

Enhancement of nonlinear susceptibility in a four-level tripod scheme

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We propose a scheme for the enhancement of nonlinear susceptibility in a four-level tripod-type atomic system in the presence of a microwave field. With a microwave field, nonlinear susceptibility can be enhanced. Nonlinearity can also be ulteriorly enhanced by controlling the coupling field under the optimal intensity of the microwave field. The physical mechanism of the obtained giant nonlinear susceptibility is mainly based on interactions between microwave field and coupling fields. We present a physical understanding of our numerical results using a dressed-state approach and an analytical explanation.

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Third-order Kerr nonlinearity ($\chi^{(3)}$) plays an important role in nonlinear optics; it has many fascinating applications in different areas of physics, ranging from phase modulation^[1], generation of optical solitons^[2], and optical switching^[3] to optical communication and computing^[4,5]. Achieving giant Kerr nonlinearity with low light powers is desirable because it can be used to realize single-photon nonlinear devices. In recent years, the study of large third-order nonlinear susceptibility with significantly reduced or even completely cancelled linear absorption has aroused much interest both theoretically^[6–10] and experimentally^[11–13]. Producing giant Kerr nonlinearity coupled with reduced absorption using quantum interference effects related to electromagnetically induced transparency (EIT) is a universal atomic scheme^[7,14,15]. Another kind of coherence, spontaneously generated coherence (SGC), can also change the nonlinear and linear response of optical media^[16]. Niu *et al.* showed that an enhanced Kerr nonlinearity coupled with vanishing linear and nonlinear absorptions can be achieved through SGC^[17,18].

In this letter, we consider the interaction of a four-level quantum system in a tripod configuration with two coupling fields, one weak probe field and a microwave field. Based on EIT, these fields were used in Ref. [19] to study the possibility of controlling the enhancement of Kerr nonlinearity through the relative phase of driven fields. Moreover, we intend to study the possibility of controlling the enhancement of Kerr nonlinearity by changing the Rabi frequency values of the coherent coupling field or the microwave field while keeping the relative phase of driven fields at zero. Results show that the magnitude of Kerr nonlinearity can be enhanced within the left transparency window by appropriately choosing the microwave field value, even when the two coupling fields are equal. This is different from Ref. [20], where the two EIT windows are equal and the enhancement of Kerr

nonlinearity is accompanied by strong linear absorption. We also study the effect of the coupling field on Kerr nonlinearity. Results show that Kerr nonlinearity can be ulteriorly enhanced by controlling the coupling field under optimal intensity of the microwave field. A dressed-state approach and an analytical explanation are developed to explicate our numerical results.

We consider a four-level tripod-type atomic system, as shown in Fig. 1. It has one excited state ($|4\rangle$) and three lower-level states ($|1\rangle$, $|2\rangle$, and $|3\rangle$). One microwave field (field 2) and two coupling fields (fields 3 and 4) with Rabi frequencies of Ω_2 , Ω_3 , and Ω_4 , respectively, are applied to $|2\rangle \leftrightarrow |3\rangle$, $|3\rangle \leftrightarrow |4\rangle$, and $|2\rangle \leftrightarrow |4\rangle$ transitions. A weak probe field (field 1) with a Rabi frequency of Ω_1 is applied to couple the ground states ($|1\rangle$ and $|4\rangle$). Spontaneous decay rates from the excited state ($|4\rangle$) to all the lower levels are denoted by γ_4 ; $\omega_1, \omega_2, \omega_3$, and ω_4 are carrier frequencies of the corresponding fields; $\Delta_1 = \omega_{41} - \omega_1$, $\Delta_2 = \omega_{32} - \omega_2$, $\Delta_3 = \omega_{43} - \omega_3$, and $\Delta_4 = \omega_{42} - \omega_4$ are the frequency detunings of probe field 1, microwave field 2, and the two coherent coupling fields (fields 3 and 4), respectively.

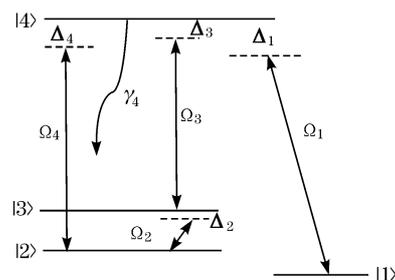


Fig. 1. Schematic diagram of the studied system. It consists of three lower levels and a single upper level. The lower states are coupled near-resonantly to the excited state by Ω_1 , Ω_3 , and Ω_4 , respectively, and a microwave field is applied to $|2\rangle \leftrightarrow |3\rangle$.

Under rotating-wave approximation, systematic density matrix in the interaction picture can be written as

$$\begin{aligned}
 \dot{\rho}_{11} &= -i\Omega_1(\rho_{14} - \rho_{41}) + \gamma_4\rho_{44}, \\
 \dot{\rho}_{12} &= i(\Delta_1 - \Delta_4)\rho_{12} + i\Omega_1\rho_{42} - i\Omega_2\rho_{13} \\
 &\quad - i\Omega_4\rho_{14}, \\
 \dot{\rho}_{13} &= i(\Delta_1 - \Delta_3)\rho_{13} + i\Omega_1\rho_{43} - i\Omega_2\rho_{12} \\
 &\quad - i\Omega_3\rho_{14}, \\
 \dot{\rho}_{14} &= i\Delta_1\rho_{14} - i\Omega_1(\rho_{11} - \rho_{44}) - i\Omega_3\rho_{13} \\
 &\quad - i\Omega_4\rho_{12} - \frac{3}{2}\gamma_4\rho_{14}, \\
 \dot{\rho}_{22} &= -i\Omega_2(\rho_{23} - \rho_{32}) - i\Omega_4(\rho_{24} - \rho_{42}) \\
 &\quad + \gamma_4\rho_{44}, \\
 \dot{\rho}_{23} &= -i(\Delta_3 - \Delta_4)\rho_{23} - i\Omega_2(\rho_{22} - \rho_{33}) \\
 &\quad - i\Omega_3\rho_{24} + i\Omega_4\rho_{43}, \\
 \dot{\rho}_{24} &= i\Delta_4\rho_{24} - i\Omega_1\rho_{21} + i\Omega_2\rho_{34} - i\Omega_3\rho_{23} \\
 &\quad + i\Omega_4(\rho_{44} - \rho_{22}) - \frac{3}{2}\gamma_4\rho_{24}, \\
 \dot{\rho}_{33} &= -i\Omega_2(\rho_{32} - \rho_{23}) - i\Omega_3(\rho_{34} - \rho_{43}) \\
 &\quad + \gamma_4\rho_{44}, \\
 \dot{\rho}_{34} &= i\Delta_3\rho_{34} - i\Omega_1\rho_{31} + i\Omega_2\rho_{24} \\
 &\quad + i\Omega_3(\rho_{44} - \rho_{33}) - i\Omega_4\rho_{32} - \frac{3}{2}\gamma_4\rho_{34}. \quad (1)
 \end{aligned}$$

The foregoing equations are constrained by $\sum_{n=1}^4 \rho_{nn}(t) = 1$ and $\rho_{ji}^* = \rho_{ij}$. In the present letter, we aim to investigate the feasibility of enhancing Kerr nonlinearity while simultaneously inhibiting the absorptions. Therefore, we need to derive the linear and third-order nonlinear susceptibilities. In the present approach, an iterative method is used, and the density matrix elements are expressed as $\rho_{ij} = \rho_{ij}^{(0)} + \rho_{ij}^{(1)} + \rho_{ij}^{(2)} + \rho_{ij}^{(3)} + \dots$. Assuming that Ω_1 is very small compared with $\Omega_2, \Omega_3, \Omega_4$, and γ_4 , the zeroth-order solution is $\rho_{11}^{(0)} = 1$ and the other elements are equal to zero. Under weak-probe approximation, we get the matrix element ρ_{14} up to the third order as follows:

Under weak-probe approximation, we get the matrix element ρ_{14} up to the third order as follows:

$$\rho_{12}^{(1)} = \frac{2\Omega_1}{\beta} [\Omega_2\Omega_3 + (\Delta_1 - \Delta_3)\Omega_4], \quad (2)$$

$$\rho_{13}^{(1)} = \frac{2\Omega_1}{\beta} [\Omega_2\Omega_4 + (\Delta_1 - \Delta_4)\Omega_3], \quad (3)$$

$$\rho_{14}^{(1)} = \frac{2\Omega_1}{\beta} [(\Delta_1 - \Delta_3)(\Delta_1 - \Delta_4) - \Omega_2^2], \quad (4)$$

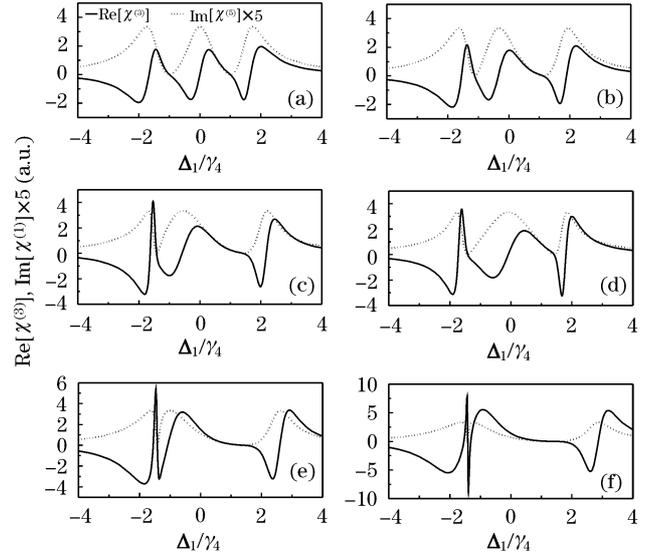


Fig. 2. Variation of $\text{Re}[\chi^{(3)}]$ (solid curves) and $\text{Im}[\chi^{(1)}]$ (dotted curve) as functions of the probe detuning Δ_1 . The parameters for the calculations are $\Delta_3 = -\Delta_4 = \gamma_4$ and $\Omega_4 = \gamma_4$. (a) $\Omega_2 = 0, \Omega_3 = \gamma_4$; (b) $\Omega_2 = 0.5\gamma_4, \Omega_3 = \gamma_4$; (c) $\Omega_2 = 1.0\gamma_4, \Omega_3 = \gamma_4$; (d) $\Omega_2 = 1.0\gamma_4, \Omega_3 = 0.5\gamma_4$; (e) $\Omega_2 = 1.0\gamma_4, \Omega_3 = 1.5\gamma_4$; (f) $\Omega_2 = 1.0\gamma_4, \Omega_3 = 1.8\gamma_4$.

$$\begin{aligned}
 \rho_{11}^{(2)} &= \frac{\Omega_1}{12\gamma_4 [(\Delta_4 - \Delta_3)\Omega_3\Omega_4 + \Omega_2(\Omega_3^2 - \Omega_4^2)]^2} \\
 &\quad \{6\gamma_4 A [(\Delta_3 - \Delta_4)\Omega_4 - 2\Omega_2\Omega_3] (\rho_{13}^{(1)} + \rho_{31}^{(1)}) + 6\gamma_4 A [(\Delta_3 - \Delta_4)\Omega_3 + 2\Omega_2\Omega_4] (\rho_{12}^{(1)} + \rho_{21}^{(1)}) \\
 &\quad + 4A i [(\Delta_3 + \Delta_4)\Omega_2\Omega_3 - (\Delta_3^2 - \Delta_3\Delta_4 + 2\Omega_2^2 - \Omega_3^2)\Omega_4 + \Omega_4^3] (\rho_{13}^{(1)} - \rho_{31}^{(1)}) \\
 &\quad - 4A i [(\Delta_4^2 - \Delta_3\Delta_4 + 2\Omega_2^2 - \Omega_4^2)\Omega_3 - (\Delta_3 + \Delta_4)\Omega_2\Omega_4 - \Omega_3^3] (\rho_{12}^{(1)} - \rho_{21}^{(1)}) \\
 &\quad - \{9i\gamma_4^2 [(\Delta_3 - \Delta_4)^2 + 4\Omega_2^2] (\Omega_3^2 + \Omega_4^2) - 4iB\} (\rho_{14}^{(1)} - \rho_{41}^{(1)})\}, \quad (5)
 \end{aligned}$$

$$\rho_{44}^{(2)} = \frac{i\Omega_1}{\gamma_4} (\rho_{14}^{(1)} - \rho_{41}^{(1)}), \quad (6)$$

$$\begin{aligned}
 \rho_{42}^{(2)} &= -\frac{\Omega_1}{6\gamma_4 A} \{-2iA(\rho_{12}^{(1)} - \rho_{21}^{(1)}) + \{3\gamma_4 [(\Delta_3 - \Delta_4)\Omega_3 + 2\Omega_2\Omega_4] \\
 &\quad - 2i[\Omega_3(\Delta_3\Delta_4 - \Delta_4^2 - 2\Omega_2^2 + \Omega_4^2) + (\Delta_3 + \Delta_4)\Omega_2\Omega_4 + \Omega_3^3]\} (\rho_{14}^{(1)} - \rho_{41}^{(1)})\}, \quad (7)
 \end{aligned}$$

$$\begin{aligned}
 \rho_{43}^{(2)} &= -\frac{\Omega_1}{6\gamma_4 A} \{-2iA(\rho_{13}^{(1)} - \rho_{31}^{(1)}) + \{3\gamma_4 [(\Delta_3 - \Delta_4)\Omega_4 - 2\Omega_2\Omega_3] \\
 &\quad + 2i[\Omega_4(\Delta_3\Delta_4 - \Delta_3^2 - 2\Omega_2^2 + \Omega_3^2) + (\Delta_3 + \Delta_4)\Omega_2\Omega_3 + \Omega_4^3]\} (\rho_{14}^{(1)} - \rho_{41}^{(1)})\}, \quad (8)
 \end{aligned}$$

$$\begin{aligned}
 \rho_{14}^{(3)} &= -\frac{\Omega_1\Omega_4}{C} \{(\Delta_1 - \Delta_4)[-(\Delta_1 - \Delta_3)(\rho_{11}^{(1)} - \rho_{44}^{(2)}) + \Omega_3\rho_{43}^{(2)}] \\
 &\quad + (\Delta_1 - \Delta_3)\Omega_4\rho_{42}^{(2)} + \Omega_2(\Omega_3\rho_{42}^{(2)} + \Omega_4\rho_{43}^{(2)}) + \Omega_2^2(\rho_{11}^{(2)} - \rho_{44}^{(2)})\}, \quad (9)
 \end{aligned}$$

with

$$\beta = (3i\gamma_4 + 2\Delta_1)(\Delta_1 - \Delta_3)(\Delta_1 - \Delta_4) - (2\Delta_1 + 3i\gamma_4)\Omega_2^2 - 2(\Delta_1 - \Delta_4)\Omega_3^2 - 2(\Delta_1 - \Delta_3)\Omega_4^2 - 4\Omega_2\Omega_3\Omega_4, \quad (10)$$

$$A = (\Delta_3 - \Delta_4)\Omega_3\Omega_4 - \Omega_2(\Omega_3^2 - \Omega_4^2), \quad (11)$$

$$B = (\Delta_4^2 + \Omega_2^2)[(\Delta_3 - \Delta_4)^2 + 4\Omega_2^2]\Omega_3^2 + [2(\Delta_3 - \Delta_4)\Delta_4 + 5\Omega_2^2]\Omega_3^4 + \Omega_3^6 - 2\Omega_2\Omega_3\Omega_4\{(\Delta_3 + \Delta_4)[(\Delta_3 - \Delta_4)^2 + 4\Omega_2^2] + (7\Delta_3 - 11\Delta_4)\Omega_3^2\} + \{(\Delta_3^2 + \Omega_2^2)[(\Delta_3 - \Delta_4)^2 + 4\Omega_2^2] + [7(\Delta_3 - \Delta_4)^2 - 26\Omega_2^2]\Omega_3^2 + 3\Omega_3^4\}\Omega_4^2 + 2(11\Delta_3 - 7\Delta_4)\Omega_2\Omega_3\Omega_4^3 + [2\Delta_3(-\Delta_3 + \Delta_4) + 5\Omega_2^2 + 3\Omega_3^2]\Omega_4^4 + \Omega_4^6, \quad (12)$$

$$C = -\frac{1}{2}[(\Delta_1 - \Delta_4)\Omega_3 + \Omega_2\Omega_4][(3i\gamma_4 + 2\Delta_1)\Omega_2 + 2\Omega_3\Omega_4] + [\Omega_2\Omega_3 + (\Delta_1 - \Delta_3)\Omega_4][\frac{1}{2}(3i\gamma_4 + 2\Delta_1)(\Delta_1 - \Delta_4) - \Omega_4^2]. \quad (13)$$

Therefore, the first- and third-order susceptibilities ($\chi^{(1)}$ and $\chi^{(3)}$) are

$$\chi^{(1)} = \frac{-2N|\vec{\mu}_{14}|^2}{\varepsilon_0\Omega_1}\rho_{14}^{(1)}, \quad (14)$$

$$\chi^{(3)} = \frac{-2N|\vec{\mu}_{14}|^4}{3\varepsilon_0\Omega_1^3}\rho_{14}^{(3)}, \quad (15)$$

and χ is defined as

$$\chi = \chi^{(1)} + 3|E_1|^2\chi^{(3)}. \quad (16)$$

The absorption of the probe field and the Kerr nonlinearity can be described by the imaginary part of $\chi^{(1)}$ and the real part of $\chi^{(3)}$, respectively.

Now, we focus on the dependence of third-order susceptibility on the intensities of the coherent coupling field and the microwave field. Based on Eqs. (14) and (15), Fig. 2 shows the linear absorption and the refractive part of the third-order susceptibility as functions of probe detuning. For simplicity, all parameters are scaled by the decay rate γ_4 , setting $\Delta_3 = -\Delta_4 = \gamma_4$, $\Omega_4 = \gamma_4$. In Figs. 2(a)–(c), $\Omega_3 = 1.0\gamma_4$, and Rabi frequencies of the microwave fields are $\Omega_2 = 0, 0.5\gamma_4$, and $1.0\gamma_4$. In Figs. 2(d)–(f), $\Omega_2 = 1.0\gamma_4$, and Rabi frequencies of the coupling fields are $\Omega_3 = 0.5\gamma_4, 1.5\gamma_4$, and $1.8\gamma_4$. Drawing from this figure, when $\Omega_2 = 0$, which means the microwave field is absent, there are two identical EIT windows, and the maximal Kerr nonlinearity is accompanied by remarkable linear absorption (Fig. 2(a)). This is not desirable for all-optical switch applications because the accompanying thermal effect of the devices is not negligible. When Ω_2 changes from 0 to $1.0\gamma_4$, it is surprising to see that the two identical EIT windows become different: the left window narrows while the right one broadens. Within the left narrow transparency window, the nonlinear dispersion curve is very steep, suggesting that a giant Kerr nonlinearity with negligible linear absorption can be realized simultaneously, as shown in Fig. 2(c). On the other hand, when $\Omega_2 = 1.0\gamma_4$, the left EIT window becomes narrower, and the strength of Kerr nonlinearity becomes stronger as Ω_3 changes from $0.5\gamma_4$ to $1.8\gamma_4$. In Fig. 2(f), Kerr nonlinearity is clearly dramatically enhanced while linear absorption is suppressed. Compared

with the case of $\Omega_2 = 0$, the maximal value of Kerr nonlinearity is enhanced about five times. In this letter, we do not present the effect of Ω_4 on Kerr nonlinearity because its effect is the same as that of Ω_3 . Therefore, we can achieve giant Kerr nonlinearity with a vanishing linear absorption by manipulating the intensities of the coupling and microwave fields.

To understand the effect of the intensities of the coherent coupling field and the microwave field on EIT window and Kerr nonlinearity, we consider the dressed-state approach. Working in an interaction picture and taking into account only the strong coupling fields and the microwave field, we find that the effective Hamiltonian can be written as

$$H_{\text{eff}} = -(\Delta_4|2\rangle\langle 2| + \Delta_3|3\rangle\langle 3| + \Omega_2|3\rangle\langle 2| + \Omega_3|4\rangle\langle 3| + \Omega_4|4\rangle\langle 2|) + H.C. \quad (17)$$

Thus, we can obtain the secular equation:

$$f(\lambda) = \lambda^3 + (\Delta_3 + \Delta_4)\lambda^2 + (\Delta_3\Delta_4 - \Omega_2^2 - \Omega_3^2 - \Omega_4^2)\lambda - 2\Omega_2\Omega_3\Omega_4 - \Delta_3\Omega_4^2 - \Delta_4\Omega_3^2, \quad (18)$$

where λ is the eigenenergy of the Hamiltonian, which gives the positions of dressed sublevels $|k\rangle$ ($k = +, 0$, and $-$) generated by the two coupling fields and the microwave field. When this transition (i.e., $|1\rangle \leftrightarrow |k\rangle$) is probed by the weak probe field, resonances occur at points where the probe frequency matches the energy-level difference between $|1\rangle$ and $|k\rangle$. If the probe field detuning (Δ_1) is chosen such that it is in resonance with a dressed state, then it experiences absorption maxima^[21]. Hence, the EIT window is dependent on the difference of the two neighboring eigenvalues. We see from Eq. (18) that the three eigenvalues (λ_+, λ_0 , and λ_-) are dependent on the three coherent fields (Ω_2, Ω_3 , and Ω_4) and can easily be obtained. For example, for the parameters in Fig. 2(c), $\lambda_+ = 2.22\gamma_4, \lambda_0 = -0.54\gamma_4$, and $\lambda_- = -1.68\gamma_4$; for the parameters in Fig. 2(e), $\lambda_+ = 2.62\gamma_4, \lambda_0 = -\gamma_4$, and $\lambda_- = -1.62\gamma_4$. To determine the influence of the coupling and microwave fields more clearly, we respectively plot the evolutions of the three eigenvalues (λ_+, λ_0 , and λ_-) versus Ω_2 and Ω_3 in Fig. 3 taking the same parameters as those in Fig. 2. In Fig. 3(a), note that the eigenvalue difference between λ_+ and λ_0 is increasing, while that

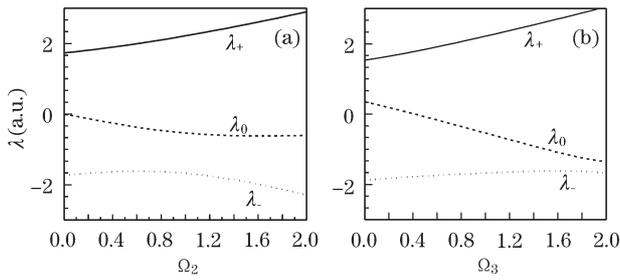


Fig. 3. Evolutions of eigenvalues of the three dressed states λ_+ (solid lines), λ_0 (dashed lines), λ_- (dotted lines) versus (a) Ω_2 and (b) Ω_3 , respectively. Other parameters are the same as those in Fig. 2.

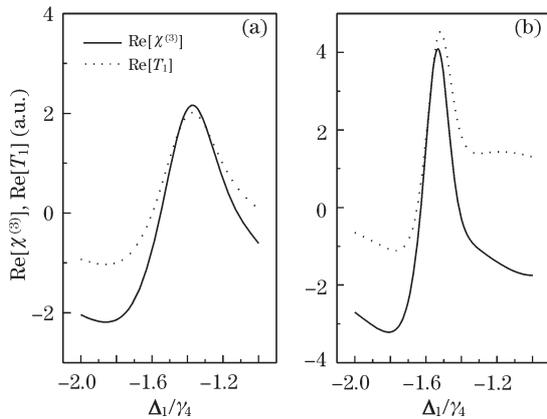


Fig. 4. Variation of $\text{Re}[\chi^{(3)}]$ (solid curves) and $\text{Re}[T_1]$ (dotted curves) versus the probe detuning Δ_1 with $\Omega_3 = \gamma_4$. (a) $\Omega_2 = 0.5\gamma_4$, (b) $\Omega_2 = 1.0\gamma_4$. Other parameters are the same as those in Fig. 2.

between λ_- and λ_0 is decreasing, corresponding exactly to the narrowing of the left EIT window and the broadening of the right EIT window when Ω_2 changes from 0 to $1.0\gamma_4$ (Figs. 2(a)–(c)). However, the reverse phenomenon is present when $\Omega_2 > 1.0\gamma_4$, that is, the left EIT window is wide and the right one is narrow (not shown in Fig. 2). In Fig. 3(b), the stronger Ω_3 is, the smaller the eigenvalue difference between λ_- and λ_0 is. This corresponds to the narrowing of the left EIT window, as shown in Fig. 2(f). Within the narrower window, the steep nonlinear dispersion profile of the probe field makes it possible to enhance Kerr nonlinearity accompanied by vanishing absorption^[22]. Dressed-state analysis suggests that the widths of the EIT window can be controlled by the intensities of the coupling and microwave fields, leading to enhanced Kerr nonlinearity.

In the following, we mainly discuss the effect of Ω_2 on Kerr nonlinearity by using an analytical explanation. In Eq. (9), the last two terms are the products of Ω_2 and the coupling fields. When the microwave field is applied to $|2\rangle \leftrightarrow |3\rangle$, Ω_3 and Ω_4 interact with Ω_2 ; consequently, Kerr nonlinearity is clearly enhanced. Here, the last two terms in Eq. (15) are designated as T_1 and the other terms as T_2 , then $\chi^{(3)} = T_1 + T_2$. We plot $\text{Re}[T_1]$ as a function of Δ_1 (Fig. 4) and display $\text{Re}[\chi^{(3)}]$ for comparison. They are approximately coincidental in the narrow region of the giant enhancement of Kerr nonlinearity. As a result, we consider from the analytical aspect that

the giant enhancement of third-order susceptibility is undoubtedly caused by interactions between the microwave and the coupling fields. In the scheme proposed in Ref. [19], giant Kerr nonlinearity is obtained by controlling the relative phase of coherent driven fields. On the other hand, in the present scheme, nonlinear property is manipulated by controlling the intensities of the microwave and coupling fields. The proper choice of Rabi frequency of the microwave field enhances Kerr nonlinearity, despite 1 being the ratio of the two coupling fields. This finding is different from that in Ref. [20], where the enhancement of Kerr nonlinearity is accompanied by strong linear absorption.

In conclusion, we have investigated the effects of the intensities of the coupling field and the microwave field on Kerr nonlinearity and on double EIT in a tripod-configuration four-level atomic system. Results show that the properties of EIT and Kerr nonlinearity can be significantly modified by changing the intensities. Moreover, the proper choice of intensity of the microwave field leads to a giant Kerr nonlinearity accompanied with vanishing linear absorption, despite 1 being the ratio of the two coupling fields. This is different from that in Ref. [20], where the enhancement of Kerr nonlinearity is accompanied by strong linear absorption. Dressed-state analysis suggests that the widths of the EIT window can be controlled by the intensities of the coupling and microwave fields. Close inspection of the analytical expression undoubtedly shows that interactions between the microwave and coupling fields are responsible for the large enhancement of Kerr nonlinearity. This system may potentially be applied to ultraslow optical solitons, frequency conversion, and other information processes.

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