Nonlinear Schrödinger equation for optical media with quadratic nonlinearity

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Wave propagation in optical media with strong dispersion and weak quadratic nonlinearity is analyzed using the method of multiple scales. This method shows that the evolution of the envelope for a single nondepleted pump wave is described by the nonlinear Schrödinger equation. Hence various self-modulation effects, due to an effective intensity-dependent refractive index, are possible to observe in materials with quadratic nonlinearity. That is, materials that are known to generate $\chi^{(2)}$ wave processes may also support, for example, soliton propagation. Physical conditions and numerical examples are given for observing solitons, self-defocusing, and spectral broadening. Other self-modulation effects are also discussed as well.
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I. INTRODUCTION

In nonlinear optics, for waves propagating in a strongly dispersive dielectric with weak quadratic nonlinearity, it has been generally accepted that various three-wave-mixing processes are obtained, such as second-harmonic generation. Intensity-dependent effects are usually expected in cubic nonlinear materials but not in purely quadratic nonlinear materials. The conventional reason that intensity-dependent refraction effects are not expected in quadratically nonlinear materials is that the phase of the fundamental harmonic cannot be phase aligned with the higher harmonics within the slowly-varying-envelope approximation (SVEA) framework [1–3]. The SVEA is a conventional first-order $[O(\epsilon)]$ perturbation method used in optics to derive simplified evolution equations for the slowly modulated envelopes of the propagating waves. However, due to recent developments using unconventional methods (in nonlinear optics), it has been shown that intensity-dependent effects occur for at least two distinct boundary-value problems in quadratically nonlinear materials. It is possible with the appropriate boundary value problem to derive the nonlinear Schrödinger equation [4,5] with an effective intensity-dependent refactive index.

In one-boundary-value problem, two time-independent envelopes [with efficient $O(\epsilon)$ phase matching], propagating at the carrier frequency and second harmonic in a lossless medium, are studied by the standard SVEA [6,7]. By utilizing the Manley-Rowe relations, the problem is reduced to a Duffing-type nonlinear oscillator with a cubic nonlinearity that describes the spatial dependence of the fundamental harmonic envelope. Here the specific nature of the boundary-value problem does not permit the derivation of the nonlinear Schrödinger equation. The theory in [6, 7] does not apply for two waves propagating at different group velocities nor two waves with inefficient $O(1)$ phase matching nor waves in a lossy material. Also the problem is truncated at first-order perturbation theory with only two spatial scales since it is analyzed within the SVEA framework.

The SVEA is a useful method as long as the physical problem may be truncated at first-order perturbation theory and only two time or distance scales are assumed. However, some problems may require proceeding to higher-order perturbation theory and may require several time and distance scales. It is convenient to utilize a perturbation method that is self-consistent to any perturbation order. One such method devised by Cole [8], Sturrock [9], and Sandri [10] is the method of multiple scales (MMS). We will use MMS to derive the nonlinear Schrödinger equation for quadratic nonlinear optical materials. MMS and several other equivalent methods have been used to analyze weakly nonlinear physical systems to extract the dominant canonical equations arising from various asymptotic limits. It must be noted that if MMS is restricted to only two time scales, it reproduces the first-order perturbation theory results obtained by SVEA.

The nonlinear Schrödinger equation is canonical in the sense that it is obtained as the asymptotic far field in the strongly dispersive, weakly nonlinear limit and describes the evolution of envelope wave packets over long times and distances [11,12]. If, on the other hand, another limit is applied, a different dominant canonical equation can be derived. For example, one usually obtains the Korteweg–deVries equation in the weakly dispersive, weakly nonlinear limit. The nonlinear Schrödinger equation arises in many physical systems such as water waves [13], plasmas [14–16], and fiber optics [17, 18]. It is also one of the equations that can be solved exactly by the inverse scattering transform [19, 20]. The idea behind MMS and other equivalent methods is to systematically eliminate artificially growing forced terms by imposing secularity conditions that, in turn, lead to the asymptotic far fields. The generalized Krylov-Bogoliubov-Mitropolsky method

[21], Taniuti's reductive perturbation method for wave propagation [22], and a version of Whitham's averaged Lagrangian method [23] are equivalent to MMS because the same secularity conditions are obtained. In this paper we prefer MMS because it has the advantage of deriving the secularity conditions in a clear and explicit manner requiring the least amount of algebra and the fewest assumptions.

In this paper, we choose to illustrate in detail the mathematical machinery of MMS applied to the simplest signaling (boundary-value) problem, that of a single time-dependent input wave incident upon a lossy, dispersive quadratically nonlinear material. Assuming three time and distance scales, and proceeding to second-order $[O(\epsilon^2)]$ perturbation theory, the nonlinear Schrödinger equation is derived for the fundamental harmonic in the nondepleted pump approximation. The phase mismatch is $O(1)$, so that all higher harmonics that are generated remain small $[O(\epsilon)$ or smaller]. Many self-modulation properties of quadratic nonlinear materials are analogous to those found in optical fibers made from low attenuation glasses. However, the effective intensity-dependent refractive index derived here is different from the intrinsic refractive index of cubic materials (and is different from [6]). The solutions of the nonlinear Schrödinger equation for cubic media such as optical fibers [24, 25] may be used for quadratic nonlinear materials by replacing the intrinsic refractive index by the effective index. For example, under appropriate conditions, temporal optical envelope solitons may be launched in quadratic nonlinear materials. Other self-phase modulation effects may also be observed.

We now summarize what is done in the following sections of this paper. In Sec. II, we state the mathematical problem and nondimensionalize the governing equations. The equations for the optical material are analogous to the Boussinesq equation that arises in water wave theory [23] and plasmas [26]. The optical medium equations are slightly more complicated, but under the appropriate asymptotic limit they may be reduced to the Boussinesq equation. Since we are solving a signaling problem, the time and spatial derivatives are interchanged from the water wave boundary-value problem.

In Sec. III, we apply MMS to the governing equations in the strongly dispersive and weakly nonlinear asymptotic limit and proceed to third-order perturbation theory. The signaling problem is solved to $O(\epsilon)$, but secularity conditions are imposed up to $O(\epsilon^3)$. In Sec. III C, we derive the nondimensionalized nonlinear Schrödinger equation from second-order $O(\epsilon^2)$ perturbation secularity conditions. This is the main result of this paper. We then include a third-order $[O(\epsilon^3)]$ correction to the effective intensity-dependent refractive index that arises from secularity conditions from a small rectified electric field.

In Sec. IV, the coefficients of the nonlinear Schrödinger equation that were derived in Sec. III are related to physical parameters. It is then shown by numerical example that the third-order perturbation correction to the effective index may be neglected. Therefore, second-order perturbation theory (of Sec. III C) provides the essential results for the dynamics of the slowly modulated envelope. In Sec. V, new experiments to perform for quadratically nonlinear materials that demonstrate self-modulation are suggested. For example, conditions for soliton propagation and self-defocusing are given.

We conlude the paper in Sec. VI and briefly discuss other pertinent boundary-value problems for quadratically nonlinear materials that may be analyzed using MMS. We would like to mention that for readers who readily accept the idea that the nonlinear Schrödinger equation exists for quadratic media due to second-order perturbation theory, and would like to dismiss the mathematical details, may skip Sec. III. That is, one may read Sec. II and proceed to Secs. IV and V to concentrate on the physical effects and experimental conditions.

II. FORMULATION OF THE SIGNALING PROBLEM

A single slowly modulated input wave of arbitrary shape propagating in free space at carrier frequency ω encounters a semi-infinite $\chi^{(2)}$ material at $z = 0$. We assume normal incidence at the boundary and the direction of propagation is in the z direction, with the electric field in the x direction and the magnetic field in the y direction. The electric and magnetic fields are transverse to the direction of propagation and to each other, but they are tangential to the boundary. Therefore, we impose boundary conditions that the tangential electric and magnetic fields be continuous across the boundary at $z = 0$. We also impose a radiation condition so that there are no incoming waves from infinity in the $\chi^{(2)}$ material. The boundary conditions imply a signaling problem instead of an initial-value problem.

We consider nonlinear materials that can be described by an ensemble of identical classical anharmonic oscillators with small quadratic restoring forces and a resonant frequency ω_0 far from ω. From the ensemble we obtain a macroscopic polarization p that is coupled to the electric field E. In the medium, we also assume that we are in the nondepleted pump approximation so that there is no phase matching. That is, all harmonics that are generated are small compared to the input beam because the phase mismatch between harmonics and fundamental is $O(1)$.

The quadratic medium is described by the following anharmonic-oscillator equation that relates the induced polarization to the applied electric field [27]:

$$\frac{\partial^2}{\partial t^{*2}}p^* + \Gamma^* \frac{\partial}{\partial t^*}p^* + \omega_0^{*2}p^* + \frac{dp^{*2}}{Ne} = \frac{Ne^2}{m}E^*. \quad (1)$$

The electric field being modified by the medium is described by Maxwell's wave equation

$$\left[c^{*2}\frac{\partial^2}{\partial z^{*2}} - \frac{\partial^2}{\partial t^{*2}}\right]E^* = \frac{1}{\epsilon_0}\frac{\partial^2}{\partial t^{*2}}p^*. \quad (2)$$

Here p^* denotes polarization, E^* the electric field in medium, Γ^* the damping coefficient, ω_0^* the resonant frequency, c^* the speed of light, d the nonlinear restoring force coefficient, N the number of atoms per unit volume, e the electron charge, m the electron mass, and

ϵ_0 the permittivity of free space. The variables with an asterisk are notations devised to refer to the fact that the variables do have physical dimensions. These will be normalized later to make all variables dimensionless. The electric field in free space $\tilde{E}^*$ is described by the homogeneous wave equation

$$\left[c^{*2}\frac{\partial^2}{\partial z^{*2}} - \frac{\partial^2}{\partial t^{*2}}\right]\tilde{E}^* = 0. \tag{3}$$

The boundary conditions which must be satisfied at the interface are that both the tangential electric fields and the tangential magnetic fields be continuous. This means

$$\tilde{E}^*(t^*,0)\Big|_{\substack{\text{free}\\\text{space}}} = E^*(t^*,0)\Big|_{\text{medium}}, \tag{4}$$

$$\frac{\tilde{B}^*(t^*,0)}{\mu_0}\Big|_{\substack{\text{free}\\\text{space}}} = \frac{B^*(t^*,0)}{\mu_0}\Big|_{\text{medium}}, \tag{5}$$

where $\tilde{B}^*$ and B^* are, respectively the magnetic fields in free space and the medium and μ_0 is the permeability of free space. The magnetic fields may be eliminated from the problem by using

$$\frac{\partial}{\partial z^*}E^* = -\frac{\partial B^*}{\partial t^*}. \tag{6}$$

In order to perform perturbation expansions, the above equations must be at first cast into dimensionless variables. A perturbation parameter must be chosen from a dimensionless ratio of physical variables. We define the following dimensionless quantities: $p = p^*/p_0^*, E = E^*/E_0^*, t = t^*/T, z = z^*/Z, \Gamma = \Gamma^* T$, and $\omega_0 = \omega_0^* T$. Here p, E, t, z, Γ, and ω_0 are, respectively, the dimensionless polarization, electric field, time, spatial variable, damping coefficient, and resonant frequency. The dimensionless frequency is chosen to be one so that T is defined as $(\omega_0^*)^{-1}$. We let $p_0^* = \epsilon_0 E_0^*$ and choose the dimensionless speed of light $c = c^* T/Z$. We let $c = 1$, which defines Z. Thus, in the quadratic medium, Eqs. (1) and (2) may be rewritten as

$$\left(\frac{\partial^2}{\partial t^2} + \Gamma\frac{\partial}{\partial t} + \omega_0^2\right)p = fE - \epsilon p^2, \tag{7}$$

$$\left(c^2\frac{\partial^2}{\partial z^2} - \frac{\partial^2}{\partial t^2}\right)E = \frac{\partial^2}{\partial t^2}p. \tag{8}$$

Let $f \equiv \frac{Ne^2}{p_0^* m}E_0^* T^2 \equiv \omega_p^2 T^2$, where $\sqrt{f}$ is the dimensionless plasma frequency. The perturbation parameter ϵ is chosen to be

$$\epsilon \equiv \frac{dT^2 p_0^*}{Ne} = \frac{dT^2\epsilon_0 E_0^*}{Ne}. \tag{9}$$

In free space, Eq. (3) is nondimensionalized as

$$\left[c^2\frac{\partial^2}{\partial z^2} - \frac{\partial^2}{\partial t^2}\right]\tilde{E} = 0, \tag{10}$$

with $\tilde{E} = \frac{\tilde{E}^*}{E_0^*}$. Defining $H^* = \frac{B^*}{\mu_0}, \tilde{H}^* = \frac{\tilde{B}^*}{\mu_0}$, and $H = \frac{H^*}{H_0}, \tilde{H}^* = \frac{\tilde{H}^*}{H_0}$ we find that the boundary conditions become

$$\tilde{E}(0,t) = E(0,t), \tag{11}$$

$$\tilde{H}(0,t) = H(0,t), \tag{12}$$

and Eq. (6) becomes

$$c\frac{\partial}{\partial z}E(0,t) = -\frac{\partial}{\partial t}H(0,t). \tag{13}$$

Since we are interested in solving a boundary-value problem for the electric field, it is convenient to operate on Eq. (8) by $(\frac{\partial^2}{\partial t^2} + \omega_0^2)$ to obtain a fourth-order equation for the electric field that has weak polarization sources. Here we assume that $\Gamma \equiv \epsilon^2\gamma$. Notice that the polarizations come in at higher perturbation orders. We replace Eq. (8) by the following:

$$\left[c^2\frac{\partial^4}{\partial t^2\partial z^2} - \frac{\partial^4}{\partial t^4} + c^2\omega_0^2\frac{\partial^2}{\partial z^2} - (\omega_0^2 + f)\frac{\partial^2}{\partial t^2}\right]E = -\epsilon\frac{\partial^2}{\partial t^2}p^2 - \epsilon^2\gamma\frac{\partial^3}{\partial t^3}p. \tag{14}$$

Equation (14) is important because the dispersion properties of the medium are included and since the polarization terms come in at higher perturbation orders, they behave as known source terms. More important, all secularity conditions on the shortest and fastest scales for MMS are determined from the ϵ independent left-hand side.

We will solve Eq. (14) in conjunction with Eqs. (7) and (10) and boundary conditions (11) and (12) using MMS [4]. The nonlinear Schrödinger equation for the medium will be derived from Eqs. (7) and (14) at second-order perturbation theory. We will also show that at third-order perturbation, a small correction term to the effective intensity-dependent refractive index of the Schrödinger equation will be added. However, for most cases of physical interest, this third-order correction term may be neglected. Therefore, second-order perturbation theory using MMS provides the main results of this paper, as shown in the next section. Afterwards, in Sec. IV, coefficients of the nonlinear Schrödinger equation will be related to well-tabulated physical parameters.

III. THE METHOD OF MULTIPLE SCALES

We choose to utilize the derivative expansion version of MMS [10]. The derivative expansion method suggests to us to extend the two independent variables z, t to the sets of independent variables

$$z_0, z_1, z_2, \ldots, z_n\ ,\quad t_0, t_1, t_2, \ldots, t_n\ ,$$

where $z = z_0, t = t_0$ and $z_i = \epsilon^i z, t_i = \epsilon^i t$. There are $2n$ independent variables. However, since we shall proceed to only second-order perturbation theory, we will have three spatial scales (z_0, z_1, z_2) and three time scales (t_0, t_1, t_2). Thus there are six independent variables with z_0 the shortest distance scale and t_0 the fastest time scale. The other variables are longer distance or slower time

scales. Accordingly, the dependent variables are regarded as functions of the six independent variables. For example, $p = p(z_0, z_1, z_2; t_0, t_1, t_2)$. Similar expressions exist for E and $\tilde{E}$. The derivative operators $\frac{\partial}{\partial z}$ and $\frac{\partial}{\partial t}$ are expanded as

$$\frac{\partial}{\partial z} = \frac{\partial}{\partial z_0} + \epsilon \frac{\partial}{\partial z_1} + \epsilon^2 \frac{\partial}{\partial z_2} + \cdots, \tag{15}$$

$$\frac{\partial}{\partial t} = \frac{\partial}{\partial t_0} + \epsilon \frac{\partial}{\partial t_1} + \epsilon^2 \frac{\partial}{\partial t_2} + \cdots. \tag{16}$$

It is assumed further that the $p, E, \tilde{E}$ dependent variables have an asymptotic representation of the following form: for instance,

$$\begin{aligned} p(z_0, z_1, z_2; t_0, t_1, t_2) = {} & p_0(z_0, z_1, z_2; t_0, t_1, t_2) \\ & + \epsilon p_1(z_0, z_1, z_2; t_0, t_1, t_2) \\ & + \epsilon^2 p_2(z_0, z_1, z_2; t_0, t_1, t_2) + \cdots. \end{aligned} \tag{17}$$

Representations similar to (17) exist for E and $\tilde{E}$. We substitute expansions such as (15)–(17) into Eqs. (14), (7), and (10) and boundary conditions (11) and (12) and then collect orders of (ϵ).

The idea behind MMS is to seek uniform perturbation expansions by the systematic elimination of secularly growing forcing terms which enter at higher perturbation orders. Such elimination of secular terms will also show how the dependent variables $p, E, \tilde{E}$ vary with respect to the slow distance and slow time scales $(z_1, z_2; t_1, t_2)$.

We are primarily interested in what happens in the semi-infinite $\chi^{(2)}$ material, once a slowly modulated electric field in free space encounters the medium at $z = 0$. We shall solve for the reflection and transmission coefficients to $O(\epsilon)$. The following form is assumed for the electric field in free space:

$$\tilde{E} = \tilde{E}_0(z_0, z_1; t_0, t_1) + \epsilon \tilde{E}_1(z_0, z_1; t_0, t_1) + \cdots, \tag{18}$$

with

$$\begin{aligned} \tilde{E}_0 = {} & \left[\tilde{a}_0\left(t_1 - \frac{z_1}{c}\right) e^{i\omega\left(\frac{z_0}{c} - t_0\right)} + \text{c.c.}\right] \\ & + \left[\tilde{b}_0\left(t_1 + \frac{z_1}{c}\right) e^{-i\omega\left(\frac{z_0}{c} + t_0\right)} + \text{c.c.}\right] \end{aligned} \tag{19}$$

and

$$\begin{aligned} \tilde{E}_1 = {} & \left[\tilde{b}_{1(1)}(t_1 + z_1) e^{-i\omega\left(\frac{z_0}{c} + t_0\right)} + \text{c.c.}\right] \\ & + \left[\tilde{b}_{1(2)}(t_1 + z_1) e^{-i2\omega\left(\frac{z_0}{c} + t_0\right)} + \text{c.c.}\right]. \end{aligned} \tag{20}$$

From the $O(1)$ electric field in free space $\tilde{E}_0$, we see from Eq. (19) that $\tilde{a}_0\left(t_1 - \frac{z_1}{c}\right)$ is the given slowly modulated amplitude of arbitrary shape that depends on only the (z_1, t_1) scales. It is multiplied by an exponential phase factor that depends on the (z_0, t_0) scales and the carrier frequency ω. Since there is a material discontinuity at $z = 0$, we have a reflected wave with unknown function b_0 that depends on $\left(t_1 + \frac{z_1}{c}\right)$. The functional form of b_0 will be determined by applying the boundary conditions.

The $O(\epsilon)$ electric field in free space $\tilde{E}_1$ consists of unknown reflected waves $\tilde{b}_{1(1)}, \tilde{b}_{1(2)}$ at the fundamental and second harmonics. Notice that the first subscript denotes perturbation order and the second one denotes harmonic component. The functional form of the $\tilde{E}_1$ reflected waves must be found from the $O(\epsilon)$ boundary conditions. One may verify that the free-space solutions of $\tilde{E}_0$ and $\tilde{E}_1$ in (19) and (20) do indeed satisfy the sequence of perturbation wave equations obtained from MMS by expanding Eq. (10). We proceed to examine the medium.

A. The $O(1)$ problem

The lowest-order perturbation problem corresponds to having set $\epsilon = 0$ after expansions (15)–(17) are substituted into Eqs. (14) and (7) and the boundary conditions. For the nonlinear medium we have

$$\begin{aligned} L^{(0)} E_0 \equiv {} & \left[c^2 \frac{\partial^4}{\partial t_0^2 \partial z_0^2} - \frac{\partial^4}{\partial t_0^4} + c^2 \omega_0^2 \frac{\partial^2}{\partial z_0^2} \right. \\ & \left. - (\omega_0^2 + f) \frac{\partial^2}{\partial t_0^2} \right] E_0 = 0 \end{aligned} \tag{21}$$

and

$$\left(\frac{\partial^2}{\partial t_0^2} + \omega_0^2\right) p_0 = f E_0. \tag{22}$$

The electric field E_0 has an unknown slowly varying amplitude $a_0(z_1, z_2; t_1, t_2)$ that depends on the slower scales and the amplitude is multiplied by an exponential phase factor. The functional form of E_0 is assumed to be

$$\begin{aligned} E_0 = {} & a_0(z_2, z_1; t_2, t_1) e^{i(kz_0 - \omega t_0)} \\ & + a_0^*(z_2, z_1; t_2, t_1) e^{-i(kz_0 - \omega t_0)}, \end{aligned} \tag{23}$$

with ω the known carrier frequency.

Note that the radiation condition was used to eliminate backward traveling waves in the medium. The $(z_1, z_2; t_1, t_2)$ scales enter this $O(1)$ problem as parameters. The wave vector k is unknown and must be determined by substituting (23) into (21). We then obtain the dispersion relation that shows how k depends on ω,

$$D(k, \omega) \equiv c^2 k^2 \omega^2 - c^2 \omega_0^2 k^2 - \omega^4 + (\omega_0^2 + f)\omega^2 = 0\,. \tag{24}$$

It can easily be shown that for k^2 to be an $O(1)$ quantity, the difference $(\omega_0 - \omega) \sim O(1)$. This implies that the applied frequency ω is far from the resonant frequency ω_0, which in turn ensures that the MMS expansion is valid. The polarization p_0 may be found since the electric field E_0 has been determined. Substituting (23) into (22) the polarization response is

$$p_0 = \frac{f E_0}{\omega_0^2 - \omega^2} = \frac{f a_0}{\omega_0^2 - \omega^2} e^{i(kz_0 - \omega t_0)} + \text{c.c.} \tag{25}$$

The polarization p_0 will enter as a source term for the electric field for the $O(\epsilon)$ calculation.

The electric fields in free space and the nonlinear medium must now be matched at the interface at $z_0 = z_1 = z_2 = 0$. From the boundary conditions (11) and (12) we respectively obtain the transmission and reflection coefficients

$$a_0(t_1) = \frac{2\omega}{\omega + ck(\omega)}\tilde{a}_0(t_1), \tag{26}$$

$$\tilde{b}_0(t_1) = \frac{\omega - ck(\omega)}{\omega + ck(\omega)}\tilde{a}_0(t_1). \tag{27}$$

Since $\tilde{a}_0(t_1)$ was chosen to depend on only t_1, then at the boundary, a_0 must depend on only t_1. Away from the boundary, a_0 may also depend on z_1, z_2, but cannot depend on t_2. The transmission and reflection coefficients (26,27) appear to be the standard linear results for harmonic waves. However, they have been generalized to include arbitrary slowly varying pulse shapes that are parametrized by the t_1 scale. The (z_1, t_1) scales will not enter only as parameters in the $O(\epsilon)$ calculation. There they will have to be treated as independent variables that will help eliminate secular behavior.

B. The $O(\epsilon)$ problem

The electric field and polarization in the medium satisfy the following $O(\epsilon)$ perturbation equations:

$$L^{(0)}E_1 = L^{(1)}E_0 - \frac{\partial^2}{\partial t_0^2}p_0^2, \tag{28}$$

$$\left(\frac{\partial^2}{\partial t_0^2} + \omega_0^2\right)p_1 = fE_1 - p_0^2 - 2\frac{\partial^2 p_0}{\partial t_1 \partial t_0}. \tag{29}$$

The operator $L^{(0)}$ was defined in Eq. (21) and $L^{(1)}$ is defined as

$$L^{(1)} \equiv \left[-2c^2\frac{\partial^4}{\partial z_0 \partial z_1 \partial t_0^2} - 2c^2\frac{\partial^4}{\partial t_0 \partial z_0^2 \partial t_1} + \frac{4\partial^4}{\partial t_0^3 \partial t_1} - 2c^2\frac{\omega_0^2\partial^2}{\partial z_0 \partial z_1} + 2(\omega_0^2 + f)\frac{\partial^2}{\partial t_0 \partial t_1}\right]. \tag{30}$$

The $L^{(1)}E_0$ term in Eq. (28) acts as a forced source term which satisfies the $L^{(0)}$ operator. It will grow as $\theta e^{i\theta}$ with $\theta \equiv k(\omega)z_0 - \omega t_0$ unless it is eliminated. Therefore set

$$L^{(1)}E_0 = \left[\frac{\partial D}{\partial k}\frac{\partial a_0}{\partial z_1} - \frac{\partial D}{\partial \omega}\frac{\partial a_0}{\partial t_1}\right]ie^{i(kz_0 - \omega t_0)} + \text{c.c.} = 0. \tag{31}$$

After substituting the form (23) for E_0 into (31) we then differentiate out the known (z_0, t_0) behavior given by the exponential phase factor. The coefficients of every exponential phase factor must be set to zero. Therefore Eq. (31) becomes an envelope equation so that a_0 obeys the following first-order linear partial differential equation on the (z_1, t_1) slow scales:

$$\frac{\partial a_0}{\partial z_1} + \frac{1}{V_g(\omega)}\frac{\partial a_0}{\partial t_1} = 0. \tag{32}$$

$V_g(\omega)$ is the group velocity and is related to derivatives of the dispersion relation (24): $\frac{\partial k}{\partial \omega} = -\frac{\partial D}{\partial \omega}/\frac{\partial D}{\partial k} = 1/V_g(\omega)$. Solving Eq. (32) shows that the envelope a_0 propagates with the group velocity on the (z_1, t_1) scales and the envelope energy $|a_0|^2$ is conserved. We have $a_0(z_1, t_1; z_2) = a_0(s_1, z_2)$, with the retarded time s_1:

$$s_1 \equiv t_1 - \frac{1}{V_g(\omega)}z_1. \tag{33}$$

The $O(\epsilon)$ problem in the medium with bounded periodic terms becomes

$$L^0(E_1) = -\frac{\partial^2}{\partial t_0^2}p_0^2 = \frac{4\omega^2 f^2 a_0^2}{(\omega_0^2 - \omega^2)^2}e^{2i(k(\omega)z_0 - \omega t_0)} + \text{c.c.} \tag{34}$$

The p_0^2 source term remains bounded because the exponential phase factor with $2k(\omega)z_0 - 2\omega t_0$ dependence does not give the dispersion relation (24) when ω is replaced by 2ω. This is because $2k(\omega) \neq k(2\omega)$. The polarization source p_0 excites a second harmonic in the medium. The electric field E_1 is composed of a homogeneous solution and a particular solution at the second harmonic. The electric field E_1 also has a source term at the interface at the fundamental harmonic. This will be shown when the boundary conditions will be applied. We let E_1 in the medium consist of

$$E_1 = E_{1H} + E_{1p}, \tag{35}$$

with E_{1H} = unknown homogeneous solution and E_{1p} = known particular solution,

$$E_{1H} = a_{1(2)}(z_1, z_2; t_1)e^{i(k(2\omega)z_0 - 2\omega t_0)} + a_{1(1)}(z_1, z_2; t_1)e^{i(k(\omega)z_0 - \omega t_0)} + \text{c.c.}, \tag{36}$$

$$E_{1p} = +\frac{fa_0^2(s_1)}{3\omega^2(\omega_0^2 - \omega^2)}e^{i(2k(\omega)z_0 - 2\omega t_0)} + \text{c.c.} \tag{37}$$

In Eq. (36) the envelopes $a_{1(2)}, a_{1(1)}$ are unknown. The exponential phase factors satisfy $L^{(0)}E_{1H} = 0$. In Eq. (37), a_0 is already known from $O(1)$ theory.

We finally match the electric fields in free space and the quadratic medium at the boundary $z = 0$ to find the above unknown envelopes. The boundary conditions (11) and (12) are expanded to $O(\epsilon)$. Applying the boundary conditions for the fundamental harmonics in $(\tilde{E}_1, E_1)$, we find

$$\tilde{b}_{1(1)}(t_1) = a_{1(1)}(t_1) = \frac{i2c[k(\omega) - \omega k'(\omega)]}{[\omega + ck(\omega)]^2}\frac{\partial \tilde{a}_0(t_1)}{\partial t_1}. \tag{38}$$

Similarly for the second harmonics we have

$$a_{1(2)}(t_1) = \frac{-8f\tilde{a}_0^2(t_1)}{3[2\omega + ck(2\omega)][\omega + ck(\omega)](\omega_0^2 - \omega^2)}, \tag{39}$$

$$\tilde{b}_{12}(t_1) = -\frac{4}{3}\frac{c[2k(\omega) - k(2\omega)]f\tilde{a}_0^2(t_1)}{[2\omega + ck(2\omega)][\omega + ck(\omega)]^2(\omega_0^2 - \omega^2)}. \quad (40)$$

Now both $\tilde{E}_1$ given by (20) and E_1 given by (35) are completely specified by the given incident field $\tilde{a}_0(t_1)$. The polarization p_1 must be determined since it will enter as a source term at $O(\epsilon^2)$. From Eq. (29) we find that p_1 is the sum of the following terms:

$$\begin{aligned} p_1 = {} & \frac{-f^2}{(\omega_0^2-\omega^2)^2}\left[\frac{a_0^2 e^{2i\theta}}{\omega_0^2-4\omega^2} + \frac{2a_0a_0^*}{\omega_0^2} + \frac{a_0^{*2}e^{-2i\theta}}{\omega_0^2-4\omega^2}\right] - \frac{2f}{(\omega_0^2-\omega^2)^2}\left[-i\omega\frac{\partial a_0}{\partial t_1}e^{i\theta} + i\omega\frac{\partial a_0^*}{\partial t_1}e^{-i\theta}\right] \\ & + \frac{f}{\omega_0^2-4\omega^2}a_{1(2)}e^{i[k(2\omega)z_0-2\omega t_0]} + \left(\frac{fa_{1(1)}e^{i\theta}}{\omega_0^2-\omega^2}\right) + \frac{f^2a_0^2e^{2i\theta}}{3\omega^2(\omega_0^2-\omega^2)(\omega_0^2-4\omega^2)} \\ & + \frac{f}{\omega_0^2-4\omega^2}a_{1(2)}^*e^{-i[k(2\omega)z_0-2\omega t_0]} + \left(\frac{fa_{1(1)}^*e^{-i\theta}}{\omega_0^2-\omega^2}\right) + \frac{f^2a_0^{*2}e^{-2i\theta}}{3\omega^2(\omega_0^2-\omega^2)(\omega_0^2-4\omega^2)}. \end{aligned} \quad (41)$$

C. The $O(\epsilon^2)$ problem

We need to determine the secularity conditions for each harmonic of the following equation:

$$L^{(0)}E_2 = L^{(1)}E_1 + L^{(2)}E_0 - 2\frac{\partial^2}{\partial t_0^2}(p_1p_0) - \gamma\frac{\partial^3}{\partial t_0^3}p_0 - 2\frac{\partial^2 p_0^2}{\partial t_0\partial t_1}. \quad (42)$$

We do not need to expand the oscillator equations or boundary conditions since they will not be used. Here $L^{(0)}$ and $L^{(1)}$ were defined respectively by (21) and (30). The operator $L^{(2)}$ is given as

$$\begin{aligned} L^{(2)} \equiv \Bigg[& -2c^2\frac{\partial^4}{\partial t_0^2\partial z_0\partial z_2} - 2c^2\omega_0^2\frac{\partial^2}{\partial z_0\partial z_2} - 2c^2\frac{\partial^4}{\partial t_0\partial z_0^2\partial t_2} + \frac{4\partial^4}{\partial t_0^3\partial t_2} + 2(\omega_0^2+f)\frac{\partial^2}{\partial t_0\partial t_2} - c^2\frac{\partial^4}{\partial t_0^2\partial z_1^2} \\ & -c^2\omega_0^2\frac{\partial^2}{\partial z_1^2} - c^2\frac{\partial^4}{\partial z_0^2\partial t_1^2} + \frac{6\partial^4}{\partial t_0^2\partial t_1^2} + (\omega_0^2+f)\frac{\partial^2}{\partial t_1^2} - 4c^2\frac{\partial^4}{\partial t_0\partial z_0\partial t_1\partial z_1}\Bigg]. \end{aligned} \quad (43)$$

The terms that are secular in the medium have to be set to zero. This means from Eq. (42) that

$$L^{(1)}E_1 + L^{(2)}E_0 - 2\frac{\partial^2}{\partial t_0^2}(p_1p_0) - \gamma\frac{\partial^3}{\partial t_0^3}p_0 = 0 \quad (44)$$

for second harmonics with $e^{i[k(2\omega)z_0-2\omega t_0]}$ phase factors and fundamental harmonics with $e^{i[k(\omega)z_0-\omega t_0]}$ phase factors. It is easily shown that the secularity condition gives the result that $a_{1(2)}$ propagates at the group velocity of the second harmonic. However, the secularity condition for the fundamental harmonic will provide the most interesting result. We have a little work to do in simplifying Eq. (44). First of all, in the p_1p_0 term we are interested in only terms with proper phase. We multiply p_1, as shown by Eq. (41), by p_0 given by Eq. (25). We keep terms with phase $\theta \equiv k(\omega)z_0 - \omega t_0$. We find from Eq. (41) that only the two $e^{2i\theta}$ terms and the dc term in p_1 will contribute to the p_1p_0 product that has the proper phase. Note that p_1 contains terms from E_1 and p_0^2, as seen by Eq. (29). E_1 itself has terms proportional to $\frac{\partial^2}{\partial t_0^2}p_0^2$, so only contributes second harmonics. The p_0^2 term contributes second harmonics and a dc polarization. After some algebra and collecting terms with $e^{i\theta}$ phase, we are able to show that

$$-2\frac{\partial^2}{\partial t_0^2}(p_1p_0)\Big|_{\text{phase}} = \frac{2f^3(\omega_0^2-6\omega^2)}{3(\omega_0^2-\omega^2)^3\omega_0^2}|a_0|^2a_0e^{i\theta}. \quad (45)$$

The p_1 term has quadratic polarization effects as solutions, so that when p_1 is multiplied by p_0, we find that p_1p_0 can align itself with $e^{i\theta}$ phase.

Determining $L^{(2)}E_0$ also requires work. Using (43), we at first differentiate out the (z_0, t_0) dependences, transform from (z_1, t_1) to s_1 coordinates, and after extensive algebra the result is

$$\begin{aligned} L^{(2)}\left(a_0e^{i\theta}\right) = {} & i\left(\frac{\partial D}{\partial k}\frac{\partial a_0}{\partial z_2} - \frac{\partial D}{\partial\omega}\frac{\partial a_0}{\partial t_2}\right)e^{i\theta} \\ & -\frac{1}{2}\frac{\partial D}{\partial k}\frac{\partial^2 k}{\partial\omega^2}\left(\frac{\partial^2}{\partial s_1^2}a_0\right)e^{i\theta}. \end{aligned} \quad (46)$$

The (z_0, t_0) behavior can easily be differentiated out of the $L^{(1)}E_1$ and damping terms in Eq. (44). Therefore, Eq. (44) reduces to the following envelope equation:

$$\begin{aligned} & i\left[\frac{\partial D}{\partial k}\frac{\partial}{\partial z_1} - \frac{\partial D}{\partial\omega}\frac{\partial}{\partial t_1}\right]a_{1(1)} + i\left[\frac{\partial D}{\partial k}\frac{\partial}{\partial z_2} - \frac{\partial D}{\partial\omega}\frac{\partial}{\partial t_2}\right]a_0 - \frac{1}{2}\frac{\partial D}{\partial k}\frac{\partial^2 k}{\partial\omega^2}\frac{\partial^2}{\partial s_1^2}a_0 \\ & \qquad + \frac{2}{3}\left(\frac{f^3\left(\omega_0^2-6\omega^2\right)}{\left(\omega_0^2-\omega^2\right)^3\omega_0^2}\right)|a_0|^2a_0 - \frac{i\omega^3\gamma f a_0}{\omega_0^2-\omega^2} = 0. \end{aligned} \quad (47)$$

We notice that the $O(1)$ field a_0 is also a solution of Eq. (32), which contains the same operator $(\frac{\partial D}{\partial k}\frac{\partial}{\partial z_1} - \frac{\partial D}{\partial \omega}\frac{\partial}{\partial t_1})$ that is in front of the $O(\epsilon)$ field $a_{1(1)}$ term. Thus, in Eq. (47) we can consider terms in a_0 as a resonant forcing term. In the medium at $O(\epsilon^2)$ we have a secularity where a_0 acts like a source term for $a_{1(1)}$ on the (z_1, t_1) scales. For $a_{1(1)}$ to be bounded, the "coefficients" in front of a_0 should vanish. Otherwise, $a_{1(1)}$ grows as $z_1 a_0$. Eliminating secularity on the (z_1, t_1) scales, we find that a_0 obeys the nonlinear Schrödinger equation [4], which we define by the operator $S^+(a_0)$:

$$S^+(a_0) = i\left(\frac{\partial}{\partial z_2} + \frac{1}{V_g(\omega)}\frac{\partial}{\partial t_2}\right)a_0 - \frac{1}{2}\frac{\partial^2 k}{\partial \omega^2}\frac{\partial^2}{\partial s_1^2}a_0 - \frac{2f^3\left(\omega_0^2 - 6\omega^2\right)}{6c^2k\left(\omega_0^2 - \omega^2\right)^4\omega_0^2}|a_0|^2 a_0 + \frac{i\gamma\omega^3 f a_0}{2c^2k\left(\omega_0^2 - \omega^2\right)^2} = 0. \tag{48}$$

Here a_0 was assumed independent of t_2. If a_0 did depend on t_2, Eq. (48) can be transformed to coordinates moving with the group velocity so that t_2 drops out. From this multiple scale derivation, we find that the applied single input wave [with $O(1)$ phase matching requirements for inefficient harmonic generation] propagating in a quadratic nonlinear material exhibits self-modulation.

This result in quadratically nonlinear dielectric materials may also be used for pulse shaping and compression as it has been used for the usual cubic materials; this is discussed in Sec. V. The SVEA never obtained this result because it is restricted to only $O(\epsilon)$ perturbation theory whereas MMS may be computed to any-order perturbation in a self-consistent manner. Here we proceeded to second-order perturbation theory to obtain the dominant asymptotic far field evolution equation, which is the nonlinear Schrödinger equation represented by Eq. (48). This is the main result of this paper. The quadratic nonlinearity forces us to examine secularity conditions at $O(\epsilon^3)$ which in turn forces us to consider the possibility of a propagating "rectified" electric field that will constitute a small correction to the intensity-dependent refractive index of Eq. (48). We will show that this correction term may be neglected for most cases of physical interest in optics.

D. The dc electric field boundary value problem

At third-order perturbation theory we have $2\frac{\partial^2}{\partial t_1^2}|p_0|^2$ as a dc polarization source term. If we allow the $O(\epsilon)$ field E_1 to have a dc component in addition to the fundamental and second harmonics, we can balance the polarization term to obtain the following secularity condition:

$$L^{(2)}E_{1\mathrm{dc}} = 2\frac{\partial^2}{\partial t_1^2}|p_0|^2. \tag{49}$$

This will ensure that the $O(\epsilon^3)$ field will remain bounded.

The $E_{1\mathrm{dc}}$ term is independent of the (t_0, z_0) scales so that only part of the $L^{(2)}$ operator defined in (43) with derivatives depending on only (z_1, t_1) is used. The secularity condition may be rewritten as

$$\left[\frac{\partial^2}{\partial z_1^2} - \frac{1}{v^2}\frac{\partial^2}{\partial t_1^2}\right]E_{1\mathrm{dc}} = -\frac{2}{c^2\omega_0^2}\frac{\partial^2}{\partial t_1^2}|p_0(s_1)|^2 \tag{50}$$

and

$$v^2 = \frac{c^2\omega_0^2}{\omega_0^2 + f}. \tag{51}$$

We notice that the left-hand side of Eq. (50) for the $E_{1\mathrm{dc}}$ field is a dispersionless wave equation in the limit as $\omega \to 0$. The speed of propagation v given in (51) is independent of ω and is equal to the phase velocity $\frac{\omega}{k}$ and group velocity $\frac{d\omega}{dk}$ in the limit as $\omega \to 0$.

We have to be careful in applying the limit as $\omega \to 0$ in Eqs. (50) and (51). Remember that (50) was obtained from the fourth-order equation (II), which in turn was constructed in part from the oscillator equation (7), which is inherently restricted to the visible frequencies. The dielectric medium has several resonance bands which may be represented by a set of oscillators with each oscillator corresponding to a region near a particular resonance band. The physical mechanism responsible for the resonance depends on the frequency range. For example, infrared resonances are due to molecular vibrations whereas optical resonances occur because of electronic displacement. As the frequency decreases, the refractive index increases and so does $1/v$ across a resonance band [28]. This is quite different from the dispersion mechanisms arising from water wave theory. Even though v in (51) was derived from parameters in the visible regime, we allow the parameters to change depending on what frequency regime we are in. We define v such that

$$\frac{1}{v} \equiv \frac{n(\tilde{\omega})}{c} \tag{52}$$

and $n(\tilde{\omega})$ is the refractive index at $\tilde{\omega}$. Here we pick $\tilde{\omega}$ corresponding to the inverse of the input signal pulse width. That is, $\tilde{\omega}$ is the bandwidth of the pulse. There is no well defined carrier frequency for the rectified electric field so that $\tilde{\omega}$ is an upper bound for the frequency components which in turn gives a lower bound on the inverse of v. If the spectral distribution of the electric field is known, then a frequency corresponding to the centroid of the wave packet may be used in place of the carrier frequency, and the value of this frequency should be smaller than the total bandwidth. Thus the limit $\omega \to 0$ actually means $\omega \to \tilde{\omega}$, where $\tilde{\omega} \ll \omega$. For example, if we have a 1-ps pulse, then $\tilde{\omega} \sim 10^{12}$ Hz, which is about three orders of magnitude smaller than the carrier frequency in the visible regime.

The general solution of (50) will consist of a forced field propagating at $V_g(\tilde{\omega})$ and a homogeneous term propagating at v. The homogeneous terms must be determined from the boundary conditions that come in at different perturbation orders.

Three different perturbation orders were utilized to extract the signaling problem for the rectified electric field. For $V_g(\omega) \neq v$, the solution to Eq. (50) and boundary conditions imply that the electric field in the medium is

$$E_{1\mathrm{dc}} = \frac{-2}{c^2\omega_0^2\left(\frac{1}{V_g^2(\omega)} - \frac{1}{v^2}\right)}\left[|p_0(s_1)|^2 - \frac{v}{V_g(\omega)}\left[\frac{c+V_g(\omega)}{v+c}\right]\left|p_0\left(t_1 - \frac{1}{v}z_1\right)\right|^2\right], \tag{53}$$

with s_1 defined by Eq. (33).

The corresponding reflected field in free space is then

$$\tilde{E}_{1\mathrm{dc}} = \frac{1}{\left(\frac{1}{V_g^2(\omega)} - \frac{1}{v^2}\right)} \times \left(\frac{2}{c^2\omega_0^2}\right)\left[\frac{c(v - V_g(\omega))}{V_g(\omega)(v+c)}\right]\left|p_0\left(t_1 + \frac{z_1}{c}\right)\right|^2. \tag{54}$$

For many materials the $\left(\frac{1}{V_g^2(\omega)} - \frac{1}{v^2}\right)$ factor will be a large quantity since $n(\tilde{\omega})$ increases as ω crosses lower frequency resonance bands and we are far away from optical resonances; in fact $\frac{1}{v^2} \gg \frac{1}{V_g^2(\omega)}$. This means that the $E_{1\mathrm{dc}}$ and $\tilde{E}_{1\mathrm{dc}}$ fields are small when compared to other $O(\epsilon)$ terms and may be neglected.

The $E_{1\mathrm{dc}}$ terms given by (53) imply extra terms will have to be included at $O(\epsilon^2)$ for the product $p_1 p_0$ since it will now have terms proportional to $E_{1\mathrm{dc}}p_0$. This means that at $O(\epsilon^2)$, the nonlinear Schrödinger equation (48) defined as $S^+(a_0)$ is corrected to

$$S^+(a_0) + \frac{2f^4\omega^2}{kc^4\omega_0^4(\omega_0^2 - \omega^2)^4}\frac{1}{\left[\frac{1}{V_g^2(\omega)} - \frac{1}{v^2}\right]}|a_0(s_1)|^2 a_0(s_1) = 0. \tag{55}$$

Note also that the $E_{1\mathrm{dc}}$ field helps drive the E_1 field at the fundamental harmonic so that $a_{1(1)}$ given in Eq. (38) is corrected to

$$a_{1(1)} = a_{1(1)}^H(s_1) + \frac{2f^4\omega^2 v[c+V_g(\omega)]a_0(s_1)\int^{z_1}|a_0\left(s_1 + \frac{z}{V_g(\omega)}\right)|^2 dz}{c^4\omega_0^4 k(\omega_0^2 - \omega^2)^4\left(\frac{1}{V_g^2(\omega)} - \frac{1}{v^2}\right)V_g(\omega)(v+c)}, \tag{56}$$

where the variable s_1 is defined in Eq. (33). The $a_{1(1)}^H$ term is the uncorrected homogeneous solution. The second term in (56) is the corrected forced term and it is small compared to the other boundary terms at the interface for the fundamental harmonic of the E_1 field. Note that the forced term in Eq. (56) was neglected when obtaining Eq. (38) from $O(\epsilon)$ boundary conditions.

We will examine under what conditions the $O(\epsilon^3)$ corrections can be neglected after we convert the dimensionless Schrödinger Eq. (55) back to physical variables.

IV. THE NONLINEAR SCHRÖDINGER EQUATION AND PHYSICAL PARAMETERS

Second-order perturbation theory was used to derive the nonlinear Schrödinger equation, Equation (48), which is the main result of this paper. A small correction term due to third-order perturbation theory was then added to the Schrödinger equation that resulted in Eq. (55). Equation (54) differs slightly from Eq. (48) only in the intensity-dependent refractive index. We will show Eq. (55) reduces to Eq. (48) for most cases of physical interest by changing from dimensionless units to physical variables. The dimensionless coefficients of the nonlinear Schrödinger Eq. (55) may be dimensionalized and transformed to characteristic physical parameters that describe the medium. At first substitute the variables (defined in Sec. II) $z_2 = \epsilon^2 z^*/Z$, $s_1 = \epsilon\tau^*/T$, $a_0 = E^*/E_0$, and $\omega = \omega^* T$ back into Eq. (55). Then multiply by ϵ^2 (also defined in Sec. II) and perform some algebra so that Eq. (55) may be rewritten as

$$i\frac{\partial}{\partial z^*}E^* - \frac{1}{2}\frac{\partial^2 k^*}{\partial\omega^{*2}}\frac{\partial^2}{\partial\tau^{*2}}E^* - \beta[1+g]|E^*|^2E^* = \frac{i\sigma}{2}E^*. \tag{57}$$

Each variable in Eq. (57) now has the appropriate physical dimensions. The electric field E^* has units of V/m. The absorption coefficient σ is defined as

$$\sigma \equiv \frac{\omega^{*3}\Gamma^*\omega_p^{*2}}{c^{*2}k^*(\omega_0^2 - \omega^{*2})^2} \tag{58}$$

and has units of m^{-1}. The parameters β and g are respectively defined as

$$\beta = \frac{4}{3}\left[d^{(2\omega)}\right]^2\frac{(\omega_0^{*2} - 4\omega^{*2})^2(\omega_0^{*2} - 6\omega^{*2})}{\omega_p^{*2}\omega_0^{*2}k^*c^{*2}}, \tag{59}$$

$$g \equiv \frac{6\omega^{*2}\omega_p^{*2}\omega_0^{*-2}}{(\omega_0^{*2} - 6\omega^{*2})c^{*2}\left[\frac{1}{v^{*2}} - \frac{1}{V_g^2(\omega^*)}\right]}. \tag{60}$$

The parameter g denotes the third-order perturbation correction to the effective refractive index coefficient β. The parameter β was obtained from second-order perturbation theory and has in it, the coefficient of second harmonic generation $d^{(2\omega)}$ "squared." We used the result from Ref. [27]:

$$d^{(2\omega)} \equiv \frac{dNe^3}{2m^2(\omega_0^{*2} - \omega^{*2})^2(\omega_0^{*2} - 4\omega^{*2})\epsilon_0}. \tag{61}$$

We would like to examine the size of g (due to third-order perturbation effects) for a typical nonlinear material such as potassium titanyl phosphate (KTP). We use the following material parameters [29,30]: $\omega_p^* = \omega_0^* = 7.9 \times 10^{15}$ Hz and $v^{*2} = c^{*2}/15.4$ (at $\tilde{\omega} \simeq 10^9$ Hz). Here we assume that the value of $\tilde{\omega}$ does not change much from the gigahertz to the far infrared regime because there are no resonance bands in between.

The given carrier frequency $\omega^* = 1.78 \times 10^{15}$ Hz so that $V_g^2(\omega^*) = c^{*2}/3.486$. We find the value of g in Eq. (57) to be $g = 0.037$. Thus $g \ll 1$ so that for the given operating frequency, we can neglect this term. We expect that $[\frac{1}{v^{*2}} - \frac{1}{V_g^2(\omega^*)}]$ will remain large for cases away from material resonances.

It turns out that the E_{1dc} field obtained from third-order perturbation theory is a small effect. Therefore, second-order perturbation theory is sufficient enough to capture the essential nonlinear behavior of the system away from any material resonances. We will ignore the effect of g as we did in Ref. [5] and design experiments to test for self-modulation effects due to the $\beta|E|^2$ term.

In order to observe self-modulation effects from the material's quadratic nonlinearity, we are interested in $\chi^{(2)}$ media where

$$\beta \gg \frac{n_2 k^*}{n}. \tag{62}$$

That is, we are interested in materials where the $\frac{n_2 k}{n}$ factor arising from the materials intrinsic cubic nonlinearity is negligible compared to the effective index coefficient β arising from the quadratic nonlinearity. The $\frac{n_2 k}{n}$ term is derived from a cubic polarization instead of a quadratic one in Eq. (7) or (14). Replacing ϵp^2 by $\epsilon^2 p^3$ in Eqs. (7) and (14) and performing the same multiple scale analysis as before, we find that the intrinsic nonlinear refractive index coefficient is

$$n_2 = \frac{3}{2}\left(\frac{\epsilon_0^2 b}{N^2 e^2}\right) \frac{\omega_p^{*6}}{(\omega_0^{*2} - \omega^{*2})^4} \left(\frac{\omega^*}{c^* k^*}\right).$$

This nonlinearity usually arises in centrosymmetric materials such as optical fibers. Here ϵ_0 is the permitivity of free space and b is the cubic nonlinear restoring force.

V. PROPOSED EXPERIMENTS

We present a small list of experiments that can be performed to demonstrate self-modulation in $\chi^{(2)}$ materials for propagating single input waves that are not phase matched. The experiments and solutions already developed for optical fibers [24,25] may now be applied to $\chi^{(2)}$ materials.

One experiment that may be performed is to determine under what conditions spectral broadening may occur. If the input pulse is wide and depends on the t_2 scale instead of t_1, so that the group velocity dispersion term is neglected, we find that instead of Eq. (57) we have

$$i\left(\frac{\partial}{\partial z^*} + \frac{1}{V_g(\omega)}\frac{\partial}{\partial t^*}\right) E^* - \beta|E^*|^2 E^* = -\frac{i\sigma}{2} E^*. \tag{63}$$

The solution to (63) is given in Ref. [25] and will not be presented here. We keep in mind that Agrawal used $\frac{n_2 k}{n}$ arising from $\chi^{(3)}$ materials instead of β. In order to utilize his solution, we replace $\frac{n_2 k}{n}$ with β. The solution to (63) gives rise to an intensity-dependent phase shift. Spectral broadening is a consequence of the time dependence of the phase shift.

Experiments for demonstrating the existence of bright soliton and dark soliton envelope propagation may be performed for sufficiently narrow input pulses that depend on the t_1 scale. Here we must take into consideration whether the product $(-\frac{\partial^2 k^*}{\partial \omega^{*2}})$ and $(-\beta)$ in Eq. (57) is greater than or less than zero. For a product greater than zero we have bright soliton propagation. For a product less than zero we have dark soliton propagation. Soliton solutions to Eq. (57) are presented in Ref. [24]. In order to utilize them, we replace $\frac{n_2 k}{n}$ by β.

Self-steepening and pulse peak flattening may be observed for a bright pulse incident upon a medium where the product of $(-\frac{\partial^2 k^*}{\partial \omega^{*2}})$ and $(-\beta)$ is less than zero. This effect deforms a Gaussian shaped pulse into a nearly square pulse. The resulting square pulse can be passed through a dispersive delay line to compress it.

Equation (57) was the result of an infinite plane wave propagating in a dispersive nonlinear material. It is a one-dimensional problem. However, if diffraction effects are included to account for finite beam radius, we should be able to observe self-focusing or self-defocusing of the beam for input wave envelopes with a radial Gaussian profile that is independent of time. Here group velocity dispersion is neglected.

A study of how the combined effects of dispersion and diffraction versus nonlinearity may be performed. Various stability problems may be investigated. We expect that for KTP, the dispersion and nonlinearity are negative with respect to the diffraction term. The possibility of observing symmetric spatial-temporal collapse of the pulse or "light bullets" (pulses that propagate without change in temporal or spatial shape) as suggested in [31] will not be possible unless $\omega^* > \omega_0^*$. Then the dispersion, diffraction, and nonlinear terms are all positive so that there is symmetry between the time and spatial variables. However, since ω^* is greater than the resonance frequency, we are in the ultraviolet region and expect a lot of absorption. The problems involved in the visible spectrum will always have positive diffraction and negative dispersion. The nonlinearity may change sign depending if ω^* is greater than or less than $\frac{\omega_0^*}{\sqrt{6}}$.

Out of this list of experiments to demonstrate self-phase modulation in $\chi^{(2)}$ materials, we consider only two experiments in more detail. We will provide conditions for (temporal) bright soliton propagation in KTP and calculate a soliton length. We then briefly show that for the same carrier frequency used in the soliton problem, a beam of finite aperture in KTP self-defocuses.

We decide to put Eq. (57) in standard normalized form by defining the following variables:

$$\zeta \equiv \left|\frac{\partial^2 k^*}{\partial \omega^{*2}}\right| T^{-2} z^*, \tag{64}$$

$$s \equiv T^{-1}\tau^*, \tag{65}$$

$$q \equiv T\left[\frac{\beta}{|\frac{\partial^2 k^*}{\partial\omega^{*2}}|}\right]^{1/2} E^* \tag{66}$$

and substitute them into (57) to obtain

$$i\frac{\partial q}{\partial\zeta} - \frac{1}{2}\frac{k^{*\prime\prime}}{|k^{*\prime\prime}|}\frac{\partial^2}{\partial s^2}q - |q|^2 q = -i\tilde{\sigma}q, \tag{67}$$

with

$$\tilde{\sigma} \equiv \frac{\sigma T^2}{2|k^{*\prime\prime}|}. \tag{68}$$

The variable T is the chosen pulse width. For bright soliton propagation, we must have $(-\frac{k''}{|k''|})(-1) > 0$. We also assume that $\sigma \ll 1$, but $\sigma \neq 0$. Under these conditions, it is well known that a damped soliton solution may be obtained using perturbation theory on the inverse scattering transform [24]. However, for $\sigma = 0$, the standard undamped one-soliton solution

$$q = M \text{ sech } (Ms). \tag{69}$$

We now estimate a soliton length for the undamped soliton. If the input pulse has $M > 1$, then the sech solution of (57) is periodic in ζ with period $\zeta_0 = \frac{\pi}{2}$. Using this fact, we rewrite (64) as

$$z_0^* = \frac{\pi}{2}T^2\left|\frac{\partial^2 k^*}{\partial\omega^{*2}}\right|^{-1}, \tag{70}$$

with z_0^* the soliton period and effective material length. From (70) we see that z_0^* depends on the pulse width T, which is related to the peak amplitude as seen from (66). We now let $M = 1$ and assume the fundamental solution $q = \text{sech } s$. The peak power of E^* is E_0^* and from (66) we find

$$T^2 = \left|\frac{\partial^2 k^*}{\partial\omega^{*2}}\right|\beta^{-1}E_0^{*-2}. \tag{71}$$

To compute the soliton length, we substitute (71) into (70), since β, E_0^*, and $k^{*\prime\prime}$ are known. Note that

$$k^{*\prime\prime} = \frac{\omega_0^{*2}\omega^*[3\omega_0^{*2} + \omega^{*2}](\omega_0^{*2} - \omega^{*2})\left(\frac{c^*k^*}{\omega^*}\right)^2 - \omega_0^{*4}\omega^{*3}}{c^*(\omega_0^{*2} - \omega^{*2})^4\left(\frac{c^*k^*}{\omega^*}\right)^3}. \tag{72}$$

Material properties for KTP are obtained from Bierlein [29]. We find that near the prescribed wavelength $\lambda = 1.064\ \mu$m, the linear refractive index in the z direction is given as

$$n_z^2 = \frac{c^{*2}k^{*2}}{\omega^{*2}} \approx 2.31 + \frac{1}{1 - \left(\frac{0.238}{\lambda}\right)^2} = 2.31 + \frac{\omega_p^{*2}}{\omega_0^{*2} - \omega^{*2}}. \tag{73}$$

From the refractive index, we see that the resonance frequency and the plasma frequency are the same $\omega_0^* = \omega_p^* = 7.9 \times 10^{15}$ Hz. The applied frequency $\omega^* = 1.78 \times 10^{15}$ Hz. The absorption coefficient is given as $\sigma = 0.6/m$. at our chosen wavelength. The coefficient of second-harmonic generation is also given as $d^{(2\omega)} = 13.7 \times 10^{-12}$ m/V. In order to launch a soliton, the nonlinear term in Eq. (57) must be greater than the absorption coefficient. Thus, we have the condition

$$\beta|E^*|^2 \gg \frac{\sigma}{2}. \tag{74}$$

Substituting in the material parameters given by the absorption and the second-harmonic generation coefficients at the applied frequency, we find that (74) becomes (in MKS units) $|E^*|^2 0.694 \times 10^{-15} > 0.3$; the electric field must be at least 6.5×10^6 V m^{-1} to satisfy this condition. We choose the electric field E^* to range from 5×10^7 to 10^8 V m^{-1} so that the absorption has negligible effects on the soliton. We note that KTP can withstand 10^9 V m^{-1} because the damage threshold is 30 GW/cm^2 for 30-ps pulses at a 10-Hz repetition rate.

For KTP we find $\frac{c^*k^*}{\omega^*} = 1.83, k^{*\prime\prime} = 1.87 \times 10^{-25}$, and $\beta = 0.694 \times 10^{-15}$. Thus for $E_0^* = 5 \times 10^7$ V/m, then $T = 0.1$ ps and the soliton period $z_0^* = 8.9$ cm. If $E_0^* = 10^8$ V/m, then $T = 0.052$ ps, and $z_0^* = 2.3$ cm. These lengths are longer than that for efficient second-harmonic generation. Usually, harmonic generation lengths are about 5 mm. We should be able to observe self-modulation effects in non-phase-matched $\chi^{(2)}$ materials that are several centimeters long. If lower intensities are used, then we require a quadratic nonlinear material with lower absorption. There is a great deal of activity in developing materials, especially polymers that have low absorption, high damage threshold and large quadratic nonlinearity. Thus in the near future, $\chi^{(2)}$ materials may find other uses in optical signal processing besides wave mixing.

We shall investigate under what conditions an incoming cylindrical beam of finite aperture will self-focus or self-defocus in KTP at the applied frequency $\omega^* = 1.78 \times 10^{15}$ Hz. The following dimensionless equation describes a beam of finite but large aperture propagating in the medium:

$$\left[c^2\frac{\partial^4}{\partial t^2\partial z^2} - \frac{\partial^4}{\partial t^4} + c^2\omega_0^2\frac{\partial^2}{\partial z^2} - (\omega_0^2 + f)\frac{\partial^2}{\partial t^2}\right]E$$

$$= -\epsilon\frac{\partial^2}{\partial t^2}p^2 - \epsilon^2\gamma\frac{\partial^3}{\partial t^3}p - \epsilon^2c^2\left(\frac{\partial^2}{\partial t^2} + \omega_0^2\right)\nabla_\perp^2 E. \tag{75}$$

Diffraction is now included by the $\nabla_\perp^2 E = (\frac{\partial^2}{\partial r^2} + \frac{1}{r}\frac{\partial}{\partial r})E$ term in (V). The boundary-value problem and multiple scales expansion is the same as before, but we assume that the incoming wave is continuous and has a cylindrically symmetric radial profile so that

$$E_0 = A(z_2, r)e^{i[k(\omega)z_0 - \omega t_0]} + \text{c.c.} \tag{76}$$

The slowly varying envelope is time independent but depends on the slowly varying coordinate $z_2 = \epsilon^2 z_0$ and radial coordinate r. After applying $O(\epsilon^2)$ multiple scale theory and converting to dimensional coordinates, we find

$$i\frac{\partial}{\partial z_2}A^* - \beta|A^*|^2A^* + \frac{1}{2k^*}\nabla_\perp^{*2}A^* = -i\frac{\sigma}{2}A^*. \quad (77)$$

By examining Eq. (77) we see that the nonlinearity and diffraction terms have opposite signs. At the given frequency, $\beta > 0$ and the beam self-defocuses.

VI. SUMMARY AND DISCUSSION

MMS is a useful technique that can be applied to physical systems with weak nonlinearities to extract the dominant asymptotic equations. One may proceed to any perturbation order in a self-consistent manner. Using MMS, we were able to derive the nonlinear Schrödinger equation for a single input wave [with $O(1)$ phase mismatch] propagating in a dispersive quadratically nonlinear optical material. We were able to solve the signaling problem for any time-dependent input pulse shape. We assumed three time and distance scales and proceeded to second-order perturbation theory. This resulted in Eq. (48). A correction term due to an $O(\epsilon)$ rectified electric field was shown to come in at third-order perturbation theory, but the term was neglected because of its small size. Therefore Eq. (55) reduced to Eq. (48).

Changing from dimensionless to physical variables, it was shown that the nonlinear Schrödinger equation, Eq. (57), has an effective nonlinear refractive index that is proportional to the second-harmonic coefficient $d^{(2\omega)}$ squared. Therefore self-modulation effects may be observed in materials that generate $\chi^{(2)}$ wave processes. Conditions for soliton propagation in KTP, self-defocusing, and spectral broadening were given.

Our results encourage new experiments to be performed to test the validity of the theory developed by MMS. New technological applications may arise for quadratic nonlinear materials due to the existence of the nonlinear Schrödinger equation. The same multiple scale expansion just presented may be used to analyze multiple input wave problems. If two $O(1)$ input waves at different carrier frequencies and $O(1)$ phase mismatch are propagated in a quadratic material, then coupled nonlinear Schrödinger equations, analogous to cross-phase modulation equations of fiber optics, are obtained at second-order perturbation theory [4].

For $O(1)$ phase mismatch, cubic-type nonlinearities are obtained. On the other hand, if the two $O(1)$ waves have $O(\epsilon)$ phase mismatch, typical three-wave interaction equations are obtained. However, depending on the pulse width, at second-order perturbation theory, group velocity dispersion terms may be included. The equations then take the appearance of coupled nonlinear Schrödinger equations with quadratic nonlinearity which are different from the cross-phase modulation equations. For $O(\epsilon)$ phase mismatch, the quadratic nonlinearity dominates. Thus, depending on the phase mismatch, two input waves may behave as though they were propagating in a cubic or quadratic nonlinear medium.

Under appropriate physical conditions, MMS may be used to show that optical bistability may exist in quadratic nonlinear materials. Second-order perturbation theory shows that the polarization undergoes a hysteretic jump as the field is varied [4]. This problem is analogous to the intrinsic optical bistability in cubic nonlinear materials studied by Haus *et al.* [32].

All problems considered so far assumed the electric fields were far away from material resonances. As the frequency of the electric field approaches a resonance, the MMS expansion breaks down because the wave number k depends strongly on the field amplitude [4]. The behavior is that of a saturation-type nonlinearity. In this asymptotic limit, the nonlinearity is is used to limit the growth of the resonance singularity. Damping is not required. The problem must be analyzed using Whitham's averaged Lagrangian method [23] or Luke's method [33]. These techniques are useful for analyzing the slowly modulated envelopes of waves propagating in media with strong nonlinearity.

MMS may also be used to derive the Boussinesq and the Kortewig–deVries–Burger equation under the appropriate asymptotic limits [34]. Soliton solutions for these equations are for the total fields and propagate at the phase velocity. This is in contrast to the envelope solitons propagating with the group velocity encountered with the Schrödinger equation. It is interesting to note that Eqs. (7) and (14) are analogous to the Boussinesq equation and also have soliton solutions.

MMS is a very general technique that can be applied to physical systems to extract the dominant asymptotic equations for various asymptotic regimes. It provides some alternative results when compared to the conventional SVEA. The results obtained here should provide impetus for further work, in optics, on MMS as a useful and systematic method. The physical theory developed from MMS suggests new signal processing applications of quadratically nonlinear materials.

Note added. Since this article was submitted for publication several papers reporting phase modulation [35, 36] and lensing effects [37] have appeared. All the papers that have appeared to date treat the steady-state situation. Our results cover the dynamic properties of $\chi^{(2)}$ materials using a systematic expansion procedure. The steady-state experiments demonstrate that the effective $\chi^{(3)}$ nonlinear response can be quite large for these materials. In the Introduction we discussed the works of DeSalvo *et al.* [6] and Belashenkov *et al.* [7], which consider boundary-value problems with the steady-state lossless medium framework, whereas our problem is a time-dependent process and has little in common with the steady-state case with efficient phase matching. The outcome of our paper leads to physical phenomena different from these papers, as explained in the Introduction. These papers would more appropriately be discussed in the context of two-wave problems [4]; this will be the topic of a future publication.

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[1] N. Bloembergen, *Nonlinear Optics* (Benjamin/Cummings, New York, 1965).
[2] Y. R. Shen, *The Principles of Nonlinear Optics* (John Wiley and Sons, New York, 1984).
[3] P. N. Butcher and D. Cotter, *The Elements of Nonlinear Optics* (Cambridge University Press, Cambridge, 1990).
[4] A. G. Kalocsai, Ph.D. thesis, Rensselaer Polytechnic Institute, New York, 1991 (unpublished).
[5] A. G. Kalocsai and J. W. Haus, Opt. Commun. **97**, 239 (1993).
[6] J. DeSalvo, D. J. Hagan, M. Shiek-Bahal, G. Stegemen, and E. W. VanStryland, Opt. Lett. **17**, 28 (1992).
[7] N. R. Belashenkov, S. V. Gagarskii, and M. V. Inochkin, Opt. Spektrosk. **66**, 1383 (1989) [Opt. Spectrosc. (USSR) **66**, 806 (1989)].
[8] J. D. Cole, *Perturbation Methods in Applied Mathematics* (Blaisdell, New York, 1968).
[9] P. A. Sturrock, Proc. R. Soc. London Ser. A **242**, 277 (1957).
[10] G. Sandri, Ann. Phys. (N.Y.) **24**, 332 (1963).
[11] D. J. Benney and A. C. Newell, J. Math. Phys. **46**, 133 (1967).
[12] R. K. Dodd, J. C. Eilbeck, J. D. Gibbon, and H. C. Morris, *Solitons and Nonlinear Wave Equations* (Academic Press, New York, 1982).
[13] A. Davey, J. Fluid Mech. **51**, 477 (1972).
[14] H. Washimi and T. Taniuti, Phys. Rev. Lett. **21**, 209 (1966).
[15] V. E. Zakharov, Zh. Eksp. Teor. Fiz. **62**, 1745 (1972) [Sov. Phys.–JETP **35**, 908 (1972)].
[16] T. Kawahara, J. Plasma Phys. **18**, 305 (1977).
[17] A. Hasegawa and F. Tappert, Appl. Phys. Lett. **23**, 142 (1973).
[18] R. H. Stolen and J. E. Bjorkholm, IEEE J. Quantum Electron. **QE-18**, 1062 (1982).
[19] V. E. Zakharov and A. B. Shabat, Zh. Eksp. Teor. Fiz. **61**, 118 (1971) Sov. Phys.–JETP **34**, 62 (1972)].
[20] M. J. Ablowitz and H. Segur, *Solitons and the Inverse Scattering Transform* (SIAM, Philadelphia 1981).
[21] T. Kakutani and N. Sugimoto, Phys. Fluids **17**, 1617 (1974).
[22] T. Taniuti and C. C. Wei, J. Phys. Soc. Jpn. **24**, 941 (1968).
[23] G. D. Whitham, *Linear and Nonlinear Waves* (Wiley, New York, 1974).
[24] A. Hasagawa, *Optical Solitons in Fibers* (Springer-Verlag, New York, 1989).
[25] G. P. Agrawal, *Nonlinear Fiber Optics* (Academic Press, New York, 1989).
[26] V. G. Makhankov, Phys. Rep. **35**, 1 (1978).
[27] A. Yariv, *Quantum Electronics* (John Wiley and Sons, New York, 1989).
[28] J. D. Jackson, *Classical Electrodynamics* (Wiley, New York, 1975).
[29] J. D. Bierlein, SPIE Proc. **1104**, 2 (1989).
[30] J. D. Bierlein and C. B. Arweiler, Appl. Phys. Lett. **49**, 917 (1986).
[31] Y. Silverberg, Opt. Lett. **15**, 1282 (1990).
[32] J. W. Haus, L. Wang, M. Scalora, and C. M. Bowden, Phys. Rev. A **38**, 4043 (1988).
[33] J. C. Luke, Proc. R. Soc. London Ser. A **292**, 403 (1966).
[34] A. G. Kalocsai and J. W. Haus (unpublished).
[35] G. I. Stegeman, M. Sheik-Bahae, E. Van Stryland, and G. Assanto, Opt. Lett. **18**, 13 (1993).
[36] A. E. Kaplan, Opt. Lett. **18**, 1223 (1993).
[37] D. Pierrottet, B. Berman, M. Vannini, and D. McGraw, Opt. Lett. **18**, 263 (1993).