Giant Kerr nonlinearity induced by interacting quantum coherences from decays and incoherent pumping*

Bai Yan-Feng(白艳锋)[a)†], Yang Wen-Xing(杨文星)[a)],
Han Ding-An(韩定安)[b)], and Yu Xiao-Qiang(喻小强)[a)]

[a)] *Department of Physics, Southeast University, Nanjing* 210096, *China*
[b)] *Department of Physics, Foshan University, Foshan* 528000, *China*

(Received 16 February 2012; revised manuscript received 25 April 2012)

A scheme for generating the giant enhancement of the Kerr nonlinearity in a four-level system with the quantum coherences from the decays and the incoherent pumping is proposed. Compared with that generated in a general four-level system, the Kerr nonlinearity can be enhanced by several orders of magnitude with vanishing linear absorption. By using the numerical results, we show that the remarkable enhancement should be attributed to the interaction of the quantum coherences from the decays and the incoherent pumping.

Keywords: Kerr nonlinearity, coherence induced by incoherent pumping, spontaneously generated coherence

PACS: 42.50.Gy, 42.65.–k **DOI:** 10.1088/1674-1056/21/11/114208

1. Introduction

Electromagnetically induced transparency (EIT) is a phenomenon that allows a beam of electromagnetic radiation to propagate through a medium almost as if the medium is not there.[1] The generic EIT is studied with three-level atoms, then the researchers extend their interest to multi-level atomic systems, which can be used to enhance the nonlinear optical processes, such as frequency conversion[2] and four-wave mixing.[3] The enhanced third-order Kerr nonlinearity in the multi-level systems due to the atomic coherence together with the vanishing absorption is responsible for such efficient nonlinear optical processes. In addition, the enhanced Kerr nonlinearity has various applications, such as in the self-phase modulation for the generation of optical solitons[4] and in the cross-phase modulation for quantum phase gates.[5] Recently, many schemes of EIT-enhanced Kerr nonlinearity in multi-level atomic systems have been studied both theoretically and experimentally.[6−14] Nakajima showed that the third-order nonlinearity can be enhanced with vanishing absorption via the autoionizing resonance.[8] Matsko *et al.* proposed a new method for the resonant enhancement of the optical Kerr nonlinearity, which uses the multi-level atomic coherence.[10] A scheme for the giant enhancement of the Kerr nonlinearity in a four-level system with double dark resonances was proposed.[11] Sun *et al.* investigated the enhancement of Kerr nonlinearity in an asymmetric GaAs double quantum well via the Fano interference.[12] Very recently, Niu and Gong showed that the enhanced Kerr nonlinearity with zero absorption can even be obtained in the general three-level systems via the spontaneously generated coherence (SGC).[15]

As we know, the coherent process is not the unique condition to generate the quantum coherence effects, the incoherent processes, such as the spontaneous emission[16,17] and the incoherent pumping,[18,19] can induce interferences in particular circumstances and thus modify the stimulated processes like absorption and refraction. Enlightened by these studies, we here demonstrate a giant enhancement of the Kerr nonlinearity induced by interacting the quantum coherences from the decays and the incoherent pumping. In a four-level system, the change of Kerr nonlinearity is not obvious by only considering the SGC or the coherence induced by the incoherent pumping (IPC). While the Kerr nonlinearity enhanced

*Project supported by the National Natural Science Foundation of China (Grant Nos. 10904015, 61008063, 11004030, and 11074036).

†Corresponding author. E-mail: yanfengbai@gmail.com

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by the interaction between the SGC and the IPC can be several orders of magnitude larger than that generated in a general four-level system.

2. Model and analytical results

The atomic system considered here is the same as that in Ref. [18] and is shown in Fig. 1. A coherent field E with Rabi frequency Ω is used to couple state $|3\rangle$ to both states $|1\rangle$ and $|2\rangle$ by choosing a frequency tuned halfway between the two states. The energy level splitting of $|1\rangle$ and $|2\rangle$ is donated as ω_{12}. The spontaneous decay rates from $|1\rangle$ and $|2\rangle$ to $|3\rangle$ are γ_1' and γ_2', respectively. Incoherent pumping processes r_1 and r_2 are used to drive the transitions $|1\rangle \leftrightarrow |4\rangle$ and $|2\rangle \leftrightarrow |4\rangle$, respectively. The decay from the upper state $|4\rangle$ to the lower state $|i\rangle$ is denoted by γ_i $(i = 1, 2, 3)$. When states $|1\rangle$ and $|2\rangle$ are closely spaced so that the two transitions to state $|3\rangle$ interact with the same vacuum mode, the two decay pathways are not independent, and the SGC should be considered. In principle, the dipole moments corresponding to transitions $|1\rangle \leftrightarrow |4\rangle$ and $|2\rangle \leftrightarrow |4\rangle$ can have different directions. However, the electric field E can be chosen in such a polarization mode that it couples to both transitions, so the coherence induced by the incoherent pumping can be presented.[18]

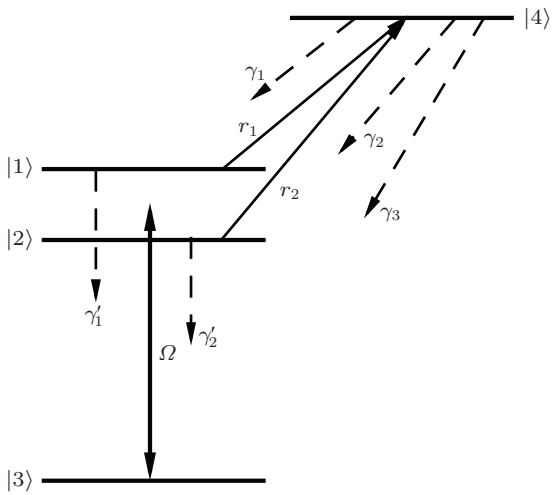


Fig. 1. Schematic diagram of the four-level system with doublet $|1\rangle$ and $|2\rangle$, state $|4\rangle$ is the auxiliary level coupled to the doublet via incoherent pumpings r_1 and r_2.

After some complicated derivation, we can obtain the density matrix equations of motion (see Sec. II in Ref. [18])

$$
\begin{aligned}
\dot{\rho}_{32} =& \Big[-\frac{1}{2}(r_2+\gamma_2')+\mathrm{i}\Delta_2\Big]\rho_{32} \\
&-\frac{1}{2}(p\sqrt{r_1r_2}+p'\sqrt{\gamma_1'\gamma_2'})\rho_{31} \\
&+\mathrm{i}[\Omega(2\rho_{22}+\rho_{11}+\rho_{44}-1)+\Omega\rho_{12}], \\
\dot{\rho}_{31} =& \Big[-\frac{1}{2}(r_1+\gamma_1')+\mathrm{i}\Delta_1\Big]\rho_{31} \\
&-\frac{1}{2}(p\sqrt{r_1r_2}+p'\sqrt{\gamma_1'\gamma_2'})\rho_{32} \\
&+\mathrm{i}[\Omega(2\rho_{11}+\rho_{22}+\rho_{44}-1)+\Omega\rho_{21}], \\
\dot{\rho}_{34} =& \Big[-\frac{1}{2}(r_1+r_2+\gamma_0+\gamma_1+\gamma_2) \\
&+\mathrm{i}(\Delta_1+\delta_1)\Big]\rho_{34}+\mathrm{i}\Omega(\rho_{24}+\rho_{14}), \\
\dot{\rho}_{22} =& -(r_2+\gamma_2')\rho_{22}+(r_2+\gamma_2)\rho_{44} \\
&-\frac{1}{2}(p\sqrt{r_1r_2}+p'\sqrt{\gamma_1'\gamma_2'})(\rho_{21}+\rho_{12}) \\
&+\mathrm{i}\Omega(\rho_{32}-\rho_{23}), \\
\dot{\rho}_{21} =& \Big[-\frac{1}{2}(r_1+r_2+\gamma_1'+\gamma_2')-\mathrm{i}(\Delta_2-\Delta_1)\Big]\rho_{21} \\
&-\frac{1}{2}(p\sqrt{r_1r_2}+p'\sqrt{\gamma_1'\gamma_2'})(\rho_{22}+\rho_{11}) \\
&+p\sqrt{r_1r_2}\rho_{44}+\mathrm{i}\Omega(\rho_{31}-\rho_{23}), \\
\dot{\rho}_{24} =& \Big[-\frac{1}{2}(r_1+2r_2+\gamma_0+\gamma_1+\gamma_2+\gamma_2')+\mathrm{i}\delta_2\Big]\rho_{24} \\
&-\frac{1}{2}(p\sqrt{r_1r_2}+p'\sqrt{\gamma_1'\gamma_2'})\rho_{14}+\mathrm{i}\Omega\rho_{34}, \\
\dot{\rho}_{11} =& -(r_1+\gamma_1')\rho_{11}+(r_1+\gamma_1)\rho_{44} \\
&-\frac{1}{2}(p\sqrt{r_1r_2}+p'\sqrt{\gamma_1'\gamma_2'})(\rho_{21}+\rho_{12}) \\
&+\mathrm{i}\Omega(\rho_{31}-\rho_{13}), \\
\dot{\rho}_{14} =& \Big[-\frac{1}{2}(r_2+2r_1+\gamma_0+\gamma_1+\gamma_2+\gamma_1')+\mathrm{i}\delta_1\Big]\rho_{14} \\
&-\frac{1}{2}(p\sqrt{r_1r_2}+p'\sqrt{\gamma_1'\gamma_2'})\rho_{24}+\mathrm{i}\Omega\rho_{34}, \\
\dot{\rho}_{44} =& -(r_1+r_2+\gamma_0+\gamma_1+\gamma_2)\rho_{44}+r_1\rho_{11} \\
&+r_2\rho_{22}+p\sqrt{r_1r_2}(\rho_{12}+\rho_{21}), \qquad (1)
\end{aligned}
$$

where $\Delta_1 = \omega_{13} - \nu$, $\Delta_2 = \omega_{23} - \nu$, $\delta_1 = \omega_{41} - \nu_\mathrm{p}$, and $\delta_2 = \omega_{42} - \nu_\mathrm{p}$ are the corresponding detunings, $p' = \mu_{13}\cdot\mu_{23}/|\mu_{13}||\mu_{23}|$ and $p = \mu_{14}\cdot\mu_{24}/|\mu_{14}||\mu_{24}|$ indicate the existence or not of the quantum coherences from the decays and the incoherent pumping, respectively. Here, ω_{ij} is the frequency of transition $|i\rangle \leftrightarrow |j\rangle$, ν (ν_p) is the frequency of the coherent (incoherent) field, and $\mu_{ij}(=\langle i|\mu|j\rangle)$ is the dipole matrix element.

The single atom polarization is given by $\langle\mu\rangle = (\mu_{13}\rho_{31}+\mu_{23}\rho_{32})\,\mathrm{e}^{-\mathrm{i}\nu t}+\mathrm{H.c.}$, while the macroscopic polarization with N atoms is described by $P(\nu) = N\langle\mu\rangle = \varepsilon_0\chi^{(1)}E+\varepsilon_0\chi^{(3)}EEE/2+\cdots$. To derive the linear and the nonlinear susceptibilities, we need to obtain the steady state solutions for the density matrix equations. In the present approach, an iterative

method is used, and the density matrix elements are expressed as $\rho_{mn} = \rho_{mn}^{(0)} + \rho_{mn}^{(1)} + \rho_{mn}^{(2)} + \rho_{mn}^{(3)} + \cdots$, where each successive approximation is calculated using the matrix elements of one order less than the one being calculated. In the limit of weak coherent field, the zero-order solution will be $\rho_{33}^{(0)} = 1$, and the other elements are equal to zero. Under the weak-probe approximation, we can obtain the matrix elements of the first order

$$\begin{aligned}
&\rho_{31}^{(1)} + \rho_{21}^{(1)} \\
&= \frac{2\mathrm{i}\Omega}{A}\Big\{(p+p'-2)[3(\rho_{11}^{(0)} + \rho_{22}^{(0)}) + 2(\rho_{44}^{(0)} - 1) \\
&\quad + (\rho_{21}^{(0)} + \rho_{12}^{(0)})] + 2\mathrm{i}\Delta_1[\rho_{22}^{(0)} - \rho_{11}^{(0)} - (\rho_{21}^{(0)} - \rho_{12}^{(0)})]\Big\} \\
&= -\frac{4\mathrm{i}\Omega}{A}(p+p'-2). \qquad (2)
\end{aligned}$$

Here all parameters are reduced to dimensionless ones by scaling with γ. For simplicity, we set $\Delta_2 = -\Delta_1$, $\delta_1 = -\delta_2 = -\gamma$, $r_i = \gamma_i' = \gamma$, and $\gamma_i = 0$ during the calculation, because the expression of $\mathrm{Re}[\chi^{(3)}]$ will be quite tedious when all parameters are considered. To obtain the third-order terms in $\rho_{21} + \rho_{31}$, we need to know ρ_{11}, ρ_{22}, ρ_{44}, and ρ_{12} to the second order

$$\begin{aligned}
\rho_{11}^{(2)} &= \rho_{22}^{(2)} = \frac{4\Omega^2}{AB}[p^3 + p(p+1)(p'-2) \\
&\quad + (p'-2)^2 - 2(p-2)\Delta_1^2], \qquad (3) \\
\rho_{44}^{(2)} &= \frac{4\Omega^2}{AB}[(1+p)(p+p'-2)^2 - 4\Delta_1^2(p-1)], \qquad (4) \\
\rho_{21}^{(2)} &= \frac{4\Omega^2}{AB}[-(p'-1)(p+p'-2) \\
&\quad + \mathrm{i}\Delta_1(p+p^2-4+3p'-pp') + 2\Delta_1^2]. \qquad (5)
\end{aligned}$$

With the above procedure, the linear absorption and the refractive part of the third-order susceptibility can be written as

$$\begin{aligned}
\mathrm{Im}[\chi^{(1)}] &= \frac{4}{A}(p+p'-2), \qquad (6) \\
\mathrm{Re}[\chi^{(3)}] &= \frac{-8\Delta_1}{A^2B}[p(12-7p^2+21p+20\Delta_1^2) \\
&\quad + p'(40-7p') \\
&\quad - pp'(10p+2+3p') - 4(11+7\Delta_1^2)], \qquad (7)
\end{aligned}$$

where

$$\begin{aligned}
A &= (p+p')^2 - 4(1+\Delta_1^2), \\
B &= p^2 + p'^2 - 2(1+\Delta_1^2),
\end{aligned}$$

and some unessential constant factors have been omitted. It is shown by Eq. (7) that the SGC, the IPC, and their interaction can affect the refractive part of the third-order susceptibility.

3. Numerical results

In the following, we focus on the third-order susceptibility. Firstly, we plot in Fig. 2 the refractive part of the third-order susceptibility obtained in the four-level system. It is shown that the Kerr nonlinearity is enhanced by about several orders of magnitude in comparison with that generated in a general four-level system without IPC and SGC. To show the property of the corresponding linear absorption, $\mathrm{Im}[\chi^{(1)}]$ is also displayed in Fig. 2 (see the dashed line). Note that for a given probe detuning (M), the giant enhancement of $\mathrm{Re}[\chi^{(3)}]$ corresponds to the strong linear absorption. This is not desirable for the applications of low-intensity nonlinear optics. While another giant enhanced Kerr nonlinearity (N) is accompanied by the vanishing linear absorption. In other words, the giant Kerr nonlinearity with zero linear absorption can be obtained in the four-level system. From Fig. 2, we can draw the elementary conclusion that the presence of quantum coherences from the decays and the incoherent pumping results in the giant enhancement of Kerr nonlinearity.

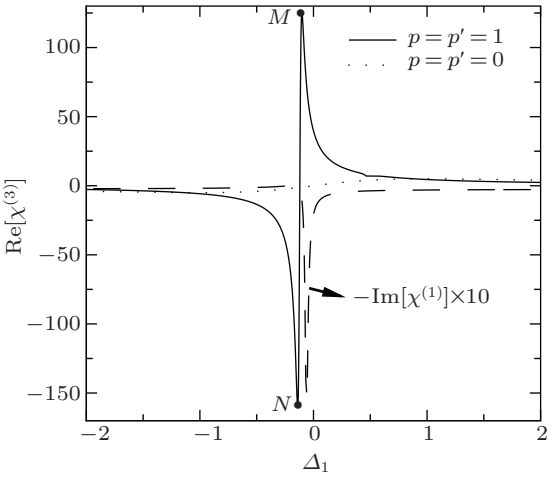


Fig. 2. Kerr nonlinearity $\mathrm{Re}[\chi^{(3)}]$ versus probe detuning Δ_1. The other parameters are chosen as $\gamma_1 = \gamma_2 = \gamma_3 = \gamma$, $r_1 = r_2 = r = 2\gamma$, $\gamma_1' = \gamma_2' = \gamma' = 2\gamma$, $\delta_2 = -\delta_1 = \gamma$, and $\omega_{12} = \gamma$.

Now we investigate the physical mechanism leading to the giant Kerr nonlinearity. From the results given in Ref. [15], we know that the Kerr nonlinearity can be enhanced by the presence of SGC in a general three-level V-type system. Is the mechanism applicable for our scheme? To answer this question, we depict $\mathrm{Re}[\chi^{(3)}]$ with only the SGC considered in Fig. 3(a), $\mathrm{Re}[\chi^{(3)}]$ in the conventional four-level system without

the quantum coherences from the decays and the incoherent pumping is also displayed for comparison. It is shown that small enhancement of the Kerr nonlinearity can be achieved by considering only the SGC. Then we focus on the effect of the quantum coherence from the incoherent pumping on the Kerr nonlinearity (see the dashed line). The variation of the Kerr nonlinearity is not obvious when only $p = 1$ is considered. So the giant Kerr nonlinearity can not be obtained by considering only the quantum coherence from the decays or the incoherent pumping. While the presence of both effects causes a giant enhancement of the Kerr nonlinearity (see the solid line in Fig. 3(b)). To explain the interesting phenomenon, we turn to the analysis of Eq. (7). Here $\mathrm{Re}[\chi^{(3)}]$ is composed of four terms, the first two terms $T_1(p)$ and $T_2(p')$ are functions of p and p', respectively. The third term $T_3(pp')$ is related to the product of p and p', and the last term T_4 is independent of p and p'. Obviously, $T_3(pp')$ exists only when both $p \neq 0$ and $p' \neq 0$ are satisfied. We plot $T_3(pp')$ obtained in the numerical simulation in Fig. 3(b). It is evident that $\mathrm{Re}[\chi^{(3)}]$ and the contribution from $T_3(pp')$ are approximately coincident in the narrow region of the giant enhancement of the Kerr nonlinearity. In other words, the giant Kerr nonlinearity is undoubtedly caused by the interaction of the quantum coherences from the decays and the incoherent pumping. Our scheme is different from the previous schemes mentioned above. So we here propose a new scheme for the giant Kerr nonlinearity by interacting the quantum coherences from the decays and the incoherent pumping.

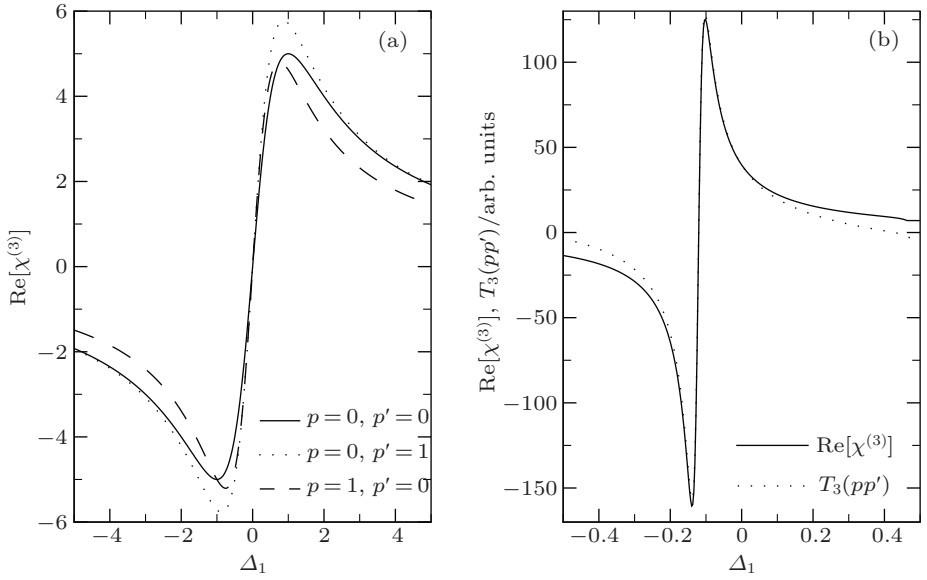


Fig. 3. (a) Kerr nonlinearity $\mathrm{Re}[\chi^{(3)}]$ for different p and p'. (b) Variation of $\mathrm{Re}[\chi^{(3)}]$ with $p = p' = 1$ (solid line) and the contribution from $T_3(pp')$ each as a function of Δ_1. The other parameters are the same as those in Fig. 2.

Next we discuss the methods of enhancing the Kerr nonlinearity in the four-level system. Based on the results given above, the presence of both coherent effects leads to the giant enhancement of the Kerr nonlinearity. Thus the changes of quantum coherences from the decays and the incoherent pumping could affect the third-order Kerr nonlinearity. The corresponding numerical results are depicted in Fig. 4. With r and γ' changing from $r = \gamma' = \gamma$ (dashed line) to $r = \gamma' = 2\gamma$ (solid line), which implies the stronger coherence effects, the refractive part of $\chi^{(3)}$ is enhanced by about 20 times.

Note that the quantum coherence of the decays is very sensitive to the space between $|1\rangle$ and $|2\rangle$. So the change of the energy level splitting ω_{12} will affect spontaneously the generated coherence, thus vary the third-order susceptibility. To demonstrate the result, the energy level splitting is decreased from $\omega_{12} = 2\gamma$ (solid line) to $\omega_{12} = \gamma$ (dotted line) in Fig. 4. The maximal Kerr nonlinearity is enhanced by about five times. While it should be noted that the effect from ω_{12} strictly depends on the presence of the SGC. By comparing the dashed curve in Fig. 4 with the dotted curve in Fig. 2, it is obvious that the enhanced Kerr nonlinearity can not be obtained with a decreasing ω_{12} when the SGC is absent.

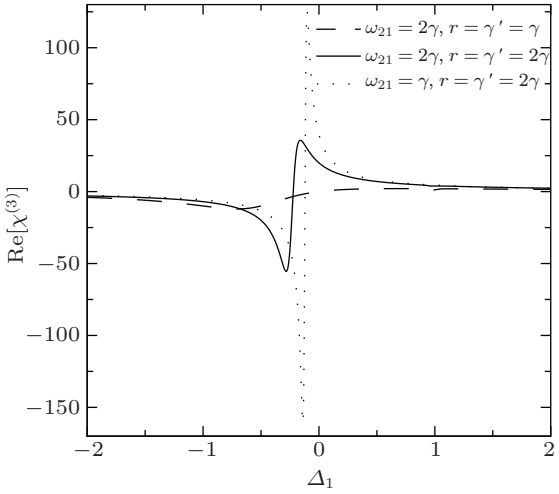


Fig. 4. The refractive part of the third-order susceptibility for different ω_{12}, decay rate γ', and incoherent pumping rate r.

It should be noticed that it is difficult to realize the quantum interference of two spontaneous decay channels in natural atomic systems experimentally, because the existence of such coherence relies on the nonorthogonality of the two transition dipole moments. Only one experiment implemented the interference.[17] However, the repetition of that result failed.[20] Very recently, there were experimental evidences that the SGC plays a role in the charged GaAs quantum dots.[21] The semiconductor microstructure allows the creation of artificial quantum systems with desirable properties, based on which we can fabricate a quantum dot with the model shown in Ref. [18] to implement our results.

4. Conclusion

In conclusion, we have investigated the giant enhancement of the Kerr nonlinearity in a four-level system with quantum coherences from the decays and the incoherent pumping. The result shows that the presence of both coherence effects can result in the giant Kerr nonlinearity with vanishing linear absorption. By analyzing the analytical expression of the third-order susceptibility, the giant enhancement is attributed to the interaction of two coherence effects. In addition, the methods of enhancing the Kerr nonlinearity in the four-level system are also discussed.

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