Enhanced Kerr nonlinearity in a negative refractive atomic medium

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The Kerr nonlinearity of a left-handed material is analyzed in a four-level atomic system. It is shown that, due to the effect of quantum interference, a large enhanced Kerr nonlinearity accompanied by vanishing absorption can be realized via choosing appropriate parameters in this negative refraction atomic medium. It not only shows the large nonlinearity but also acts as the phase and amplitude compensating effects.

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Recently, an interesting research about left-handed material (LHM)[1] has attracted considerable attention because of its surprising and counterintuitive electromagnetic and optical properties, such as the reversals of both Doppler shift and Cerenkov effect[1], amplification of evanescent waves[2], subwavelength focusing[2,3], abnormal longitudinal and lateral shifts[4,5]. This material has a negative refractive index when the permittivity and permeability are negative simultaneously[1]. In LHM, the wave vector is opposite to the direction of energy propagation, so that this material acts as a phase compensating effect in contrast to the ordinary materials[6]. This makes it have the potential applications in perfectlens[7,8] and spatial filtering[9]. Up to now, there are several approaches to the realization of LHM, including artificial composite metamaterials[10,11], photonic crystal structures[12,13], and transmission line simulation[14] as well as photonic resonant materials (coherent atomic vapor)[15−19]. Very recently, some nonlinear properties of LHM have been studied in metamaterials, such as the hysteresis-type dependence of the magnetic permeability on the field intensity[20], second-harmonic generation (SHG)[21], optical parametric amplification (OPA)[22], and optical bistability (OB)[23]. It opens a new branch of nonlinear optics.

Although some linear properties of LHM have been studied in atomic systems[15−19], to our knowledge, there are few literatures to investigate the nonlinearity of a negative refractive atomic medium. In this Letter, we investigate the Kerr nonlinearity of LHM in a four-level atomic system, where the negative refraction can be realized by quantum coherence[18]. We find that the nonlinear susceptibilities can be controlled by the control and pump fields, and the focusing or defocusing effects occur in this left-handed material. We also find that a large enhanced Kerr nonlinearity accompanied by vanishing absorption can be realized via choosing appropriate parameters. Therefore, we may not only obtain the large nonlinearity but also realize the phase compensating effect to the transmission wave and amplitude compensating effect to the evanescent wave.

We consider a four-level atomic system as that in Ref. [18], which is shown in Fig. 1. The two lower levels $|1\rangle$ and $|2\rangle$, have the same parity with the magnetic dipole element $\mu_{12} = \langle 1|\hat{\vec{\mu}}|2\rangle \neq 0$. The two upper levels $|3\rangle$ and $|4\rangle$, which have opposite parity with electric dipole element $d_{34} = \langle 3|\hat{\vec{d}}|4\rangle \neq 0$, are coupled by a weak probe electric field $\Omega_{\rm p}$. The levels $|1\rangle$ and $|3\rangle$ are coupled by a control field $\Omega_{\rm c}$ and levels $|2\rangle$ and $|4\rangle$ are coupled by a strong pump field $\Omega_{\rm s}$. Here, $\Omega_i(i = {\rm p,c,s})$ are the Rabi frequencies of the probe, control, and pump fields respectively and the transition $|3\rangle \leftrightarrow |1\rangle$ is assumed to be a two-photon process.

The Hamiltonian of the system with rotating-wave approximations reads

$$H = \hbar \begin{bmatrix} 0 & 0 & -\Omega_{\rm c} & 0 \\ 0 & \delta_{12} & 0 & -\Omega_{\rm s} \\ -\Omega_{\rm c} & 0 & \delta_{13} & -\Omega_{\rm p} \\ 0 & -\Omega_{\rm s} & -\Omega_{\rm p} & \delta_{14} \end{bmatrix}, \quad (1)$$

where $\delta_{12} = \Delta_{\rm p} + \Delta_{\rm c} - \Delta_{\rm s}$, $\delta_{13} = \Delta_{\rm c}$ and $\delta_{14} = \Delta_{\rm p} + \Delta_{\rm c}$, in which $\Delta_{\rm p} = \nu_{\rm p} - (\omega_4 - \omega_3)$, $\Delta_{\rm c} = 2\nu_{\rm c} - (\omega_3 - \omega_1)$, and $\Delta_{\rm s} = \nu_{\rm s} - (\omega_4 - \omega_2)$ denote the detunings of the probe, control, and pump fields, respectively, and they satisfy the relation $\Delta_{\rm c} = \Delta_{\rm s}$; $\omega_{ij} = \omega_i - \omega_j$ is the transition frequency from levels $|i\rangle$ to $|j\rangle$ $(i, j = 1, 2, 3, 4)$; ν_i $(i = {\rm p,c,s})$ are the frequencies of the probe, control,

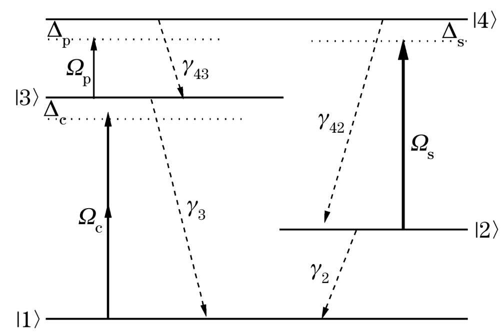

Fig. 1. Scheme of a four-level atom interacting with the probe, pump, and control fields.

and the pump fields, respectively, in which the frequencies of the fields are assumed to satisfy $\nu_s = 2\nu_c$[18].

It is well known that the response of the atomic medium to the probe field is governed by its polarization and magnetization. Applying the perturbative method, we solve the density matrix equations and ultimately get the linear electric and magnetic susceptibilities:

$$\alpha_e^{(1)} = \frac{|d_{34}|^2}{\varepsilon_0 \hbar} \times \frac{\Omega_c \Omega_s}{(\delta_{13} - i\Gamma_{13})[(\delta_{12} - i\Gamma_{12})(\delta_{14} - i\Gamma_{14}) - |\Omega_s|^2]}, \quad (2)$$

$$\alpha_m^{(1)} = \frac{c\mu_0 |\mu_{12}| |d_{34}|}{\eta \hbar} \times \frac{\Delta |\Omega_c|^2}{(\delta_{13} + i\Gamma_{13})[(\delta_{12} - i\Gamma_{12})(\delta_{14} - i\Gamma_{14}) - |\Omega_s|^2]}, \quad (3)$$

where $\Delta = \frac{(\delta_{12}-i\Gamma_{12})(\delta_{12}-\delta_{13}-i\Gamma_{23})+|\Omega_s|^2}{(\delta_{12}-\delta_{13}-i\Gamma_{23})(\delta_{14}-\delta_{13}-i\Gamma_{34})-|\Omega_s|^2}$, μ_0 is the permeability of vacuum, c is the speed of light in vacuum and η is a unitary complex number depending on the polarization of the probe field $\vec{E}_p$; the damping rates $\Gamma_{ij} = (\gamma_i + \gamma_j)/2 (i, j = 1-4, i \neq j)$, where γ_i $(i = 2, 3)$ and γ_{4j} $(j = 2, 3)$ denote the spontaneous emission rate from levels $|i\rangle$ to $|1\rangle$ and $|4\rangle$ to $|j\rangle$, respectively, and $\gamma_4 = \gamma_{43} + \gamma_{42}$, $\gamma_1 = 0$.

In order to obtain the left-handed material, the atomic gas should be dense[15,16], so one should consider the local field effect that results from the dipole-dipole interaction between neighboring atoms[24]. According to the Clausius-Mossotti relation[24,25], the macroscopic linear electric permittivity and magnetic permeability for a gas with atomic density N become

$$\varepsilon_r^L = 1 + \chi_e^{(1)} = 1 + \frac{N\alpha_e^{(1)}}{1 - \frac{N\alpha_e^{(1)}}{3}}, \quad (4)$$

$$\mu_r^L = \frac{1}{1 - \chi_m^{(1)}} = \frac{1 + \frac{2N\alpha_m^{(1)}}{3}}{1 - \frac{N\alpha_m^{(1)}}{3}}. \quad (5)$$

Using the same method, we get the third-order susceptibility

$$\chi^{(3)} = \frac{N |d_{34}| \rho_{43}^{(3)}}{\varepsilon_0 |E_p|^2 E_p} = \frac{N |d_{34}|^4 \rho_{43}^{(3)}}{\varepsilon_0 \hbar^3 |\Omega_p|^2 \Omega_p}, \quad (6)$$

where $\rho_{43}^{(3)}$ is the third-order coherent term, whose general form is very tedious, so we only present the numerical analysis subsequently. Without loss of the generality, here we only consider the electric nonlinearity because the magnetic response is much smaller than the electric response of the atomic medium.

In the following, we will numerically calculate the response of the medium to the probe field for the case of neon atoms. All the parameters are scaled by $\gamma_{43} = \gamma = 100$ MHz. The wavelength of resonant probe field transition is 5.4 μm. The pump field is resonant at 352 nm and the control field drives a two-photon transition at 704 nm. Typical values for $\gamma_{42} = \gamma_3 = \gamma$ and $\gamma_2 = 0.01\gamma$ are used. The detunings of the control and pump fields $\Delta_c = \Delta_s = -0.3\gamma$. The atomic density is $N = 2.5 \times 10^{25}$ m^{-3}. The Rabi frequency of the pump field is $\Omega_s = \gamma$.

Firstly, we investigate the linear response of the medium to the probe field. The probe-detuning dependence of the real and imaginary parts of linear permittivity ε_r^L and permeability μ_r^L are plotted in Fig. 2. It is shown that $\mathrm{Re}[\varepsilon_r^L]$ and $\mathrm{Re}[\mu_r^L]$ are negative simultaneously at the region $-1.2\gamma < \Delta_p < 1.9\gamma$ (grey region in Fig. 2). Compared with Ref. [18], the negative refraction band is widened because the different parameters are chosen. We also find that the linear absorption may be reduced to zero when negative refraction occurs. It is identical to the result of Ref. [18].

In addition, we focus on the nonlinear response of the medium in the negative refraction region (grey region in Fig. 3). Figure 3 shows the variation of the Kerr nonlinearity $\mathrm{Re}[\chi^{(3)}]$, nonlinear absorption $\mathrm{Im}[\chi^{(3)}]$, and linear absorption $\mathrm{Im}[\varepsilon_r^L]$ as a function of the probe detuning Δ_p. We find that the linear absorption $\mathrm{Im}[\varepsilon_r^L]$ is very small in a wide frequency band. Especially, $\mathrm{Im}[\varepsilon_r^L]$ and $\mathrm{Im}[\chi^{(3)}]$ respectively have two zero points, and the right ones overlap in negative refraction region. That is to say, there is no absorption in this frequency point

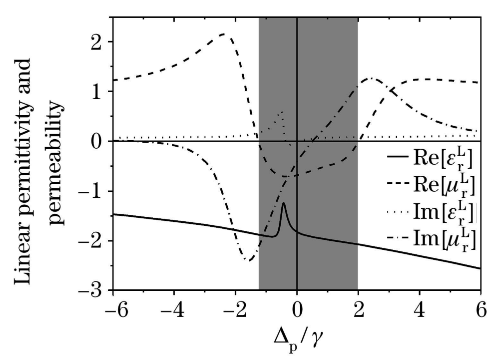

Fig. 2. Probe-detuning dependence of the real and imaginary parts of linear permittivity ε_r^L and permeability μ_r^L. The parameters are: $\gamma_{42} = \gamma_3 = \gamma$, $\gamma_2 = 0.01\gamma$, $\Delta_c = \Delta_s = -0.3\gamma$, $\Omega_s = \gamma$, $\Omega_c = 0.4\gamma$, $N = 2.5 \times 10^{25}$ m^{-3}. The grey region is the negative refraction band.

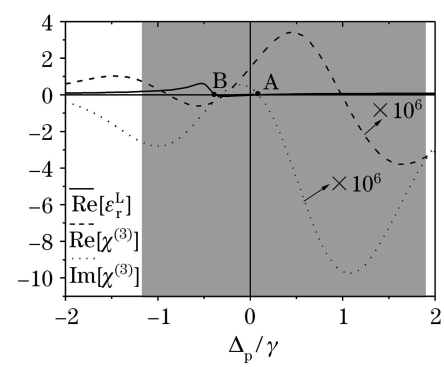

Fig. 3. Variation of the Kerr nonlinearity $\mathrm{Re}[\chi^{(3)}]$ (dashed curve), nonlinear absorption $\mathrm{Im}[\chi^{(3)}]$ (dotted curve), and linear absorption $\mathrm{Im}[\varepsilon_r^L]$ (solid curve) as a function of the probe detuning Δ_p. The parameters are the same to Fig. 2. The grey region is the negative refraction band.

(dot A in Fig. 3). We also find that the third-order susceptibility $\mathrm{Re}[\chi^{(3)}]$ reaches 10^{-6} times of the linear susceptibility at this frequency. Its amplitude is dramatically enhanced 9 orders than that in the conventional system, in which the radio of third-order to first-order susceptibility, $\mathrm{Re}[\chi^{(3)}]/\mathrm{Re}[\chi^{(1)}]$, is about 10^{-15}[26]. As we known, the refractive index can be enhanced by the atomic coherence[27]. Here, this enhanced nonlinear effect is also caused by the intense quantum interference as mentioned in Schmidt's scheme[28]. Hence, a large enhanced Kerr nonlinearity with vanishing absorption can be obtained in this negative refraction atomic medium. Moreover, we find that it shows different nonlinear responses at different frequencies in negative refraction region. It is evident that the third-order susceptibility shows different signs in two zero points of linear absorption: $\mathrm{Re}[\chi^{(3)}] < 0$ in the left zero absorption point (dot B in Fig. 3), while $\mathrm{Re}[\chi^{(3)}] > 0$ in the right one (dot A in Fig. 3). It is well known that focusing effect occurs when the third-order nonlinearity is positive, while defocusing effect takes place when the third-order nonlinearity is negative. Thus, the focusing or defocusing effects with vanishing linear absorption could take place at different frequencies in this negative refraction atomic medium. In addition, due to the negative refraction, phase compensating effect for propagating wave and amplitude compensating effect for evanescent wave are established in LHMs[6]. Owing to these effects, both propagating and evanescent waves could propagate in LHMs. Therefore, our scheme may not only realize the enhanced Kerr nonlinearity with vanishing absorption but also act as the phase and amplitude compensating effects.

Secondly, we will discuss the modulation effect of the control field to the Kerr nonlinearity. The real and imaginary parts of the nonlinear susceptibility $\chi^{(3)}$ as a function of probe-detuning Δ_p with different control fields are plotted in Fig. 4. Adjusting the Rabi frequency of the control field Ω_c from 0.1γ to 0.5γ, it exhibits different third-order dispersion and absorption behaviors. With

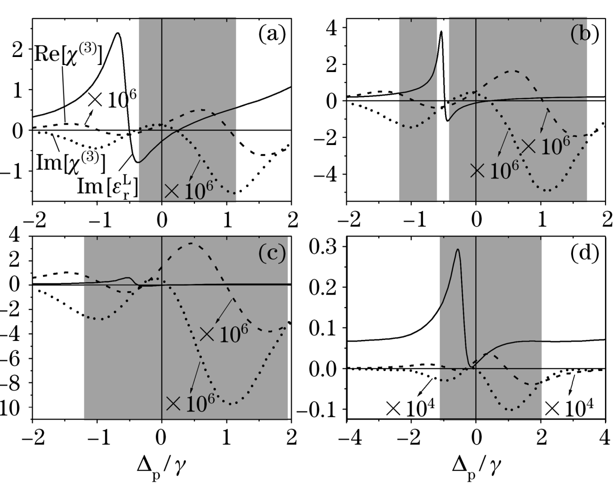

Fig. 4. Kerr nonlinearity $\mathrm{Re}[\chi^{(3)}]$ (dashed curve), nonlinear absorption $\mathrm{Im}[\chi^{(3)}]$ (dotted curve), and linear absorption $\mathrm{Im}[\varepsilon_\mathrm{r}^\mathrm{L}]$ (solid curve) as a function of the probe detuning Δ_p with different control fields Ω_c. (a) 0.1γ; (b) 0.2γ; (c) 0.4γ; (d) 0.5γ. The other parameters are the same to Fig. 2. The grey region is the negative refraction band.

the increase of the control filed, the third nonlinearity $\mathrm{Re}[\chi^{(3)}]$ increases, while the third-order absorption also increases in the negative refraction region. Fortunately, the linear absorption decreases with the increase of control field. It is shown that the third-order absorption is much smaller than the linear absorption from Fig. 4. So the total absorption decreases with the increase of control field. However, we find that the linear absorption is impossible to be zero when Ω_c approaches to 0.5γ (see Fig. 4(d)). Compared these four cases, the best value of the Rabi frequency of the control field is $\Omega_\mathrm{c} = 0.4\gamma$. In this case, a large third-order nonlinearity accompanied by vanishing absorption may be obtained in negative refractive region (see Fig. 4(c)). Therefore, we can get a large Kerr nonlinearity with vanishing absorption via choosing appropriate parameters in this negative refractive atomic medium.

In conclusion, we have investigated the Kerr nonlinearity in a negative refractive atomic system. It is shown that the nonlinear susceptibility could be controlled by the control and pump fields, and the focusing or defocusing effect occurs in this left-handed material. We also find that a large enhanced Kerr nonlinearity accompanied by vanishing absorption can be realized via choosing appropriate parameters. It may not only show the large Kerr nonlinearity but also act as the phase and amplitude compensating effects. It will have the potential applications in nonlinear left-handed lens[29], frequency conversion[30], the generation of the optical solitons[31], and so on.

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