Nonlinear wave mixing and susceptibility properties of negative refractive index materials

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We present an analysis of second-order and third-order nonlinear susceptibilities and wave-mixing properties of negative refractive index materials. We show that the nonlinear susceptibilities for noncentrosymmetric and centrosymmetric media may be positive or negative and away from resonance depending on the frequency of interest relative to the resonant frequencies of the material. Manipulation of the signs of the nonlinear susceptibilities is important in the field of optics, particularly for solitons and compensation of nonlinear effects. We also show that three- and four-wave mixing can be naturally phase matched in the material.

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Beginning with the early studies of Mandelstam [1,2], considerable attention has been given to the concept of negative index of refraction. Fundamental features of negative index materials have been explored in these early references and elsewhere [3], and basic issues related to negative group velocity, negative permittivity, and permeability, and consequently to negative index of refraction, have also been addressed [4,5]. Moreover, a variety of novel concepts such as refraction without reflection, the reversed Doppler effect, and subdiffraction imaging have been proposed and explored [3]. Negative refraction and its related ideas have been experimentally realized and extensively analyzed in recent years [6–9]. Nonlinear effects in negative index materials have also received attention. Among them we mention here structures with metamaterials embedded in nonlinear positive dielectrics [10], derivation of a generized nonlinear Schrödinger equation that can be applied to left-handed metamaterials [11], and studies of solitons in left-handed media [12,13]. In this paper, we explore another aspect of nonlinear wave properties of negative index materials, namely their effective nonlinear susceptibilities and wave-mixing properties based on the Lorentz anharmonic oscillator model.

Nonlinear media with negative index of refraction have a variety of potential applications. We propose that negative index media may be used to perform nonlinearity compensation without optical phase conjugation if the sign of $\chi^{(3)}$ is negative. The concept of nonlinearity compensation using a medium with a negative nonlinear refractive index coefficient was suggested in Ref. [14]. One of the major advantages of using negative index media for nonlinearity compensation is that there need not be wavelength translation, which is one of the accompanying features in optical phase conjugation: wavelength translation forces communication systems to have more complicated schemes for wavelength management. Devices based on such nonlinear media may be of great importance for nonlinearity management in optical communication systems for the purpose of increasing the transmission reach and system performance [15]. Another potential application is the use of the negative index medium to support bright solitons [16].

In this paper, we explore the effective nonlinear susceptibility and wave-mixing properties of negative refractive index centrosymmetric and noncentrosymmetric media. We present the second-order $\chi^{(2)}$ and third-order $\chi^{(3)}$ nonlinear susceptibilities of the aforementioned media based on the classical anharmonic oscillator model and then go on to discuss the phase-matching properties that arise from nonlinear wave mixing in such media. An important result that has emerged from our analysis is that perfect phase matching is possible in three- and four-wave mixing if at least one of the interacting waves exhibits a negative index of refraction. Furthermore, we find that the signs of the susceptibilities can be engineered by specifying appropriately the frequencies of the interacting waves.

We examine here one-dimensional electromagnetic wave propagation in a lossless optical medium that we model in terms of nonlinear dipole oscillators. Both quadratic and cubic nonlinearities of noncentrosymmetric and centrosymmetric media will be explored. We assume that the propagation is parallel to the z axis of an (x,y,z) Cartesian coordinate system. The electric field, $\mathbf{E}(z,t)=E(z,t)\hat{\mathbf{x}}$, is parallel to the x axis, while the magnetic field, $\mathbf{B}(z,t)=B(z,t)\hat{\mathbf{y}}$, is parallel to the y axes. The following equation is found to govern $E(z,t)$:

$$\frac{\partial^2 E}{\partial z^2}-\epsilon_0\frac{\partial^2}{\partial t^2}\int_{-\infty}^{\infty}d\tau\mu(t-\tau)\left[E(z,\tau)+\int_{-\infty}^{\infty}d\bar{\tau}\chi(\tau-\bar{\tau})E(z,\bar{\tau})\right]$$
$$=\frac{\partial^2}{\partial t^2}\int_{-\infty}^{\infty}d\tau\mu(t-\tau)P_{\mathrm{NL}}(z,\tau), \tag{1}$$

where ϵ_0 is the vacuum permittivity, $P_{\mathrm{NL}}(z,t)$ is the nonlinear part of the polarization, and the frequency-dependent magnetic permeability, $\mu(\omega)$, and electric susceptibility, $\chi(\omega)$, are the Fourier transforms of $\mu(t)$ and $\chi(t)$, respectively,

$$\mu(\omega)=\int_{-\infty}^{\infty}dt\mu(t)e^{-i\omega t},\quad \chi(\omega)=\int_{-\infty}^{\infty}dt\chi(t)e^{-i\omega t}. \tag{2}$$

The electric permittivity is $\epsilon(\omega)=\epsilon_0[1+\chi(\omega)]$, where ϵ_0 is the vacuum permittivity. Causality requires that $\mu(t-\tau)$ and $\chi(t-\tau)$ vanish for $\tau>t$.

Our description of negative refractive index media is based on the lossless anharmonic oscillator Lorentz model similar to that of Owyoung [17] and Boyd [18] for positive index media. The Lorentz oscillator model may apply not

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only for homogeneous media but also for composite media [19]. In this model, the polarization $P(z,t)$ is related to the wave electric field by the following equations:

$$P = -qNz, \quad \frac{d^2z}{dt^2} + \omega_0^2 z + az^2 = -\frac{q}{m}E$$

$$\text{for noncentrosymmetric media,} \tag{3}$$

$$P = -qNz, \quad \frac{d^2z}{dt^2} + \omega_0^2 z - bz^3 = -\frac{q}{m}E$$

$$\text{for centrosymmetric media,} \tag{4}$$

where q and m are, respectively, the charge and mass of the oscillating particle, N is the particle density, ω_0 is the oscillation frequency of the linear dipole motion, and a and b are constants that govern the strength of the nonlinear restoring forces acting on the particle. In Eqs. (3) and (4), we used the macroscopic electric field that appears in Maxwell's equations. This is a simplifying assumption that is reasonable for a low density of dipoles. For dense media, local field corrections would have to be taken into account [18]. If nonlinear effects are neglected in Eq. (1) by setting $P_{\mathrm{NL}}=0$, the resulting linear equation has harmonic solutions of the form $E(z,t) \sim \exp[i(\omega t - kz)]$, where the frequency ω and the wave number k satisfy the dispersion relation,

$$k^2 = \frac{\omega^2 n^2(\omega)}{c_0^2}, \quad n(\omega) = \sqrt{\frac{\epsilon(\omega)}{\epsilon_0}\frac{\mu(\omega)}{\mu_0}}. \tag{5}$$

In Eq. (5), c_0 is the vacuum speed of light and $n(\omega)$ is the index of refraction of the dispersive medium. For a given ω, the dispersion relation yields two roots for the wave number, $k(\omega) = \pm \omega n(\omega)/c_0$, one of which is a propagating mode with a positive phase velocity and the other is a propagating mode with a negative phase velocity. The permittivity that follows from the linearized Lorentz equations and the permeability that has typically been used in past studies of left-handed materials [20,21] read, respectively,

$$\epsilon(\omega) = \epsilon_0 \frac{\omega^2 - \omega_a^2}{\omega^2 - \omega_0^2}, \quad \mu(\omega) = \mu_0 \frac{\omega^2 - \omega_b^2}{\omega^2 - \Omega^2}, \tag{6}$$

where $\omega_a^2 = \omega_0^2 + \omega_p^2$, ω_p is the plasma frequency of the medium, and ω_b and Ω are real characteristic frequencies of the permeability. Equations (5) and (6) then yield the wave number and the index of refraction,

$$k(\omega) = \pm \frac{\omega}{c}\sqrt{\frac{(\omega^2 - \omega_a^2)(\omega^2 - \omega_b^2)}{(\omega^2 - \omega_0^2)(\omega^2 - \Omega^2)}},$$

$$n(\omega) = \sqrt{\frac{(\omega^2 - \omega_a^2)(\omega^2 - \omega_b^2)}{(\omega^2 - \omega_0^2)(\omega^2 - \Omega^2)}}. \tag{7}$$

A plot of the dispersion relation ω versus k is presented in Fig. 1 for the case $\Omega < \omega_0 < \omega_b < \omega_a$. It is seen that ω_a and ω_b are cutoff frequencies, while ω_0 and Ω are resonances. Important for our analysis is the existence of two negative wave branches in the frequency range $\omega_0 < \omega < \omega_b$, one associated with each sign of Eq. (7). The index of refraction $n(\omega)$ in the

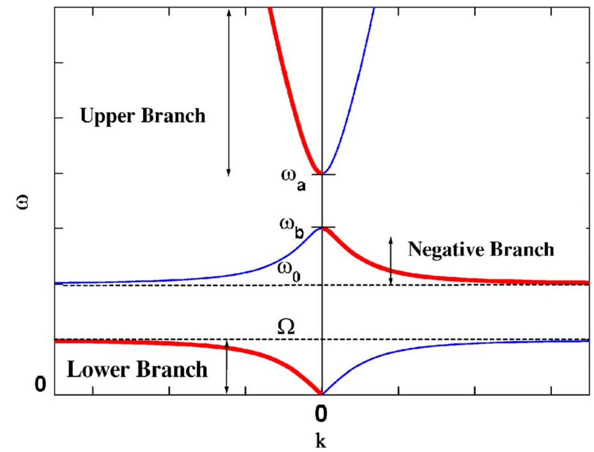

FIG. 1. (Color online) Graphical depiction of the dispersion diagram ω vs k defined by Eq. (7) with $\Omega < \omega_0 < \omega_b < \omega_a$. Three propagating branches exist: upper branch, negative index branch, and lower branch.

negative wave branches is negative, while the group speed $d\omega/dk$ and the phase speed ω/k are of opposite signs. The index of refraction is positive for $0 < \omega < \Omega$ and $\omega_a < \omega$. The sign of $n(\omega)$ is readily established by assuming that ω in Eq. (7) is a complex variable with a negative imaginary part and letting $\mathrm{Im}(\omega) \to 0$, a procedure that implies causality. Finally, there are two frequency ranges with evanescent waves, $\omega_b < \omega < \omega_a$ and $\Omega < \omega < \omega_0$.

We now explore nonlinear interactions in a negative index medium. Assuming a linear combination of harmonic waves with slowly varying envelopes $A(z,\omega_m)$, $E(z,t) = \Sigma_{\omega_m} A(z,\omega_m) e^{i[\omega_m t - k_m z]}$, where the frequency ω_m and the wave number k_m are related by the dispersion characteristics shown in Fig. 1. If a finite number of mode frequencies are in resonance, the waves interact strongly. Under this condition, the associated envelopes are governed by coupled first-order nonlinear differential equations. The focus of our analysis will be on the frequency resonance conditions and nonlinear susceptibilities when at least one mode exhibits a negative index of refraction. This occurs if the wave frequency lies between ω_0 and ω_b in Fig. 1.

We begin with the nonlinear interaction of three waves with frequencies $\omega_1(k_1)$, $\omega_2(k_2)$, and $\omega_3(k_3)$ that satisfy the energy and momentum resonance conditions, respectively,

$$\omega_3(k_3) = \omega_1(k_1) + \omega_2(k_2), \quad k_3 = k_1 + k_2. \tag{8}$$

The condition for phase matching (momentum) cannot be satisfied in normal dispersive materials because the index of refraction increases with ω. Although birefringence, quasi-phase-matching [22], and M-waveguides [23] have been successfully applied to achieve phase matching, it would be preferable that phase matching be realizable without the necessity of the aforementioned techniques. We have carried out a series of computations that demonstrate that the frequency resonance condition and perfect phase matching can be satisfied in our model if at least one wave has a negative refractive index. Results are presented in Figs. 2–6 as plots of $\bar{\omega}_1$ versus $\bar{\omega}_2$ such that Eq. (8) is satisfied. A bar over a

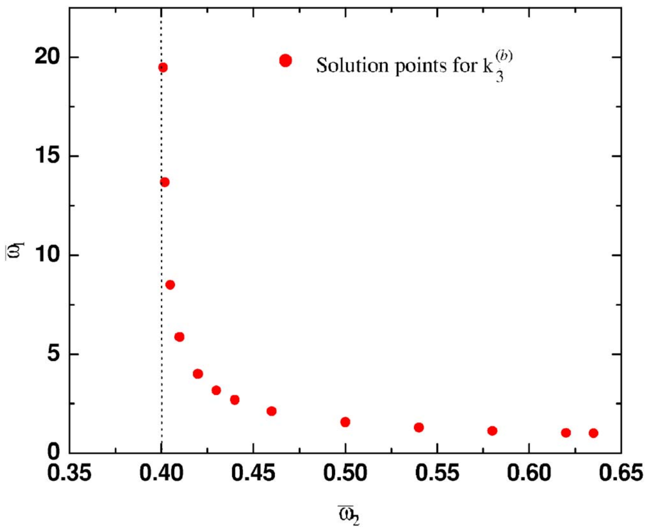

FIG. 2. (Color online) Plot of $\bar{\omega}_1$ vs $\bar{\omega}_2$ such that the resonance conditions given by Eq. (8) are satisfied. Wave 1 is on the upper branch, wave 2 is on the negative branch, and wave 3 is on the upper branch with negative group velocity. The medium parameters are $\bar{\Omega}=0.3$, $\bar{\omega}_0=0.4$, $\bar{\omega}_b=0.9$, and $\bar{\omega}_a=1.0$.

frequency variable signifies normalization with respect to ω_a. To construct these plots, frequencies ω_1 and ω_2 are assumed to lie on any of the three propagating frequency bands shown in Fig. 1. With k_2 held fixed, k_1 is varied until the frequency $\omega_3(k_3)=\omega_3(k_1+k_2)$ also lies in a propagating band. The wave numbers k_1 and k_2, as well as $k_3=k_1+k_2$, can take on positive and negative values. This process is tantamount to solving the nonlinear algebraic equation $\omega_3(k_1+k_2)=\omega_1(k_1)+\omega_2(k_2)$ for k_1 for a specified value of k_2. A color code is used in the plots to identify sections where wave 3 is either a forward or a backward propagating wave. *We adopt the definition that a wave is forward propagating if its group velocity is positive and backward propagating if its group velocity is negative*. Along a red section labeled with solid points (●), wave 3 is backward propagating and the wave number is labeled $k_3^{(b)}$; along a blue sector labeled with open points (○), wave 3 is forward propagating and the wave number is $k_3^{(f)}$.

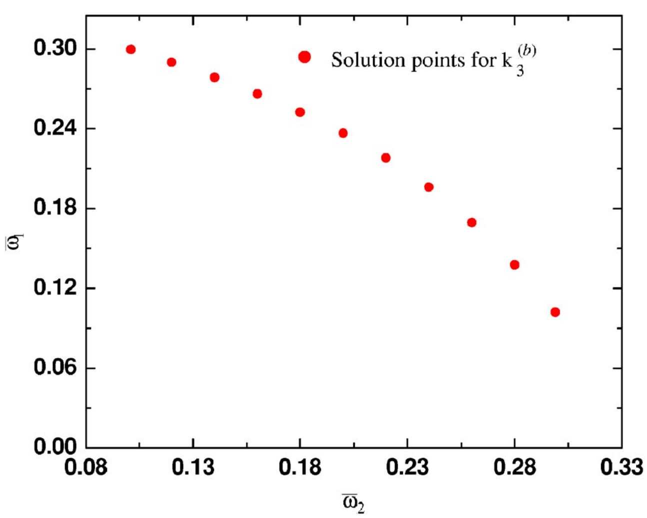

FIG. 3. (Color online) Plot of $\bar{\omega}_1$ vs $\bar{\omega}_2$ such that the resonance conditions given by Eq. (8) are satisfied. Waves 1 and 2 are on the lower branch, while wave 3 is on the negative branch with negative group velocity. The medium parameters are $\bar{\Omega}=0.3$, $\bar{\omega}_0=0.4$, $\bar{\omega}_b=0.9$, and $\bar{\omega}_a=1.0$.

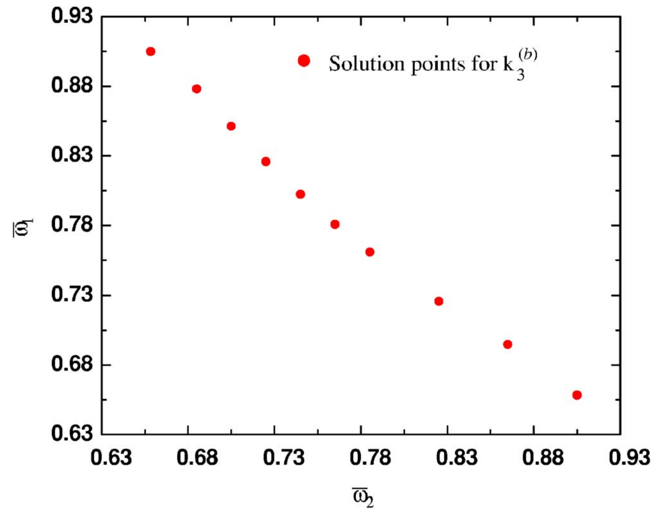

FIG. 4. (Color online) Plot of $\bar{\omega}_1$ vs $\bar{\omega}_2$ such that the resonance conditions given by Eq. (8) are satisfied. Wave 1 is on the negative branch, wave 2 is on the negative branch, and wave 3 is on the upper branch with negative group velocity. The medium parameters are $\bar{\Omega}=0.3$, $\bar{\omega}_0=0.4$, $\bar{\omega}_b=0.9$, and $\bar{\omega}_a=1.0$.

Figure 2 depicts a case with wave 1 on the upper positive index branch and wave 2 on the negative index branch, while wave 3 appears on the upper positive index branch. Wave 3 in this case is backward propagating. In this wave configuration, a positive index wave is excited by mixing a positive

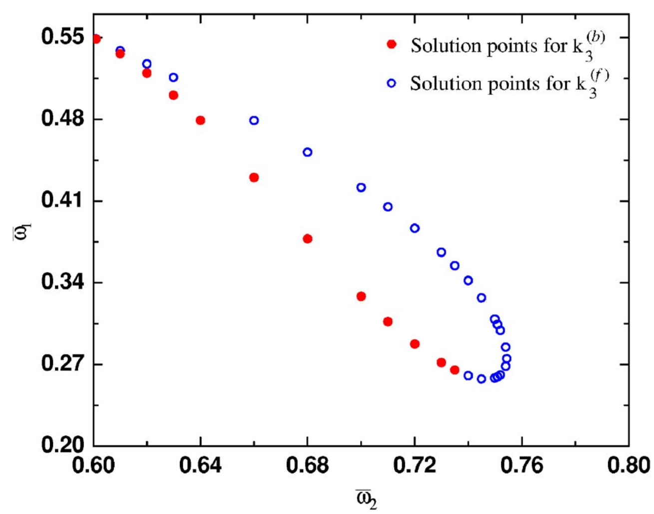

FIG. 5. (Color online) Plot of $\bar{\omega}_1$ vs $\bar{\omega}_2$ such that the resonance conditions given by Eq. (8) are satisfied. Wave 1 is on the lower branch, wave 2 is on the negative branch, and wave 3 is on the upper branch. Note the "teardrop" shape of the curve. Also note that the group velocity for wave 3 may be positive or negative depending on the input frequencies. The medium parameters are $\bar{\Omega}=0.55$, $\bar{\omega}_0=0.6$, $\bar{\omega}_b=0.8$, and $\bar{\omega}_a=1.0$.

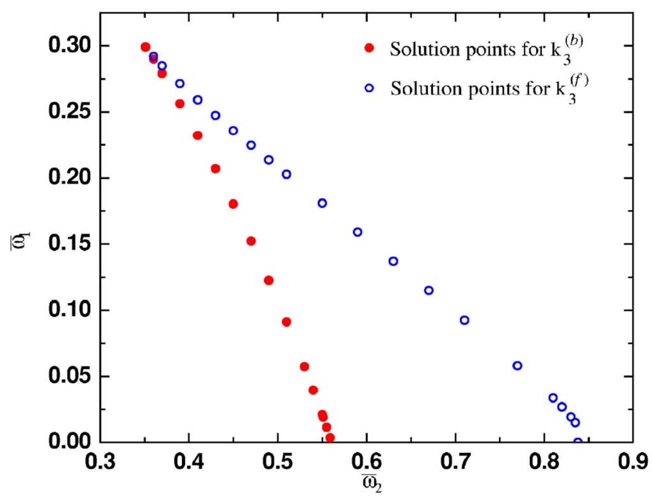

FIG. 6. (Color online) Plot of $\bar{\omega}_1$ vs $\bar{\omega}_2$ such that the resonance conditions given by Eq. (8) are satisfied. Wave 1 is on the lower branch, wave 2 is on the negative branch, and wave 3 is on the negative branch. Note that the group velocity for wave 3 may be positive or negative. The medium parameters are $\bar{\Omega}=0.3$, $\bar{\omega}_0=0.4$, $\bar{\omega}_b=0.9$, and $\bar{\omega}_a=1.0$.

index wave with a negative index wave. Figure 3 illustrates the mixing of two positive index waves to produce a negative index wave. Waves 1 and 2 are both on the lower positive index branch, while the resulting wave 3 is on the negative index branch. Two negative index waves can also mix to excite a positive index wave. This case is illustrated in Fig. 4, where waves 1 and 2 are on the negative index branch. The excited wave 3 is backward propagating on the upper positive index branch. The configuration in Fig. 5 is interesting because the $\bar{\omega}_1$ versus $\bar{\omega}_2$ plot is a closed curve in the form of a "teardrop." Waves 1 and 2 are on the lower positive index branch and the negative index branch, respectively. Wave 3 is on the upper positive index branch. Observe that both forward and backward propagating waves can be excited in this case. Figure 6 illustrates the mixing of a positive index wave and a negative index wave to produce a negative index wave. Wave 1 is on the lower positive index branch, wave 2 is on the negative index branch, and the resulting wave 3 is on the negative index branch. Forward and backward propagating waves exist in this case.

The interaction of the three waves in resonance is governed by the nonlinear susceptibilities of the medium. These nonlinear coefficients may be positive or negative depending on the frequencies of the interacting waves relative to the resonant frequency ω_0 of the material. For the noncentrosymmetric medium governed by Eq. (3), the second-order nonlinear susceptibility $\chi^{(2)}(\omega_m,\omega_n,\omega_q)$ for three-wave mixing can be derived as [18]

$$\chi^{(2)}(\omega_q,\omega_m,\omega_n)=\frac{a(Nq^3/m^2)}{D(\omega_m)D(\omega_n)D(\omega_q)}, \tag{9}$$

where $\omega_q=\omega_m+\omega_n$ and $D(\omega)\equiv\omega_0^2-\omega^2$. The sign of $\chi^{(2)}(\omega_q,\omega_m,\omega_n)$ is determined by the sign of the triple product $D(\omega_m)D(\omega_n)D(\omega_q)$, which in turn is determined by the sign of the $D(\omega)$: $D(\omega)$ is negative if $\omega^2>\omega_0^2$ and positive if $\omega^2<\omega_0^2$. Thus, the sign of the second-order nonlinear susceptibility can be engineered by specifying appropriately the values of the input frequencies ω_m and ω_n relative to the resonance frequency ω_0. For example, if ω_m^2 and ω_n^2 are both less than ω_0^2 while ω_q^2 exceeds ω_0^2, $\chi^{(2)}(\omega_q,\omega_m,\omega_n)$ will be negative. On the other hand, $\chi^{(2)}(\omega_q,\omega_m,\omega_n)$ will be positive if ω_m^2 and ω_n^2 exceed ω_0^2 while ω_q^2 is less than ω_0^2. It is readily established that $\chi^{(2)}(\omega_q,\omega_m,\omega_n)$ is negative for the wave-mixing processes depicted in Figs. 2–4 and positive for the processes depicted in Figs. 5 and 6.

The preceding analysis treats three-wave mixing in a noncentrosymmetric dispersive medium with a negative index wave. Four-wave mixing in the presence of a negative index wave is also possible, in either centrosymmetric or noncentrosymmetric media. The analysis of four-wave mixing is similar to that of three-wave mixing. A linear combination of interacting waves is assumed. Four waves interact strongly when the frequencies satisfy the resonance conditions,

$$\omega_4(k_4)=\omega_1(k_1)+\omega_2(k_2)+\omega_3(k_3),\quad k_4=k_1+k_2+k_3. \tag{10}$$

For a centrosymmetric nonlinear medium, the mixing process is governed by Eq. (4). The third-order nonlinear susceptibility that is derived from this equation reads [18]

$$\chi^{(3)}(\omega_s,\omega_m,\omega_n,\omega_q)\equiv\frac{b(Nq^4/m^3)}{D(\omega_m)D(\omega_n)D(\omega_q)D(\omega_s)},$$

$$\omega_s=\omega_m+\omega_n+\omega_q. \tag{11}$$

For a noncentrosymmetric medium, which Eq. (3) governs, $\chi^{(3)}(\omega_s,\omega_m,\omega_n,\omega_q)$ has a form similar to but somewhat more complicated than Eq. (11). We treat here only the centrosymmetric medium. Most general results that we find can readily be extended to the noncentrosymmetric medium. We have performed computations of Eq. (10) with waves 1, 2, and 3 on the lower positive index branch of Fig. 1. The wave numbers of the three waves are positive. The resonance conditions of Eq. (10) yield a fourth wave that lies on the negative index branch between with $k_4>0$. Because k_4 is positive, the negative index wave has a negative group velocity. The numerical values of the normalized frequencies and wave numbers in our computations are the following: $(\bar{\omega}_1,\bar{k}_1)=(0.10,0.83)$, $(\bar{\omega}_2,\bar{k}_2)=(0.15,1.37)$, $(\bar{\omega}_3,\bar{k}_3)=(0.20,2.22)$, and $(\bar{\omega}_4,\bar{k}_4)=(0.45,4.42)$. For these computations, the four characteristic frequencies of the model, normalized to the cutoff frequency ω_a, have the following values: $\bar{\Omega}=0.3$, $\bar{\omega}_0=0.4$, $\bar{\omega}_b=0.9$, and $\bar{\omega}_a=1.0$. The negative index branch lies in the frequency range $0.4<\bar{\omega}<0.9$.

It is important to determine the sign of $\chi^{(3)}(\omega_4,\omega_1,\omega_2,\omega_3)$ because of its strong effect on the wave amplitudes. From Eq. (11), we can see that $\chi^{(3)}(\omega_4,\omega_1,\omega_2,\omega_3)$ can take on positive or negative values depending on the net sign of the product $D(\omega_1)D(\omega_2)D(\omega_3)D(\omega_4)$. In the aforementioned numerical example of four-wave mixing, the coefficients $D(\omega_1)$, $D(\omega_2)$, and $D(\omega_3)$ are positive, while $D(\omega_4)$ is negative. Therefore,

$\chi^{(3)}(\omega_4,\omega_1,\omega_2,\omega_3)$ is negative. Other wave configurations that achieve a negative third-order nonlinear susceptibility are also possible. In summary we find that the sign of $\chi^{(3)}(\omega_4,\omega_1,\omega_2,\omega_3)$ can be reversed if a negative index wave is present in the third-order mixing process.

Negative $\chi^{(3)}$ produced by the aforementioned technique can be useful in reversing pulse distortion due to nonlinearities, as mentioned earlier. Negative $\chi^{(3)}$ has been experimentally produced in semiconductors [24]. However, to do this, it is necessary to operate in the spectral vicinity of the fundamental absorption edge, which has the disadvantage of two-photon absorption effects. Our technique based on propagation in negative index media avoids these disadvantages.

In conclusion, we have shown novel nonlinear wave-mixing and susceptibility properties of negative refractive index materials and the potential benefits in the areas of optical communication and solitons. As negative index media move more toward the optical regime [25] and research continues in reducing their losses [26], the nonlinear properties of these media will be of great interest. As research progresses in nonlinearities of negative refractive index media, novel nonlinear optical signal processing capabilities will emerge that either have no counterparts or are very difficult to realize in the positive index world. As a final comment, we point out that the nonlinear interactions with negative index waves that we have explored here are not unique to dispersive optical media. They will likely occur in any media that have gaps in the linear dispersion characteristics resulting from periodicities. An example of such media would be a hot magnetized plasma [27].

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