Long Wave–Short Wave Resonance in Nonlinear Negative Refractive Index Media

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We show that long wave–short wave resonance can be achieved in a second-order nonlinear negative refractive index medium when the short wave lies on the negative index branch. With the medium exhibiting a second-order nonlinear susceptibility, a number of nonlinear phenomena such as solitary waves, paired solitons, and periodic wave trains are possible or enhanced through the cascaded second-order effect. Potential applications include the generation of terahertz waves from optical pulses.

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In recent years, negative refractive index media [1] under the influence of nonlinearities have stimulated great interest. Areas of detailed theoretical studies are metamaterials in a positive nonlinear dielectric [2], second harmonic generation [3], generalized nonlinear Schrödinger equation [4], solitons [5,6], nonlinear susceptibilities, and three- and four-wave mixing and their applications [7]. Experimental realization of second harmonic generation in a magnetic metamaterial was also recently demonstrated [8]. Nonlinear metamaterials in the optical regime are very attractive owing to the availability of high intensity light sources. Significant experimental progress has been made in the optical regime with a variety of structures and negative index wavelengths [9].

In this Letter, we present the nonlinear phenomena of long wave–short wave (LWSW) resonance in negative refractive index media. Long wave–short wave resonance occurs when the group velocity of a short wave (high-frequency wave) is equal to the phase velocity of a long wave (low-frequency wave). This resonance can be satisfied when the short wave lies on the negative branch of the dispersion curve. We will see that the governing nonlinear equations of the wave amplitudes are driven by a ponderomotive force [10]. However, our work introduces the concept of using a second-order nonlinearity for efficient resonant coupling, which is different from past work done in the area of slow light where the ponderomotive force alone is the local nonlinearity [11,12]. The source of the second-order nonlinearity in a metamaterial may be from the background dielectric or from the inherent response of the negative index medium [7]. We show that the governing nonlinear equations have a number of interesting solutions depending on the effective material parameters of the medium. The solutions include solitary waves, paired solitons, and periodic wave trains. A potential application of such a resonance is the generation of terahertz waves from an input optical wave.

The general theory describing the interaction between long waves and short waves was first formalized by Benney [13], but these and related equations appeared earlier in plasma physics in studies of nonlinear Langmuir wave phenomena [14,15]. In the field of fluid mechanics, we refer to research on capillary-gravity and long gravity waves [16]. Newell [17] studied a broader class of nonlinear wave equations that are similar to the LWSW equations and identified those that can be solved via the inverse scattering technique. Explicit wave solutions of the LWSW equations, including solitons, have been derived and studied by Ma [18], and Ma and Redekopp [19]. Finally, Dodd *et al.* [20], and Moloney and Newell [21] present expansion methods that can be used to derive and solve LWSW equations.

Our analysis of nonlinear wave activity in negative index media focuses on one-dimensional wave propagation parallel to the z axis of an (x, y, z) Cartesian coordinate system with the electric field directed parallel to the x axis. The time domain nonlinear wave equation that governs the electric field $E(z, t)$ is given by

$$\begin{aligned} LE \equiv \frac{\partial^2 E}{\partial z^2} - \varepsilon_0 \frac{\partial^2}{\partial t^2} \int_{-\infty}^{\infty} d\tau \mu(t-\tau)\Big[E(z,\tau) \\ + \int_{-\infty}^{\infty} d\varsigma \chi(\tau-\varsigma) E(z,\varsigma) \Big] \\ = \frac{\partial^2}{\partial t^2} \int_{-\infty}^{\infty} d\tau \mu(t-\tau) P_{\rm NL}(z,\tau), \end{aligned} \quad (1)$$

where $P_{\rm NL}(z, t)$ is the second-order nonlinear polarization given by the integral expression $P_{\rm NL}(z, t) = \varepsilon_0 \int_{-\infty}^{\infty} d\tau_1 \int_{-\infty}^{\infty} d\tau_2 \chi^{(2)}(t-\tau_1, t-\tau_2) E(z, \tau_1) E(z, \tau_2)$. In Eq. (1), ε_0 is the vacuum permittivity, the electric flux density $\mathbf{D}$ has been related to the electric field $\mathbf{E}$, the magnetic flux density $\mathbf{B}$ has been related to the magnetic intensity $\mathbf{H}$ via convolution integrals, and the integral kernels $\mu(t)$ and $\chi(\tau)$ are, respectively, inverse Fourier transforms of the frequency-dependent magnetic permeability $\mu(\omega)$ and electric susceptibility $\chi(\omega)$. In the expression for the nonlinear polarization, the $\chi^{(2)}(t_1, t_2)$ is the inverse Fourier transform of $\chi^{(2)}[\omega_1 + \omega_2, \omega_1, \omega_2]$, the second-order nonlinear susceptibility. Equation (1) with $\mu(\omega) = 1$ is treated in Ref. [21]. The analytical techniques introduced in Ref. [21] will be used in this Letter to explore the consequences of dispersive effects in both the electric permittivity and magnetic permeability. We adopt here the expressions for the electric permittivity

$\varepsilon(\omega)$ [$\equiv 1 + \chi^{(1)}(\omega)$] and $\mu(\omega)$ that are used in Ref. [7]

$$\varepsilon(\omega) = \varepsilon_0 \frac{\omega^2 - \omega_a{}^2}{\omega^2 - \omega_0{}^2}, \qquad \mu(\omega) = \mu_0 \frac{\omega^2 - \omega_b{}^2}{\omega^2 - \Omega^2}, \quad (2)$$

where μ_0 is the vacuum permeability, and $(\omega_a, \omega_0, \omega_b, \Omega)$ are characteristic frequencies of the medium. Plane wave solutions, $E(z, t) \sim \exp[i(\omega t - kz)]$, of $LE = 0$ are governed by the dispersion relation $k^2(\omega) = \omega^2 \varepsilon(\omega)\mu(\omega)$. For a given frequency, two signs of the wave number $k(\omega)$, and therefore of the index of refraction $n(\omega)$, are possible. The correct sign is determined by causality, which is equivalent to the condition that plane waves decay as $z \to \infty$ in the presence of small dissipation. The causal dispersion diagram calculated with Eq. (2) is shown in Fig. 1, where $\Omega < \omega_0 < \omega_b < \omega_a$. For a given wave number, three modes of plane wave propagation exist. The central mode, $\omega_0 < \omega < \omega_b$, has a negative index of refraction, while each of the upper ($\omega_a < \omega$) and lower ($0 < \omega < \Omega$) branches has a positive index of refraction. The negative index branch appears over the frequency range where $\varepsilon(\omega)$ and $\mu(\omega)$ are simultaneously negative. If absorption is introduced in the permittivity and permeability, the dispersion curves in Fig. 1 will be altered in two key ways. First, for real ω, the wave number will be complex with a negative imaginary part, reflecting wave damping. Second, the positive and negative branches will be joined by a curve that will exhibit anomalous dispersion.

If $P_{\rm NL}(z, t)$ is retained in Eq. (1), nonlinear mixing of plane waves is possible. The three-wave mixing process of frequency down-conversion that we addressed in Ref. [7] and will develop further in this Letter satisfies the respective frequency and wave number resonance conditions $\omega_3 = \omega_1 - \omega_2$ and $k_3(\omega_3) = k_1(\omega_1) - k_2(\omega_2)$, where $k(\omega) = \omega\sqrt{\varepsilon(\omega)\mu(\omega)}$, waves 1 and 2 lie on the negative index branch, and wave 3 lies on the low-frequency positive index branch. The wave numbers $k_1(\omega_1)$ and $k_2(\omega_2)$ are negative, and $k_3(\omega_3)$ is positive. Let ω_s be a frequency between ω_1 and ω_2. As ω_1 and ω_2 approach ω_s, ω_3 can attain arbitrary small values. It is through this process that terahertz wave phenomena can be produced via nonlinear mixing of negative index waves.

It is convenient to express ω_1 and ω_2 in terms of ω_3 and ω_s, $\omega_1 = \omega_s + \omega_3/2$, and $\omega_2 = \omega_s - \omega_3/2$. The frequency resonance condition is then identically satisfied, while the momentum resonance condition is

$$k_{\rm NI}(\omega_s + \omega_3/2) - k_{\rm NI}(\omega_s - \omega_3/2) = k_3(\omega_3). \quad (3)$$

Equation (3) is a nonlinear algebraic equation that determines the central frequency as a function of ω_3. A numerical solution of Eq. (3) is presented in Fig. 2 as a plot of ω_s vs ω_3 for $\Omega = 0.3\omega_a$, $\omega_0 = 0.4\omega_a$, and $\omega_b = 0.9\omega_a$. Each curve is labeled by a value proportional to Δk [$\equiv k_{\rm NI}(\omega_s + \omega_3/2) - k_{\rm NI}(\omega_s - \omega_3/2) - k_3(\omega_3)$], where Δk represents the wave number mismatch. For exact resonance, $\Delta k = 0$, the lower curve in Fig. 2 satisfies Eq. (3). If $\Delta k \neq 0$, $\omega_s(\omega_3)$ follows the upper curves.

As $\omega_3 \to 0$, ω_1 and ω_2 approach a common value, $\omega_s(0)$. This limit has important implications that we now explore. For small ω_3, we expand $k_{\rm NI}(\omega_s + \omega_3/2) - k_{\rm NI}(\omega_s - \omega_3/2)$ in powers of ω_3, then substitute the result in Eq. (3). To lowest order in ω_3, the resulting expression is $dk_{\rm NI}/d\omega = k_3(\omega_3)/\omega_3$ at $\omega = \omega_s$ as $\omega_3 \to 0$. At the value of ω_s on the negative branch that satisfies this criterion, the group speed on the negative index branch equals the phase speed of the wave at the origin on the lower positive index branch. This condition, which is depicted graphically in Fig. 1 with tangent lines, is the criterion for the long wave–short wave resonance [13]. It is a resonance between a low-frequency mode and two degenerate high-frequency

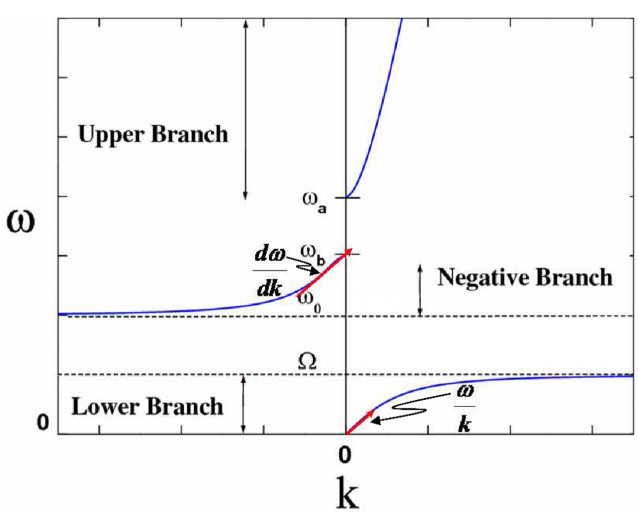

FIG. 1 (color online). Depiction of LWSW resonance on a dispersion diagram ω vs k based on Eq. (3) with $\Omega < \omega_0 < \omega_b < \omega_a$. The parallel lines show the matching of the group and phase velocities on the negative and lower branches, respectively.

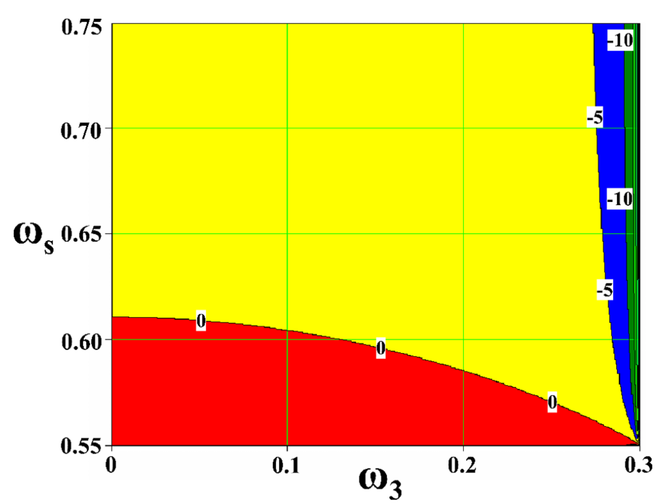

FIG. 2 (color online). Contour plot of the normalized phase mismatch ($c_0\Delta k$) with ω_s and ω_3 normalized to ω_a. The lower curve with $\Delta k = 0$ is the curve satisfying Eq. (4). The characteristic frequencies are $\Omega = 0.3\omega_a$, $\omega_0 = 0.4\omega_a$, and $\omega_b = 0.9\omega_a$.

modes. With Eq. (2) the LWSW criterion reduces to $\frac{dk_{\mathrm{NI}}}{d\omega}(\omega_s) = \frac{1}{c}\frac{\omega_a\omega_b}{\omega_0\Omega}$. The solution of this equation for ω_s is the LWSW frequency, which is $\omega_s(0)$ in Fig. 2.

In the limit of the LWSW resonance, it is useful to consider the interaction of two wave packets, one packet propagating in the negative index mode about the LWSW frequency and the second propagating in the low-frequency positive index mode. A wave packet is a group of plane waves with frequencies and wave numbers centered about and near a central frequency ω and a central wave number $k(\omega)$. The packet waves interfere to produce a plane wave with a slowly varying envelope given by

$$E_1(z, t) = A(Z, T)\exp[i(\omega t - kz)] + \text{c.c.}, \quad (4)$$

where $Z \equiv \eta z$, $T \equiv \eta t$, and η is a small parameter. The scaled spatial and temporal coordinates imply that derivatives of $A(Z, T)$ with respect to z and t are of order η and therefore small. By allowing the amplitudes to depend on multiple spatial scales $A(Z, T) \equiv A(Z_1, Z_2, Z_3, \ldots; T)$, where $Z_1 \equiv \eta z$, $Z_2 \equiv \eta^2 z$, $Z_3 \equiv \eta^3 z, \ldots$, the spatial derivative $\partial A/\partial Z$ can be expanded with respect to η, $\partial A/\partial Z = \partial A/\partial Z_1 + \eta \partial A/\partial Z_2 + \eta^2 \partial A/\partial Z_3 + \ldots$.

A solution of Eq. (1) for interacting wave packets is derived by expanding the electric field $E(z, t)$ in terms of the small scaling parameter η [21],

$$E(z, t) = \eta E_1(z, t) + \eta^2 E_2(z, t) + \eta^3 E_3(z, t)\ldots, \quad (5)$$

where the first-order term $E_1(z, t)$ is the wave packet represented by Eq. (4). The higher-order terms arise from the nonlinear polarization. Because the nonlinear polarization is quadratic in the electric field, $E_2(z, t)$ can be represented as a linear superposition of wave packets at zero and second harmonics of the phase $(\omega t - kz)$,

$$E_2(z, t) = D(Z, T) + C(Z, T)\exp[i2(\omega t - kz)] + \text{c.c.}, \quad (6)$$

where the amplitudes are slowly varying functions of space and time, $D(Z, T) \equiv D(Z_1, Z_2, Z_3, \ldots; T)$ and $C(Z, T) \equiv C(Z_1, Z_2, Z_3, \ldots; T)$. Higher harmonics of $(\omega t - kz)$ will appear in the higher-order terms of Eq. (5). Analysis reveals that $A(\eta z, \eta t)$ and $D(\eta z, \eta t)$ are coupled by two nonlinear partial differential equations, and $C(\eta z, \eta t)$ is proportional to $A^2(Z, T)$. In our application, $A(z, t)$ and $D(\eta z, \eta t)$ are, respectively, the amplitude of the high-frequency wave packet on the negative index branch and the low-frequency wave packet on the lower positive index branch. Equation (5) is now substituted in Eq. (1) followed by an expansion of the left and right hand sides with respect to η. The expansion of the left hand side is

$$LE(z, t) = \eta LE_1(z, t) + \eta^2 LE_2(z, t) + \ldots, \quad (7)$$

where each factor, $LE_1(z, t), LE_2(z, t), \ldots$, must also be expanded with respect to η. At order one, $E_1(z, t)$ is given by Eq. (4). The expansion of $LE_1(z, t)$ is then

$$LE_1(z, t) = [L_0(\omega, k)A + \eta L_1 A + \eta^2 L_2 A + \ldots] \times \exp[i(\omega t - kz)] + \text{c.c.}, \quad (8)$$

where $L_0(\omega, k) \equiv -k^2 + \omega^2\varepsilon(\omega)\mu(\omega)$, and $L_1, L_2, \ldots$ are linear differential operators with respect to the scaled variables $(Z_1, Z_2, \ldots, T)$. Substitution of Eqs. (7) and (8) in Eq. (1) yields

$$\eta^2 LE_2 + \eta^3 LE_3 \ldots = -\eta\exp[i(\omega t - kz)][L_0(\omega, k)A + \eta L_1 A + \eta^2 L_2 A + \ldots] + \text{c.c.} + \frac{\partial^2}{\partial t^2}\int_{-\infty}^{\infty} d\tau \mu(t-\tau)P_{\mathrm{NL}}(z, \tau). \quad (9)$$

The expansion of LE_2 has the form $LE_2 = (LE_2)_0 + \eta(LE_2)_1 + \eta^2(LE_2)_2 + \ldots$ and similarly for $LE_3 \ldots$. We equate terms of equal order in η in the left hand side of Eq. (9) to the corresponding terms on the right hand side. This process leads to a sequence of partial differential equations that govern $A(Z_1, Z_2, Z_3, \ldots; T)$, $C(Z_1, Z_2, Z_3, \ldots; T)$, and $D(Z_1, Z_2, Z_3, \ldots; T)$. It should be noted that $P_{\mathrm{NL}}(z, t)$ defined earlier is of order η^2. At order η, Eq. (9) yields $L_0(\omega, k) = 0$, which is the plane wave dispersion relation. The order η^2 terms of Eq. (9) yield

$$(LE_2)_0 = -L_1 A\exp[i(\omega t - kz)] + \text{c.c.} + \left[\frac{\partial^2}{\partial t^2}\int_{-\infty}^{\infty} d\tau\mu(t-\tau)P_{\mathrm{NL}}(z, \tau)\right]_2, \quad (10)$$

where $[\ldots]_2$ designates the order η^2 of the enclosed quantity. Because the first harmonic term $L_1 A\exp[i(\omega t - kz)]$ lies in the null space of the linear operator $(L\ldots)_0$, it must be set equal to zero to avoid secular components in E_2. This solvability condition implies $L_1 A = 0$, which is explicitly, $\partial A/\partial T + v_g(\omega)\partial A/\partial Z_1 = 0$. This expression implies that the amplitude $A(Z_1, Z_2, Z_3, \ldots; T)$ propagates with speed $v_g(\omega)$ with respect to the scaled coordinates Z_1 and T: $A(Z_1, Z_2, Z_3, \ldots; T) \equiv A[Z_2, Z_3, \ldots; T - Z_1/v_g(\omega)]$. The general solution of Eq. (10) is Eq. (6), where the amplitude of the second harmonic term is $C(Z, T) = \frac{\mu[2\omega]/\mu_0}{n^2[\omega] - n^2[2\omega]}\chi^{(2)}[2\omega, \omega, \omega]A^2(Z, T)$. The zero harmonic amplitude $D(Z, T)$ is determined at order η^4 in the expansion. The order η^3 and η^4 terms of Eq. (9) are expressed as $(LE_3)_0 = F_3$ and $(LE_4)_0 = F_4$ where F_3 and F_4 are functions of lower order fields. Analogous to Eq. (10), spatial secularities will not be present in their solutions if the following solvability conditions are satisfied: the first harmonic term in F_3 and the zero harmonic term in F_4 must be absent. These conditions yield the coupled equations

$$-i\frac{\partial A}{\partial Z_2}-\frac{1}{2}\frac{d^2k}{d\omega^2}\frac{\partial^2 A}{\partial T^2}+\varepsilon_0{}^2c_0{}^2\frac{\omega^2}{k}\frac{\mu[\omega]\mu[2\omega]}{n^2[\omega]-n^2[2\omega]}\chi^{(2)}[2\omega,\omega,\omega]\chi^{(2)}[\omega,2\omega,-\omega]|A|^2A=-\varepsilon_0\mu[\omega]\frac{\omega^2}{k}\chi^{(2)}[\omega,\omega,0]DA, \tag{11}$$

$$\frac{\partial^2 D}{\partial Z_1{}^2}-\frac{n^2[0]}{c_0{}^2}\frac{\partial^2 D}{\partial T^2}=\varepsilon_0\mu[0]\chi^{(2)}[0,\omega,-\omega]\frac{\partial^2|A|^2}{\partial T^2}, \tag{12}$$

where c_0 represents the vacuum speed of light. Note that the product $\chi^{(2)}[2\omega,\omega,\omega]\chi^{(2)}[\omega,2\omega,-\omega]$ in Eq. (11) is actually a cascaded second-order nonlinearity and is equivalent to an effective third-order nonlinear susceptibility, $\chi^{(3)}_{\rm eff}[\omega,\omega,\omega,-\omega]$. It is also important to note that the coefficient of $|A|^2A$ in Eq. (11) is singular if $n^2[\omega]=n^2[2\omega]$. Under this particular condition, it is necessary to introduce higher-order multiple spatial and temporal scales when reducing Eq. (1). When the LWSW resonance condition is satisfied, $v_g^2(\omega)=c_0{}^2/n^2[0]$, Eq. (12) can be reduced to a lower order equation,

$$\frac{\partial D}{\partial Z_2}=-\frac{v_g(\omega)}{2\eta}\varepsilon_0\mu[0]\chi^{(2)}[0,\omega,-\omega]\frac{\partial|A|^2}{\partial T_R}, \tag{13}$$

where T_R is the retarded time, $T_R\equiv T-Z_1/v_g(\omega)$. The term $\partial|A|^2/\partial T_R$ is related to the ponderomotive force that drives the system. In Eq. (13) the coefficient of the derivative with respect to T_R must be of order 1, $v_g(\omega)\varepsilon_0\mu[0]\chi^{(2)}[0,\omega,-\omega]\sim O(\eta)$, which places a restriction on the material parameters.

Equations (11) and (13) govern the amplitudes of high- and low-frequency wave packets that satisfy the LWSW resonance. The variety of solutions of these equations requires further study, which we cannot undertake here. However, we can briefly discuss some of these solutions. If the medium and wave parameters are such that the term proportional to $|A|^2A$ in the left hand side of Eq. (11) can be neglected, the resulting equation coupled with Eq. (13) forms a system that is solved in Refs. [18,19]. Among possible solutions that have been derived via the inverse scattering technique are solitons, which will also be applicable to negative index media. A class of solutions that can be readily derived for our nonlinear LWSW system, Eqs. (11) and (13), are wave structures of the form $D(Z_2,T_R)=D(\vartheta)$ and $A(Z_2,T_R)=Q(\vartheta)\exp[i(\Lambda T_R-KZ_2)]$, where $\vartheta\equiv T_R-Z_2/C$, $D(\vartheta)$ and $Q(\vartheta)$ are real amplitudes, C is a speed that must be determined, and Λ and K are, respectively, a real frequency and a real wave number. Substituting this particular wave form in Eqs. (11) and (13) yields the following important results. First, the speed C is related to the frequency Λ, $\Lambda C=-(2\beta)^{-1}$, where β is the coefficient of the second derivative with respect to T in Eq. (11). Second, $D(\vartheta)$ is proportional to $Q^2(\vartheta)$ while $Q(\vartheta)$ is proportional to $\mathrm{sech}(\alpha\vartheta)$, where $\alpha\equiv\sqrt{(K+\beta\Lambda^2)/\beta}$, i.e., $D(\vartheta)$ and $Q(\vartheta)$ propagate as paired nondispersive pulses with speed C. Other solutions for $D(\vartheta)$ and $Q(\vartheta)$ exist, including wave trains with $Q(\vartheta)$ as a periodic function of ϑ. In summary, Eqs. (11) and (13) are rich in nonlinear wave solutions that may have important applications in negative index media.

In conclusion, the LWSW resonance could be a mechanism of terahertz wave generation from optical waves. The fields A and D that Eqs. (11) and (13) couple would represent the optical wave envelope and the terahertz wave, respectively. Energy from the optical wave transfers to the terahertz wave through the action of a ponderomotive force produced by the optical wave. Currently, a promising metamaterial candidate for nonlinear phenomena in general and terahertz generation via LWSW resonance in particular is the 3D semiconductor negative index metamaterial recently demonstrated [22], as its third dimension now permits us to explore propagation-dependent nonlinear phenomena as opposed to nonlinear scattering from just single layer 2D metamaterials.

In addition, the nonlinear LWSW resonance phenomenon affords us the opportunity of realizing solitary waves, paired solitons, and periodic wave trains.

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