Enhanced Kerr nonlinearity in a quantized four-level graphene nanostructure

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In this paper, a new model is proposed for manipulating the Kerr nonlinearity of right-hand circular probe light in a monolayer of graphene nanostructure. By using the density matrix equations and quantum optical approach, the third-order susceptibility of probe light is explored numerically. It is realized that the enhanced Kerr nonlinearity with zero linear absorption can be provided by selecting the appropriate quantities of controllable parameters, such as Rabi frequency and elliptical parameter of elliptical polarized coupling field. Our results may be useful applications in future all-optical system devices in nanostructures.

Keywords: Kerr nonlinearity, graphene nanostructure, linear absorption

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1. Introduction

Electromagnetically induced transparency (EIT),[1] in atomic systems and semiconductor quantum wells and quantum dot structures has led to many unforeseen phenomena such as lasing without inversion,[2] four-wave mixing,[3,4] and so on.[5–15] EIT exploits destructive quantum interference to make an otherwise opaque medium almost transparent. This feature of EIT medium (transparent to the applied fields) makes it suitable for application in nonlinear optical science. In nonlinear optics, it is favorable to have a strong nonlinear medium in low light intensities. Therefore, Kerr nonlinearity of such medium plays essential roles for studying some other phenomena which require strong nonlinear medium such as quantum nondemolition measurements,[16] quantum bit regeneration,[17] quantum state teleportation,[18] and the generation of the optical solitons.[19] The Kerr nonlinearity of the medium corresponds to the real part of third-order susceptibility of weak probe light which propagates through it. The features of Kerr nonlinearity of weak probe light in multilevel atomic systems have been reported by many research groups.[20–29] It has been shown that the Kerr nonlinearity can be manipulated by many different mechanisms. For instance, Niu *et al.* proposed a model for giant Kerr-nonlinearity-based spontaneously generated coherence (SGC) in a usual three-level atomic system.[26] In another study done by Niu *et al.*, the role of the double dark resonances on Kerr nonlinearity has been discussed.[27] Asadpour *et al.*[28] proposed a model for studying the enhanced Kerr nonlinearity via quantum interference from spontaneous emission. The properties of Kerr nonlinearity in a semiconductor quantum well and quantum dot-based different mechanisms have also been investigated.[30–35] For instance, phase control of Kerr nonlinearity in a quantum well nanostructure has been suggested by Asadpour *et al.*[30] The role of biexciton coherence on Kerr nonlinearity of probe light propagated in a multiple quantum well nanostructure has also been reported.[33] Moreover, it has been found that exciton spin relaxation can lead to manipulating of Kerr nonlinearity in a multiple quantum well nanostructure.[34] The giant Kerr nonlinearity in a crystal of molecular magnet has also been discussed.[35]

On the other hand, the optical and electronic features of graphene nanostructure due to its important usages in condense matter and optical physics have been investigated recently.[36–39] The properties of electrons near the Dirac points make it possible for interacting by electromagnetic fields.[40–42] The nonlinear optical features of graphene nanostructure in the presence of strong magnetic field has been discussed very recently in four-wave mixing,[43,44] generation of polarized-entangle photon, nonlinear frequency conversion of THz surface plasmons based on the nonlinear optical interaction,[45,47] optical bistability,[48,49] and slow and fast light propagating.[50] To the best of our knowledge, the enhanced Kerr nonlinearity of graphene with zero linear and nonlinear absorption has not been reported theoretically or experimentally. In this paper, we suggest a model based on the quantized four-level graphene nanostructure for enhancing the Kerr nonlinearity by using a probe and an elliptical polarized control field. We show that by selecting appropriate values for Rabi frequency and elliptical parameter of control field, the Kerr nonlinearity can be enhanced at certain values of probe field detuning.

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2. Model and equations

A single layer of graphene system in the presence of strong magnetic field is presented in a four-level scheme (Fig. 1). Due to the presence of external magnetic field, the unbroken energy bands near Dirac points split into individual Landau levels. It is well known that the selection rules in graphene obey from $\Delta|n| = \pm 1$, where n is the energy quantum number. It should be noted that the optical transitions between Landau levels in the range of 0.01 T–10 T can be located in the infrared and terahertz regions due to $\hbar\omega_c \simeq 36\ \mathrm{B}^{1/2}\cdot\mathrm{T}^{1/2}\cdot\mathrm{meV}$. The mentioned graphene system interacts with a probe and an elliptical polarized control field. The elliptically polarized control field can be regarded as a combination of the right- and left-circularly polarized components which can be obtained by using a quarter wave plate (QWP).[51] After passing a vertical polarized coupling field with amplitude E_0 through QWP, the elliptical polarized coupling field can be obtained with $E_0 = E_+\sigma^+ + E_-\sigma^-$, where $E_+ = E_0/\sqrt{2}(\cos\theta + \sin\theta)\,\mathrm{e}^{\mathrm{i}\theta}$, and $E_- = E_0/\sqrt{2}(\cos\theta - \sin\theta)\,\mathrm{e}^{-\mathrm{i}\theta}$. Parameter θ is the ellipticity parameter. The unit vectors of right- and left-circularly polarized basis are denoted by σ^+ and σ^-, respectively. The probe light with right-hand circular polarization, frequency ω_p, and Rabi frequency Ω_p drives transition $|1\rangle \leftrightarrow |4\rangle$ and elliptical polarized coupling field with frequency ω_0 and Rabi frequency Ω_c act on transitions $|4\rangle \leftrightarrow |3\rangle$ and $|3\rangle \leftrightarrow |2\rangle$, respectively. The Rabi frequencies for transitions $|4\rangle \leftrightarrow |3\rangle$ and $|3\rangle \leftrightarrow |2\rangle$ are equal to $\Omega_3 = \Omega_c(\cos\theta - \sin\theta)\,\mathrm{e}^{-\mathrm{i}\theta}$ and $\Omega_3 = \Omega_c(\cos\theta + \sin\theta)\,\mathrm{e}^{\mathrm{i}\theta}$, respectively. It is assumed that $\mu_{43} = \mu_{32} = \mu$ and $\Omega_c = \mu E_0/2\sqrt{2}\hbar$. The corresponding optical transition between $|m\rangle$ and $|n\rangle$ denoted by $\mu = \langle m|\,\mu\,|n\rangle = e\langle m|\,\boldsymbol{r}\,|n\rangle = \mathrm{i}\hbar/\varepsilon_m - \varepsilon_n\langle m|\,v_F\boldsymbol{\sigma}\,|n\rangle$, where $\boldsymbol{\sigma} = (\sigma_x, \sigma_y)$ is the Pauli matrix vector and $v_F = 3\gamma_0/2\hbar a \simeq 10^6$ m/s is Fermi velocity ($\gamma_0 \simeq 2.8$ eV and $a = 1.42$ Å are the nearest neighbor hopping energy and C–C spacing).[52] The corresponding Hamiltonian for a monolayer of graphene system is governed by

$$\hat{H}_0 = v_F \begin{pmatrix} 0 & \hat{\pi}_x - \mathrm{i}\hat{\pi}_y & 0 & 0 \\ \hat{\pi}_x + \mathrm{i}\hat{\pi}_y & 0 & 0 & 0 \\ 0 & 0 & 0 & \hat{\pi}_x + \mathrm{i}\hat{\pi}_y \\ 0 & 0 & \hat{\pi}_x - \mathrm{i}\hat{\pi}_y & 0 \end{pmatrix}, \quad (1)$$

where $\hat{\boldsymbol{\pi}} = \hat{\boldsymbol{p}} + e\boldsymbol{A}/c$ denotes the generalized momentum operator, $\hat{\boldsymbol{p}}$ is the electron momentum operator, e is the electron charge, and $\boldsymbol{A}$ is the vector potential, which is equal to $(0, Bx)$ for a static magnetic field. For considering the incident optical field on the Hamiltonian of the system, one needs to add the vector potential of the optical field ($\boldsymbol{A}_{opt} = \mathrm{i}c\boldsymbol{E}/\omega$ and $\boldsymbol{E} = \boldsymbol{E}_p + \boldsymbol{E}_0$) to the vector potential of the magnetic field in the generalized momentum operator $\hat{\boldsymbol{\pi}}$. After this interchange, the interaction Hamiltonian can be written in the following form:

$$\hat{H}_{int} = v_F\boldsymbol{\sigma}\cdot\frac{e}{c}\boldsymbol{A}_{opt}. \quad (2)$$

A standard dynamic equation for the density matrix of Dirac electrons in graphene by using the Liouville's equation

$$\frac{\partial\hat{\rho}}{\partial t} = -\frac{\mathrm{i}}{\hbar}[\hat{H}_{int}\hat{\rho}] + \hat{R}(\hat{\rho})$$

can be obtained. Here, $\hat{R}(\hat{\rho})$ indicates incoherent relaxation which may originate from disorder, interaction with phonons, and carrier–carrier interactions. The density matrix elements of the system are governed by

$$\begin{aligned}
\dot{\rho}_{22} &= -\gamma_2\rho_{22} + \mathrm{i}\Omega_2\rho_{32} - \mathrm{i}\Omega_2\rho_{23},\\
\dot{\rho}_{33} &= -\gamma_3\rho_{33} + \mathrm{i}\Omega_2\rho_{23} + \mathrm{i}\Omega_3\rho_{43} - \mathrm{i}\Omega_2\rho_{32} - \mathrm{i}\Omega_3\rho_{34},\\
\dot{\rho}_{44} &= -\gamma_4\rho_{44} + \mathrm{i}\Omega_p\rho_{14} + \mathrm{i}\Omega_3\rho_{34} - \mathrm{i}\Omega_p\rho_{41} - \mathrm{i}\Omega_3\rho_{43},\\
\dot{\rho}_{21} &= -\left(\mathrm{i}\Delta_p - \mathrm{i}\Delta_3 - \mathrm{i}\Delta_2 + \frac{\gamma_2}{2}\right)\rho_{21} + \mathrm{i}\Omega_2\rho_{31} - \mathrm{i}\Omega_p\rho_{24},\\
\dot{\rho}_{31} &= -\left(\mathrm{i}\Delta_p - \mathrm{i}\Delta_3 + \frac{\gamma_3}{2}\right)\rho_{31} + \mathrm{i}\Omega_2\rho_{21} + \mathrm{i}\Omega_3\rho_{43} - \mathrm{i}\Omega_p\rho_{34},\\
\dot{\rho}_{41} &= -\left(\mathrm{i}\Delta_p + \frac{\gamma_4}{2}\right)\rho_{41} + \mathrm{i}\Omega_p(\rho_{11} - \rho_{44}) + \mathrm{i}\Omega_3\rho_{31},\\
\dot{\rho}_{32} &= -\left(\mathrm{i}\Delta_2 + \frac{\gamma_3 + \gamma_2}{2}\right)\rho_{32} + \mathrm{i}\Omega_2(\rho_{22} - \rho_{33}) + \mathrm{i}\Omega_3\rho_{42},\\
\dot{\rho}_{42} &= -\left(\mathrm{i}\Delta_3 + \mathrm{i}\Delta_2 + \frac{\gamma_4 + \gamma_2}{2}\right)\rho_{42} + \mathrm{i}\Omega_p\rho_{12}\\
&\quad + \mathrm{i}\Omega_3\rho_{32} - \mathrm{i}\Omega_2\rho_{43},\\
\dot{\rho}_{43} &= -\left(\mathrm{i}\Delta_3 + \frac{\gamma_4 + \gamma_3}{2}\right)\rho_{43} + \mathrm{i}\Omega_p\rho_{13}\\
&\quad + \mathrm{i}\Omega_3(\rho_{33} - \rho_{44}) - \mathrm{i}\Omega_2\rho_{42}, \qquad (3)
\end{aligned}$$

where $\Delta_p = (\varepsilon_{n=1} - \varepsilon_{n=-2})/\hbar - \omega_p$, $\Delta_2 = (\varepsilon_{n=0} - \varepsilon_{n=-1})/\hbar - \omega_0$, and $\Delta_3 = (\varepsilon_{n=1} - \varepsilon_{n=0})/\hbar - \omega_0$ represent the corresponding frequency detunings, and $\varepsilon_n = \mathrm{sgn}(n)\hbar\omega_c\sqrt{|n|}$ is the energy of the Landau level for electrons near the Dirac point, with $n = 0, \pm1, \pm2, \ldots$, $\omega_c = \sqrt{2}v_F/l_c$, and $l_c = \sqrt{\hbar c/eB}$ implies the magnetic length. For achieving the third-order of susceptibility, we assume that the probe field is weaker than the elliptical coupling field. In this case density matrix elements are expanding as $\rho_{ij} = \rho_{ij}^{(0)} + \rho_{ij}^{(1)} + \rho_{ij}^{(2)} + \cdots$. The zeroth order solution of ρ_{11} will be identical, i.e., $\rho_{11}^{(0)} = 1$, and other elements are set to be zero. The first- and third-order susceptibilities $\chi^{(1)}$ and $\chi^{(3)}$ of the medium can be determined by coherence terms $\rho_{41}^{(1)}$ and $\rho_{41}^{(3)}$, respectively, as

$$\chi^{(1)} = \frac{2N|\mu_{14}|^2}{\varepsilon_r\hbar\Omega_p}\rho_{41}^{(1)}, \quad (4)$$

$$\chi^{(3)} = \frac{2N|\mu_{14}|^4}{3\varepsilon_r\hbar^3\Omega_p^3}\rho_{41}^{(3)}, \quad (5)$$

where N and ε_r are the sheet electron density of graphene and the substrate dielectric constant, respectively.

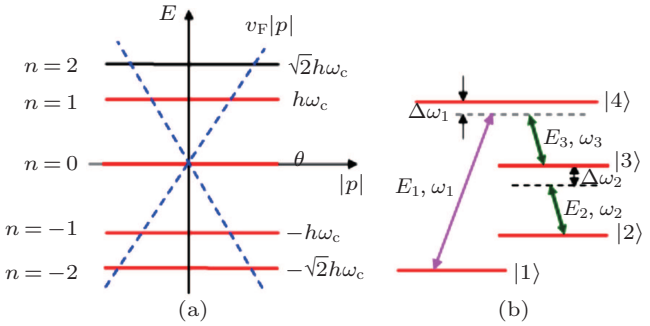


Fig. 1. (color online) (a) The LLs near the K point superimposed on the electronic energy dispersion without a magnetic field $E = \pm v_F|p|$. The magnetic field condenses the original states in the Dirac cone into discrete energies. The LLs in graphene are unequally spaced: $\propto \sqrt{B}$. (b) Energy level diagram and optical transitions in a monolayer graphene interacting with probe and elliptical polarized coupling field. Graphene monolayer is a one-atom-thick monolayer of carbon atoms arranged in a hexagonal lattice, which we will treat as a perfect two-dimensional (2D) crystal structure in the x–y plane.

3. Results and discussion

In the following section, we will analyze the third-order susceptibility through numerical simulations in detail. According to the numerical estimate based on Refs. [43]–[50], we can take a reasonable value for the decay rate and all parameters are scaled by decay rate γ. This value depends on the sample quality and the substrate used in experiments.[51,52] The electron concentration is $N \simeq 5 \times 10^{12}$ cm^{-2} and the substrate dielectric constant is $\varepsilon_r \simeq 4.5$.[53–55] The linear and nonlinear features of the susceptibility for parameter $\theta = 0$ are explored in Fig. 2. By choosing $\theta = 0$, the Rabi-frequencies of elliptical polarized field will become $\Omega_+ = \Omega_- = \Omega_c$. In this case, we will expect that due to the double dark resonances, the medium is accompanied with strong linear and nonlinear absorption. From Fig. 2, one can find that the Kerr nonlinearity of the medium (dashed line of Fig. 2(b)) corresponds to strong linear and nonlinear absorption (solid line of Figs. 2(a) and 2(b)). These properties are not suitable for using in nonlinear optics where enhanced Kerr nonlinearity with zero linear absorption is needed. For overcoming this deficiency, we examine the effect of elliptical parameter θ on the behaviors of linear and nonlinear susceptibilities. The properties of linear and nonlinear susceptibilities for $\theta = \pi/6$ are presented in Fig. 3. The linear properties are shown in Fig. 3(a) and nonlinear features are presented in Fig. 3(b). It can be easily seen that the linear absorption (solid line of Fig. 3(a)) of weak probe light at $\Delta_p = \pm 0.41\gamma$ is zero. This means that the medium for these quantities of detuning is transparent for the probe light. The slope of dispersion curve in this situation is positive corresponding to slow light propagation. Until now, these features of linear susceptibility are favorable for applications in nonlinear optics. The nonlinear behaviors of susceptibility for $\theta = \pi/6$ are presented in Fig. 3(b). It can be realized that the nonlinear absorption converts to nonlinear amplification in two

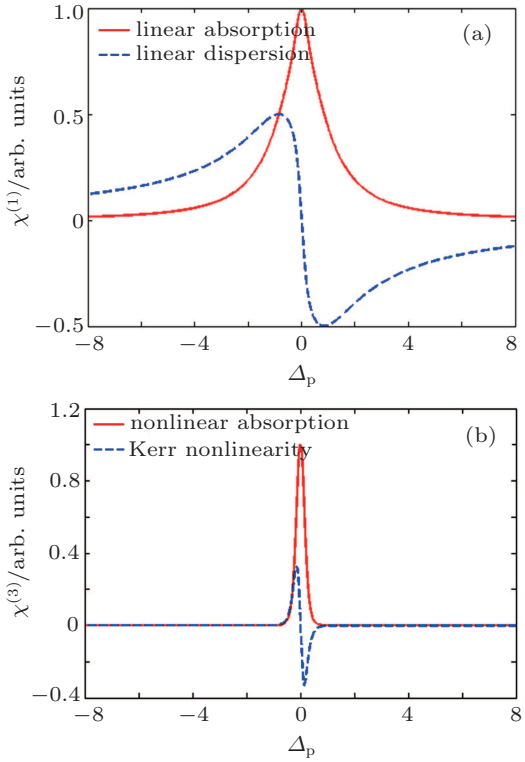


Fig. 2. (color online) (a) Linear susceptibility $\chi^{(1)}$ versus probe light detuning. Solid line corresponds to linear absorption and dashed line corresponds to dispersion. (b) Nonlinear susceptibility $\chi^{(3)}$ versus probe light detuning. Solid line corresponds to nonlinear absorption and dashed line corresponds to Kerr nonlinearity. The used parameters are $\Omega_p = 0.01\gamma$, $\Omega_c = 0.4\gamma$, $\Delta_2 = \Delta_3 = 0$, and $\theta = 0$.

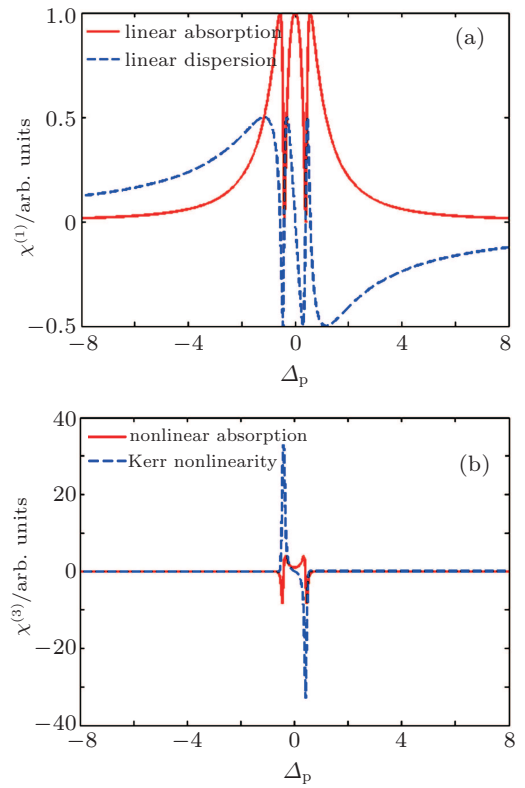


Fig. 3. (color online) (a) Linear susceptibility $\chi^{(1)}$ versus probe light detuning. Solid line corresponds to linear absorption and dashed line corresponds to dispersion. (b) Nonlinear susceptibility $\chi^{(3)}$ versus probe light detuning. Solid line corresponds to nonlinear absorption and dashed line corresponds to Kerr nonlinearity. Parameter $\theta = \pi/6$ and others are the same as Fig. 2.

detunings of probe field. In this situation, the Kerr nonlinearity is enhanced and reaches to maximum quantity at $\Delta_p = \pm 0.41\gamma$. These properties of the medium due to zero linear absorption and enhanced Kerr nonlinearity are very suitable for application in nonlinear optical technologies, where the enhanced Kerr nonlinearity at low light level is needed. Compared with the single dark resonance case, the interacting double dark resonances cause an enhancement of the Kerr nonlinearity with vanishing linear absorption. In the dressed-state picture, this case corresponds to the profound interference situation because the eigenvalues of two of the dressed states intersect.[56] For further discussion about the role of elliptical parameter θ on linear absorption and Kerr nonlinearity of the medium, we display the features of linear absorption and Kerr nonlinearity of medium versus θ in Fig. 4. It is realized that the enhanced Kerr nonlinearity is accompanied by vanishing absorption at different values of parameter θ. These results have good agreement with our previous results. At the final step, the properties of linear absorption (solid line) and Kerr nonlinearity (dashed line) versus Rabi frequency Ω_c for $\theta = 0$ (Fig. 4(a)), and $\theta = \pi/6$ (Fig. 4(b)) are plotted in Fig. 5. By careful attention to the obtained results, one can find that when we choose $\theta = 0$, by enhancing the Rabi frequency Ω_c, the Kerr nonlinearity increases and linear absorption reduces, however, the linear absorption never reaches zero. In addition, when we select $\theta = \pi/6$, the Kerr nonlinearity is enhanced and simultaneously linear absorption is vanished when Ω_c reaches about 0.4γ. This is very interesting for us to reach enhanced Kerr nonlinearity with vanishing absorption. As a result, we find that the enhanced Kerr nonlinearity with vanishing absorption is caused by simultaneous effects of elliptical parameter and Rabi frequency Ω_c. In fact, the elliptical parameter θ leads to modifying intensities of the Rabi frequencies Ω_2 and Ω_3, respectively. This means that the enhanced Kerr nonlinearity with zero linear absorption can be tuned by elliptical parameter.

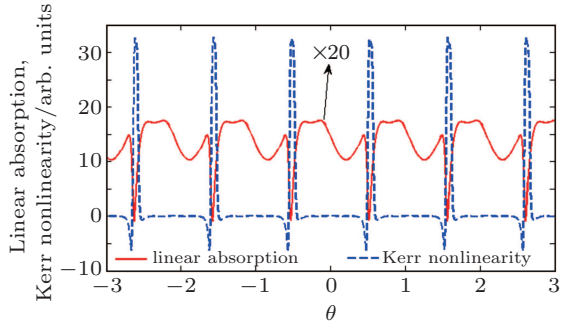


Fig. 4. (color online) Linear absorption (solid line) and Kerr nonlinearity (dashed line) versus elliptical parameter θ. The selected parameters are the same as Fig. 2.

Similar to the laser-driven atomic medium or ensemble consisting of many single atoms,[57] here we only consider a graphene monolayer in the graphene ensemble interacting with optical fields for the sake of simplification of the calculation. It should be emphasized that the optical conductivity of a graphene layer is proportional to its effective thickness or surface density of atoms.[58,59] This is in spite of the fact that the effective thickness of a graphene layer is around one angstrom and the vacuum wavelength of the optical waves under consideration is around one micron. However, through our calculation and analysis, it is possible to observe such spatial solitons by use of the graphene ensemble. In view of rapid advances in graphene material, we believe that the formation of matched infrared soliton pairs will be accessible in experiments in the near future.

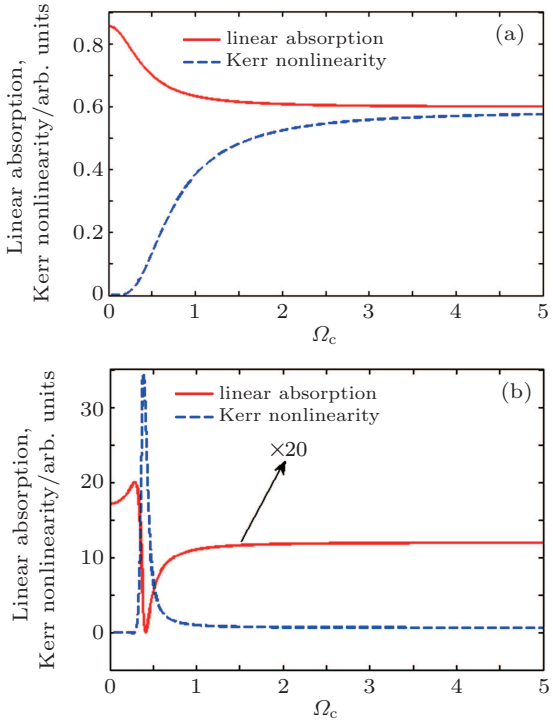


Fig. 5. (color online) Linear absorption (solid line) and Kerr nonlinearity (dashed line) versus different quantities of Rabi frequency Ω_c for (a) $\theta = 0$ and (b) $\theta = \pi/6$. The other parameters are the same as Fig. 2.

4. Conclusion

In summary, the linear and nonlinear susceptibilities of the monolayer graphene system driven by a probe and elliptical polarized coupling fields are explored by using the density matrix formalism. Our numerical results show that by tuning the elliptical parameter of elliptical polarized coupling field, the Rabi frequencies of coupling fields can be modified. Therefore, the enhanced Kerr nonlinearity with vanishing of linear absorption can be provided in two detunings of probe field.

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