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Generalized Nonlinear Schrödinger Equation for Dispersive Susceptibility and Permeability: Application to Negative Index Materials

Michael Scalora,¹ Maxim S. Syrchin,² Neset Akozbek,³ Evgeni Y. Poliakov,¹ Giuseppe D'Aguanno,³ Nadia Mattiucci,^{3,4} Mark J. Bloemer,¹ and Aleksei M. Zheltikov²

¹Charles M. Bowden Research Center, AMSRD-AMR-WS-ST, RDECOM, Redstone Arsenal, Alabama 35898-5000, USA

²Physics Department and International Laser Center, M V Lomonosov Moscow State University, Vorob'evy Gory, 119899 Moscow, Russian Federation

³Time Domain Corporation, Cummings Research Park, 7057 Old Madison Pike, Huntsville, Alabama 35806, USA

⁴Università "Roma Tre," Dipartimento di Fisica "E. Amaldi," Via Della Vasca Navale 84, I-00146 Rome, Italy

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A new generalized nonlinear Schrödinger equation describing the propagation of ultrashort pulses in bulk media exhibiting frequency dependent dielectric susceptibility and magnetic permeability is derived and used to characterize wave propagation in a negative index material. The equation has new features that are distinct from ordinary materials ($\mu = 1$): the linear and nonlinear coefficients can be tailored through the linear properties of the medium to attain any combination of signs unachievable in ordinary matter, with significant potential to realize a wide class of solitary waves.

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Nonlinear wave propagation in optics has been widely studied in the framework of the nonlinear Schrödinger equation (NLSE). Its use in describing the propagation of picosecond pulses has led to innumerable innovations in many fields, the most notable of which is perhaps fiber optics [1]. The NLSE describes the evolution of an envelope function, which is assumed to vary slowly over an optical cycle. Typical pulse durations are now routinely below 50 femtoseconds (fs), a regime where pulse envelopes can no longer be assumed to always vary slowly in space and/or time, especially near resonance or unusual dispersive conditions [2]. The advent of fs lasers and recent demonstrations of attosecond pulses [3] highlight a need for a tailored, more measured theoretical approach to study specific problems, to address new, unusual materials, and to go beyond well-established approximations. The filamentation of intense fs pulses in air and supercontinuum (SC) generation can be described by a properly managed, modified NLSE [4]. The same is true for the case of photonic crystal fibers [5], where a SC is generated under conditions of transverse light confinement [6]. It has been shown that the slowly varying envelope approximation (SVEA) breaks down even for initial pulses that are many optical cycles long [7]. In this respect, a modified NLSE is used that includes correction terms that go beyond the SVEA, such as the shock term [8] and coupled temporal and transverse spatial derivatives [7,9]. Other corrections are derived from including the second order spatial derivatives, leading to a pseudo- $\chi^{(5)}$ -like effect and modifications of the shock term [10].

In this Letter we discuss the propagation of pulses at least a few tens of optical cycles in duration, within the context of a new NLSE derived for the general case of dispersive dielectric susceptibility ε and magnetic permeability μ , and focus our attention on the unusual character-

istics of uniform bulk, negative index materials [11], in the absence of feedback. When ε and μ are simultaneously dispersive and negative, the index of refraction $n = \pm\sqrt{\varepsilon\mu}$ allows the negative root as its solution [11], leading to unusual refraction of the beam, as if the index of refraction were negative. Our approach evolves through the narrow-band constraints imposed by the SVEA [2], and leads to an equation of motion where group velocity and group velocity dispersion (GVD) can easily be identified and quantified without ambiguity when the medium is relatively transparent. Then, eliminating the magnetic field and introducing a nonlinear polarization or magnetization leads to a NLSE similar to the usual NLSE, but different in some important aspects. We find that while the group velocity is always positive, GVD, the transverse Laplacian, and non-SVEA corrections can all have the same sign, a positive or negative sign, or a variety of combinations, depending on the specifics of the linear dispersion curves and the sign of the nonlinear coefficient. The available combination of signs leads to new, richer dynamical characteristics compared to the case of ordinary nonlinear dynamics [1], and immediately suggests the existence of temporal and spatial, bright and dark solitons in bulk NIMs.

Nonlinear pulse propagation in NIMs of both finite and infinite lengths remains essentially unexplored. In the linear regime authors have investigated group delay and superluminal propagation [12–14], and the anomalous refraction process for wave packets of finite spatial and temporal extent [15–17]. For structures of finite length bright and dark *gap* solitonlike solutions can be dynamically excited and made to propagate within a single layer of NIM [18]. Here we use the same formal approach used in Ref. [17] to derive a new wave equation that can be used to study pulse propagation in uniform, bulk materials under the conditions of a nonlinear polarization and/or magneti-

zation [19]. We go beyond the usual SVEA to obtain only qualitative understanding of how higher order terms contribute to the dynamics when the material is magnetically active, and compare with the dynamics that ensues in ordinary materials. For simplicity we first consider one longitudinal spatial coordinate and time, and define the fields as follows:

$$\mathbf{D}(z, t) = \hat{\mathbf{i}} \left[\int_{-\infty}^{\infty} \varepsilon(\omega) E_x(z, \omega) e^{-i\omega t} d\omega + P_{nl}(z, t) \right], \quad (1)$$

$$\mathbf{B}(z, t) = \hat{\mathbf{j}} \int_{-\infty}^{\infty} \mu(\omega) H_y(z, \omega) e^{-i\omega t} d\omega. \quad (2)$$

P_{nl} is the nonlinear polarization. Substitution into Maxwell equations yields

$$\frac{\partial E_x(z, t)}{\partial z} = \frac{i}{c} \int_{-\infty}^{\infty} [\omega \mu(\omega)] H_y(z, \omega) e^{-i\omega t} d\omega, \quad (3)$$

$$\begin{aligned} \frac{\partial H_y(z, t)}{\partial z} &= \frac{i}{c} \int_{-\infty}^{\infty} [\omega \varepsilon(\omega)] E_x(z, \omega) e^{-i\omega t} d\omega - \frac{1}{c} \\ &\times \frac{\partial P_{nl}(z, t)}{\partial t}. \end{aligned} \quad (4)$$

The symmetry of the equations suggests that a nonlinear magnetization produces qualitatively similar effects. In Eqs. (3) and (4) we have expanded ε and μ as follows:

$$\omega \varepsilon(\omega) = \sum_{n=0}^{\infty} \left\{ \frac{\partial^n [\omega \varepsilon(\omega)]}{\partial \omega^n} \right\}_{\omega=\omega_0} \frac{(\omega - \omega_0)^n}{n!}; \quad (5)$$

$\omega \mu(\omega)$ is written in a similar way. ω_0 is the carrier frequency of the incident pulse. Substituting the expansions for $\omega \varepsilon(\omega)$ and $\omega \mu(\omega)$ into Eqs. (3) and (4) yields

$$\frac{\partial E_x}{\partial z} = \frac{i}{c} e^{ikz-i\omega_0 t} \sum_{n=0}^{\infty} \left\{ i^n \frac{\partial^n (\omega \mu)}{\partial \omega^n} \right\}_{\omega=\omega_0} \frac{1}{n!} \frac{\partial^n H_y}{\partial t^n}, \quad (6)$$

$$\frac{\partial H_y}{\partial z} = \frac{i}{c} e^{ikz-i\omega_0 t} \sum_{n=0}^{\infty} \left\{ i^n \frac{\partial^n (\omega \varepsilon)}{\partial \omega^n} \right\}_{\omega=\omega_0} \frac{1}{n!} \frac{\partial^n E_x}{\partial t^n} - \frac{1}{c} \frac{\partial P_{nl}}{\partial t}. \quad (7)$$

where all the fields are understood to be explicit functions of z and t . Equations (6) and (7) are very general because they include dispersion effects up to any desired order. For ultrashort pulse propagation one requires an expansion of ε and μ at least up to second order to account for GVD effects. Accordingly, we decompose the fields as a general envelope function (not necessarily slowly varying), multiplied by carrier wave vector and frequency: $[E(z, t), H(z, t)] e^{i(kz-\omega t)}$, and substitute in Eqs. (6) and (7) to obtain

$$\begin{aligned} \alpha \frac{\partial E}{\partial \tau} + \frac{i\alpha'}{4\pi} \frac{\partial^2 E}{\partial \tau^2} &= i\beta \varepsilon E - i\beta n H - \frac{\partial H}{\partial \xi} \\ &+ i\beta \chi^{(3)} |E|^2 E - \chi^{(3)} \frac{\partial}{\partial \tau} (|E|^2 E), \end{aligned} \quad (8)$$

$$\gamma \frac{\partial H}{\partial \tau} + \frac{i\gamma'}{4\pi} \frac{\partial^2 H}{\partial \tau^2} = i\beta \mu H - i\beta n E - \frac{\partial E}{\partial \xi}, \quad (9)$$

where $\alpha = \frac{\partial[\tilde{\omega}\varepsilon(\tilde{\omega})]}{\partial\tilde{\omega}}$, $\alpha' = \frac{\partial^2[\tilde{\omega}\varepsilon(\tilde{\omega})]}{\partial\tilde{\omega}^2}$ and $\gamma = \frac{\partial[\tilde{\omega}\mu(\tilde{\omega})]}{\partial\tilde{\omega}}$, $\gamma' = \frac{\partial^2[\tilde{\omega}\mu(\tilde{\omega})]}{\partial\tilde{\omega}^2}$. We have adopted the following scaling: $\xi = z/\lambda_p$; $\tau = ct/\lambda_p$, $\beta = 2\pi\tilde{\omega} = 2\pi\omega/\omega_p$; $n = \sqrt{\varepsilon\mu} = n(\tilde{\omega})$, ω_p is the plasma frequency and λ_p its corresponding wavelength. We now combine Eqs. (8) and (9) and eliminate the magnetic field. For relatively transparent materials, the fact that energy should always be positive [20] imposes the following conditions: $\alpha = \frac{\partial[\tilde{\omega}\varepsilon(\tilde{\omega})]}{\partial\tilde{\omega}} > 0$ and $\gamma = \frac{\partial[\tilde{\omega}\mu(\tilde{\omega})]}{\partial\tilde{\omega}} > 0$ simultaneously. Although absorption is an issue one must deal with in any bulk medium and at any wavelength, including the visible range [21], we are mainly interested in seeing how magnetic activity manifests itself in the dynamics. Therefore, we assume that linear absorption remains small [22]. Substituting and retaining linear derivatives up to second order, and neglecting nonlinear second order temporal derivatives, we can write

$$\begin{aligned} \frac{\partial E}{\partial \xi} + \frac{(\varepsilon\gamma + \mu\alpha)}{2n} \frac{\partial E}{\partial \tau} &= \frac{i}{2\beta n} \left\{ \frac{\partial^2 E}{\partial \xi^2} - \alpha\gamma \frac{\partial^2 E}{\partial \tau^2} \right\} + \frac{i\beta\mu\chi^{(3)}}{2n} \\ &\times |E|^2 E - \frac{\gamma\chi^{(3)} + \mu\chi^{(3)}}{2n} \frac{\partial}{\partial \tau} (|E|^2 E) \\ &- i \frac{(\varepsilon\gamma' + \mu\alpha')}{8\pi n} \frac{\partial^2 E}{\partial \tau^2}. \end{aligned} \quad (10)$$

Ordinarily, propagation takes place in low density gases or other materials such that the index of refraction is greater than one. Our system displays metallic behavior, and so one has to be careful about extending the range of validity of Eq. (10) to regions where $n \rightarrow 0$ [18]. In that case, one should solve Eqs. (8) and (9) simultaneously.

The form of Eq. (10) suggests a group velocity: $V_g = \frac{2n}{(\varepsilon\gamma + \mu\alpha)}$. It can easily be shown that given $n^2 = \varepsilon\mu$, then $V_g = \frac{1}{n + \omega \frac{\partial n}{\partial \omega}} = \left(\frac{\partial k}{\partial \omega} \right)^{-1}$, which is the usual expression for the group velocity in units of c . Because n is negative when both ε and μ are simultaneously negative, and given $\alpha > 0$ and $\gamma > 0$ [20], it follows that the group velocity is always positive without ambiguity.

Transforming to the retarded coordinates: $\frac{\partial}{\partial z} = \frac{\partial}{\partial \xi} - \frac{1}{V_g} \frac{\partial}{\partial \tau}$, and $\frac{\partial}{\partial t} = \frac{\partial}{\partial \tau}$ and substituting into Eq. (10) yields

$$\begin{aligned} \frac{\partial E}{\partial z} &= \frac{i}{2\beta n} \left(\frac{1}{V_g^2} - \alpha\gamma - \beta \frac{\varepsilon\gamma' + \mu\alpha'}{2} \right) \frac{\partial^2 E}{\partial \tau^2} \\ &+ \frac{i}{2\beta n} \left(\frac{\partial^2 E}{\partial z^2} - \frac{2}{V_g} \frac{\partial}{\partial z} \frac{\partial E}{\partial \tau} \right) + \frac{i\beta\mu\chi^{(3)}}{2n} |E|^2 E \\ &- \frac{\gamma\chi^{(3)} + \mu\chi^{(3)}}{2n} \frac{\partial}{\partial \tau} (|E|^2 E). \end{aligned} \quad (11)$$

The change of coordinates immediately reveals a GVD coefficient k'' [1]. Although it is not obvious from its form,

one can show that $k'' = \frac{\partial}{\partial \omega} \left(\frac{1}{V_g} \right) = \frac{1}{\beta n} \left(\frac{1}{V_g} - \alpha \gamma - \beta \frac{\varepsilon \gamma' + \mu \alpha'}{2} \right)$. Because we are assuming propagation in bulk media and no feedback, and pulses at least a few tens of wave cycles in duration, all higher order derivatives should give negligible contributions. We now calculate the first order non-SVEA correction terms by using Eq. (11) to evaluate $\frac{\partial^2 E}{\partial z^2}$ and $\frac{\partial E}{\partial t}$ [10]. Neglecting higher order derivatives, we estimate $\frac{\partial^2 E}{\partial z^2} \approx \frac{i\beta\mu\chi^{(3)}}{2n} \frac{\partial}{\partial z} (|E|^2 E)$ and $\frac{\partial^2 E}{\partial z \partial t} \approx \frac{i\chi^{(3)}\beta\mu}{2n} \frac{\partial}{\partial t} \times (|E|^2 E)$. Therefore,

$$\frac{\partial E}{\partial z} \approx \frac{ik''}{2} \frac{\partial^2 E}{\partial t^2} + \frac{i\beta\mu\chi^{(3)}}{2n} \left[1 - \frac{\mu\chi^{(3)}}{4n^2} |E|^2 \right] |E|^2 E + \chi^{(3)} \left\{ \frac{\mu}{2V_g n^2} - \frac{\gamma + \mu}{2n} \right\} \frac{\partial (|E|^2 E)}{\partial t}. \quad (12)$$

Equation (12) is the generalized NLSE we sought. It can be solved using any of a number of equivalent numerical techniques [1].

There are some qualitative aspects of this modified NLSE that are remarkable. First, the sign of the GVD can be positive or negative, depending on the particular choice of parameters. Second, assuming a *positive* $\chi^{(3)}$, the sign of the leading nonlinear coefficient is always positive because the ratio μ/n is positive. On the other hand, the pseudo- $\chi^{(5)}$ correction term is proportional to $(-\mu^2/n^3)$, which makes the coefficient positive, and its effect is to enhance the nonlinearity. For ordinary materials the effect of this term is to quench the nonlinearity. Finally, magnetic contributions to the shock term are also evident. Usually ($\mu = 1$ and $n > 1$) this coefficient is negative [1], but its form in Eq. (12) and the specific model used may allow for it to be positive in a frequency range where $0 < n < 1$. In optical fibers the shock term causes the pulse to steepen along its trailing edge and the spectrum to split asymmetrically with larger identifiable redshifted peaks [1]. In NIMs the opposite occurs, with self-steepening characterizing the front of the pulse (see Fig. 3 below).

We now illustrate the dynamics with a representative example. We use a Drude model described by $\varepsilon(\tilde{\omega}) = 1 - \frac{1}{\tilde{\omega}^2 + i\tilde{\omega}\gamma}$ and $\mu(\tilde{\omega}) = 1 - \frac{\omega_m^2/\omega_p^2}{\tilde{\omega}^2 + i\tilde{\omega}\gamma}$. In our scaled units, we choose $\gamma \approx 5 \times 10^{-4}$, which results in negligible absorption [12]. In Fig. 1 we show ε , μ , and the index of refraction n when $\omega_m^2/\omega_p^2 = 0.64$. Using these dispersion curves, we calculate the group velocity, GVD, and the shock term and show them in Fig. 2. The figure shows that the shock term can be zero, which means shorter pulses can evolve into solitary waves, at least compared to ordinary media. In general, the dispersion curves can be engineered by choosing the size of split-ring resonator circuit elements [21,23], which have been shown to exhibit negative refraction under microwave illumination [23]. In Fig. 3 we illustrate the effect of a positive shock coefficient. The figure shows that the pulse self-steepens along its leading edge, and the peaks with the largest amplitude are blueshifted (inset).

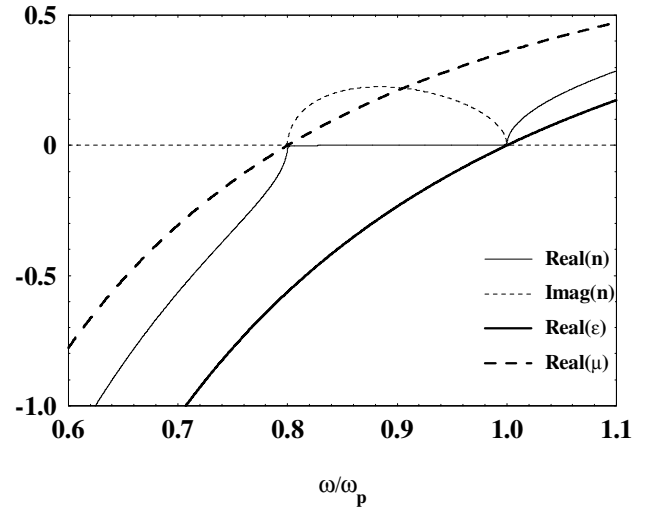


FIG. 1. Dispersion of ε , μ , and resulting n . The region $0.8 \leq \tilde{\omega} \leq 1$ is characterized by metal-like reflections, as n becomes almost purely imaginary. $n < 0$ in the region $\tilde{\omega} \leq 0.8$.

Magnetic nonlinearities can also play a role, and indeed their contribution may be more pronounced than electric nonlinearities [19]. A magnetic nonlinearity would produce additional terms, and Eq. (12) should be modified accordingly. If only a magnetic nonlinearity is considered, the qualitative aspects would remain the same thanks to the symmetry properties of Eqs. (6) and (7). We note that a negative $\chi^{(3)}$ may be used to make similar arguments, with appropriate sign changes applied to the other coefficients.

We can gain further insight if we consider Eq. (12) in its simplest form, but with the addition of transverse coordinates. For the simple case of linearly polarized fields, and to the extent that one can neglect the $\vec{\nabla}(\vec{\nabla} \cdot \mathbf{E})$ in the vector equations, the analysis yields the expected transverse Laplacian that describes diffraction. The result is

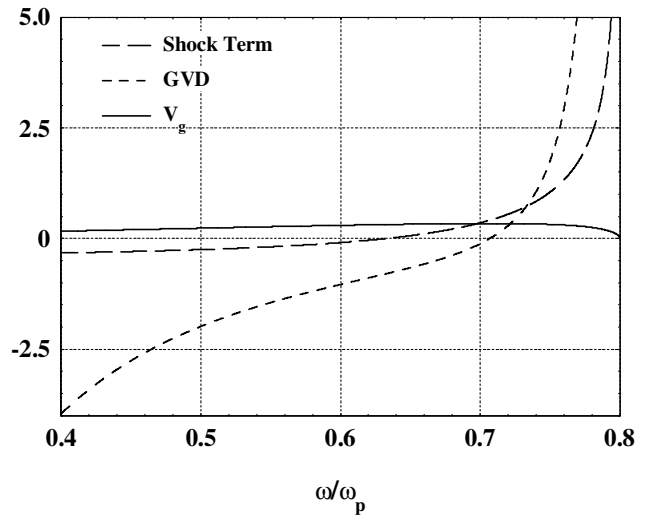


FIG. 2. $V_g = \frac{2n}{\varepsilon\gamma + \mu\alpha'}$, $\text{GVD} = \frac{k''}{\beta n} = \frac{1}{\beta n} \times \left(\frac{1}{V_g} - \alpha\gamma - \beta \frac{\varepsilon\gamma' + \mu\alpha'}{2} \right)$, and shock term $S = \frac{\mu}{2V_g n^2} - \frac{\gamma + \mu}{2n}$ in units of $\chi^{(3)}$.

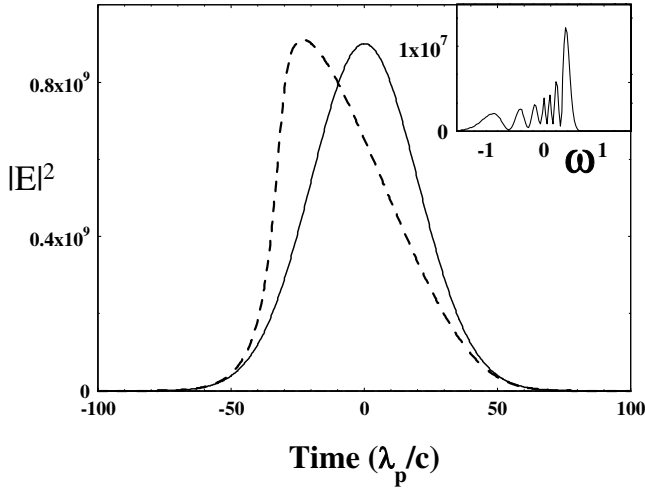


FIG. 3. Field intensity profiles with $\tilde{\omega} \approx 0.707$, where $k'' \sim 0$, and $S = 0.41$. The pulse self-steepens with larger blueshifted spectral components (inset). The input field is $E(t) = E_0 e^{-t^2/\tau_p^2}$, $E_0 = 3 \times 10^4$ (Statvolts/cm), and $\tau_p = 20$, $\chi^{(3)} = 10^{-10}$ (esu). Propagation distance is $\sim 200\lambda_p$.

$$\frac{\partial E}{\partial z} = \frac{i\nabla_{\perp}^2 E}{2\beta n} + \frac{ik''}{2} \frac{\partial^2 E}{\partial t^2} + \frac{i\beta\mu\chi^{(3)}}{2n} |E|^2 E. \quad (14)$$

Although the sign of the nonlinear coefficient is unchanged in NIMs, the GVD term can have the same or opposite sign. Bright (dark) soliton solutions thus emerge when both coefficients are positive (negative), with solutions that are identical to those discussed for ordinary materials [1]. Because the temporal derivative and the transverse Laplacian are on equal footing, the same arguments we have made for temporal solitons may be made for transverse or spatial solitons. Finally, the index of refraction also rescales the Fresnel number (the coefficient in front of the transverse Laplacian) and determines its sign. This provides a simple dynamical explanation for the negative refraction process.

In conclusion, a generalized nonlinear Schrödinger equation for a dispersive dielectric susceptibility and permeability was derived from first principles, and was used to describe pulse propagation in a NIM. We find that the linear properties of the medium can be tailored to change both the linear and nonlinear effective properties of the medium leading to a new class of dynamic behavior and solitary waves. We hope our findings will further stimulate the study of nonlinear wave dynamics in NIMs, in the visible range [21] in particular and in the field of solitary waves in general.

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