Giant Kerr nonlinearity induced by tunneling in triple quantum dot molecules

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Received January 16, 2014; revised April 21, 2014; accepted April 28, 2014; posted May 2, 2014 (Doc. ID 204425); published June 4, 2014

A scheme for giant enhancement of the Kerr nonlinearity in linear triple quantum dot molecules is proposed. In such a system, the tunneling-induced transparency window obtained in double quantum dot molecules splits into two windows, due to the coupling with the third quantum dot. And most important, the Kerr nonlinearity can be enhanced by several orders of magnitude, compared with that generated in double quantum dot molecules. With proper detuning of the tunneling, giant Kerr nonlinearity accompanied by vanishing absorption can be realized, which opens the possibility to enhance self-phase modulation in tunneling controllable semiconductor nanostructures under conditions of low light levels. Quantitative analysis shows that the giant Kerr nonlinearity is attributed to the interacting double dark resonances induced by the tunneling between the triple quantum dots, therefore no extra laser fields are required.

OCIS codes: (270.0270) Quantum optics; (190.3270) Kerr effect; (230.5590) Quantum-well, -wire and -dot devices.

http://dx.doi.org/10.1364/JOSAB.31.001436

1. INTRODUCTION

The Kerr nonlinearity, corresponding to the refractive part of the third-order susceptibility in optical media, plays an important role in the field of nonlinear optics [1], such as self focusing [2], the generation of optical solitons [3], polarization phase gates [4], and so on. The enhanced nonlinear susceptibility with suppressed absorption has attracted tremendous interest in nonlinear optics at low light levels. The electromagnetically induced transparency (EIT) scheme is capable of producing enhanced Kerr nonlinearity and, at the same time, suppressing the linear absorption [5]. And in order to enhance the Kerr nonlinearity, several multiple-level atomic schemes (such as N- [6,7], M- [8], double-Λ [9], inverted-Y [10], and tripod-type [11,12] systems) have been proposed. But in all these atomic systems, it is crucial to have additional coupling lasers to modify the linear and nonlinear optical properties.

Semiconductor quantum dots (QDs) are collections of thousands of atoms, whose electronic degrees of freedom are discretized, similar to those of an atom, owing to confinement of electrons and holes. QDs coherently driven by electromagnetic fields have been used to investigate quantum coherence phenomena, such as Autler–Townes splitting (ATS) and Mollow triplets [13], EIT [14], and resonance fluorescence [15,16]. Moreover, two or more QDs coupled by tunneling can form quantum dot molecules (QDMs), in which one can control the tunneling of electrons or holes by an external electric field and create a multilevel structure of excitonic states. And both vertical [17] and lateral [18] double quantum dots (DQDs) have been realized experimentally. Many studies have been carried out about such DQDs, such as optical spectroscopy [19], excitonic entanglement [20], single photon and spin storage [21], and coherent population trapping (CPT) [22].

Most recently, Borges *et al.* have proposed a scheme to use tunneling to induce transparency in DQDs, which is called tunneling-induced transparency (TIT) [23]. Through TIT, the linear and nonlinear optical properties of DQDs media can be modified by using the electric gates. Inspired by this, in this paper we investigate the Kerr nonlinearity in a linear triple quantum dots (TQDs). Such molecules have been achieved in much experimental progress [24–27]. In the presence of only one tunneling, the system reduces to DQDs and the enhancement of Kerr nonlinearity is obtained. While under the coupling of two tunnelings between the TQDs, the Kerr nonlinearity can be dramatically enhanced compared to that of DQDs. By proper control of the tunneling, a giant Kerr nonlinearity accompanied by vanishing absorption can be realized. And unlike the atomic system, in TQDs the enhanced Kerr nonlinearity is induced by the tunneling between the dots, requiring no coupling lasers. The system considered here opens the possibility to enhance self-phase modulation with vanishing linear absorption in semiconductor nanostructures controlled by electric gates.

The remaining of this paper is organized as follows: In Section 2, we introduce the model and the basic equations. In Section 3, we describe the numerical results and explain the corresponding features. Section 4 is the conclusions.

2. TRIPLE QUANTUM DOT SYSTEM

We show the schematic of the band structure and level configuration of a TQD system in Fig. 1. At nanoscale interdot separation, the hole states are localized in the QDs and the electron states are rather delocalized. From Fig. 1(a), in the absence of a gate voltage, the conduction-band electron levels are out of resonance and the electron tunneling between the QDs is very weak. In contrast, in the presence of a gate voltage, the conduction-band electron levels come close to resonance and the electron tunneling between the QDs is greatly enhanced, as shown in Fig. 1(b). In such system, the tunneling can be controlled by placing a gate electrode between the neighboring dots. And in the latter case the hole tunneling can be neglected because of the more off-resonant valence-band energy levels. Therefore, we can give the schematic of the level configuration of a TQD system, as shown in Fig. 1(c). Without the excitation of the laser, no excitons are inside all QDs, which corresponds to state $|0\rangle$. When a laser field is applied, a direct exciton is created inside the QD1, a condition represented by the state $|1\rangle$. Under the tunneling couplings, the electron can tunnel from QD1 to the QD2, and from QD2 to QD3. And we denote these indirect excitons as state $|2\rangle$ and state $|3\rangle$, respectively.

Under the rotating-wave and the electric-dipole approximations, and after performing the unitary transformation $U = e^{-i\omega_p t}(|1\rangle\langle 1| + |2\rangle\langle 2| + |3\rangle\langle 3|)$, which removes the time-dependent oscillatory terms, the Hamiltonian under the basis $\{|0\rangle, |1\rangle, |2\rangle, |3\rangle\}$ can be written as (assumption of $\hbar = 1$)

$$H_I = \begin{pmatrix} 0 & -\Omega_p & 0 & 0 \\ -\Omega_p & \delta_p & -T_1 & 0 \\ 0 & -T_1 & \delta_p - \delta_1 & -T_2 \\ 0 & 0 & -T_2 & \delta_p - \delta_1 - \delta_2 \end{pmatrix}. \quad (1)$$

Here, $\Omega_p = \boldsymbol{\mu}_{01} \cdot \mathbf{e} \cdot E_p$ is the Rabi frequency of the transition $|0\rangle \to |1\rangle$, with $\boldsymbol{\mu}_{01}$ being the associated dipole transition-matrix element, $\mathbf{e}$ the polarization vector, and E_p the electric-field amplitude of the probe laser. And T_1 and T_2 are the tunneling couplings, which depends on the barrier characteristics and the external electric field. $\delta_p = (\omega_{10} - \omega_p)$, $\delta_1 = \delta_p - \omega_{12}$, and $\delta_2 = \delta_p - \omega_{12} - \omega_{23}$ are the detuning of the probe field and the tunneling couplings, respectively, with ω_{mn} being the transition frequency between $|m\rangle$ and $|n\rangle$ states, and ω_p being the frequency of the probe field. Experimentally, the value of δ_1 and δ_2 are controlled by varying the value of δ_p and the frequency transition ω_{12} and ω_{23}, which can be done by manipulation of the external electric field that changes the effective confinement potential.

At any time t, the state vector can be written as

$$|\Psi_I(t)\rangle = a_0(t)|0\rangle + a_1(t)|1\rangle + a_2(t)|2\rangle + a_3(t)|3\rangle. \quad (2)$$

The evolution of the state vector obeys the Schrödinger equation

$$\frac{d}{dt}|\Psi_I(t)\rangle = -iH_I(t)|\Psi_I(t)\rangle. \quad (3)$$

Substituting Eqs. (1) and (2) into Eq. (3), and then using the Weisskopf–Wigner theory [28–30], we can obtain the following dynamical equations for atomic probability amplitudes in the interaction picture:

$$i\dot{a}_0 = -\Omega_p a_1, \quad (4a)$$

$$i\dot{a}_1 = -\Omega_p a_0 - T_1 a_2 + (\delta_p - i\gamma_1)a_1, \quad (4b)$$

$$i\dot{a}_2 = -T_1 a_1 - T_2 a_3 + (\delta_p - \delta_1 - i\gamma_2)a_2, \quad (4c)$$

$$i\dot{a}_3 = -T_2 a_2 + (\delta_p - \delta_1 - \delta_2 - i\gamma_3)a_3, \quad (4d)$$

$$|a_1|^2 + |a_2|^2 + |a_3|^2 + |a_4|^2 = 1, \quad (4e)$$

where $\gamma_i = (1/2)\Gamma_{i0} + \gamma_{i0}^d$ ($i = 1$–3) is the typical effective decay rate, with Γ_{i0} being the radiative decay rate of populations from $|i\rangle \to |0\rangle$ and γ_{i0}^d being the pure dephasing rates.

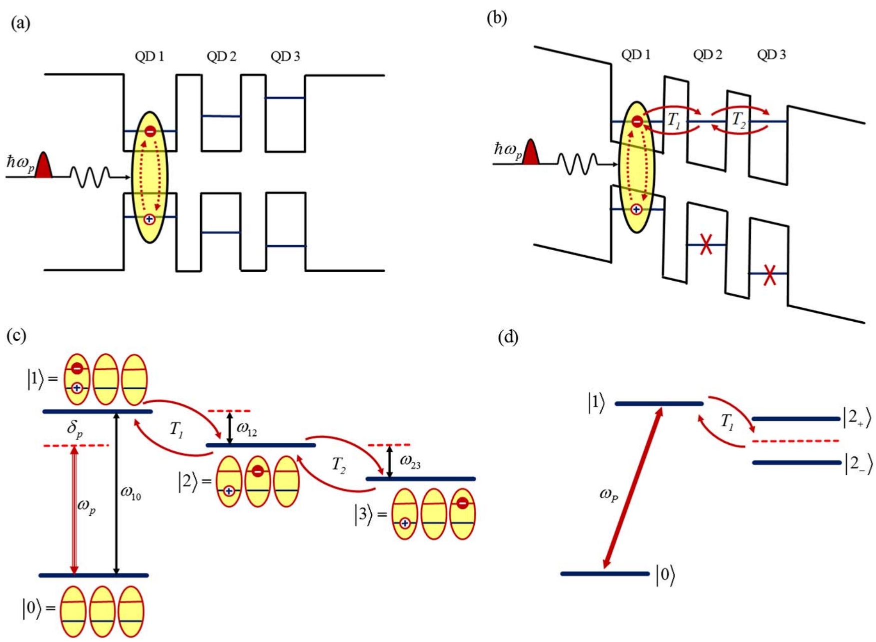

Fig. 1. (a) Schematic of band structure without a gate voltage. (b) Schematic of band structure with a gate voltage. (c) Schematic of the level configuration of a TQD system. (d) Dressed states under the tunneling coupling T_2.

It is well known that the response of the QDs media to the probe field is governed by its polarization χ. We solve Eq. (4) and obtain the analytical expressions of the first- and third-order susceptibilities (see Appendix A):

$$\chi^{(1)} = \frac{\Gamma|\boldsymbol{\mu}_{01}|^2}{V\varepsilon_0\hbar}\frac{1}{\Gamma_1 - \frac{T_1^2\Gamma_3}{\Gamma_2\Gamma_3 - T_2^2}}, \tag{5a}$$

$$\chi^{(3)} = \frac{\Gamma|\boldsymbol{\mu}_{01}|^4}{3V\varepsilon_0\hbar^3}\frac{1}{\Gamma_1 - \frac{T_1^2\Gamma_3}{\Gamma_2\Gamma_3 - T_2^2}}\frac{1}{\left|\Gamma_1 - \frac{T_1^2\Gamma_3}{\Gamma_2\Gamma_3 - T_2^2}\right|^2} \times \left(1 + T_1^2\frac{\Gamma_3^2}{|\Gamma_2\Gamma_3 - T_2^2|^2} + (T_1T_2)^2\frac{1}{|\Gamma_2\Gamma_3 - T_2^2|^2}\right). \tag{5b}$$

Here,

$$\Gamma_1 = \delta_p - i\gamma_1, \tag{6a}$$

$$\Gamma_2 = \delta_p - \delta_1 - i\gamma_2, \tag{6b}$$

$$\Gamma_3 = \delta_p - \delta_1 - \delta_2 - i\gamma_3, \tag{6c}$$

and Γ is the optical confinement factor, V is the volume of a single QD, ε_0 is the dielectric constant, and $\boldsymbol{\mu}_{01}$ is the associated dipole transition-matrix element [31]. According to Eq. (5), we can obtain the linear absorption $\mathrm{Im}[\chi^{(1)}]$ and the refractive part of the third-order susceptibility $\mathrm{Re}[\chi^{(3)}]$.

3. RESULTS AND DISCUSSION

In TQDs, the tunneling couplings T_1 and T_2 depend on the barrier characteristics and the external electric field. Frequency transition ω_{12} and ω_{23} can be done by manipulation of the external electric field that changes the effective confinement potential. In addition, in this investigation we work in the low temperature regime, and consider both the population decay rates and the dephasing rates. The realistic values of the parameters are listed in Table 1, which are according to Ref. [23] and references therein [32–43]. And for simplicity, all the parameters are scaled by the decay rate γ_1. (Though some of the value of parameters are for DQDs, it can be inferred that the tunneling, frequency transition, and decay rates of TQDs have the same values as that of DQDs. And such TQDs have already been achieved in much experimental progress [24–27].)

First, in the absence of the tunneling couplings, the electron cannot tunnel from QD 1 to the other QDs. Thus the system is reduced to a single QD with one exited state $|1\rangle$ and one ground state $|0\rangle$. When the probe field passes through such QDs media, the normal absorption and nonlinearity curves of the two-level system are obtained (dotted line in Fig. 2). In the presence of the tunneling coupling T_1, the system turns to be a DQD system and one transparency window appears, as shown in Fig. 2(a) by the solid line. The transparency of the probe field, so-called TIT in [23], is owning to the dark state induced by the tunneling T_1, which can been written as $|\mathrm{Dark}\rangle = (1/\sqrt{\Omega_p^2 + T_1^2})(T_1|0\rangle - \Omega_p|2\rangle)$ under the resonant coupling. And simultaneously the nonlinearity is enhanced in the region of the transparency window [solid line in Fig. 2(b)]. The origination of the enhanced nonlinearity can be seen below.

Table 1. Experimental Values of the Parameters in TQDs

Parameters	Values Regime	Values/γ_1	Reference
$\hbar\omega_{10}$	1.6 eV	—	[32,33]
$\hbar\omega_{12}$, $\hbar\omega_{23}$	−0.01–0.01 meV	−1–1	[23,34]
Ω_p	1 μeV	0.1	[23,35] ($\Omega_p \ll T_1, T_2$)
T_1, T_2	0–10 μeV	0–1	[23,35,36] (TIT regime)
Γ_{10}	0–6.6 μeV	0–0.66	[37]
Γ_{20}, Γ_{30}	$10^{-4}\Gamma_{10}$	$10^{-4}\times(0\text{–}0.66)$	[38,39]
γ_1	2–10 μeV	—	[40–42]
γ_2, γ_3	$10^{-3}\gamma_1$	10^{-3}	[43]

When considering the DQD system, i.e., $T_1 \neq 0$, $T_2 = 0$, Eq. (5b) is simplified as

$$\chi^{(3)} = \frac{\Gamma|\boldsymbol{\mu}_{01}|^4}{3V\varepsilon_0\hbar^3}\frac{1}{\Gamma_1 - \frac{T_1^2}{\Gamma_2}}\frac{1}{\left|\Gamma_1 - \frac{T_1^2}{\Gamma_2}\right|^2}\left(1 + \frac{T_1^2}{|\Gamma_2|^2}\right). \tag{7}$$

We denote the second term of Eq. (7) as $F_1(T_1)$, which is proportional to the square of the tunneling T_1. While we denote the first term of Eq. (7) as F_2, which is independent of the square of T_1. Then the Kerr nonlinearity can be written as $\chi^{(3)} = F_2 + F_1(T_1)$. In Fig. 3, we plot both $\mathrm{Re}[F_1]$ and $\mathrm{Re}[\chi^{(3)}]$ as a function of δ_p. It can be seen from the figure that in the region of TIT window the two curves are approximately coincident. Therefore, it can be concluded that the enhancement of the third-order susceptibility is mainly caused by the tunneling coupling T_1.

Next we apply both tunneling T_1 and T_2, thus the electrons can tunnel from QD1 to QD2, then from QD2 to QD3, which creates a four-level Λ type system. We can analyze this TQD system by diagonalizing the interaction with the tunneling T_2 and show the corresponding dressed states in Fig. 1(d). Under

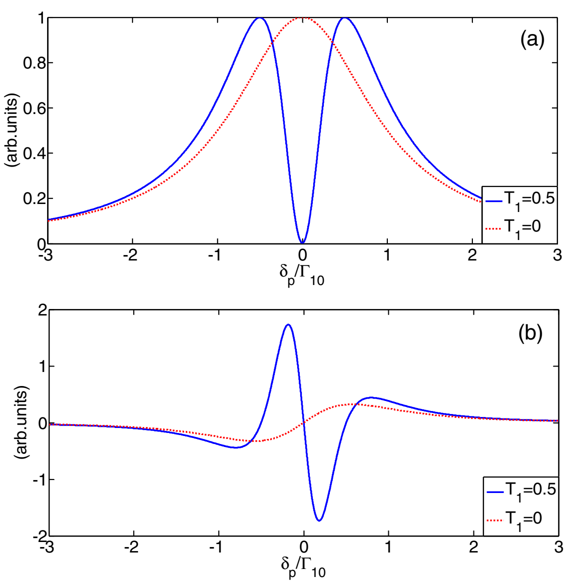

Fig. 2. Variation of $\mathrm{Re}[\chi^{(3)}]$ and $\mathrm{Im}[\chi^{(1)}]$ as a function of the probe detuning δ_p with $T_1 = 0$ (dotted line) and $T_1 = 0.5$ (solid line), (a) $\mathrm{Im}[\chi^{(1)}]$, (b) $\mathrm{Re}[\chi^{(3)}]$. Other parameters are $T_2 = 0$, $\delta_1 = 0$, $\delta_2 = 0$, $\gamma_1 = 1$, $\gamma_2 = \gamma_3 = 10^{-3}\gamma_1$.

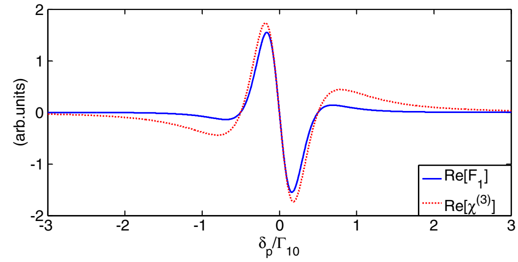

Fig. 3. Variation of Re[F_1] (solid line) and Re[$\chi^{(3)}$] (dotted line) as a function of the probe detuning δ_p. Parameters are the same as those in Fig. 2.

the coupling of tunneling T_2, the state $|2\rangle$ splits into two dressed levels $|2_\pm\rangle$ with splitting of $\sqrt{4T_2^2+\delta_2^2}$ and two Λ type subsystems can be created. In such two Λ type subsystems, the existence of two distinct dark states $|\mathrm{Dark}_\pm\rangle = (1/\sqrt{\Omega_p^2+T_{1\pm}^2})(T_{1\pm}|0\rangle - \Omega_p|2_\pm\rangle)$ is clear at hand, where $T_{1\pm}$ denote the tunneling coupling between the state $|1\rangle$ and $|2_\pm\rangle$. Each dark state corresponds to a two-photon resonance between the state $|0\rangle$ and $|2_+\rangle$, and the state $|0\rangle$ and $|2_-\rangle$, respectively. In this case, the single TIT window obtained in the DQD system splits into two TIT windows, and the position of these two transparency windows is $\delta_p = (\delta_2/2) \pm \sqrt{(\delta_2/2)^2+T_2^2}$ (dotted line in Fig. 4).

When detuning $\delta_2 = 0$, the linear absorption is symmetrical and two distinct transparency windows induced by the tunneling T_2 can be found at $\delta_p = \pm T_2$, as shown in Fig. 4(a) by the dotted line. In the vicinity of the resonance, the third-order susceptibility Re[$\chi^{(3)}$] is now significantly enhanced by about 2 orders of magnitude compared with the DQD system [solid line in Fig. 4(a)]. But such enhanced Re[$\chi^{(3)}$] is accompanied by a strong linear absorption, which is not desirable for applications of low-intensity nonlinear optics. Fortunately, one can tune the tunneling to obtain the enhanced Kerr nonlinearity with vanishing absorption.

We show in Figs. 4(b) and 4(c) the variation of the linear absorption Im[$\chi^{(1)}$] and the Kerr nonlinearity Re[$\chi^{(3)}$] with nonzero value of detuning δ_2. As detuning δ_2 is increased from 0 to 0.4, one can see from the figure that the linear absorption Im[$\chi^{(1)}$] configuration changes from symmetrical to unsymmetrical, with one broadened EIT window (left region) and one narrowed EIT window (right region). Simultaneously, the enhanced Re[$\chi^{(3)}$] gradually enters the narrowed EIT window, which means the Kerr nonlinearity is dramatically enhanced with suppressed linear absorption. The results can be analyzed in two regions. In the left region, Im[$\chi^{(1)}$] and Re[$\chi^{(3)}$] of the TQD system are similar to those of the DQD system (Fig. 2). Thus, for certain probe detuning between δ_A and δ_B in Fig. 4(c), the enhanced Re[$\chi^{(3)}$] corresponds to fractional linear absorption. While in the right region, for certain probe detuning between δ_C and δ_D, the giant enhanced Re[$\chi^{(3)}$] is accompanied by suppressed absorption, which cannot be obtained in the DQD system.

Now we provide a qualitative explanation for the origination of the enhanced Kerr nonlinearity. As can be seen, the third term of Eq. (5b) is proportional to the product of the two tunneling T_1 and T_2, which is denoted as $F_1'(T_1T_2)$. While the other two terms are independent of the product, which is

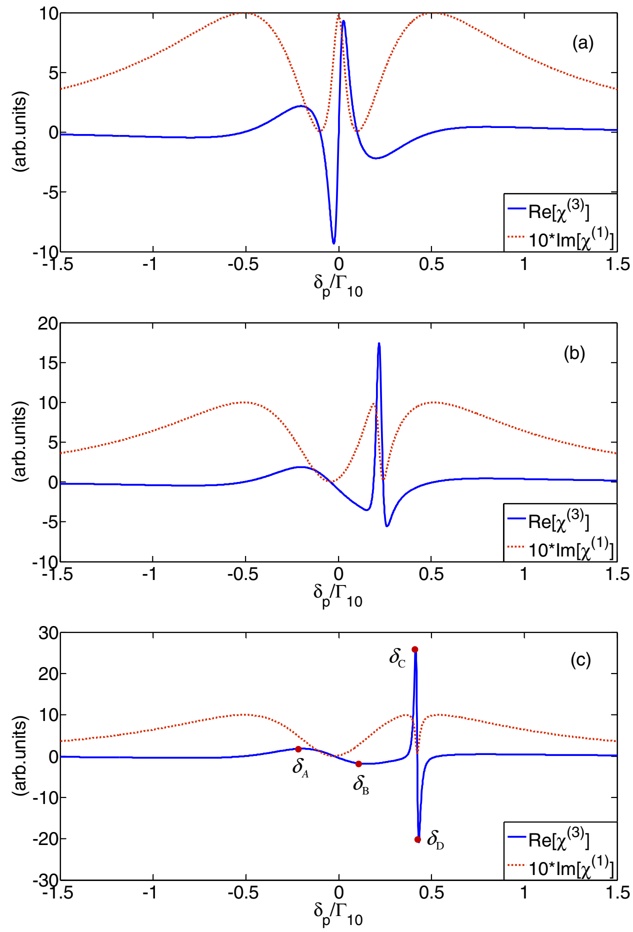

Fig. 4. Variation of Re[$\chi^{(3)}$] (solid line) and Im[$\chi^{(1)}$] (dotted line) as a function of the probe detuning δ_p with different values of detuning δ_2: (a) $\delta_2 = 0$, (b) $\delta_2 = 0.2$, (c) $\delta_2 = 0.4$. Other parameters are $T_1 = 0.5$, $\delta_1 = 0$, $T_2 = 0.1$, $\gamma_1 = 1$, $\gamma_2 = \gamma_3 = 10^{-3}\gamma_1$.

denoted as F_2'. Then $\chi^{(3)} = F_2' + F_1'(T_1T_2)$. For comparison, we plot the both the term Re[F_1'] and Re[$\chi^{(3)}$] in Fig. 5 as a function of δ_p, with $\delta_2 = 0$ and $\delta_2 = 0.4$, respectively. From the figure one can see that the profile of Re[F_1'] is almost the same with the third-order susceptibility Re[$\chi^{(3)}$] in the vicinity of the resonance. So the giant enhancement of the third-order susceptibility undoubtedly originates from the interaction of the two tunneling coupling T_1 and T_2. So here we propose a new possibility for giant enhancement of Kerr nonlinearity by combination of two tunneling between the TQDs.

To understand the detuning-dependent behavior, we consider the dressed state approach. Working in an interaction picture and taking into account only the strong tunneling coupling T_1 and T_2, the effective Hamiltonian under the basis $\{|1\rangle, |2\rangle, |3\rangle\}$ can be written as

$$H = \begin{bmatrix} 0 & -T_1 & 0 \\ -T_1 & -\delta_1 & -T_2 \\ 0 & -T_2 & -\delta_1-\delta_2 \end{bmatrix}. \tag{8}$$

And the eigenstates of this interaction Hamiltonian is the set of three linear combinations of the energy eigenstates $|1\rangle$, $|2\rangle$ and $|3\rangle$, and they are given by the formulas

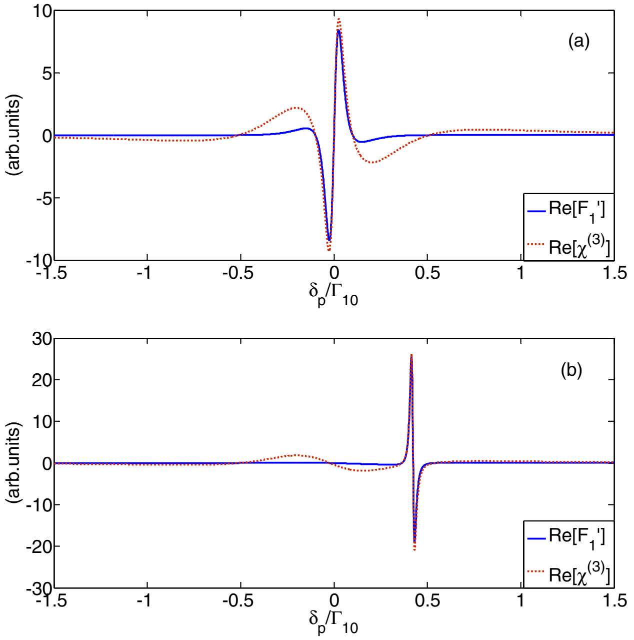

Fig. 5. Variation of Re[F_1'] (solid line) and Re[$\chi^{(3)}$] (dotted line) as a function of the probe detuning δ_p with different values of detuning δ_2: (a) $\delta_2 = 0$, (b) $\delta_2 = 0.4$. Other parameters are the same as those in Fig. 4.

$$|i\rangle = \cos\varphi \cos\theta|1\rangle + \sin\varphi|2\rangle + \cos\varphi \sin\theta|3\rangle, \tag{9}$$

where

$$\tan\varphi = \frac{AB}{\sqrt{A^2 + B^2}}, \tag{10a}$$

$$\tan\theta = \frac{A}{B}, \tag{10b}$$

$$A = \frac{\lambda_i}{T_1}, \tag{10c}$$

$$B = \frac{\lambda_i - (\delta_1 + \delta_2)}{T_2}. \tag{10d}$$

Here, λ_i is the eigenvalue of the dressed level $|i\rangle$ $(i = a, b, c)$, giving the relative energy of the dressed sublevels $|i\rangle$ $(i = a, b, c)$ generated by T_1 and T_2. Therefore, the weak probe field couples the transition from state $|0\rangle$ to the dressed state $|i\rangle$ $(i = a, b, c)$, as shown in Fig. 6(a). If the frequency of the probe field is chosen such that it is in resonance with one of the transition $|0\rangle \leftrightarrow |i\rangle$, then the maximal absorption is obtained. And the destructive interference between the three absorptive channels $|0\rangle \rightarrow |a\rangle$ and $|0\rangle \rightarrow |b\rangle$, also $|b\rangle \leftrightarrow |0\rangle$ and $|0\rangle \rightarrow |c\rangle$ can lead to the two dark states, which correspond to the two transparency windows.

From Eqs. (9) and (10), one can note that as the eigenvalues of the three dressed sublevels are dependent on the detuning δ_2, so is the position of the absorption peaks. In order to see the influence of the detuning δ_2 on the position of the dressed states, we plot the eigenenergies (λ_i, $i = a, b, c$) as a function of the detuning δ_2 [see Fig. 6(b)]. When the detuning $\delta_2 = 0$,

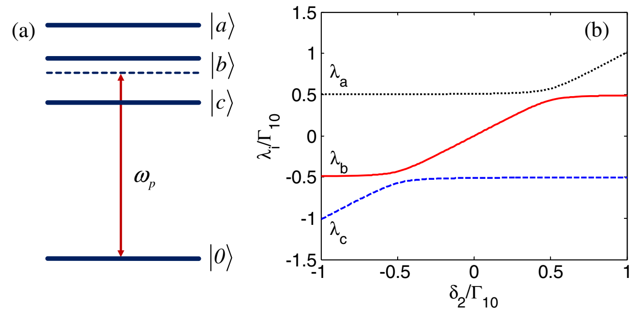

Fig. 6. (a) Dressed states under the coupling of two tunneling T_1 and T_2. (b) The eigenenergies λ_i $(i = a, b, c)$ as a function of the detuning δ_2. The parameters are the same as those in Fig. 4.

from the figure one can see that the energy difference between the dressed state $|a\rangle$ and $|b\rangle$ is equal to that of between $|b\rangle$ and $|c\rangle$. Therefore, the absorption profile is symmetrical, with the same width of two transparency windows. In this case although the Kerr nonlinearity is enhanced, it is accompanied by strong linear absorption. As changing the detuning δ_2 from 0 to 0.4, the energy difference between the dressed states $|a\rangle$ and $|b\rangle$ decreases, while the energy difference between the dressed states $|b\rangle$ and $|c\rangle$ increases. As a result, the absorption profile is unsymmetrical, and the width of the left transparency window becomes wider, and that of the right one becomes narrower. Within the narrower window, the steep dispersion profile of the probe field makes it possible to enhance the Kerr nonlinearity accompanied by vanishing absorption. The result demonstrates that the widths of the transparency window and the Kerr nonlinearity can be significantly modified by the detuning of the tunneling.

4. CONCLUSIONS

In this paper, we demonstrate that it is possible to obtain giant enhancement of Kerr nonlinearity in linear TQDs. Compared with single QD, the Kerr nonlinearity obtained in DQDs is enhanced in the TIT window. The analytical expression clearly demonstrates that the enhancement of the Kerr nonlinearity is owing to the tunneling between the DQDs. When considering the TQDs, two TIT windows are obtained and the Kerr nonlinearity can be dramatically enhanced compared to that of DQDs. And the properties of TIT windows and the Kerr nonlinearity are significantly modified by changing the detuning of the tunneling. With the proper detuning of the tunneling, the giant Kerr nonlinearity can be accompanied by vanishing linear absorption. Quantitative analysis shows that the giant enhancement of the Kerr nonlinearity is attributed to the interacting double dark resonances induced by the tunneling in the TQDs. Potential applications of such semiconductor nanostructures are to the enhancement of self-phase modulation at low light levels, such as optical solitons and self focusing.

APPENDIX A

The analytical expressions of the first- and third-order susceptibilities can be obtained by solving Eq. (4). Under the steady-state condition, Eqs. (4a)–(4d) can be set to zero; then substitute Eq. (6) into Eqs. (4a)–(4d):

$$-\Omega_p a_1 = 0, \tag{A1a}$$

$$-\Omega_p a_0 - T_1 a_2 + \Gamma_1 a_1 = 0, \tag{A1b}$$

$$-T_1 a_1 - T_2 a_3 + \Gamma_2 a_2 = 0, \tag{A1c}$$

$$-T_2 a_2 + \Gamma_3 a_3 = 0, \tag{A1d}$$

$$|a_1|^2 + |a_2|^2 + |a_3|^2 + |a_4|^2 = 1. \tag{A1e}$$

$\Gamma_3 \times$ (A1c) $- T_2 \times$ (A1d),

$$a_2 = \frac{T_1\Gamma_3}{\Gamma_2\Gamma_3 - T_2^2} a_1. \tag{A2}$$

Substituting Eq. (A2) into Eq. (A1b), then

$$a_1 = \frac{\Omega_p}{\Gamma_1 - \frac{T_1^2\Gamma_3}{\Gamma_2\Gamma_3 - T_2^2}} a_0. \tag{A3}$$

Substituting Eq. (A3) into Eq. (A2), then

$$a_2 = \frac{T_1\Gamma_3}{\Gamma_2\Gamma_3 - T_2^2} \frac{\Omega_p}{\Gamma_1 - \frac{T_1^2\Gamma_3}{\Gamma_2\Gamma_3 - T_2^2}} a_0. \tag{A4}$$

Substituting Eq. (A4) into Eq. (A1d), then

$$a_3 = \frac{T_1 T_2}{\Gamma_2\Gamma_3 - T_2^2} \frac{\Omega_p}{\Gamma_1 - \frac{T_1^2\Gamma_3}{\Gamma_2\Gamma_3 - T_2^2}} a_0. \tag{A5}$$

Substituting Eq. (A3)–(A5) into Eq. (A1e), then

$$|a_0|^2 = \frac{1}{1 + \frac{\Omega_p^2}{\left|\Gamma_1 - \frac{T_1^2\Gamma_3}{\Gamma_2\Gamma_3 - T_2^2}\right|^2}\left(1 + T_1^2 \frac{\Gamma_3^2}{|\Gamma_2\Gamma_3 - T_2^2|^2} + (T_1T_2)^2 \frac{1}{|\Gamma_2\Gamma_3 - T_2^2|^2}\right)}. \tag{A6}$$

$\Omega_p \to 0$ (TIT condition), so $(1/1 + x) \to 1 - x$. Thus,

$$|a_0|^2 = 1 - \frac{\Omega_p^2}{\left|\Gamma_1 - \frac{T_1^2\Gamma_3}{\Gamma_2\Gamma_3 - T_2^2}\right|^2} \times \left(1 + T_1^2 \frac{\Gamma_3^2}{|\Gamma_2\Gamma_3 - T_2^2|^2} + (T_1T_2)^2 \frac{1}{|\Gamma_2\Gamma_3 - T_2^2|^2}\right). \tag{A7}$$

The coherence element between state $|0\rangle$ and $|1\rangle$ is

$$\rho_{01} = a_1 a_0^* = \frac{\Omega_p}{\Gamma_1 - \frac{T_1^2\Gamma_3}{\Gamma_2\Gamma_3 - T_2^2}} |a_0|^2. \tag{A8}$$

Substituting Eq. (A7) into Eq. (A8), then

$$\rho_{01} = \frac{\Omega_p}{\Gamma_1 - \frac{T_1^2\Gamma_3}{\Gamma_2\Gamma_3 - T_2^2}} \left[1 - \frac{\Omega_p^2}{\left|\Gamma_1 - \frac{T_1^2\Gamma_3}{\Gamma_2\Gamma_3 - T_2^2}\right|^2} \times \left(1 + T_1^2 \frac{\Gamma_3^2}{|\Gamma_2\Gamma_3 - T_2^2|^2} + (T_1T_2)^2 \frac{1}{|\Gamma_2\Gamma_3 - T_2^2|^2}\right)\right]. \tag{A9}$$

The susceptibility χ is proportional to ρ_{01}; then

$$\chi = \frac{\Gamma|\boldsymbol{\mu}_{01}|^2}{V\varepsilon_0\hbar\Omega_p}\rho_{01} = \frac{\Gamma|\boldsymbol{\mu}_{01}|^2}{V\varepsilon_0\hbar} \frac{1}{\Gamma_1 - \frac{T_1^2\Gamma_3}{\Gamma_2\Gamma_3 - T_2^2}} \times \left[1 - \frac{\Omega_p^2}{\left|\Gamma_1 - \frac{T_1^2\Gamma_3}{\Gamma_2\Gamma_3 - T_2^2}\right|^2}\left(1 + T_1^2 \frac{\Gamma_3^2}{|\Gamma_2\Gamma_3 - T_2^2|^2} + (T_1T_2)^2 \frac{1}{|\Gamma_2\Gamma_3 - T_2^2|^2}\right)\right]. \tag{A10}$$

This expression includes all the linear and nonlinear contributions due to the two tunneling couplings. And susceptibility χ can be written as [6–12]

$$\chi = \chi^{(1)} + \chi^{(3)}\Omega_p^2. \tag{A11}$$

That is to say, the first-order susceptibilities are proportional to $\Omega_p^{(0)}$, and third-order susceptibilities are proportional to Ω_p^2; thus

$$\chi^{(1)} \propto \frac{1}{\Gamma_1 - \frac{T_1^2\Gamma_3}{\Gamma_2\Gamma_3 - T_2^2}}, \tag{A12a}$$

$$\chi^{(3)} \propto \frac{1}{\Gamma_1 - \frac{T_1^2\Gamma_3}{\Gamma_2\Gamma_3 - T_2^2}} \frac{1}{\left|\Gamma_1 - \frac{T_1^2\Gamma_3}{\Gamma_2\Gamma_3 - T_2^2}\right|^2} \times \left(1 + T_1^2 \frac{\Gamma_3^2}{|\Gamma_2\Gamma_3 - T_2^2|^2} + (T_1T_2)^2 \frac{1}{|\Gamma_2\Gamma_3 - T_2^2|^2}\right). \tag{A12b}$$

ACKNOWLEDGMENTS

This project received financial support from the National Basic Research Program of China (grant 2013CB933300), the National Natural Science Foundation of China (grants 11304308 and 61176046), Jilin Provincial Natural Science Foundation (grant 20140101203JC), and the Hundred Talents Program of the Chinese Academy of Sciences.

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