INDUCED SUPERCONTINUUM AND STEEPENING OF AN ULTRAFAST LASER PULSE

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Received 17 June 1985; accepted in revised form 21 October 1985

The problem of the propagation of an intense ultrashort pulse in a cubic (χ^3) nonlinear medium is generalized to include coupling between the primary and second harmonics signals. It is shown that the presence of a strong primary signal induces the superbroadening of the spectrum of a weak second harmonic signal and the deformation of its pulse shape.

The study of the propagation of an ultrashort pulse in a nonlinear medium provides the underlying understanding to the problems of the generation of the supercontinuum [1] and to the distortion of pulses in long optical waveguides [2]. The outstanding experimental feature in both problems is the existence of asymmetry. In the supercontinuum spectrum, the spectral extents on the Stokes and anti-Stokes sides differ by factors of nearly two [3]. In experiments on pulse propagation in long optical fibers, asymmetry in the pulse shape between the leading and trailing edges of the pulse have been observed [4]. In recent publications [5] we considered the propagation of an ultrashort pulse in a nonlinear cubic medium by the method of multiple scales [6]. We derived a set of quasilinear partial differential equations, correct to second order in the nonlinear coupling constant, that interrelates the pulse distortion and the supercontinuum spectral distribution.

In this letter, we extended our formalism to investigate the pulse distortion and supercontinuum generated in a weak probe signal of a second harmonics pulse induced by the presence of a strong primary mode pump signal. The formalism is outlined, the single mode results are reviewed, and the induced pulse shape and generated supercontinuum of the second harmonics are presented.

Nonlinear wave equation. The Maxwell equation in a nonlinear medium, where the dispersion of the index of refraction and its imaginary part are neglected, is given by

$$\nabla^2 E - \frac{n^2}{c^2}\frac{\partial^2 E}{\partial t^2} = \frac{2nn_2}{c^2}\frac{\partial^2}{\partial t^2}\langle E\cdot E\rangle E\,, \qquad (1)$$

where n is the linear index of refraction and n_2 is the nonlinear index of refraction. The wave equation reduces to

$$\frac{\partial^2 E}{\partial z^2} - \frac{1}{v_g^2}\frac{\partial^2 E}{\partial t^2} = \frac{nn_2}{c^2}\frac{\partial^2}{\partial t^2}|E|^2 E\,, \qquad (2)$$

under the assumption that one component of E is present and the transverse variation of E is neglected, and $\langle E\cdot E\rangle = |E|^2/2$.

To reduce the differential equation to a dimensionless form, we introduce the new dimensionless variables Φ, T, Z defined as

$$T = t/\tau_L\,, \quad Z = z/v_g\tau_L\,, \quad E = E_0\Phi\,, \qquad (3)$$

where τ_L is the characteristic time associated with the pulse (pulse width), v_g is the group velocity in the medium, and E_0 is the peak amplitude of the electric field at the entrance plane of the medium. Introducing the dimensionless nonlinear coupling constant ϵ, defined as

$$\epsilon \equiv n_2|E_0|^2/n \qquad (4)$$

the nonlinear wave equation in dimensionless form reduces to

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$$(\partial^2/\partial Z^2 - \partial^2/\partial T^2)\Phi = \epsilon(\partial^2/\partial T^2)|\Phi|^2\Phi . \qquad (5)$$

The functional dependence of Φ on Z, T and ϵ, in the solution of (5), is not disjoint. To first order in ϵ, Φ depends on the combinations of ϵT and ϵZ as well as the individual T, Z and ϵ. Carrying the perturbation to higher orders, Φ depends additionally on $\epsilon^2 T$, $\epsilon^2 Z$, $\epsilon^3 T$, $\epsilon^3 Z$, ... Hence it is convenient to write Φ as $\Phi(Z_0, T_0, Z_1, T_1, Z_2, T_2, ...; \epsilon)$ where the new scaled variables Z_1, T_1, Z_2, T_2 etc. are defined as $T_n = \epsilon^n T$ and $Z_n = \epsilon^n Z$. Next, we seek a uniform expansion solution to Φ in the form

$$\Phi = \Phi_0 + \epsilon\Phi_1 + \epsilon^2\Phi_2 + ... , \qquad (6)$$

where the Φ_n are expressed as functions of the scaled variables.

Substituting (6) in (5) and using the scaled variables, the terms multiplying ϵ^0, ϵ^1, and ϵ^2 give the following equations:

$$(\partial^2/\partial Z_0^2 - \partial^2/\partial T_0^2)\Phi_0 = 0 , \qquad (7)$$

$$(\partial^2/\partial Z_0^2 - \partial^2/\partial T_0^2)\Phi_1 + 2(\partial^2/\partial Z_1\partial Z_0 - \partial^2/\partial T_1\partial T_0)\Phi_0 = (\partial^2/\partial T_0^2)|\Phi_0|^2\Phi_0 , \qquad (8)$$

$$(\partial^2/\partial Z_0^2 - \partial^2/\partial T_0^2)\Phi_2 + 2(\partial^2/\partial Z_1\partial Z_0 - \partial^2/\partial T_1\partial T_0)\Phi_1 + (\partial^2/\partial Z_1^2 - \partial^2/\partial T_1^2 + 2\partial^2/\partial Z_0\partial Z_2 - 2\partial^2/\partial T_0\partial T_2)\Phi_0 = 2(\partial^2/\partial T_1\partial T_0)|\Phi_0|^2\Phi_0 + (\partial^2/\partial T_0^2)|\Phi_1|^2\Phi_1 . \qquad (9)$$

In what follows it is convenient to use a new coordinate system which is moving with the pulse. Denoting the new coordinates by U_n and V_n, where $U_n = Z_n - T_n$ and $V_n = Z_n$, the above equations can respectively be written in the new variables as:

$$\left(\frac{\partial^2}{\partial V_0^2} + 2\frac{\partial}{\partial U_0}\frac{\partial}{\partial V_0}\right)\Phi_0 = 0 , \qquad (7a)$$

$$\left(\frac{\partial^2}{\partial V_0^2} + 2\frac{\partial}{\partial U_0}\frac{\partial}{\partial V_0}\right)\Phi_1 + 2\left(\frac{\partial}{\partial U_1}\frac{\partial}{\partial V_0} + \frac{\partial}{\partial U_0}\frac{\partial}{\partial V_1} + \frac{\partial}{\partial V_1}\frac{\partial}{\partial V_0}\right)\Phi_0 = \frac{\partial^2}{\partial U_0^2}|\Phi_0|^2\Phi_0 , \qquad (8a)$$

$$\left(\frac{\partial^2}{\partial V_0^2} + 2\frac{\partial}{\partial U_0}\frac{\partial}{\partial V_0}\right)\Phi_2 + 2\left(\frac{\partial}{\partial U_1}\frac{\partial}{\partial V_0} + \frac{\partial}{\partial U_0}\frac{\partial}{\partial V_1} + \frac{\partial}{\partial V_1}\frac{\partial}{\partial V_0}\right)\Phi_1 + \left(\frac{\partial^2}{\partial V_1^2} + 2\frac{\partial}{\partial U_1}\frac{\partial}{\partial V_1} + 2\frac{\partial}{\partial U_0}\frac{\partial}{\partial V_2} + 2\frac{\partial}{\partial U_2}\frac{\partial}{\partial V_0} + 2\frac{\partial}{\partial V_2}\frac{\partial}{\partial V_0}\right)\Phi_0 = 2\frac{\partial}{\partial U_1}\frac{\partial}{\partial U_0}|\Phi_0|^2\Phi_0 + \frac{\partial^2}{\partial U_0^2}|\Phi_1|^2\Phi_1 . \qquad (9a)$$

These equations will serve as the basis for obtaining specific solutions to physical problems under consideration.

Single-mode. In this section we review the results for the pulse distortion and the spectral extent of the generated supercontinuum for an ultrashort pulse propagating in the medium.

The incoming pulse is described by

$$\Phi_{in} = f(U)\exp(iKU) , \quad \text{where } f(U) = \mathrm{sech}(U) . \qquad (10)$$

f is the pulse form function, which we will assume for simplicity to be hyperbolic secant, and $W = \omega\tau = K = k/v_g\tau$.

Eqs. (7a), (8a) and (9a) can be solved through the ansatz

$$\Phi_0 = A(U_1, V_1, U_2, V_2)\, e^{iKU_0} , \quad \Phi_1 = 0 ,$$

$$\Phi_2 = \sum_n E_n(U_1, V_1, U_2, V_2)\, e^{inKU_0} . \qquad (11)$$

The specific form of E_n cannot be obtained from the above equations, however for cases under consideration, where $\epsilon < 1$, Φ can be adequately approximated by Φ_0.

Denoting A by

$$A \equiv a\,\mathrm{e}^{\mathrm{i}\alpha}\,, \tag{12}$$

where a is the amplitude and α is the phase of the pulse, a and α are functions of (U_1, V_1, U_2, V_2). The quasilinear partial differential equations deduced from the set (7a), (8a), (9a) are given by

$$\partial a/\partial V - \tfrac{3}{2}\epsilon a^2 \partial a/\partial U = 0\,, \tag{13}$$

$$\partial\alpha/\partial V - \tfrac{1}{2}\epsilon a^2 \partial\alpha/\partial U = \tfrac{1}{2}\epsilon K a^2 - \tfrac{1}{8}\epsilon^2 K a^4\,. \tag{14}$$

The solution of (13) with the boundary condition (10) is given by [2]

$$a^2 = \mathrm{sech}^2(U + \tfrac{3}{2}\epsilon V a^2)\,, \tag{15}$$

where $\epsilon V = n_2|E_0|^2 Z/c\tau_\mathrm{L}$. This solution derived under the condition of no dispersion and no absorption is valid for all values of $V < V_\mathrm{crit}$, where V_crit is the value for which $\partial a/\partial U$ is not finite. For values of $V > V_\mathrm{crit}$, the above solution needs to be smoothed at the discontinuity. The value of V_crit is given by

$$\epsilon V_\mathrm{crit} \sim \tfrac{1}{2}\sqrt{3} \approx 0.866\,. \tag{16}$$

The spectral distribution for the signal at the exit plane of the medium is given by

$$S(\omega', z_\mathrm{e}) \propto |\tilde{E}(\omega', z_\mathrm{e})|^2 \tag{17}$$

where $\tilde{E}(\omega', z_\mathrm{e})$ is the time Fourier transform of the electric field at $z = z_\mathrm{e}$, the exit plane,

$$\tilde{E}(\omega', z_\mathrm{e}) = \frac{1}{2\pi}\,\mathrm{Re}\int \mathrm{e}^{\mathrm{i}\omega' t} E(t, z_\mathrm{e})\,\mathrm{d}t\,. \tag{18}$$

In the U–V coordinates, where $W = \omega\tau$ and $W' = \omega'\tau$, the expression for $\tilde{E}$ is given by

$$\tilde{E}(W', V_\mathrm{e}) = \frac{E_0\tau}{2\pi}\,\mathrm{Re}\int_{-\infty}^{\infty} \mathrm{d}U\,\mathrm{e}^{\mathrm{i}(W-W')U} \times a(U, V_\mathrm{e})\,\mathrm{e}^{\mathrm{i}\alpha(U, V_\mathrm{e})}\,, \tag{19}$$

where V_e corresponds to the V of the exit plane.

The extent of the spectral distribution can be estimated through the method of stationary phase, specifically:

$$W' - W|_\text{anti-Stokes} \sim \mathrm{Max}(\partial\alpha/\partial U)_V\,,$$

$$W' - W|_\text{Stokes} \sim \mathrm{Min}(\partial\alpha/\partial U)_V\,. \tag{20}$$

The general solution of α satisfying eq. (14) is given by

$$\alpha = -KU - \tfrac{1}{2}K\epsilon \int_0^U a^2(p, V)\,\mathrm{d}p - \tfrac{3}{8}K\epsilon^2 \int_0^V a^4(0, q)\,\mathrm{d}q + KF(U, V)\,, \tag{21}$$

where the function $F(U, V)$ is a solution of the equation

$$\partial F/\partial V - \tfrac{1}{2}\epsilon a^2 \partial F/\partial U = 0\,. \tag{22}$$

The general solution of (22) is given by

$$F(f) = F\left(\frac{\epsilon}{2}\int_0^V a^3(U, q)\,\mathrm{d}q + \int_0^U a(p, 0)\,\mathrm{d}p\right)\,, \tag{23}$$

and the particular solution of α satisfying the boundary condition $\alpha(U, 0) = 0$ is written as

$$\alpha = -KU - \tfrac{1}{2}K\epsilon \int_0^U a^2(p, V)\,\mathrm{d}p - \tfrac{3}{8}K\epsilon^2 \int_0^V a^2(0, q)\,\mathrm{d}q + K\tanh^{-1}[\sin f(U, V)] + \tfrac{1}{2}K\epsilon \sin f(U, V)\,, \tag{24}$$

where

$$f(U, V) = \tfrac{1}{2}h(U, V) + \sin^{-1}\tanh U\,,$$

$$h(U, V) = \epsilon \int_0^V a^3(U, q)\,\mathrm{d}q\,.$$

The corresponding expression for the frequency sweep $\partial\alpha/\partial U$ is given by

$$\partial\alpha/\partial U = -K - \tfrac{1}{2}K\epsilon a^2(U, V) + Ka(U, V)[\cos f(U, V)]^{-1} + \tfrac{1}{2}K\epsilon a(U, V)\cos f(U, V)\,. \tag{25}$$

Primary and second harmonics coupling. In this section we consider the case of two pulses with frequency distribution centered respectively at ω and 2ω simultaneously entering the medium. The boundary condition at the entrance plane is [‡1]

$$\Phi_{\text{in}} = \operatorname{sech} U\, e^{iKU} + \delta \operatorname{sech}(1.76U)\, e^{2iKU}\,, \tag{26}$$

where δ measures the relative strength of the second-harmonic signal to the primary frequency signal.

Eqs. (7a), (8a) and (9a) can be solved for this case through

$$\Phi_0 = \tilde{A}(U_1, V_1, U_2, V_2)\, e^{iKU_0} + \delta B(U_1, V_1, U_2, V_2)\, e^{2iKU_0}\,,$$

$$\Phi_1 = C(V_0, U_1, V_1, U_2, V_2)\, e^{3iKU_0}\,,$$

$$\Phi_2 = \sum_n D_n(U_1, V_1, U_2, V_2)\, e^{niKU_0}\,, \tag{27}$$

where A, B and C are the complex amplitudes associated with the ω, 2ω and 3ω signals respectively. Assuming the velocity of propagation of the wave to be the same for ω and 2ω in the medium, i.e., we are neglecting more dispersion terms, the quasilinear partial differential equations satisfied by $\tilde{a}$, $\tilde{\alpha}$, b, and β defined by $\tilde{A} = \tilde{a}\, e^{i\tilde{\alpha}}$ and $B = b\, e^{i\beta}$, are given by [‡2]

$$\partial a/\partial V - \tfrac{1}{2}\epsilon[(3\tilde{a}^2 + 2\delta^2 b^2)\partial a/\partial U + 4\delta^2 \tilde{a} b \partial b/\partial U] = 0\,, \tag{28}$$

$$\partial\tilde{\alpha}/\partial V - \tfrac{1}{2}\epsilon(\tilde{a}^2 + 2\delta^2 b^2)\partial\tilde{\alpha}/\partial U = \tfrac{1}{2}K\epsilon(\tilde{a}^2 + 2\delta^2 b^2) - \tfrac{1}{8}\epsilon^2 K(\tilde{a}^2 + 2\delta^2 b^2)^2\,, \tag{29}$$

$$\partial b/\partial V - \tfrac{1}{2}\epsilon[(2\delta^2 b^2 + 2\tilde{a}^2)\partial b/\partial U + 4ab\partial\tilde{a}/\partial U] = 0\,, \tag{30}$$

$$\partial\beta/\partial V - \tfrac{1}{2}\epsilon(\delta^2 b^2 + 2\tilde{a}^2)\partial\beta/\partial U = K\epsilon(2\tilde{a}^2 + \delta^2 b^2) - \tfrac{1}{4}\epsilon^2 K(\delta^2 b^2 + 2\tilde{a}^2)^2\,. \tag{31}$$

In the limit $\delta^2 \ll 1$, the above equations reduce to

(i) for the strong wave

$$\partial\tilde{a}/\partial V - \tfrac{3}{2}\epsilon\tilde{a}^2\partial\tilde{a}/\partial U = 0\,, \tag{28a}$$

$$\partial\tilde{\alpha}/\partial V - \tfrac{1}{2}\epsilon\tilde{a}^2\partial\tilde{\alpha}/\partial U = \tfrac{1}{2}K\epsilon\tilde{a}^2 - \tfrac{1}{8}K\epsilon^2\tilde{a}^4\,, \tag{29a}$$

(ii) and for the weak wave

$$\partial b/\partial V - \epsilon\tilde{a}^2\partial b/\partial U - 2\epsilon\tilde{a}b\partial\tilde{a}/\partial U = 0\,, \tag{30a}$$

$$\partial\beta/\partial V - \epsilon\tilde{a}^2\partial\beta/\partial U = 2K\epsilon\tilde{a}^2 - \epsilon^2 K\tilde{a}^4\,. \tag{31a}$$

Note that both the amplitude and the phase of the weak pulse are driven by the amplitude of the strong wave. Eqs. (28a) and (29a) are identical to (13) and (14) and consequently $\tilde{a} = a$ and $\tilde{\alpha} = \alpha$.

The equations which determine C [defined in (27)] have their source terms proportional to δ^2. The initial condition for C is $C(U, 0) = 0$. In the limit $\delta^2 \ll 1$, $C = 0$ is then the solution.

The general solutions for (30a) and (31a) are given by

$$b(U, V) = L_1(U, V)a^4(U, V) \tag{32}$$

and

$$\beta(U, V) = -2KU + 4\epsilon K\int_0^U a^2(p, V)\,dp + 3K\epsilon^2\int_0^V a^4(0, q)\,dq + KL_2(U, V)\,, \tag{33}$$

and where L_1 and L_2 satisfy the partial differential equation

$$\partial L_i/\partial V - \epsilon a^2\partial L_i/\partial U = 0\,. \tag{34}$$

The solution of (34) is given by:

$$L(U, V) = L(l) = L\left(\epsilon\int_0^V a^6(U, q)\,dq + \int_0^U a^4(p, 0)\,dp\right) = L(g(U, V) + \tanh U - \tfrac{1}{3}\tanh^3 U)\,, \tag{35}$$

where $g(U, V) = \epsilon\int_0^V a^6(U, q)\,dq$. The particular solutions for L_1 and L_2 satisfying the boundary conditions (26) and $\beta(U, 0) = 0$ are given respectively by the following parametric representation (where s is the parameter):

$$L_1(x) = \cosh^4 s\, \operatorname{sech}(1.76s)\,,$$

[‡1] The incoming primary mode pulse form function is assumed to be given by sech(U) and the incoming second harmonic pulse form function is assumed to be given by sech(1.76U). See for example, ref. [7].

[‡2] The solutions presented here correspond to a nonlinear index of refraction $\delta n_{\text{NL}} = n^{(1)} + n^{(2)}\cos\omega t$. If $\delta n_{\text{NL}} = n^{(1)}$, the solution, for $\delta \ll 1$, is $\beta = 2\alpha$.

$$L_2(x) = 2s - 4\epsilon \tanh s\,, \quad x = \tanh s - \tfrac{1}{3}\tanh^3 s\,. \tag{36}$$

Using the above relations $b(U, V)$ is plotted in fig. 1 for different $\epsilon V = n_2 E_0^2 z / C\tau_L$. Notice that the induced steepening of the probe (weak signal) depends on the intensity ($\propto E_0^2$) of the strong wave.

The frequency extent of the supercontinuum centered at 2ω is given by the maximum and the minimum of $(1/2K)\partial\beta/\partial U$, where

$$(1/2K)\partial\beta/\partial U = 1 + 2\epsilon a^2(U, V) + \tfrac{1}{2}[\partial L_2(l)/\partial l]\, a^4(U, V)\,, \tag{37}$$

and L_2 in parametric form is

$$\frac{\partial L_2}{\partial x} = \frac{2 - 4\epsilon\,\mathrm{sech}^2 s}{\mathrm{sech}^2 s - \tanh^2 s\,\mathrm{sech}^2 s}\,. \tag{38}$$

The frequency extents for the induced supercontinuum are plotted from eq. (37) in fig. 2 as a function of ϵV.

It is worth noting that for $\epsilon V \ll 1$,

$$\beta/2K = 2(\alpha/K) \sim \mathrm{sech}^2 U\, (\epsilon V)\,, \tag{39}$$

which is the result of traditional SPM [1]. For larger ϵV the induced frequency extents grow faster than the primary pulse frequency extents.

In this letter, we derived the quasilinear partial differential equations that describe the propagation of a pulse in a non-linear cubic medium, and the effects of a strong primary signal on a weak second harmonics signal propagating in the same medium. In our calculations, we made no assumptions on the relative magnitude of the derivative of the amplitude versus the phase derivative as found in Yang and Shen [8], and the derivatives from all terms in (1) are consistently retained at each order of the expansion contrary to the SPM traditional calculation [1] and the slowly varying amplitude (SVA) [2] approximation where the contributions from the cubic term are not consistently treated. (See table 1 for the equations.) The physical results that emanate from these calculations are that (i) the pulse shape is distorted producing an asymmetry skewed towards the trailing edge; (ii) the pulse distortion and the spectral asymmetry can be inter-related, and there is no need to invoke contributions from plasmas and/or the time response of the nonlinear index of refraction to explain the Stokes/anti-Stokes asymmetry; (iii) the primary strong pulse

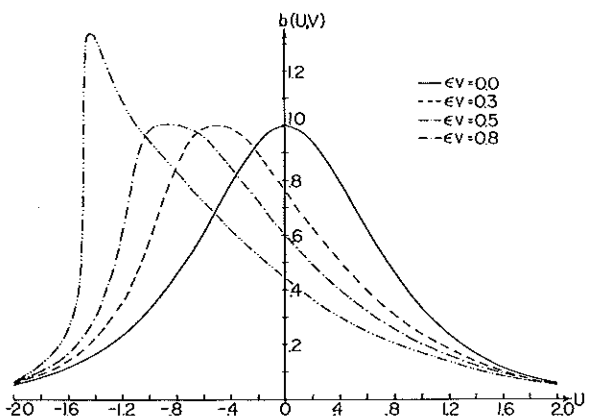

Fig. 1. Induce shape of a second harmonic pulse due to the presence of an intense primary mode pulse ($\epsilon V = n_2 |E_0|^2 z / C\tau_L$).

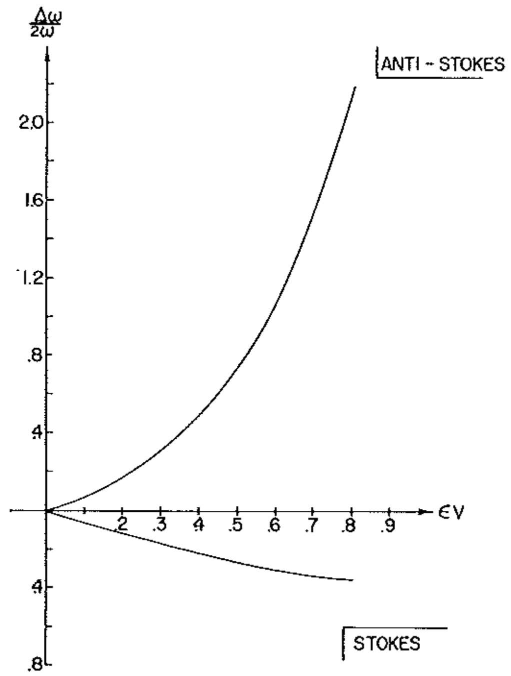

Fig. 2. Induced Stokes and anti-Stokes spectral extents for a second harmonic pulse due to the presence of an intense primary mode pulse.

Table 1
Quasi-linear partial differential equations for the amplitude and phase of a pulse propagating in a cubic nonlinear medium.

	Amplitude equation	Phase equation
Traditional SPM theory	$\partial a/\partial V = 0$	$\partial \alpha/\partial V = \frac{1}{2}\epsilon K a^2$
Slowly varying amplitude	$\partial a/\partial V - 3\epsilon a^2 \partial a/\partial U = 0$	$\partial \alpha/\partial V - \epsilon a^2 \partial \alpha/\partial U = \frac{1}{2}\epsilon K a^2$
Yang and Shen	$\partial a/\partial V - \frac{1}{2}\epsilon a^2 \partial a/\partial U = 0$	$\partial \alpha/\partial V - \frac{1}{2}\epsilon a^2 \partial \alpha/\partial U = \frac{1}{2}\epsilon K a^2$
This paper	$\partial a/\partial V - \frac{3}{2}\epsilon a^2 \partial a/\partial U = 0$	$\partial \alpha/\partial V - \frac{1}{2}\epsilon a^2 \partial \alpha/\partial U = \frac{1}{2}\epsilon K a^2 - \frac{1}{8}\epsilon^2 K a^4$

induces an index of refraction in the medium that modulates the second harmonics weak pulse. Specifically, the mixed signal input of eq. (26) produced by combining the primary signal with the output from a second harmonic generator [7] coupling in the nonlinear medium produces an output signal eqs. (32) and (37) for the second harmonics portion distorted and modulated by the distortion of the primary pulse confirming the picture of the induced index of refraction created by the primary pulse. The results obtained are large and should be experimentally observable.

We thank Jacques Beneviste for discussions and we acknowledge the partial support of the National Science Foundation (NSF-84 13144) for this research.

Note added. Recent experiments conducted by our group [9] gave data in agreement with eq. (25) in particular, the observed asymmetry in the supercontinuum of the single mode fits the theoretically predicted values. On-going experiments, using an intense primary beam with a weak second harmonic beam greatly enhanced the supercontinuum of the secondary beam giving intensities ten times larger than those observed for the single mode secondary beam.

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