ continues to move to the right, finally it spans the entire region of integration and the integral limits become γz and αz . This condition begins at $t + \frac{T}{2} = \alpha z$ and lasts until $t - \frac{T}{2} = \gamma z$. This condition is shown in Figure A-IV(c).

After $t - \frac{T}{2} = \gamma z$, $\xi_f(o, t - \tau)$ begins emerging from the region of integration and until $t - \frac{T}{2} = \alpha z$, the integral limits are $t - \frac{T}{2}$ and αz . This is shown in Figure A-IV(d). The regions depicted in Figures A-IV(b), (c), and (d) are called Regions I, II and III respectively and are the regions in which $\xi_g(z, t)$ is not necessarily zero. For $t - \frac{T}{2} > \alpha z$, $\xi_f(o, t - \tau)$ has completely emerged from the region of integration and $\xi_g(z, t)$ is zero. This is shown in Figure A-IV(e).

The results of this exercise and the remaining cases involving αz , γz and T are tabulated in Table A-IV for an integral as shown here:

$$\xi_g(z, t) = \int_{r_1}^{r_2} \xi_f(o, t - \tau)^3 d\tau .$$

$$\xi_g(z,t) = \int_{\tau_1}^{\tau_2} \xi_f(0,t-\tau)^3 d\tau$$

<u>Case</u>	<u>Region</u>	<u>Interval</u>	<u>τ_1</u>	<u>τ_2</u>
$\alpha > \gamma$ $T \geq \alpha - \gamma z$	I	$\gamma z - \frac{T}{2} \leq t \leq \alpha z - \frac{T}{2}$	γz	$t + \frac{T}{2}$
	II	$\alpha z - \frac{T}{2} \leq t \leq \gamma z + \frac{T}{2}$	γz	αz
	III	$\gamma z + \frac{T}{2} \leq t \leq \alpha z + \frac{T}{2}$	$t - \frac{T}{2}$	αz
$\alpha > \gamma$ $T \leq \alpha - \gamma z$	I	$\gamma z - \frac{T}{2} \leq t \leq \gamma z + \frac{T}{2}$	γz	$t + \frac{T}{2}$
	II	$\gamma z + \frac{T}{2} \leq t \leq \alpha z - \frac{T}{2}$	$t - \frac{T}{2}$	$t + \frac{T}{2}$
	III	$\alpha z - \frac{T}{2} \leq t \leq \alpha z + \frac{T}{2}$	$t - \frac{T}{2}$	αz
$\gamma > \alpha$ $T \geq \alpha - \gamma z$	I	$\alpha z - \frac{T}{2} \leq t \leq \gamma z - \frac{T}{2}$	$t + \frac{T}{2}$	αz
	II	$\gamma z - \frac{T}{2} \leq t \leq \alpha z + \frac{T}{2}$	γz	αz
	III	$\alpha z + \frac{T}{2} \leq t \leq \gamma z + \frac{T}{2}$	γz	$t - \frac{T}{2}$
$\gamma > \alpha$ $T \leq \alpha - \gamma z$	I	$\alpha z - \frac{T}{2} \leq t \leq \alpha z + \frac{T}{2}$	$t + \frac{T}{2}$	αz
	II	$\alpha z + \frac{T}{2} \leq t \leq \gamma z - \frac{T}{2}$	$t + \frac{T}{2}$	$t - \frac{T}{2}$
	III	$\gamma z - \frac{T}{2} \leq t \leq \gamma z + \frac{T}{2}$	γz	$t - \frac{T}{2}$

Table A-IV.

Convolution Integral Limits.

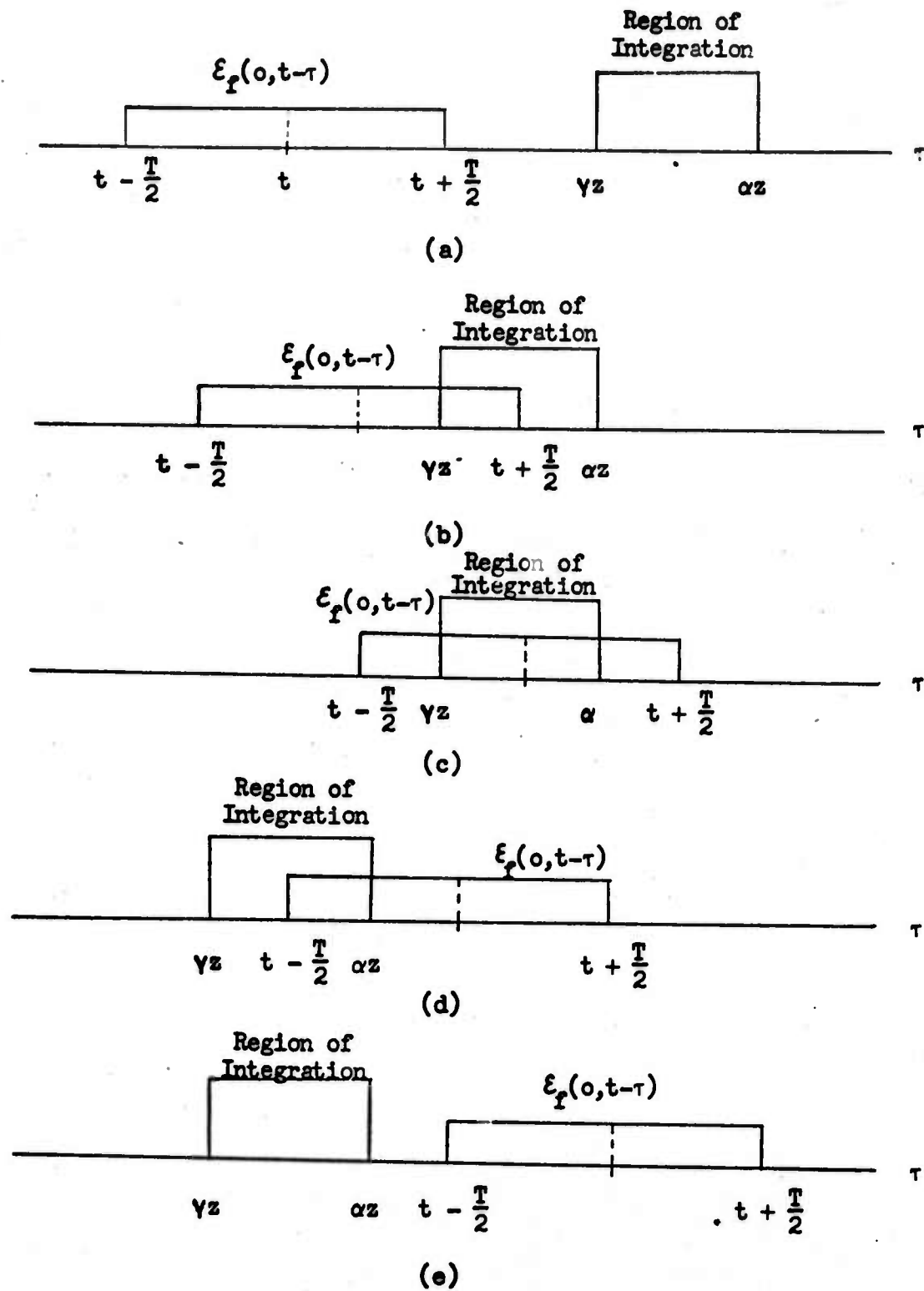


FIGURE A-IV. Convolution Integral Limits Relationship.

REFERENCES

1. Bernfeld, M., and C. E. Cook, Radar Signals: An Introduction to Theory and Application, New York: Academic Press (1967).
2. Bey, P. P., J. F. Giuliani, and H. Rabin, "Phase-Matched Optical Harmonic Generation in Liquid Media Employing Anomalous Dispersion," IEEE Journal of Quantum Electronics, Vol. QE-4, No. 11, pp 932-939, November (1968).
3. DeMaria, A. J., D. A. Stetser, and W. H. Glenn, Jr., "Ultrashort Light Pulses," Science, Vol. 156, No. 3782, pp 1557-1568, 23 June (1967).
4. Glenn, W. H., "Second Harmonic Generation by Picosecond Optical Pulses," IEEE Journal of Quantum Electronics, Vol. QE-5, No. 6, pp 284-290, June (1969).
5. Jenkins, F. A., and H. E. White, Fundamentals of Optics, New York: McGraw-Hill (1957).
6. Lathi, B. P., Communication Systems, New York: John Wiley (1968).
7. Treacy, E. B., "Direct Demonstration of Picosecond-Pulse Frequency Sweep, App. Phys. Letters, Vol. 17, No. 1, pp 14-16, July (1970).