Solutions of nonlinear-optics equations found outside the approximation of slowly varying amplitudes and phases

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A class of new nonlinear effects in the interaction of an electromagnetic field with a medium has been established: the existence of "half-wave" electromagnetic-field solitons, which is induced by a field of "Josephson" currents, which effectively multiply the light frequency.

1. Progress[1] in methods for forming light pulses a few wavelengths long (as short as a single wavelength) with power densities of 10^9–10^{18} W/cm^2 has opened up some completely new topics in the optics of nonlinear media. The methods of traditional nonlinear optics—the method of slowly varying amplitudes and phases for the field and constitutive variables of the medium[2,3]—are becoming ineffective in describing wave processes at such small spatial and temporal scales and at such high fields. We are reporting here the solution of several equations for pulse propagation in nonlinear media in situations in which the method of slowly varying amplitudes and phases is inapplicable. We show, in particular, that the equations of nonlinear optics permit the existence of some new wave entitles: unipolar pulses which might be called "pulses half a wavelength long" by analogy with the terminology used in the description of bipolar Čerenkov pulses.[1] In contrast with the corresponding pulses in linear media, in which their "lifetime" is limited by the dispersive-spreading length, these are stable entities. One might say that these local concentrations of electromagnetic field in a nonlinear medium are endowed with the properties of particles to a large extent.

To describe the evolution of the pulse, we use the equations for the density matrix of the medium, [2] with the components ρ_{mn} (m and n specify the levels of the system), and the wave equation for the field ϵ:

$$\text{curl curl}\,\vec{\epsilon} + \frac{1}{c^2}\frac{\partial^2\vec{\epsilon}}{\partial t^2} = -\frac{4\pi}{c^2}\frac{\partial^2\vec{\mathcal{P}}}{\partial t^2}\ . \tag{1}$$

The polarization of the medium, $\vec{\mathcal{P}}$, is related to the matrix elements of the dipole moments, μ_{mn}, and the particle density N by $\vec{\mathcal{P}} = N\Sigma_{m,n}\,\rho_{mn}\vec{\mu}_{nm}$. The equations for the density matrix ρ_{mn} contain the frequencies (ω_{mn}) of transitions between levels, but not terms with relaxation constants, which we are omitting. In solving (1), we formulate the problem in a very simple way: An electromagnetic wave with the field which is independent of the coordinates y and z and with a polarization $\vec{e}$ along the z axis is propagating along the x direction in a slab $|z| < a$. The space $|z| > a$ is filled by a medium with a conductivity $\sigma = \infty$.

2. We assume that the following condition holds for the time scale of the variation of the pulse field, τ_p:

$$\tau_p^{-1} \ll \omega_{mn} \qquad (m \neq n). \tag{2}$$

This condition means that the variation of $\epsilon(t)$ is slow in comparison with the characteristic atomic times. In other words,the pulse interacts only weakly with the system and in this sense is in a transparency region of the nonliner medium. We then write the solution for ρ_{mn} as a series in the small parameter $(\tau_p \omega_{mn})^{-1}$. For fields which are weak in comparison with $\epsilon_a \sim 10^9$ V/cm, we can find an explicit expression for the expansion by working from integral representations of the linear and nonlinear components of ρ_{mn} (Ref. 2). If the vectors $\vec{\epsilon}$ and $\vec{\mathscr{P}}$ are collinear, we find the following equation from (1) for a "quadratic" medium:

$$\frac{\partial \epsilon}{\partial x} + c_2 \epsilon \frac{\partial \epsilon}{\partial \tau} + c_1 \frac{\partial^3 \epsilon}{\partial \tau^3} = 0. \tag{3}$$

For a "cubic" medium we find the equation

$$\frac{\partial \epsilon}{\partial x} + c_3 \epsilon^2 \frac{\partial \epsilon}{\partial \tau} + c_1 \frac{\partial^3 \epsilon}{\partial \tau^3} = 0. \tag{4}$$

Since the dispersion and the nonlinearity are slight in this case, we can simplify wave equation (1) and describe the wave process by moving along the characteristic $\tau = t - x/v$. All the parameters of Eqs. (3) and (4) are determined by the low-frequency limit of the linear $(\chi^{(1)})$ and nonlinear $(\chi^{(2),(3)})$ susceptibilities of the medium:[1)]

$$v = c(1 + 4\pi\chi^{(1)}(0))^{-1/2}, \qquad c_1 = \frac{\pi v}{c^2} \frac{\partial^2 \chi^{(1)}(\omega)}{\partial \omega^2} \Big|_{\omega = 0}, \qquad c_2 = \frac{4\pi v}{c^2} \chi^{(2)}(0),$$

$$c_3 = \frac{6\pi v}{c^2} \chi^{(3)}(0). ^{1)}$$

Equation (3) is the Korteweg–de Vries equation, and Eq. (4) is a modification of it. Both fall in the category of nonlinear evolutionary equations which can be integrated by the method of the inverse scattering problem.[5] The simplest solution of (3) is a unipolar soliton

$$\epsilon(x, \tau) = \epsilon_0 \operatorname{sech}^2 \left[\frac{\tau - x/v_p}{\tau_p} \right], \tag{5}$$

whose velocity is $v_p = 3(\epsilon_0 c_2)^{-1}$, and whose length is $\tau_p = (12 c_1 / \epsilon_0 c_2)^{1/2}$.

Equation (4) has soliton solutions only in the case sign $(c_1 c_2) = 1$. The simplest of these solutions is again a unipolar pulse:

$$\epsilon(x, \tau) = \epsilon_0 \operatorname{sech} \left[\frac{\tau - x/v_p}{\tau_p} \right], \tag{6}$$

where $v_p = 6(c_3 \epsilon_0^1)^{-1}$ and $\pi_p = \epsilon_0^{-1}(6 c_1 / c_3)^{1/2}$.

3. Another limiting case in the interaction of the pulse with the medium is associated with the situation which is the direct opposite of that which we have been discussing. We assume that two or more levels, which are well-separated from other levels, can be distinguished in a quantum-mechanical system.[2)] For this group, the following condition holds:

$$\tau_p^{-1} \gg \omega_{mn}. \tag{7}$$

In this interaction regime, the state of the medium may change markedly, so the response cannot be written as an expansion in a small parameter. Let us examine this point in the example of a two-level system. From the equations for the density matrix for $m, n = 1,2$, we find the following system of equations:

$$\frac{\partial^2 P}{\partial t^2} + \omega_{21}^2 P = -\omega_{21}\frac{\mu_{12}}{\hbar} n\epsilon, \qquad \frac{\partial n}{\partial t} = -4\frac{\mu_{12}}{\hbar}\frac{\partial n}{\partial t}\epsilon, \tag{8}$$

where $P = \mathrm{Re}\,\rho_{12}$, and $n = \rho_{22} - \rho_{11}$. According to (7), we can eliminate the term $\omega_{21}^2 P$ from the first equation of system (8). It is then an elementary matter to solve (8). In particular, the current density induced by the field is

$$j = \frac{\partial \mathcal{P}}{\partial t} = j_c \sin\Psi, \qquad j_c = N\mu_{12}\omega_{21}n_0 . \tag{9}$$

Wave equation (1), which cannot be simplified in this very nonlinear case, reduces to a sine-Gordon equation for the phase $\Psi(t) = (2\mu_{12}/\hbar)\int_{-\infty}^{t}\epsilon dt$:

$$\frac{\partial^2\Psi}{\partial x^2} - \frac{1}{c^2}\frac{\partial^2\Psi}{\partial t^2} = -n_0\frac{\Omega^2}{c^2}\sin\Psi, \qquad \Omega^2 = \frac{8\pi N\mu_{12}^2\omega_{21}}{\hbar}. \tag{10}$$

In the case of an absorbing medium ($n_0 = -1$), Eq. (10) has soliton solutions, the simplest of which describes a unipolar 2π pulse:

$$\epsilon(x,t) = \frac{\hbar}{\mu_{12}\tau_p}\,\mathrm{sech}\left[\frac{t - x/v_p}{\tau_p}\right], \qquad \frac{1}{v_p^2} = \frac{1}{c^2}(1 + \Omega^2\tau_p^2). \tag{11}$$

In an amplifying medium ($n_0 = +1$), Eq. (10) has the self-similar solution[6]

$$\epsilon(x,t) = xF\left(\frac{2\Omega^2}{c}x\left(t - \frac{x}{c}\right)\right),$$

where $F(z)$ is a variable-sign function of the wave-packet type. As the pulse propagates, its frequency $\omega(x) = (2\Omega^2/c)x$ shifts ($\sim x$) in the blue direction. The number of photons in the pulse does not change as the pulse is amplified.

It is pertinent to note the interaction of a short pulse with a Raman-active medium. In this case condition (7) holds: The pulse length τp is shorter than the time (ω_{21}^{-1}) of the oscillation of a stimulated-Raman scattering center. Discarding the term with the restoring force proportional to ω_{21}^2 in the equation for the Raman-scattering oscillator, we find an expression for the difference between the populations of the vibrational levels in the two-level approximation[4]: $n(t) = -\cos\Psi(t)$, where

$\Psi(t) = \int_{-\infty}^{t} \alpha\epsilon^2 dt$, and the coefficient α is determined by the parameters of the scattering center. Even without looking at the equations for the field, we can say that a pulse propagating through a stimulated-Raman-active medium may be a 2π pulse of a self-induced transparency $[\Psi(\infty) = 2\pi]$: At the leading edge, the energy of the pulse decreases because of the excitation of Stokes radiation, while at the trailing edge the anti-Stokes components restore energy to the pulse field.

A condition equivalent to (7) can be written in the form $\mu_{12}\epsilon_0/\hbar \gg \omega_{21}$, where $\epsilon_0 = \hbar/\mu_{12}\tau_p$ is a characteristic value of the field for pulse (11) (in this particular case, it is the maximum field). For a monochromatic wave with an amplitude E_0 and a frequency $\omega \sim \omega_{21}$, the condition under which the expression for j as a function of ϵ in (9) is applicable to the condition that ω_{21} be small in comparison with the Rabi frequency $\mu_{12}E_0/\hbar$.

4. Let us examine expression (9) for the current in more detail. At small values of $\Psi(\sin\Psi \approx \Psi)$, expression (9) becomes the London equation, which relates the current density j to the field ϵ in a superconductor. In general, the current given by (9) refers to a Josephson current; for example, a current of this sort is induced by a pulse propagating through a planar superconducting structure.[7] A nonlinearity of the Josephson type is unique in frequency multiplication,[3)] so it is interesting to note the efficiency of the harmonic generation by the current:

$$j = j_c \sin\left[\frac{2\mu_{12}}{\hbar\omega} E_0 \sin(\omega t - kx)\right] = j_c \sum_n J_n\left(\frac{2\mu_{12}E_0}{\hbar\omega}\right) \sin(n(\omega t - kx)) .$$

This current is induced by a given field $\epsilon = E_0 \cos(\omega t - kx)$. In contrast with the case of harmonic generation under conditions such that the method of slowly varying amplitudes and phases is applicable, and interactions which build up can be realized for a fixed number of waves (usually no more than two or three), the current in (9) can excite $\sim 10^2$–10^3 harmonics simultaneously, with comparable amplitudes. The reason for this circumstance is that the coefficients of an expansion of j in harmonics are the Bessel functions $J_n(z)$, and for certain values of the parameters n and $z = 2\mu_{12}E_0/\hbar\omega$ these coefficients fall off extremely slowly. At $z \sim n \gg 1$, for example, the asymptotic behavior is $J_n(z) \approx 0.67n^{-1/3}$.

5. In summary, if we refrain from using the approximation of slowly varying amplitudes and phases, and if we describe the wave processes in terms of real fields, we are presented with a large class of new nonlinear phenomena. The present state of the art in laser technology is quite adequate for experimental observation of these phenomena. To form solitons with a length $\tau_p \sim 10^{-14}$s in crystals of the $AgGaS_2$ type, with a large nonlinearity and a wide transparency window,[1] for example, we would need a power density $\sim 10^{11}$ W/cm^2 from a femtosecond CO_2 laser. Producing a Josephson current as in (9) in the optical range (and describing the field propagation on the basis of the sine-Gordon equation) would require power densities $\sim 10^{12}$–10^{13} W/cm^2. At $\sim 10^{14}$ W/cm^2, it would become possible to generate the tenth harmonic of a neodymium laser, i.e., light with a photon energy $\hbar\omega \sim 10$ eV.

[1)]It can be shown that the nonlinearity parameters $\chi^{(2)}(0)$ and $\chi^{(3)}(0)$ are related to the measurable nonlinear susceptibilities of the medium in the following way: $\chi^{(2)}(0) = 2\chi^{(2)}(0,\omega,-\omega)|_{\omega=0}$

$= 2\chi^{(2)}(2\omega,\omega,\omega)|_{\omega} = 0; \ \chi^{(3)}(0) = \frac{4}{3}\chi^{(3)}(3\omega,\omega,\omega,\omega)|_{\omega=0} = 4\chi^{(3)}(\omega,\omega,\omega,-\omega)|_{\omega} = 0.$

2) An example of such a system might be a group of vibrational sublevels of an electronic state of a molecule.

3) In frequency multiplication at a Josephson junction, for example, it has been found possible to generate harmonics up to the thousandth.[8]

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