Giant second-order cross nonlinearities via direct perturbation to the dark state in coherent population trapping

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(Received 8 August 2019; revised manuscript received 22 December 2019; accepted 21 January 2020; published 12 February 2020)

We study the nonlinearities due to direct perturbation to the dark state in coherent population trapping (CPT). To extract the susceptibilities of the CPT atoms with respect to probe fields, we treat the CPT dressing fields as control parameters and redefine susceptibilities with respect to the probe fields alone. With such a redefinition, we reveal that a CPT-based system displays an ultralarge, resonantly enhanced second-order cross susceptibility $\chi^{(2)}$ together with vanishing linear absorption. Physically, this effect is based on the CPT dark-state shift, which is traced to the six-photon parametric processes for all involved fields (including the dressing and probe fields). Because of the lack of the direct perturbation, this effect is absent in the electromagnetically induced transparency (EIT)-based system, in which only resonantly enhanced third-order (Kerr) susceptibility $\chi^{(3)}$ is obtainable but much weaker than the second-order one. The second-order nonlinearity, because of its stronger effect, can be more sensitive for quantum nonlinear optics at low light levels than the third-order nonlinearity.

DOI: 10.1103/PhysRevA.101.023816

I. INTRODUCTION

Optical nonlinearity plays an important role in quantum optics [1,2] and quantum information processing [3], because it lays foundation for various useful applications, such as quantum nondemolition measurements [4–10], generation of squeezing and entanglement [11–15], quantum teleportation [16–19], and so on. Generally, one expects both large nonlinear susceptibilities and vanishing linear absorption for all fields participating in the nonlinear processes [20–33]. These requirements are, however, incompatible with each other in conventional optical devices. Recently, it was demonstrated that the third-order (Kerr) nonlinearities are resonantly enhanced via a coherent perturbation to electromagnetically induced transparency (EIT) [22–24].

EIT is closely related to coherent population trapping (CPT). Both of them, as one of the most representative coherence effects in laser physics, nonlinear optics and quantum optics, are common in the dark resonance mechanism but remarkably different in conditions [34–38]. Typically, they happen when two optical fields as "dressing fields" are, respectively, coupled to two arms of three-level atoms in Λ configuration. On two-photon resonance, the atoms are not excited but oscillate between two long-lived ground states. This oscillation is called the "dark resonance" because of the absence of the excitation. When the dark resonance occurs, the atoms enter one of the coherent superposition states of the two ground states. This specific superposition state is called the "dark state." The common physics of EIT and CPT lies in the dark resonance or the dark state. The difference in conditions lies in the relative amplitudes of the two dressing fields. They are in the great disparity for EIT but have comparable or equal amplitudes for CPT. The EIT atoms stay in the state to which the weak dressing field couples, while the CPT atoms are comparably or equally populated in the two ground sates. This determines the essential difference: considerably weak coherence for EIT versus large or maximal coherence for CPT. So far it has been demonstrated that a third-order (Kerr) nonlinearity is resonantly enhanced through a perturbation to EIT [22–24]. Because of the disparity in the ground state populations, however, this perturbation is only applied to the empty ground state but not to the populated ground state. This forms an *indirect* perturbation to the EIT dark state.

A natural question arises: What happens if a *direct* perturbation is introduced to the dark state? This happens most frequently in the CPT-based systems because of the comparable or equal populations in the dark state involved ground states. Because the strong dressing fields and weak probe fields are both directly coupled to the populated states, the nonlinearities with respect to all involved fields are intertwined with each other. Particularly, different orders of nonlinearities for all involved fields can correspond to the same orders only with respect to the probe fields. Usually, the interest is put in the nonlinear response to the weak probe fields since the dressing fields are much stronger and stay unchanged during the interaction. To extract the response to the weak probe fields alone, it is reasonable to merge the dressing fields into the atoms and to treat the atoms plus the dressing fields as an integrated coherent medium. Different from the usual case, the new medium consists of not only the atoms but also the fields. By separating the dressing fields as the control parameters, we are left with the resulting nonlinearities with respect to the probe fields alone. It turns out that this provides us with a convenient way to focus on the nonlinear response to the weak probe fields only. In the generalized sense we define

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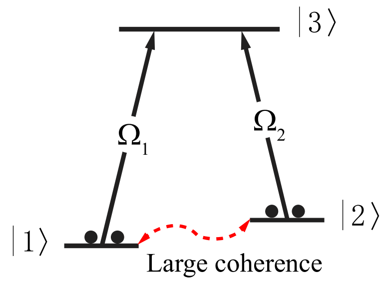

FIG. 1. A CPT atom as a dressed atom consists of a three-level atom and two dressing fields. The atom interacts with the dressing fields in Λ configuration with half Rabi frequencies $\Omega_{1,2}$. Two-photon resonance as dark resonance happens between the ground states $|1,2\rangle$ and does not contribute to the excited state. The atom is driven into dark state, which is a coherent superposition of $|1,2\rangle$. When $\Omega_1 \simeq \Omega_2$, the atom is populated comparably or equally in $|1,2\rangle$ and large or maximal coherence is established between them.

the second-order susceptibility. It is clear that this is not contrary to the fact that second-order nonlinearity vanishes for a centrosymmetric medium [21].

Here we show that the direct coherent perturbation to the CPT dark state leads to novel nonlinear effects. One of two probe fields is applied to one dark resonance arm while the other probe field is cascaded to the other arm. By examination we find a giant, resonant enhancement of second-order susceptibility with respect to the probe fields. As a physical mechanism, this revealed coherent effect originates from the CPT dark-state shift, which is dated back to six-photon parametric processes for all involved fields (including the dressing and probe fields). In view of the importance of the dark state, for consistency we use the dressed-state picture to present the essential physics and the comprehensive calculation. The essential physics is revealed by using the eigenvalue approach while the susceptibilities are derived by using the density matrix approach. These two approaches reach exactly the same results involving the nonlinear susceptibilities and interaction Hamiltonian for the probe fields. Three factors are important for the effect. The first factor is the dark state, which acts as a premise. Once formed, it stays long and there is no excitation to the excited state and subsequent spontaneous emission. The second factor is the photon exchange on one arm of the dark-resonance in Λ configuration, and the third factor is the Stark shift caused via the other arm. It is the combination of these two factors that leads to the CPT dark-state shift and the resulting second-order susceptibilities.

The remainder of this article is organized as follows. In Sec. II we compare different dark states underlying EIT and CPT. Section III presents the nonlinear response due to direct perturbation to CPT dark state, and Sec. IV compares the nonlinear susceptibilities between the EIT and CPT cases. Finally our conclusion is given in Sec. V.

II. DIFFERENT DARK STATES BETWEEN EIT AND CPT

To discriminate the indirect and direct perturbations to the dark state we first make a comparison between EIT and CPT and describe the dark state as a background. As shown in Fig. 1, an atom has two ground states $|1\rangle$ and $|2\rangle$ and the excited state $|3\rangle$. Two dressing fields resonantly interact, respectively, with the two atomic transitions $|1\rangle \leftrightarrow |3\rangle$ and $|2\rangle \leftrightarrow |3\rangle$ in Λ configuration. The Hamiltonian for the atom-field interaction is derived in the electronic dipole approximation and in an appropriate rotating frame as

$$H_0 = \hbar(\Omega_1\sigma_{31} + \Omega_2\sigma_{32} + \Omega_1^*\sigma_{13} + \Omega_2^*\sigma_{23}), \tag{1}$$

where $\hbar$ is the Planck constant, $\sigma_{kl} = |k\rangle\langle l|$ $(k, l = 1, 2, 3)$ are the projection operators for $k = l$ and the spin-flip operators $k \neq l$. By $\Omega_l = -\mu_{3l}\mathcal{E}_l/(2\hbar)$ $(l = 1, 2)$ we denote half Rabi frequencies, where μ_{3l} are the electric dipole moments and $\mathcal{E}_l$ are the amplitudes of the dressing fields. As the resonance conditions, the circular frequencies ω_l of the dressing fields are equal to the resonance circular frequencies ω_{3l} of the two arms in Λ configuration, $\omega_l = \omega_{3l}$.

From the Hamiltonian Eq. (1) we have the equation for the eigenvalue λ,

$$\begin{vmatrix} -\lambda & 0 & \Omega_1^* \\ 0 & -\lambda & \Omega_2^* \\ \Omega_1 & \Omega_2 & -\lambda \end{vmatrix} = 0. \tag{2}$$

Solving it we write the Hamiltonian in the form $H = \sum_{l=D,\pm} \hbar\lambda_l|\Psi_l\rangle\langle\Psi_l|$ with the eigenvalue states

$$\begin{aligned} |\tilde{1}\rangle &= \frac{1}{\Omega}(\Omega_2|1\rangle - \Omega_1|2\rangle), \\ |\tilde{2}\rangle &= \frac{1}{\sqrt{2}\Omega}(\Omega_1^*|1\rangle + \Omega_2^*|2\rangle + |3\rangle), \\ |\tilde{3}\rangle &= \frac{1}{\sqrt{2}\Omega}(\Omega_1^*|1\rangle + \Omega_2^*|2\rangle - |3\rangle), \end{aligned} \tag{3}$$

and the corresponding eigenvalues

$$\lambda_{\tilde{1}} = 0, \quad \lambda_{\tilde{2},\tilde{3}} = \pm\Omega, \quad \Omega = \sqrt{|\Omega_1|^2 + |\Omega_2|^2}. \tag{4}$$

We note that the state $|\tilde{1}\rangle$ remains at zero energy, while the pair of $|\tilde{2},\tilde{3}\rangle$ states are shifted up and down by an amount Ω. The states $|\tilde{2},\tilde{3}\rangle$ contain all of the bare atomic states, but in contrast, the state $|\tilde{1}\rangle$ has no contribution from $|3\rangle$ and is therefore called the "dark state." We write $|D\rangle = |\tilde{1}\rangle$ for its particularity. Once the dark state is formed there is no possibility of excitation to $|3\rangle$ and subsequent spontaneous emission. By including the spontaneous emission from $|3\rangle$ to $|1,2\rangle$ at equal rate, it is easy to find that all population indeed stays in $|D\rangle$,

$$\rho_{DD} = 1. \tag{5}$$

Since the atom is trapped in the dark state $|D\rangle$, which is at zero energy, the Hamiltonian is now

$$H_0 = 0. \tag{6}$$

The atom is transparent to the dressing fields. For this reason, the state $|D\rangle$ is usually called the dark state. This effect is usually referred to as the dark resonance, which underlies EIT and CPT. We can obtain the steady state solutions for the density matrix elements as

$$\rho_{11} = \frac{|\Omega_2|^2}{\Omega^2}, \quad \rho_{22} = \frac{|\Omega_1|^2}{\Omega^2}, \quad \rho_{12} = -\frac{\Omega_1^*\Omega_2}{\Omega^2}. \tag{7}$$

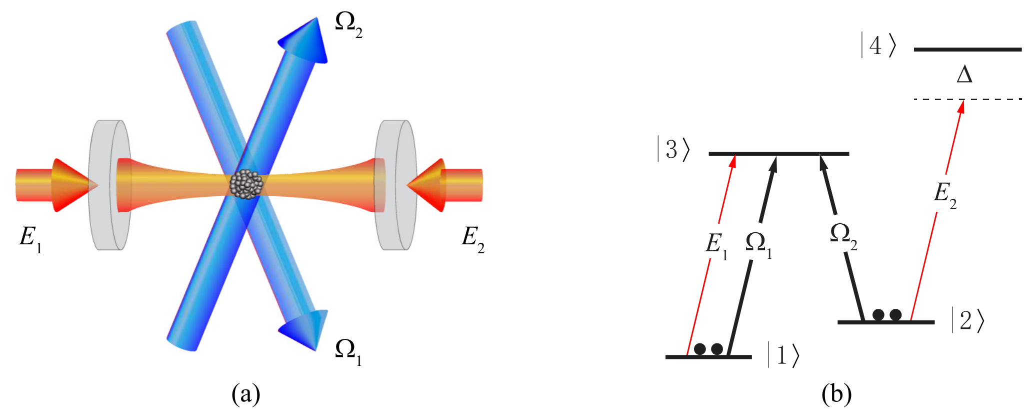

FIG. 2. (a) A possible setup for nonlinear response of the CPT atoms (the atoms plus the dressing fields $\Omega_{1,2}$ as an integrated medium) to the probe fields $E_{1,2}$. (b) Coherent perturbation to the dark state by coupling the weak probe fields $E_{1,2}$ to the populated ground states $|1,2\rangle$ involved transitions $|1\rangle - |3\rangle$ and $|2\rangle - |4\rangle$, respectively. If we remove Ω_1, then α_1 and Ω_2 ($|\Omega_2| \gg |\alpha_1|$) in EIT configuration and the atom stays mainly in $|1\rangle$. Since the empty state $|2\rangle$ is coupled to E_2, this turns out to be an indirect perturbation to the EIT dark state. In sharp contrast, however, if Ω_1 is used, $|\Omega_1| \gg |\alpha_1|$, each ground state is comparably or equally populated. Their interaction with $E_{1,2}$ turns out to be a direct perturbation to the CPT dark state. We will reveal that the direct perturbation to the CPT dark state has completely different effects from those based on the indirect perturbation to the EIT dark state.

Now we discriminate two cases. One case is for $|\Omega_1| \ll |\Omega_2|$, for which we have the EIT dark state,

$$|D\rangle \approx |1\rangle, \quad \rho_{11} \approx 1, \quad \rho_{22} \to 0, \quad \rho_{12} \to 0. \tag{8}$$

For $|\Omega_1| \ll |\Omega_2|$, the dark state is the superposition state of the extremely unbalanced populated ground states. The atom almost stays in a single state, and the coherence tends to vanish. This is just the usual EIT configuration. If a probe field is coupled to the transition involving the $|2\rangle$ state and an additional state [22,23], then the perturbation is not directly to the dark-state because the $|2\rangle$ state almost keeps empty. In sharp contrast, typically for $\Omega_1 = \Omega_2$ (real), we have the CPT dark-state

$$|D\rangle = \frac{1}{\sqrt{2}}(|1\rangle - |2\rangle), \quad \rho_{11} = \rho_{22} = -\rho_{12} = \frac{1}{2}. \tag{9}$$

Generally, when the Rabi frequencies are equal or comparable, $|\Omega_1| \simeq |\Omega_2|$, the two equally or comparably populated ground states superpose to constitute the dark-state, and we have a large or maximal coherence. Beyond the EIT case, as a discrimination, we refer to as the CPT dark-state. If any probe field is coupled to either of the two ground states, then we will have a direct perturbation to the CPT dark-state.

It should be noted that although the atom is trapped in the CPT dark-state, this does not mean the absence of the interaction of the dressing fields $\Omega_{1,2}$ with the atom. Actually, the dark-state is maintained via two-photon resonance (dark resonance) between the two ground states $|1\rangle$ and $|2\rangle$ via the excited state $|3\rangle$,

$$\begin{aligned} &|1\rangle \xrightarrow{\Omega_1} |3\rangle \xrightarrow{\Omega_2^*} |2\rangle, \\ &|1\rangle \xleftarrow{\Omega_1^*} |3\rangle \xleftarrow{\Omega_2} |2\rangle. \end{aligned} \tag{10}$$

It is for destructive interference between the two pathways that the atom is not excited to $|3\rangle$. The interfering two-photon resonances are strong nonlinear processes. Once the CPT dark-state is perturbed by a coherent perturbation, the two-photon resonances are mixed with the nonlinear processes involving in the weak probe fields. To focus on the response to the probe fields, we need to treat the atoms and the dressing fields $\Omega_{1,2}$ as an integrated coherent medium. In what follows, by "CPT atoms" we mean that the atoms and the dressing fields $\Omega_{1,2}$ act as an integrated coherent medium.

III. NONLINEARITIES VIA DIRECT PERTURBATION TO CPT DARK-STATE

To investigate the nonlinearities based on the direct perturbation to the CPT dark-state, we propose a possible setup as in Fig. 2(a) together with the atom-field interaction in N configuration as in Fig. 2(b). While CPT is established by two dressing fields (their half Rabi frequencies $\Omega_{1,2}$), applied to the CPT atoms are two probe fields (their amplitudes $E_{1,2}$). By $\alpha_1 = -\mu_{31}E_1/(2\hbar)$ and $\alpha_2 = -\mu_{42}E_2/(2\hbar)$ we denote half Rabi frequencies for the probe fields. The probe fields are much weaker than the dressing fields, $|\alpha_{1,2}| \ll |\Omega_{1,2}|$. Our purpose is to reveal the nonlinear response of the dressed atoms to the probe fields. For the convenience of discrimination, simply by $\Omega_{1,2}$ we indicate the dressing fields while by $\alpha_{1,2}$ (or $E_{1,2}$) we denote the probe fields from now on. The dressing and probe fields (Ω_1, E_1) propagate in different directions although they are of the same frequency and coupled to the common transition $|1\rangle \leftrightarrow |3\rangle$. Such a case was once employed for strong-driving-assisted multipartite entanglement in cavity QED [39,40]. The weak probe field E_2 of frequency ω_2' is far off the additional transition $|2\rangle \leftrightarrow |4\rangle$ by a large detuning $\Delta = \omega_{42} - \omega_2'$ such that the conditions are satisfied,

$$|\Delta| \gg |\Omega_{1,2}| \gg (|\alpha_{1,2}|, \Gamma), \tag{11}$$

where Γ is spontaneous decay rate for each of dipole allowed transitions $|3,4\rangle \rightsquigarrow |1,2\rangle$.

The total Hamiltonian of the system reads as $H = H_0 + H_\mathrm{I}$, where the dressing part H_0 is given in Eq. (1), and the probe part H_I takes the form in an appropriate rotating frame [1,2],

$$H_\mathrm{I} = \hbar(\alpha_1\sigma_{31} + \alpha_2\sigma_{42} + \alpha_1^*\sigma_{13} + \alpha_2^*\sigma_{24}) + \hbar\Delta\sigma_{44}. \tag{12}$$

If the dressing field Ω_1 is removed, then the present system simply reduces to the EIT-based scheme [22]. In that case, α_1 serves as a dressing field, and the atom stays mainly in the ground state $|1\rangle$. The only perturbation to EIT is the coupling of α_2 to the far detuned transition $|2\rangle - |4\rangle$, where $|2\rangle$ is almost an empty ground state. No direct perturbation is to the EIT dark state $|D\rangle \approx |1\rangle$.

However, the case differs completely for the CPT-based configuration. Since each ground state constituting the dark state is comparably or equally populated, the coherent perturbations by $\alpha_{1,2}$ are both directly applied to the dark state. Therefore, we have direct perturbation to the CPT dark state. We will focus on the response of the CPT atoms to the probe fields $\alpha_{1,2}$ and treat the dressing fields as control parameters. It will be shown that the second-order susceptibility with respect to the probe fields, which is absent in the EIT case, happens in the CPT case and dominates over the third-order one.

A. CPT dark-state shift via direct perturbation

As the essential difference, we show that the CPT dark-state shift by direct perturbation has a remarkably different dependence on the probe fields $\alpha_{1,2}$. We derive the eigenvalues and eigenstates by following the method of Zubairy *et al.* [24]. The eigenvalue equation of the total Hamiltonian $H = H_0 + H_I$ is

$$\begin{vmatrix} -\lambda & 0 & \Omega_1^* + \alpha_1^* & 0 \\ 0 & -\lambda & \Omega_2^* & \alpha_2^* \\ \Omega_1 + \alpha_1 & \Omega_2 & -\lambda & 0 \\ 0 & \alpha_2 & 0 & \Delta - \lambda \end{vmatrix} = 0, \tag{13}$$

which is written in the usual algebraic form

$$\lambda^4 + u\lambda^3 + v\lambda^2 + w\lambda + z = 0, \tag{14}$$

with parameters $u = -\Delta$, $v = -|\Omega_1 + \alpha_1|^2 - |\Omega_2|^2 - |\alpha_2|^2$, $w = \Delta(|\Omega_1 + \alpha_1|^2 + |\Omega_2|^2)$, and $z = |(\Omega_1 + \alpha_1)\alpha_2|^2$. Under the conditions Eq. (11), the Hamiltonian of the composite atom-field system can now be written in the form $H = \sum_{l=D,\tilde{2},\tilde{3},\tilde{4}} \hbar\lambda_l |l\rangle\langle l|$ with the new dressed states

$$\begin{aligned} |\tilde{1}\rangle &\approx s|1\rangle - c|2\rangle, \\ |\tilde{2}\rangle &\approx \frac{1}{\sqrt{2}}(c^*|1\rangle + s^*|2\rangle + |3\rangle), \\ |\tilde{3}\rangle &\approx \frac{1}{\sqrt{2}}(c^*|1\rangle + s^*|2\rangle - |3\rangle), \\ |\tilde{4}\rangle &\approx |4\rangle, \end{aligned} \tag{15}$$

with the corresponding eigenvalues

$$\lambda_{\tilde{1}} = -\frac{|c\alpha_2|^2}{\Delta}, \quad \lambda_{\tilde{2},\tilde{3}} = \pm\tilde{\Omega}, \quad \lambda_{\tilde{4}} = \Delta, \tag{16}$$

where we have used the parameters

$$\begin{aligned} c &= \frac{\Omega_1 + \alpha_1}{\tilde{\Omega}}, \\ s &= \frac{\Omega_2}{\tilde{\Omega}}, \\ \tilde{\Omega} &= \sqrt{|\Omega_1 + \alpha_1|^2 + |\Omega_2|^2}. \end{aligned} \tag{17}$$

It is clear that the first dressed state $|\tilde{1}\rangle$, containing no excited state, is a dark state. Without confusion we also write $|D\rangle = |\tilde{1}\rangle$ for its importance. This dark state is slightly different from that in Eq. (3) because of the addition of α_1 to Ω_1. Differently, the CPT dark state λ_D has its energy shift $\lambda_D = \lambda_{\tilde{1}}$. Including the atomic relaxations we obtain $\rho_{DD} \approx 1$, as will be verified below. The Hamiltonian can be now reduced to

$$H \approx \hbar\lambda_D |D\rangle\langle D|. \tag{18}$$

It is easy to understand that the dark-state shift λ_D is proportional to the $|2\rangle$ state population $\rho_{22} = |c|^2 = |\Omega_1 + \alpha_1|^2/\tilde{\Omega}^2$ and the linear Stark shift of an entire atom $-|\alpha_2|^2/\Delta$. Both of them combine to give the contribution of the direct perturbation to the dark state.

Let us look first at the power series expansion of the dark-state shift λ_D in the probe fields $\alpha_{1,2}$. Noting that λ_D is now already in the second order in α_2, it is enough for us to expand λ_D up to the second orders in α_1. The resulting expansion is written in the form

$$\begin{aligned} \lambda_D = &-\frac{|\Omega_1|^2}{\Delta\Omega^2}|\alpha_2|^2 - \frac{|\Omega_2|^2}{\Delta\Omega^4}(\Omega_1^*\alpha_1 + \Omega_1\alpha_1^*)|\alpha_2|^2 \\ &- \frac{|\Omega_2|^2}{\Delta\Omega^4}\left(1 - \frac{2|\Omega_1|^2}{\Omega^2}\right)|\alpha_1\alpha_2|^2 \\ &+ \frac{|\Omega_2|^2}{\Delta\Omega^6}\left(\Omega_1^{*2}\alpha_1^2 + \Omega_1^2\alpha_1^{*2}\right)|\alpha_2|^2. \end{aligned} \tag{19}$$

If $\Omega_1 = 0$, then Eq. (19) reduces to

$$\lambda_D = -\frac{|\alpha_1\alpha_2|^2}{\Delta\Omega_2^2}, \tag{20}$$

which gives just the EIT-based third-order (Kerr) susceptibility [22].

However, there appears the power series in $\Omega_{1,2}$ in the general CPT case ($|\Omega_1| \gg |\alpha_1|$, saturated by Ω). This is just a consequence of the direct perturbation to the CPT dark state. Once $\Omega_1 = 0$ all these series terms disappear. However, in the presence of Ω_1, different orders of nonlinearities for all involved fields can correspond to the same orders only with respect to the probe fields $\alpha_{1,2}$. Because we are interested in the response of the dressed CPT atoms to the probe fields $\alpha_{1,2}$, it is more interesting to treat the CPT atoms plus the dressing fields $\Omega_{1,2}$ as an integrated coherent medium. To focus on the response to the probe fields $\alpha_{1,2}$ alone, it is more reasonable to define susceptibilities only for them and to treat the dressing fields as control parameters at the same time.

In this sense, the first term in Eq. (19) corresponds to a linear response of the CPT atoms to the probe field α_2, as will be shown below. Since the ground state $|2\rangle$ has its population $\rho_{22} = |\Omega_1|^2/\Omega^2$, the direct perturbation to the populated $|2\rangle$ state gives just rise to the linear Stark shift with the proportion to $|\Omega_1|^2/\Omega^2$. Once $\Omega_1 = 0$, the population vanishes $\rho_{22} \to 0$ and this shift vanishes [22]. The CPT dark-state shift via the direct perturbation only appears for the CPT case and is absent for the EIT case [22]. The second term in Eq. (19), i.e., the $(\Omega_1^*\alpha_1 + \Omega_1\alpha_1^*)|\alpha_2|^2$ term, which appears as a nonlinear effect for the CPT case and is also absent for the EIT case, belongs to the second-order susceptibility. Obviously, this originates from the direct perturbation to the CPT dark state.

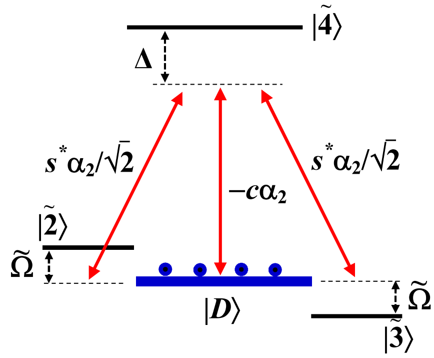

FIG. 3. The coherent perturbation to the CPT dark state $|D\rangle$ by the probe fields $\alpha_{1,2}$. α_1 is merged in the dressing field Rabi frequency dependent coupling coefficients (c, s). The direct perturbation to the dark state depends on Ω_1 in (c, s).

Certainly, because of the separation of the dressing fields, this effect is not conflicting to the previous belief that the second-order cross nonlinearities are not allowed for all involved fields. The last two terms in Eq. (19) describe the third-order nonlinearities. It is seen from the above analysis that due to the comparable or equal Rabi frequencies $|\Omega_1| \sim |\Omega_2|$, the direct perturbation to the dark state happens most frequently in the CPT-based systems.

B. Susceptibilities via direct perturbation

To confirm the above essential physics we turn to using the standard nonlinear optics method [20–22] to calculate the susceptibilities. This is done in exactly the same dressed-states picture for consistency. We rewrite the total Hamiltonian $H = H_0 + H_{\rm I}$ in the form

$$
\begin{aligned}
H &= \hbar(\tilde{\Omega}\sigma_{\tilde{2}\tilde{2}} - \tilde{\Omega}\sigma_{\tilde{3}\tilde{3}} + \Delta\sigma_{\tilde{4}\tilde{4}}) \\
&\quad + \hbar\alpha_2\left(-c\sigma_{\tilde{4}D} + \frac{s^*}{\sqrt{2}}\sigma_{\tilde{4}\tilde{2}} + \frac{s^*}{\sqrt{2}}\sigma_{\tilde{4}\tilde{3}}\right) \\
&\quad + \hbar\alpha_2^*\left(-c^*\sigma_{D\tilde{4}} + \frac{s}{\sqrt{2}}\sigma_{\tilde{2}\tilde{4}} + \frac{s}{\sqrt{2}}\sigma_{\tilde{3}\tilde{4}}\right). \qquad (21)
\end{aligned}
$$

In Fig. 3 given is the pictorial representation for the interaction of the dressed CPT atoms with the probe fields. It should be noted that the probe field α_1 is contained in the $\Omega_{1,2}$ dependent coupling coefficients (c, s).

We first analyze the the direct perturbation to the CPT dark state from Hamiltonian Eq. (21) before we solve for the susceptibilities. Since the atom is trapped in the dark state $|D\rangle$ and all other states $|\tilde{2} - \tilde{4}\rangle$ are empty, the energy shift of the dark state $|D\rangle$ is important. In the order of $|\alpha_2|^2/\Delta$, the $\sigma_{\tilde{2}\tilde{4}}$ and $\sigma_{\tilde{3}\tilde{4}}$ transitions have no contributions. The only contributions come from the dispersive $\sigma_{\tilde{4}D}$ transition. The amount of the dark-state shift can readily obtained as $\lambda_D = -|c\alpha_2|^2/\Delta$, which is exactly the same as in Eq. (16). If $\Omega_1 = 0$, i.e., $c \approx \alpha_1/\Omega_2$, then the effective Rabi frequency for the $\sigma_{\tilde{4}D}$ transition is $\alpha_1\alpha_2/\Omega_2$ and the dark-state shift is $|\alpha_1\alpha_2|^2/\Delta\Omega_2$. This originates from the fact that α_2 is only coupled to the empty state $|2\rangle$. This is the called indirect perturbation to the EIT dark state. However, if the dressing field Ω_1 is applied and $|\Omega_1| \gg |\alpha_1|$, the parameter c contains the power series in Ω_1. These power series in Ω_1 are a consequence of the direct coupling to the populated ground states $|1, 2\rangle$. Because of the populations in the ground states constituting the CPT dark state, the direct perturbation to the CPT dark state is most frequently met. More importantly, up to third-order in the probe fields $\alpha_{1,2}$, the nonlinearities contained in the dark-state shift λ_D are exactly the same as those in the following polarization of the CPT atoms.

From the above analysis and the conditions Eq. (11) we can deduce roughly that the $\sigma_{\tilde{4}D}$ transition contributes to the density matrix element $\rho_{D\tilde{4}} \propto c^*\alpha_2^*/\Delta$ in the linear order in α_2. The $\sigma_{\tilde{2}\tilde{4}}$ and $\sigma_{\tilde{3}\tilde{4}}$ transitions alone do not contribute to any response to the probe fields. However, the cascade transitions $\sigma_{D\tilde{4}}$ and $\sigma_{\tilde{4}\tilde{2}}$ will contribute to $\rho_{D\tilde{2}} \propto c^*s^*|\alpha_2|^2/\Delta\tilde{\Omega}$ in the second order, and the cascade transitions $\sigma_{D\tilde{4}}$ and $\sigma_{\tilde{4}\tilde{3}}$ will contribute to $\rho_{D\tilde{3}}$ similarly. In what follows we present the derivation in details.

The master equation of the present system takes the standard form $\dot{\rho} = -\frac{i}{\hbar}[H, \rho] + \mathcal{L}\rho$, where $\mathcal{L}\rho$ is the $\mathcal{L}\rho{=}\mathcal{L}\rho_{13}+\mathcal{L}\rho_{23} + \mathcal{L}\rho_{14} + \mathcal{L}\rho_{24}$, $\quad\mathcal{L}\rho_{kl} = \frac{\Gamma}{2}(2\sigma_{kl}\rho\sigma_{lk} - \sigma_{ll}\rho - \rho\sigma_{ll})$. Expressing the relaxations in terms of dressed states and neglecting the rapidly oscillating terms, we derive the equations for the density matrix elements. The populations $(\rho_{DD}, \rho_{\tilde{2}\tilde{2}}, \rho_{\tilde{3}\tilde{3}})$ read

$$
\begin{aligned}
\dot{\rho}_{DD} &= \frac{\Gamma}{2}(\rho_{\tilde{2}\tilde{2}} + \rho_{\tilde{3}\tilde{3}}) + \Gamma\rho_{\tilde{4}\tilde{4}} - ic\alpha_2\rho_{D\tilde{4}} + ic^*\alpha_2^*\rho_{\tilde{4}D}, \\
\dot{\rho}_{\tilde{2}\tilde{2}} &= -\frac{3\Gamma}{4}\rho_{\tilde{2}\tilde{2}} + \frac{\Gamma}{4}\rho_{\tilde{3}\tilde{3}} + \frac{\Gamma}{2}\rho_{\tilde{4}\tilde{4}} + \frac{is^*\alpha_2}{\sqrt{2}}\rho_{\tilde{2}\tilde{4}} - \frac{is\alpha_2^*}{\sqrt{2}}\rho_{\tilde{4}\tilde{2}}, \\
\dot{\rho}_{\tilde{3}\tilde{3}} &= -\frac{3\Gamma}{4}\rho_{\tilde{3}\tilde{3}} + \frac{\Gamma}{4}\rho_{\tilde{2}\tilde{2}} + \frac{\Gamma}{2}\rho_{\tilde{4}\tilde{4}} + \frac{is^*\alpha_2}{\sqrt{2}}\rho_{\tilde{3}\tilde{4}} - \frac{is\alpha_2^*}{\sqrt{2}}\rho_{\tilde{4}\tilde{3}},
\end{aligned}
\qquad (22)
$$

the equations for the off-diagonal elements $(\rho_{D\tilde{2}}, \rho_{D\tilde{3}}, \rho_{\tilde{2}\tilde{3}})$ not involving $|\tilde{4}\rangle$ are written

$$
\begin{aligned}
\dot{\rho}_{D\tilde{2}} &= -\left(\frac{\Gamma}{2} - i\tilde{\Omega}\right)\rho_{D\tilde{2}} + ic^*\alpha_2^*\rho_{\tilde{4}\tilde{2}} + \frac{is^*\alpha_2}{\sqrt{2}}\rho_{D\tilde{4}}, \\
\dot{\rho}_{D\tilde{3}} &= -\left(\frac{\Gamma}{2} + i\tilde{\Omega}\right)\rho_{D\tilde{3}} + ic^*\alpha_2^*\rho_{\tilde{4}\tilde{3}} + \frac{is^*\alpha_2}{\sqrt{2}}\rho_{D\tilde{4}}, \qquad (23) \\
\dot{\rho}_{\tilde{2}\tilde{3}} &= -\left(\frac{5}{4}\Gamma + 2i\tilde{\Omega}\right)\rho_{\tilde{2}\tilde{3}} + \frac{is^*\alpha_2}{\sqrt{2}}\rho_{\tilde{2}\tilde{4}} - \frac{is\alpha_2^*}{\sqrt{2}}\rho_{\tilde{4}\tilde{3}},
\end{aligned}
$$

and equations for the off-diagonal elements $(\rho_{D\tilde{4}}, \rho_{\tilde{2}\tilde{4}}, \rho_{\tilde{3}\tilde{4}})$ associated with $|\tilde{4}\rangle$ are listed

$$
\begin{aligned}
\dot{\rho}_{D\tilde{4}} &= -(\Gamma - i\Delta)\rho_{D\tilde{4}} + ic^*\alpha_2^*(\rho_{\tilde{4}\tilde{4}} - \rho_{DD}) \\
&\quad + \frac{is\alpha_2^*}{\sqrt{2}}\rho_{D\tilde{2}} + \frac{is\alpha_2^*}{\sqrt{2}}\rho_{D\tilde{3}}, \\
\dot{\rho}_{\tilde{2}\tilde{4}} &= -\left[\frac{3\Gamma}{2} - i(\Delta - \tilde{\Omega})\right]\rho_{\tilde{2}\tilde{4}} - ic^*\alpha_2^*\rho_{\tilde{2}D} \\
&\quad - \frac{is\alpha_2^*}{\sqrt{2}}(\rho_{\tilde{4}\tilde{4}} - \rho_{\tilde{2}\tilde{2}}) + \frac{is\alpha_2^*}{\sqrt{2}}\rho_{\tilde{2}\tilde{3}}, \\
\dot{\rho}_{\tilde{3}\tilde{4}} &= -\left[\frac{3\Gamma}{2} - i(\Delta + \tilde{\Omega})\right]\rho_{\tilde{3}\tilde{4}} - ic^*\alpha_2^*\rho_{\tilde{3}D} \\
&\quad - \frac{is\alpha_2^*}{\sqrt{2}}(\rho_{\tilde{4}\tilde{4}} - \rho_{\tilde{3}\tilde{3}}) + \frac{is\alpha_2^*}{\sqrt{2}}\rho_{\tilde{3}\tilde{2}}.
\end{aligned}
\qquad (24)
$$

We consider the solutions at the steady state, i.e., let $\dot{\rho}_{kl}=0$. Substituting the off-diagonal elements into the equations for populations, using the conditions Eq. (11) and the closure relation $\rho_{DD}+\rho_{\tilde{2}\tilde{2}}+\rho_{\tilde{3}\tilde{3}}+\rho_{\tilde{4}\tilde{4}}=1$, we obtain the only nonvanishing population $\rho_{DD}\approx 1$, which verifies indeed that the atom is trapped in its dark state.

To make a comparison and show the dominance of the second-order susceptibility, we derive the susceptibilities up to the third-order. We solve Eq. (24) and express $(\rho_{D\tilde{4}},\rho_{\tilde{2}\tilde{4}},\rho_{\tilde{3}\tilde{4}})$ in terms of $(\rho_{D\tilde{2}},\rho_{D\tilde{3}},\rho_{\tilde{2}\tilde{3}})$, and then we substitute the resulting expressions into Eq. (23). As predicted as above, nonzero elements sufficient for the third-order susceptibilities are the dark-state-involved elements, which are obtained as

$$\rho_{D\tilde{2}}=-\rho_{D\tilde{3}}=-\frac{c^*s^*|\alpha_2|^2}{\sqrt{2}\Delta\tilde{\Omega}},\quad \rho_{D\tilde{4}}=\frac{c^*\alpha_2^*}{\Delta}. \tag{25}$$

It is clear that only the dark-state-involved density matrix elements are nonvanishing. Expressing the bare state elements in terms of the nonvanishing dressed state elements

$$\rho_{13}=-\frac{s}{c}\rho_{23}=\frac{s}{\sqrt{2}}(\rho_{D\tilde{2}}-\rho_{D\tilde{3}}),\quad \rho_{24}=-c\rho_{D\tilde{4}}, \tag{26}$$

we obtain

$$\rho_{13}=-\frac{c^*|s\alpha_2|^2}{\Delta\tilde{\Omega}},\quad \rho_{23}=\frac{s^*|c\alpha_2|^2}{\Delta\tilde{\Omega}},\quad \rho_{24}=-\frac{|c|^2\alpha_2^*}{\Delta}. \tag{27}$$

It should be emphasized that the linear and nonlinear contributions of the direct perturbation to the CPT dark state are contained in (c,s).

Now we are in a position to collect the bare-state off-diagonal elements $(\rho_{13},\rho_{23},\rho_{24})$ into the total complex polarization and perform an expansion in α_1. Including the contributions from all three transitions ($|1\rangle\to|3\rangle$, $|3\rangle\to|2\rangle$, $|2\rangle\to|4\rangle$), we obtain the total complex polarization [21]

$$\boldsymbol{P}=\boldsymbol{e}_1\mu_{13}\rho_{31}+\boldsymbol{e}_2\mu_{24}\rho_{42}+\boldsymbol{e}_3\mu_{23}\rho_{32}, \tag{28}$$

where the orthogonal unit vectors $(\boldsymbol{e}_1,\boldsymbol{e}_2,\boldsymbol{e}_3)$ correspond to the dipole moments $(\mu_{13},\mu_{24},\mu_{23})$, respectively, and contain the information about the polarizations and frequencies. We have the selection rules, $\boldsymbol{E}_k\cdot\boldsymbol{e}_l=E_k\delta_{kl}$ $(k=1,2;l=1,2,3)$. Expanding to the second order in α_1 and placing the dressing field Rabi frequencies $\Omega_{1,2}$ and the atomic dipole moments (μ_{13},μ_{24}) in the susceptibilities, we express the total polarization in terms of the probe field amplitudes $E_{1,2}$,

$$\boldsymbol{P}=\epsilon_0\boldsymbol{e}_1\begin{pmatrix}\chi^{(2)*}|E_2|^2\\+\chi_1^{(3)}E_1|E_2|^2\\+2\chi_2^{(3)*}E_1^*|E_2|^2\end{pmatrix}+\epsilon_0\boldsymbol{e}_2\begin{pmatrix}\chi^{(1)}E_2\\+\chi^{(2)}E_1E_2\\+\chi^{(2)*}E_1^*E_2\\+\chi_1^{(3)}|E_1|^2E_2\\+\chi_2^{(3)}E_1^2E_2\\+\chi_2^{(3)*}E_1^{*2}E_2\end{pmatrix}+\epsilon_0\boldsymbol{e}_3\begin{pmatrix}\chi'^{(2)}|E_2|^2\\+\chi_1'^{(3)}E_1|E_2|^2\\+\chi_1'^{(3)*}E_1^*|E_2|^2\end{pmatrix}, \tag{29}$$

where the resulting susceptibilities read

$$\chi^{(1)}=\frac{|\mu_{24}|^2N|\Omega_1|^2}{2\epsilon_0\hbar\Delta\Omega^2}, \tag{30}$$

$$\chi^{(2)}=-\frac{\mu_{13}^*|\mu_{24}|^2N\Omega_1^*|\Omega_2|^2}{4\epsilon_0\hbar^2\Delta\Omega^4}, \tag{31}$$

$$\chi_1^{(3)}=\frac{|\mu_{13}\mu_{24}|^2N|\Omega_2|^2}{8\epsilon_0\hbar^3\Delta\Omega^4}\left(1-\frac{2|\Omega_1|^2}{\Omega^2}\right), \tag{32}$$

$$\chi_2^{(3)}=-\frac{\mu_{13}^{*2}|\mu_{24}|^2N\Omega_1^{*2}|\Omega_2|^2}{8\epsilon_0\hbar^3\Delta\Omega^6}. \tag{33}$$

ϵ_0 is the permittivity of free space, and the contributions of N atoms are included. We have not listed χ''s here since they do not contribute to the probe fields, $\boldsymbol{E}_{1,2}\cdot\boldsymbol{e}_3=0$. In addition, the decay of the atom can be neglected because of the negligible ratio of the dissipation to dispersion contribution $\Gamma/|\Delta|\ll 1$.

Here we have defined the orders n in the susceptibilities $\chi^{(n)}$ with respect to only the probe fields $E_{1,2}$ and have used the dressing fields $\Omega_{1,2}$ as the control parameters. Perhaps one finds immediately that the nonlinear dependencies of $\chi^{(n)}$ on the Rabi frequencies $\Omega_{1,2}$ exactly appear in the dark-state shift Eq. (19). As shown above, these dependencies of $\chi^{(n)}$ on $\Omega_{1,2}$ are simply the contributions from the direct perturbation to the CPT dark state. Indeed, the dark-state shift Eq. (19) and the atomic polarization Eq. (28) give exactly the same nonlinearities. Also the polarization Eq. (29) is expressed only in terms of the amplitudes of the probe fields, but does not display the frequency factors. This is because we work in an appropriate rotating frame, where the frequency factors are removed. Actually, if we recover all the frequency factors we have vanishing sums of the involved frequencies. This indicates that the multiphoton parametric resonances of the dressing and probe fields $(\Omega_{1,2},E_{1,2})$ with the three-level atoms are established through a round trip of cascade transitions, as will be shown in the following section. In each round trip, the energy and moment conservations are guaranteed, and the symmetry of frequency permutations is satisfied. In principle, the susceptibilities are net contributions of different multiphoton processes because the susceptibilities in the same order can come from different multiphoton processes involving the dressing fields. Matching the Rabi frequencies $\Omega_{1,2}$ and dipole moments $(\mu_{13},\mu_{23},\mu_{24})$ in the the numerators of the susceptibility expressions, we can list the dressing and probe field frequencies as a possible form

$$\chi^{(1)}=\chi_1^{(1)}(-\omega_2',\omega_2')+\chi_2^{(1)}(-\omega_2',\omega_2',-\omega_2,\omega_2), \tag{34}$$

$$\chi^{(2)}=\chi^{(2)}(-\omega_1,\omega_2,-\omega_2',\omega_2',-\omega_2,\omega_1), \tag{35}$$

$$\begin{aligned}\chi_1^{(3)}=&\ \chi_{11}^{(3)}(-\omega_1,\omega_2,-\omega_2',\omega_2',-\omega_2,\omega_1)\\&+\chi_{12}^{(3)}(-\omega_1,\omega_2,-\omega_2',\omega_2',-\omega_2,\omega_1,-\omega_1,\omega_1),\end{aligned} \tag{36}$$

$$\chi_2^{(3)}=\chi_2^{(3)}(-\omega_1,\omega_2,-\omega_2',\omega_2',-\omega_2,\omega_1,-\omega_1,\omega_1), \tag{37}$$

where we have used $\frac{|\Omega_1|^2}{\Omega^2}=1-\frac{|\Omega_2|^2}{\Omega^2}$ for $\chi^{(1)}$ and have divided it into two parts.

The probe fields have their self-consistent response to the polarization of the CPT atoms. Noting the selection rules, $\boldsymbol{E}_k \cdot \boldsymbol{e}_l = E_k \delta_{kl}$ $(k = 1, 2; l = 1, 2, 3)$, we obtain the self-consistent equations for the amplitudes [2]

$$\dot{E}_1 = -i\omega_1 \begin{pmatrix} \chi^{(2)*}|E_2|^2 \\ +\chi_1^{(3)} E_1 |E_2|^2 \\ +2\chi_2^{(3)*} E_1^* |E_2|^2 \end{pmatrix} - \frac{\kappa_1}{2} E_1, \quad (38)$$

$$\dot{E}_2 = -i\omega_2' \begin{pmatrix} \chi^{(1)} E_2 \\ +\chi^{(2)} E_1 E_2 \\ +\chi^{(2)*} E_1^* E_2 \\ +\chi_1^{(3)} |E_1|^2 E_2 \\ +\chi_2^{(3)} E_1^2 E_2 \\ +\chi_2^{(3)*} E_1^{*2} E_2 \end{pmatrix} - \frac{\kappa_2}{2} E_2, \quad (39)$$

where the cavity loss rates $\kappa_{1,2}$ are also included.

Following the standard technique we can treat the probe fields quantum mechanically [1,2]. Using the annihilation and creation operators $(a_l, a_l^\dagger; l = 1, 2)$ to express the probe fields, $E_l = a_l \varepsilon_l$ (where $\varepsilon_l = \sqrt{\hbar \nu_l / V \epsilon_0}$; $\nu_l = \omega_1, \omega_2'$ is the electric field "per photon," V is the cavity volume), from the χ's terms in Eqs. (38) and (39) we obtain immediately the effective Hamiltonian for the interaction between the probe fields

$$H_\mathrm{I} = \hbar\big(\tilde{\chi}^{(1)} + \tilde{\chi}^{(2)} a_1 + \tilde{\chi}^{(2)*} a_1^\dagger + \tilde{\chi}_1^{(3)} a_1^\dagger a_1 + \tilde{\chi}_2^{(3)} a_1^2 + \tilde{\chi}_2^{(3)*} a_1^{\dagger 2}\big) a_2^\dagger a_2, \quad (40)$$

where the susceptibilities with tildes $\tilde{\chi}^{(n)}$ (of which $\tilde{\chi}^{(1)}$ is the Stark shift and $\tilde{\chi}^{(2,3)}$ are the cross interaction strengths) take the same forms as $(\epsilon_0/\hbar)\chi^{(n)}$ in Eqs. (30)–(33) with the substitutions of $(\mu_{31}\varepsilon_1, \mu_{42}\varepsilon_2)$ for the dipole moments (μ_{31}, μ_{42}), respectively.

Actually, exactly the same Hamiltonian is obtained from the CPT dark-state shift in Eq. (19) by the above substitutions. This simply verifies that the Hamiltonian for the interaction between the probe fields is just a result of the dark-state shift.

C. Dominance of second- over third-order nonlinearity

It is easy to verify that the second-order susceptibility $\chi^{(2)}$ dominates over the third-order susceptibility $\chi_{1,2}^{(3)}$. For the present case $|\Omega_1| = |\Omega_2|$, we have $\chi_1^{(3)} = 0$. Meanwhile, the ratio of second- to third-order susceptibility can be readily obtained,

$$\frac{|\chi^{(2)}|}{|\chi_2^{(3)} E_1|} = \frac{|\Omega_1|}{|\alpha_1|} \gg 1. \quad (41)$$

Since the dressing field is much stronger than the probe field, $|\Omega_1| \gg |\alpha_1|$, we have the second-order polarization dominant over the third-order polarization. As Schmidt and Imamoglu [22] once showed, the susceptibility $\chi_1^{(3)}$ is raised even by 9 orders compared with the usual three-level systems. Since a giant, resonantly enhanced second-order nonlinearity $\chi^{(2)}$ dominates over the three-order susceptibility, the second-order susceptibility $\chi^{(2)}$ can be much more greatly increased, by several orders depending on the ratio $|\Omega_1/\alpha_1|$. Neglecting the third-order parts, we are left with the second-order complex polarization with respect to the weak probe fields,

$$\boldsymbol{P} = \epsilon_0(\boldsymbol{e}_1 \chi^{(2)*}|E_2|^2 + \boldsymbol{e}_2 \chi^{(2)} E_1 E_2 + \boldsymbol{e}_2 \chi^{(2)*} E_1^* E_2). \quad (42)$$

Correspondingly, the Hamiltonian for the second-order nonlinear interaction between the probe fields is established as

$$H_\mathrm{I} = \hbar(\tilde{\chi}^{(2)} a_1 + \tilde{\chi}^{(2)*} a_1^\dagger) a_2^\dagger a_2. \quad (43)$$

Equations (42) and (43) are our central result.

So far we have presented the nonlinear response of the dressed CPT atoms to the probe fields close to the dark state. The nonlinear susceptibilities are only confined to the probe fields, while the dressing fields are treated as the control parameters. The nonlinear dark-state shift as the physical essence is given by using the eigenvalue approach, while the nonlinear susceptibilities are calculated by using the density matrix approach. These two aspects yield exactly the same results, including the susceptibilities and the Hamiltonian for the interaction between the probe fields.

We are now in a position to summarize the three fundamental elements for the effects of the direct perturbation to the CPT dark state.

(i) The first factor is the formation of the CPT dark state $|D\rangle$ with comparable or equal Rabi frequencies $\Omega_{1,2}$. The other states are far off the dark state. The superposition states $|\tilde{2}, \tilde{3}\rangle$ are shifted by large spacing $\pm\Omega$, and the additional state $|4\rangle$ remains far off the dark state by detuning Δ. In other words, the atom always stays in the dark state during the interaction. Note that this does not mean that the dressing fields $\Omega_{1,2}$ do not interact with the dark-state atom. In fact, the two-photon processes between the two ground states $|1, 2\rangle$ always are existent in the parametric processes. It is the very dark resonance that determines the existence of parametric processes although the resonant transitions are involved. The expressions Ω^{2l} $(l = 1 - 3)$ in the denominator of χ's reflect the saturation effects. However, the saturation does not excite the atoms during the interaction, but sustains the dark state and induces the dark-state-based parametric interaction.

(ii) The second factor is the photon exchange between the probe field E_1 and the dressing field Ω_1. This is seen by noting the appearance of $\mu_{13}^* \Omega_1^*$ in the numerator of $\chi^{(2)}$, and the appearance of $\mu_{13}^{*2} \Omega_1^{*2}$ in the numerators of $\chi_2^{(3)}$. This happens for the CPT case $(|\Omega_1| \gg |\alpha_1|)$ but is absent for the EIT case $(\Omega_1 = 0)$. The photon exchange originates from the direct perturbation to the CPT dark state since the probe field E_1 is coupled to one of the dark resonant arms.

(iii) The third factor is the linear Stark shift $\chi^{(1)}$ of the dark state $|D\rangle$ due to the field E_2. Once we remove E_2 $(\mu_{24} = 0)$ the shift vanishes. Once we remove Ω_1, we come to the EIT case $(\Omega_1 = 0)$ and the dark-state shift also vanishes. For the present CPT case, although the frequency shift can be merged into the large detuning Δ, it brings us the nonlinear effects. This is seen by noting those nonlinear terms involving $|\Omega_1|^2$ in $\chi_1^{(3)}$. The shift is the consequence of the direct perturbation to the CPT dark state because the dark-state-involved ground state $|2\rangle$ coupled to the probe field E_2 is largely populated.

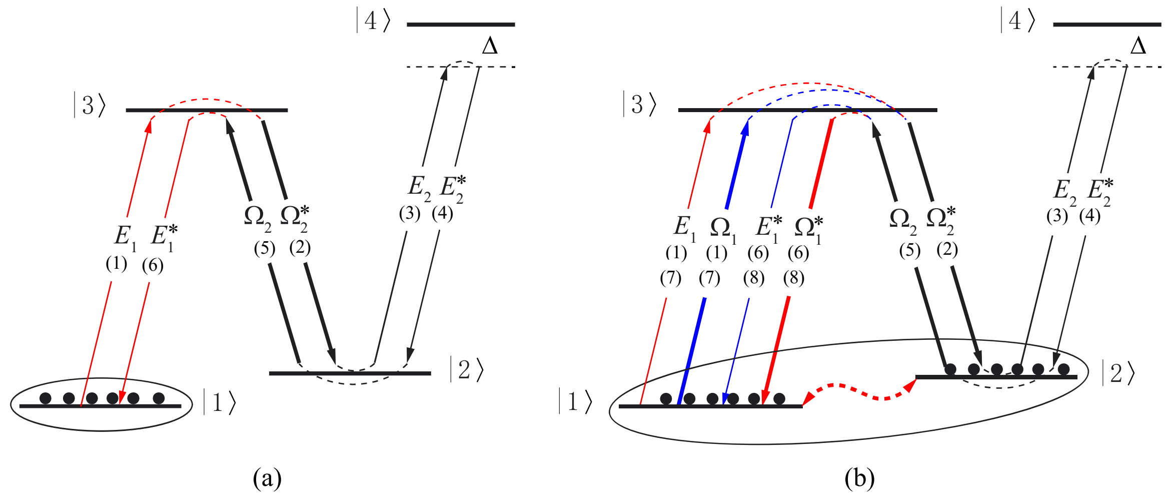

FIG. 4. Going and return lines for multiphoton parametric interactions. The numbers in the circular brackets indicate the successive steps in a round trip. As a comparison, (a) is for the indirect perturbation to the EIT dark state (i.e., to the empty state $|2\rangle$). Six-photon process of all fields $(\Omega_2, E_{1,2})$ other than Ω_1 supports the third-order nonlinearity for the probe fields $E_{1,2}$. (b) is for the direct perturbation to the CPT dark state (i.e., to the comparably or equally populated $|1, 2\rangle$ states). With the substitution of CPT transition $|1\rangle \xrightarrow[(1)]{\Omega_1} |3\rangle$ for EIT transition $|1\rangle \xrightarrow[(1)]{E_1} |3\rangle$ as in (a), and with the substitution of $|3\rangle \xrightarrow[(6)]{\Omega_1^*} |1\rangle$ for $|3\rangle \xrightarrow[(6)]{E_1^*} |1\rangle$, six-photon processes of all fields $(E_{1,2}, \Omega_{1,2})$ are responsible for the second-order nonlinearity for the probe fields $E_{1,2}$. In addition, there exist six- and eight-photon processes of all fields for the third-order nonlinearity, which is negligibly weak.

IV. COMPARISON BETWEEN EIT- AND CPT-BASED NONLINEARITIES

A comparison is made to show the essential difference in susceptibilities due to the EIT-based indirect perturbation and due to the CPT-based direct perturbation.

A. EIT-based susceptibilities

As a special case ($\Omega_1 \to 0$), we are left only with the indirect perturbation to the EIT dark state. We can immediately recover the EIT-induced Kerr nonlinearity as in Ref. [22]. When $\Omega_1 \to 0$, the probe field E_1 and the dressing field Ω_2 are in the EIT interaction with the atoms. Since $|\alpha_1| \ll |\Omega_2|$, the ground state $|1\rangle$ is simply the dark state. The only perturbation to the dark state is the additional field E_2, which is coupled to the empty state $|2\rangle$. This is only the indirection perturbation to the EIT dark state, but no direct perturbation is used. It is seen from Eqs. (30) to (33) that, as $\Omega_1 \to 0$, we have vanishing χ's parameters ($\chi^{(1)}, \chi^{(2)}, \chi_2^{(3)} \to 0$), but nonvanishing third-order susceptibility

$$\chi_1^{(3)} \approx -\frac{|\mu_{13}\mu_{24}|^2 N}{8\epsilon_0 \hbar^3 \Delta |\Omega_2|^2}, \tag{44}$$

which describes the Kerr nonlinear interaction strength. It is exactly the same result as in Ref. [22] when we note that Ω_2 is half Rabi frequency. This reflects the generality of our results.

Now we show that $\chi_1^{(3)}$ nonlinearity for $E_{1,2}$ is essentially extracted from six-photon parametric process of all fields in EIT. At first, we should note that the total order of the χ's numerators in $(\mu_{13,24}, \Omega_{1,2})$ corresponds to the steps of a round trip for multiphoton process. In the EIT case ($\Omega_1 \to 0$, $\chi_{12}^{(3)} \to 0$), it is seen from Eqs. (32) and (36) that the remaining $\chi_{11}^{(3)}$ term describes a six-photon process mediated by the dressing and probe fields $(\Omega_2, E_{1,2})$

$$\underbrace{\underbrace{|1\rangle \left\{\begin{matrix} \xrightarrow[(1)]{E_1} \\ \xleftarrow[(6)]{E_1^*} \end{matrix}\right\} |3\rangle \left\{\begin{matrix} \xrightarrow[(2)]{\Omega_2^*} \\ \xleftarrow[(5)]{\Omega_2} \end{matrix}\right\}}_{\text{EIT}} |2\rangle \left\{\begin{matrix} \xrightarrow[(3)]{E_2} \\ \xleftarrow[(4)]{E_2^*} \end{matrix}\right\} |4\rangle}_{\text{EIT + empty } |2\rangle \text{ state Stark shift}}, \tag{45}$$

which is easily seen from Fig. 4(a). The parametric multiphoton process is composed of the same going and return lines. This differs from the EIT plus two-photon transition based scheme [20], where a round contour consists of the different going and return lines. Once in the absence of the dressing field Ω_2, no such six-photon process is existent and no EIT-based Kerr nonlinearity. In the presence of the dressing field Ω_2, however, not only is vanishing the linear response to the probe field E_1 through the (1) transition but also the nonlinear response is induced by the five-photon transitions (2)–(6) in the going and return lines, which are performed by Ω_2 and $E_{1,2}$. Because the dressing field Ω_2 is much stronger than the probe fields $E_{1,2}$ and keeps unchanged during the interaction, thus we are left with the third-order nonlinearity for the probe fields $E_{1,2}$. Now we can conclude that the third-order (Kerr) nonlinearity in EIT configuration for $E_{1,2}$ is actually taken out from the six-photon parametric process of all fields, including the dressing and probe fields $(\Omega_2, E_{1,2})$. Since the dressing fields serve as the controllable parameters, it is reasonable to separate the dressing fields and to focus on the response of the dressed atoms to the probe fields. This was done but not stated in Ref. [22]. Here we would like to emphasize that the EIT-based Kerr nonlinearity for the probe fields is essentially attributed to the six-photon process that involves all fields.

B. CPT-based susceptibilities

Following exactly the same line, we compare the case with the direct perturbation to the CPT dark state. We show that $\chi^{(2)}$ nonlinearity for $E_{1,2}$ is extracted from six-photon parametric process of all fields $(\Omega_{1,2}, E_{1,2})$ in CPT. When the dark resonance as in Eq. (10) is maintained, the two ground states are populated comparably or equally with large or maximal coherence due to two equal or comparable dressing fields $|\Omega_1| \sim |\Omega_2|$. This is remarkably different from the the EIT case in that the probe fields are both directly coupled to the CPT involved ground states. Since the dressing fields are strong and keep unchanged during the interaction, we can focus on the nonlinear response of the dressed CPT atoms to the probe fields $E_{1,2}$.

(i) The susceptibility $\chi^{(1)}$ is a linear frequency shift. This arises from the interaction described by the $c\alpha_2\sigma_{\bar{4}D}$ term and its Hermitian conjugate term in Eq. (21). It is seen from Eqs. (30), (34) that, by using $\frac{|\Omega_1|^2}{\Omega^2} = 1 - \frac{|\Omega_2|^2}{\Omega^2}$ we can divide $\chi^{(1)}$ into two terms $\chi^{(1)} = \chi_1^{(1)} + \chi_2^{(1)}$. The two terms $\chi_{1,2}^{(1)}$ come, respectively, from two- and four-photon processes

$$\underbrace{|2\rangle \left\{\begin{matrix} \xrightarrow[(3)]{E_2} \\ \xleftarrow[(4)]{E_2^*} \end{matrix}\right\} |4\rangle}_{\text{Linear Stark shift}}, \tag{46}$$

and

$$\underbrace{|3\rangle \left\{\begin{matrix} \xrightarrow[(2)]{\Omega_2^*} \\ \xleftarrow[(5)]{\Omega_2} \end{matrix}\right\} |2\rangle \left\{\begin{matrix} \xrightarrow[(3)]{E_2} \\ \xleftarrow[(4)]{E_2^*} \end{matrix}\right\} |4\rangle}_{\Omega_2\text{ saturated shift}}. \tag{47}$$

These two processes give the opposite contributions and the net effect is dependent on $\frac{|\Omega_1|^2}{\Omega^2}$. This frequency shift is much smaller than the detuning Δ and can be included in the latter. However, the latter four-photon process is merged in the six-photon transitions as follows.

(ii) The $\chi^{(2)}$ and $\chi^{(2)*}$ terms stand for the second-order susceptibility for the $E_{1,2}$ fields, and are established via six-photon parametric processes of all dressing and probe fields $(\Omega_{1,2}, E_{1,2})$. As shown in Fig. 4(b), the two round trips of transitions that correspond to the conjugate terms $\chi^{(2)}$ and $\chi^{(2)*}$ are, respectively,

$$\underbrace{\underbrace{|1\rangle \left\{\begin{matrix} \xrightarrow[(1)]{E_1} \\ \xleftarrow[(6)]{\Omega_1^*} \end{matrix}\right\} |3\rangle \left\{\begin{matrix} \xrightarrow[(2)]{\Omega_2^*} \\ \xleftarrow[(5)]{\Omega_2} \end{matrix}\right\} |2\rangle}_{\text{CPT + photon exchange}} \left\{\begin{matrix} \xrightarrow[(3)]{E_2} \\ \xleftarrow[(4)]{E_2^*} \end{matrix}\right\} |4\rangle}_{\text{CPT + photon exchange + }\Omega_2\text{ saturated Stark shift}}, \tag{48}$$

and

$$\underbrace{\underbrace{|1\rangle \left\{\begin{matrix} \xrightarrow[(1)]{\Omega_1} \\ \xleftarrow[(6)]{E_1^*} \end{matrix}\right\} |3\rangle \left\{\begin{matrix} \xrightarrow[(2)]{\Omega_2^*} \\ \xleftarrow[(5)]{\Omega_2} \end{matrix}\right\} |2\rangle}_{\text{CPT + photon exchange}} \left\{\begin{matrix} \xrightarrow[(3)]{E_2} \\ \xleftarrow[(4)]{E_2^*} \end{matrix}\right\} |4\rangle}_{\text{CPT + photon exchange + }\Omega_2\text{saturated Stark shift}}. \tag{49}$$

To be exact, the latter five photon processes (2)–(6) in Eq. (48) and the former five-photon processes (1)–(5) in Eq. (49) contribute to the susceptibility $\chi^{(2)}$ and $\chi^{(2)*}$, respectively. Therefore, $\chi^{(2)}$ for $E_{1,2}$ describes actually fifth-order nonlinearity for all fields. However, the dressing fields $\Omega_{1,2}$ are much stronger than the probe fields $E_{1,2}$ and remain unchanged during the interaction. Comparing Eqs. (48) and (49) with Eq. (45), it is easy to find the substitution of $|1\rangle \xleftarrow[(6)]{\Omega_1^*} |3\rangle$ for $|1\rangle \xleftarrow[(6)]{E_1^*} |3\rangle$ and the substitution of $|1\rangle \xrightarrow[(1)]{\Omega_1} |3\rangle$ for $|1\rangle \xrightarrow[(1)]{E_1} |3\rangle$. This is the essential difference from the EIT case. The going and return steps between $|1\rangle$ and $|3\rangle$ are performed by different fields. This causes the photon exchange between the probe and dressing fields (E_1, Ω_1). The photon exchange turns out to be one essentially important factor for the second-order susceptibility $\chi^{(2)}$. In a word, it is its simultaneous introduction with the linear Stark shift into the dark resonance that leads to the second-order nonlinearity for the probe fields $E_{1,2}$.

(iii) Meanwhile, the third-order susceptibility is negligibly weak compared with the second-order susceptibility. We can see that, the first term $\chi_{11}^{(3)}$ in $\chi_1^{(3)}$ corresponds to six-photon parametric process as in Eq. (45). The second term $\chi_{12}^{(3)}$ is extracted from eight-photon parametric process

$$\underbrace{\underbrace{|1\rangle \left\{\begin{matrix} \xrightarrow[(1)]{E_1} \\ \xleftarrow[(6)]{E_1^*} \\ \xrightarrow[(7)]{\Omega_1} \\ \xleftarrow[(8)]{\Omega_1^*} \end{matrix}\right\} |3\rangle \left\{\begin{matrix} \xrightarrow[(2)]{\Omega_2^*} \\ \xleftarrow[(5)]{\Omega_2} \end{matrix}\right\} |2\rangle}_{\text{CPT + two-photon exchange}} \left\{\begin{matrix} \xrightarrow[(3)]{E_2} \\ \xleftarrow[(4)]{E_2^*} \end{matrix}\right\} |4\rangle}_{\text{CPT + two-photon exchange + }\Omega_2\text{saturated Stark shift}}, \tag{50}$$

and $\chi_2^{(3)}$ is drawn from eight-photon process

$$\underbrace{\underbrace{|1\rangle \left\{\begin{matrix} \xrightarrow[(1)]{E_1} \\ \xleftarrow[(6)]{\Omega_1^*} \\ \xrightarrow[(7)]{E_1} \\ \xleftarrow[(8)]{\Omega_1^*} \end{matrix}\right\} |3\rangle \left\{\begin{matrix} \xrightarrow[(2)]{\Omega_2^*} \\ \xleftarrow[(5)]{\Omega_2} \end{matrix}\right\} |2\rangle}_{\text{CPT + two-photon exchange}} \left\{\begin{matrix} \xrightarrow[(3)]{E_2} \\ \xleftarrow[(4)]{E_2^*} \end{matrix}\right\} |4\rangle}_{\text{CPT + two-photon exchange + }\Omega_2\text{saturated Stark shift}}. \tag{51}$$

It is seen that there contains two-photon exchange between the probe and dressing fields (E_1, Ω_1) through $|1\rangle \leftrightarrow |3\rangle$ transition. Because the dressing fields $\Omega_{1,2}$ are equally or comparably strong, the two kinds of multiphoton parametric processes in Eqs. (50) and (51) are comparable with the six-photon parametric process in Eq. (45). Since the dressing fields $\Omega_{1,2}$ remain unchanged, we are left with the third-order nonlinearity for the fields $E_{1,2}$. The preset case shows that it is necessary for us to separate the dressing fields and to focus on the response of the dressed atoms to the probe fields because different multiphoton (six- and eight-photon) processes correspond to the same order of nonlinearity for the probe fields.

V. CONCLUSION

In conclusion, we have studied the nonlinear effects based on the direct perturbation to the CPT dark state. The dressed CPT atoms serves as an integrated coherent medium, in which the dressing fields are merged into the dressed atoms. By separating the weak probe fields from the dressing fields, we are provided with a way to focus on the response of the dressed CPT atoms to the weak probe fields only. With such a background in mind, we have defined second-order susceptibility with respect to the probe fields. The dressed-state picture is employed to present the essential physics and the comprehensive calculation since the atoms are maintained in the CPT dark state during their interaction with the probe fields. A through comparison of the nonlinear susceptibilities is made between the EIT-based indirect perturbation and the CPT-based direction perturbation.

It is shown that the direct perturbation to the CPT dark state leads to the giant, resonantly enhanced second-order cross nonlinearities with respect to the weak probe fields. Further, the dark-state shift can be traced to the dark-state-based six-photon parametric processes for all involved fields, including the dressing and probe fields. A necessary premise for this effect is the formation of the dark state, which contains no excited state, and stays unchanged once formed. The dark-state-based six-photon parametric processes are established through the two combinable factors. One is the photon exchange that happens on one arm of the dark resonance in Λ configuration, while the other is the linear Stark shift that is introduced through the other arm. These two factors work together to lead to a nonlinear dispersive perturbation to the dark state. The creation and resonant enhancement of the second-order cross nonlinearities are unique for the direct perturbation to the CPT dark state since the direct perturbation to the CPT dark state establishes the photon exchange between the dressing and probe fields and the linear Stark shift of the CPT dark state. The direct perturbation is most frequently met in the CPT-based system because each dark-state-involved ground state is comparably or equally populated. The second-order nonlinearity, much more greatly enhanced than the third-order one, is remarkably more sensitive for the quantum control at low light levels.

ACKNOWLEDGMENT

This work is supported by the National Natural Science Foundation of China (Grants No. 61875067 and No. 11474118).

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