Multiphoton coherent population oscillation

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We study the bichromatic driving of a two-level system which displays long-lived coherent population oscillations (CPO). We show that under certain conditions, multiphoton parametric interaction leads to the appearance of CPO resonances at the subharmonic frequencies. In addition, in the region of the CPO resonances, there is strong parametric interaction between the weak sideband components of the electromagnetic field.

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I. INTRODUCTION

Non-linear interaction between electromagnetic fields can lead to the appearance of resonances with a bandwidth which is much narrower than the unperturbed natural linewidth. The most familiar effects that can give such narrowband response are electromagnetically induced transparency (EIT) \[1\] and coherent population oscillations (CPO) \[2\]. In the EIT and CPO cases, the strong pump can form a transparency window in the probe absorption spectrum accompanied by steep dispersion of the refractive index that leads to effects such as slow light \[3, 4\]. The narrowband medium response can also appear in the four-wave mixing (FWM) process \[5\], leading under certain conditions to narrowband biphon generation due to EIT \[4, 5\] or CPO \[8\]. Although there are similarities between EIT and CPO, these effects are actually different in nature, and the properties of the nonlinear response in each case are determined by different system parameters. For example, in the case of EIT, the minimal bandwidth of the resonance is determined by the transverse relaxation rate of the two-photon transition, whereas in the case of CPO, the minimal bandwidth is determined by the effective longitudinal relaxation rate; in the EIT case, the resonance appears at zero two-photon detuning, whereas in the CPO case, the non-linear resonance is centered at the pump frequency. The fact that in the CPO case, the weak signal is always centered at the pump frequency makes it difficult to filter out the weak signal from the strong pump and incoherent scattering.

Here, we analyze the response of a two-level system (TLS) that displays long-lived coherent population oscillations in the presence of a bichromatic pump, and one or two weak fields. The two sidebands of the pump are symmetrically displaced from the pump frequency \(\omega_0\) and in the case where the scanning is realized by two probe fields, these fields are also symmetrically displaced from \(\omega_0\) (see Fig. 1). The interaction of the TLS with a polychromatic field has been studied from many different aspects; see, for example, \[3\] and references therein. In the bichromatic fields, resonances at the subharmonic frequencies \(\omega_{\sub} = \omega_0 \pm (2n + 1) \delta\) \[10, 11\] appear where \(\delta\) is the frequency difference between the two pumps and \(n\) is an integer. Here we demonstrate that these resonances can appear under CPO conditions with a width determined by the effective longitudinal relaxation rate. We also demonstrate that there is effective parametric interaction between two weak probes tuned in the region of the two symmetrically displaced CPO resonances. Under certain conditions this parametric interaction can appear at a pump Rabi frequency well below the transverse relaxation rate of the TLS.

II. THE MODEL

Let us consider the two-level quantum system composed of levels \(|1\rangle\) and \(|2\rangle\) interacting with the electromagnetic field \(E\). It is assumed that system has an additional off-resonant quantum metastable state \(|m\rangle\) which is radiatively coupled to the TLS (see Fig. 1). The set of Bloch equations for the present system has the following form:

\[
\frac{d}{dt} [\rho_{21} + i \omega_{21}] + \frac{d}{dt} [\rho_{12} - i \omega_{12}] = 0
\]

\[
\frac{d}{dt} \rho_{12} - i V_{12} (\rho_{11} - \rho_{22}) = 0
\]

\[
V_{12} \rho_{21} - i V_{21} \rho_{12} + \frac{d \rho_{11}}{dt} - \gamma_{21} \rho_{22} - \gamma_m \rho_m = 0
\]

\[
\frac{d \rho_m}{dt} + \gamma_m \rho_m - \gamma_0 \rho_{22} = 0
\]

where \(V_{21}\) is the total Rabi frequency, and \(\Gamma_{21}\) is the transverse relaxation rate of the \(|2\rangle - |1\rangle\) transition, and \(\gamma_{21}, \gamma_0, \) and \(\gamma_m\) are the population relaxation constants from the excited state to the ground state, from the excited state to the metastable state, and from the
metastable to the ground state, respectively. Further, we assume that the system is closed and that the total population in the system is conserved so that

\[ \rho_{11} + \rho_{22} + \rho_m = 1. \]  

(5)

Let us introduce the population inversion

\[ r = \rho_{22} - \rho_{11}, \]  

(6)

and, using Eqs. (5) and (6), define \( \rho_{11} \) and \( \rho_{22} \) in terms of \( r \) and \( \rho_m \). We then rewrite Eqs. (1) - (4) and obtain the following set of equations:

\[
\begin{align*}
\frac{d}{dt} + \Gamma_{21} + i\omega_{21} \rho_{21} - iV_{21}r &= 0, \quad (7) \\
\frac{d}{dt} + \Gamma_{21} - i\omega_{21} \rho_{12} + iV_{12}r &= 0, \quad (8) \\
2iV_{12}\rho_{21} - 2iV_{21}\rho_{12} - \left( \frac{d}{dt} + \gamma_{21} + \frac{\gamma_o}{2} \right) r &+ \left( \gamma_{21} - \gamma_m + \frac{\gamma_o}{2} \right) \rho_m = \gamma_{21} + \frac{\gamma_o}{2}, \quad (9) \\
- \frac{\gamma_o}{2} r + \left( \frac{d}{dt} + \gamma_m + \frac{\gamma_o}{2} \right) \rho_m &= \frac{\gamma_o}{2}. \quad (10)
\end{align*}
\]

### A. The electromagnetic field

We now specify the electromagnetic fields acting on the system. We consider the case of the bichromatic pumping of the TLS at frequencies \( \omega_{s1} \) and \( \omega_{s2} \), and define the detuning of the pump fields from resonance in the following way

\[ \Delta = \omega_0 - \omega_{21}, \]  

(11)

where \( \omega_0 = (\omega_{s1} + \omega_{s2})/2 \) and \( \omega_{21} \) is the resonance frequency of the \( |1\rangle \rightarrow |2\rangle \) transition. We also apply a weak probe field which is a combination of two sidebands arranged symmetrically with respect to \( \omega_0 \) with frequencies \( \omega_{p1} \) and \( \omega_{p2} \). The Rabi frequency for the total electromagnetic field can be written in the form

\[ V_{21} = \left( V_{21}^1 e^{-i\delta t} + V_{21}^2 e^{i\delta t} + V_{21}^p e^{-i\omega t} + V_{21}^p e^{i\omega t} \right) e^{-i\omega_0 t}, \]  

(12)

where \( V_{21}^1, V_{21}^2, V_{21}^p \), and \( V_{21}^p \) are the Rabi frequencies of the corresponding fields. We also define the following detunings:

\[ \delta = \omega_{s1} - \omega_0 = -\left( \omega_{s2} - \omega_0 \right), \quad w = \omega_{p1} - \omega_0 = -\left( \omega_{p2} - \omega_0 \right). \]  

(13)

Next we move to the frame rotating at frequency \( \omega_0 \) and define

\[ \sigma_{21} = \rho_{21} e^{i\omega_0 t}, \quad \sigma_{12} = \rho_{12} e^{-i\omega_0 t}, \quad \sigma_0 = r, \quad \sigma_m = \rho_m e^{i\omega_0 t}. \]  

(14)

Substituting Eqs. (12) and (14) into Eqs. (7)-(10), we obtain a set of equations which we write in the matrix form

\[ BS = F, \]  

(15)

where

\[ S = \begin{pmatrix} \sigma_{21} \\ \sigma_{12} \\ \sigma_0 \\ \sigma_m \end{pmatrix}, \quad F = \begin{pmatrix} 0 \\ 0 \\ -\left( \gamma_{21} + \frac{\gamma_o}{2} \right) \end{pmatrix}, \]  

(16)

and the matrix \( B \) is written as the sum of components oscillating at the different frequencies

\[ B = O + D^+ e^{i\delta t} + D^- e^{-i\delta t} + W^+ e^{i\omega t} + W^- e^{-i\omega t}, \]  

(17)

where the matrices are defined as

\[ O = \frac{d}{dt} I + \begin{pmatrix} \Gamma_{21} - i\Delta & 0 & 0 & 0 \\ 0 & \Gamma_{21} + i\Delta & 0 & 0 \\ 0 & 0 & \gamma_{21} + \frac{\gamma_o}{2} & \gamma_m - \frac{\gamma_o}{2} - \gamma_{21} \\ 0 & 0 & -\frac{\gamma_o}{2} & \gamma_m + \frac{\gamma_o}{2} \end{pmatrix}, \]  

(18)
\[
D^{-\delta} = i \begin{pmatrix}
0 & 0 & -V_{21}^{s1} & 0 \\
0 & 0 & V_{12}^{s2} & 0 \\
-2V_{12}^{s1} & 2V_{21}^{s1} & 0 & 0 \\
0 & 0 & 0 & 0
\end{pmatrix},
\quad (19)
\]

\[
D^{+\delta} = i \begin{pmatrix}
0 & 0 & -V_{21}^{s2} & 0 \\
0 & 0 & V_{12}^{s1} & 0 \\
-2V_{12}^{s1} & 2V_{21}^{s2} & 0 & 0 \\
0 & 0 & 0 & 0
\end{pmatrix},
\quad (20)
\]

\[
W^{-w} = i \begin{pmatrix}
0 & 0 & -V_{21}^{p1} & 0 \\
0 & 0 & V_{12}^{p2} & 0 \\
-2V_{12}^{p1} & 2V_{21}^{p1} & 0 & 0 \\
0 & 0 & 0 & 0
\end{pmatrix},
\quad (21)
\]

\[
W^{+w} = i \begin{pmatrix}
0 & 0 & -V_{21}^{p2} & 0 \\
0 & 0 & V_{12}^{p1} & 0 \\
-2V_{12}^{p2} & 2V_{21}^{p2} & 0 & 0 \\
0 & 0 & 0 & 0
\end{pmatrix},
\quad (22)
\]

and \( I \) is the identity matrix of order 4.

**B. Expansion of the solution**

As can be seen from Eq. (17), the Hamiltonian of the system has a periodic time dependence with two characteristic frequencies \( \delta \) and \( w \). In order to find a solution, we use the Floquet theorem and make a harmonic expansion of the vector of the density-matrix elements \( S \)

\[
S = \sum_{n=-\infty}^{+\infty} \sum_{m=-\infty}^{+\infty} S^{n,m} e^{i(n\delta + mw)t}.
\quad (23)
\]

After substituting Eq. (23) into Eq. (15), we take the time derivative of the oscillating term and separate the equations with different time dependence to obtain the matrix recurrence relation

\[
(O + T) S^{n,m} + D^{-\delta} S^{n+1,m} + D^{+\delta} S^{n-1,m} + W^{-w} S^{n,m+1} + W^{+w} S^{n,m-1} = F \delta_{n=0,m=0},
\quad (24)
\]

where

\[
T = i (n\delta + mw) I.
\quad (25)
\]

**C. Steady state approximation**

We now make the steady-state approximation and rewrite the matrix equation of Eq. (24) as a set of four algebraic equations:

\[
\sigma_{21}^{n,m} = i P_{21}^{n,m} (V_{21}^{s1} \sigma_{0}^{n+1,m} + V_{21}^{s2} \sigma_{0}^{n-1,m} + V_{21}^{p1} \sigma_{0}^{n,m+1} + V_{21}^{p2} \sigma_{0}^{n,m-1}),
\quad (26)
\]

\[
\sigma_{12}^{n,m} = -i P_{12}^{n,m} (V_{12}^{s2} \sigma_{0}^{n+1,m} + V_{12}^{s1} \sigma_{0}^{n-1,m} + V_{12}^{p1} \sigma_{0}^{n,m+1} + V_{12}^{p2} \sigma_{0}^{n,m-1}),
\quad (27)
\]

\[
\sigma_{12}^{n,m} = \sigma_{0}^{n,m} (\gamma_{21} + \gamma_{0} \frac{m}{2} + i (n\delta + mw)) \sigma_{21}^{n,m} - (\gamma_{21} - \gamma_{0} + \frac{\gamma_{0} m}{2}) \sigma_{12}^{n,m},
\quad (28)
\]

\[
2V_{21}^{s1} \sigma_{12}^{n+1,m} + 2iV_{21}^{s2} \sigma_{12}^{n-1,m} - 2V_{21}^{s1} \sigma_{12}^{n,m+1} + 2V_{21}^{s2} \sigma_{12}^{n,m-1}.
\quad (29)
\]

where

\[
P_{21}^{n,m} = \frac{\gamma_{0}}{2} \left[ \gamma_{1} + \gamma_{0} \frac{m}{2} + i (n\delta + mw) \right]^{-1},
\quad (30)
\]

\[
P_{12}^{n,m} = \frac{\gamma_{0}}{2} \left[ \gamma_{1} + \gamma_{0} \frac{m}{2} + i (n\delta + mw) \right]^{-1},
\quad (31)
\]

We now solve these equations by writing an equation for the function \( \sigma_{0}^{n,m} \) alone. Substituting corresponding terms from Eqs. (26), (27), and (28) into Eq. (25), we obtain a two-dimensional recurrence relation for \( \sigma_{0}^{n,m} \):

\[
a_{1,n,m} \sigma_{0}^{n,m} + a_{2,n,m} \sigma_{0}^{n-2,m} + a_{3,n,m} \sigma_{0}^{n+2,m} + a_{4,n,m} \sigma_{0}^{n-1,m-1} + a_{5,n,m} \sigma_{0}^{n+1,m-1} + a_{6,n,m} \sigma_{0}^{n-1,m+1} + a_{7,n,m} \sigma_{0}^{n+1,m+1} + a_{8,n,m} \sigma_{0}^{n+2,m} + a_{9,n,m} \sigma_{0}^{n-2,m} = -R_{0,0} \delta_{n=0,m=0},
\quad (33)
\]

where we have introduced the following notation:

\[
a_{1,n,m} = 2|V_{21}^{s1}|^2 \left( P_{21}^{n-1,m} + P_{12}^{n-1,m} \right),
\quad (34)
\]

\[
a_{2,n,m} = 2V_{21}^{s2} \left[ P_{21}^{n-1,m} + P_{12}^{n-1,m} \right] + 2V_{21}^{p1} \left( P_{21}^{n-1,m} + P_{12}^{n-1,m} \right) + 2V_{21}^{p2} \left( P_{21}^{n-1,m} + P_{12}^{n-1,m} \right) + R_{n,m},
\quad (35)
\]

\[
a_{3,n,m} = 2V_{21}^{s1} V_{12}^{s2} \left( P_{21}^{n+1,m} + P_{12}^{n+1,m} \right),
\quad (36)
\]

\[
a_{4,n,m} = 2V_{21}^{s2} V_{12}^{s1} \left( P_{21}^{n+1,m} + P_{12}^{n+1,m} \right) + 2V_{12}^{s2} V_{21}^{p2} \left( P_{21}^{n-1,m} + P_{12}^{n-1,m} \right),
\quad (37)
\]

\[
a_{5,n,m} = 2V_{21}^{s1} V_{12}^{p1} \left( P_{21}^{n-1,m} + P_{12}^{n+1,m} \right) + 2V_{12}^{s2} V_{21}^{p2} \left( P_{21}^{n+1,m} + P_{12}^{n-1,m} \right),
\quad (38)
\]
\[ a_{n,m}^6 = 2V_{12}^{a1}V_{21}^p \left( P_{21}^{n,1-m} + P_{12}^{n,m+1} \right) + 2V_{21}^{\sigma 2}V_{12}^{p1} \left( P_{21}^{n,m+1} + P_{12}^{n,1-m} \right), \tag{39} \]

\[ a_{n,m}^7 = 2V_{12}^{a1}V_{21}^p \left( P_{21}^{n+1,m} + P_{12}^{n,m+1} \right) + 2V_{21}^{\sigma 2}V_{12}^{p1} \left( P_{21}^{n,m+1} + P_{12}^{n,1-m} \right), \tag{40} \]

\[ a_{n,m}^8 = 2V_{21}^{p1}V_{12}^{p2} \left( P_{21}^{n,m+1} + P_{12}^{n,1-m} \right), \tag{41} \]

\[ a_{n,m}^9 = 2V_{21}^{p1}V_{12}^{p2} \left( P_{21}^{n-1,m} + P_{12}^{n-m+1} \right), \tag{42} \]

\[ R_{n,m} = \gamma_{21} + \frac{\gamma_0}{2} + i(n\delta + mw) - P_{m}^{n,m} \left( \gamma_{21} - \gamma_m + \frac{\gamma_0}{2} \right). \tag{43} \]

The recurrence relation of Eq. (33) is solved numerically and allows us to treat probe fields of arbitrary strength.

D. Polarization

The medium polarization is determined by the non-diagonal matrix element

\[ P(t) = \mu N (\rho_{21} + \text{c.c.}) = \mu N \left( \sum_{n=-\infty}^{+\infty} \sum_{m=-\infty}^{+\infty} \sigma_{21}^{n,m} e^{-i\omega_{21}t} e^{i(n\delta + mw)t} + \text{c.c.} \right). \tag{44} \]

Thus the component of the polarization that oscillates at a particular frequency gives the medium’s response to the electromagnetic field at this frequency. As can be seen from Eq. (44), the polarization at the probe frequency \( \omega_{p1} \) is proportional to \( \sigma_{21}^{0,1-1} \) which can be found from Eq. (20)

\[ \sigma_{21}^{0,1-1} = e^{i\Delta_p} \left( V_{21}^{p1} \sigma_{21}^{0,0} + V_{21}^{p2} \sigma_{21}^{0,-2} \right). \tag{45} \]

In the next section we analyze the normalized probe absorption, which is defined as \( \alpha/\alpha_0 = -\text{Im} \sigma_{21}^{0,1-1} \Gamma_{21}/V_{21}^{p2} \), for several cases.

III. THE CPO RESONANCE IN MONOCHROMATIC PUMPING

The role of the excited-state decay’s branching ratio to the ground and metastable states is not discussed in the literature. However, it is of crucial importance in obtaining CPO at the subharmonic frequencies. Thus, in order to better understand the behavior of the system in a bichromatic driving field, we first consider the case of a monochromatic driving where it is easy to get an analytical result and analyze the role of the branching ratio. Let us consider the interaction of the TLS with a strong pump and a weak probe. In this case, we take \( V_{21}^{p2} = V_{p2}^{p2} = 0 \) in Eq. (45) so that

\[ \sigma_{21}^{0,1-1} = i\frac{p_{0,0}}{V_{21}^{p1}} \left( V_{21}^{p1} \sigma_{21}^{0,0} + V_{p1}^{p2} \sigma_{0,0} \right). \tag{46} \]

Assuming that the probe field is weak we can keep only the first-order terms in the probe Rabi frequencies. We then obtain for the functions \( \sigma_{0,0}^{1-1} \) and \( \sigma_0^{0,0} \) of Eq. (33)

\[ \sigma_{0,0}^{1-1} = -\frac{a_{1,1-1}^{0,0}}{\alpha_0}, \tag{47} \]

\[ \sigma_0^{0,0} = -\frac{a_{1,1-1}^{0,0}}{\alpha_0} = -\left( 1 - \frac{\kappa}{1 + \kappa} \right), \tag{48} \]

where

\[ \kappa = \kappa_{TLS} (\gamma_0/2\gamma_m + 1) \tag{49} \]

is the saturation parameter of the system and

\[ \kappa_{TLS} = -\frac{\mu N V_{12}^{12} \left( 1 + \Delta_s^2 / \Gamma_{21}^2 \right)}{\gamma_2 \Gamma_{21}^2 + \Delta_s^2 / \Gamma_{21}^2} \tag{50} \]

is the saturation parameter of the ordinary two-level system (if there is no additional channel for the decay via intermediate state). The functions \( a_{1,1-1} \) and \( a_{1,1-1}^{0,0} \) are found from Eqs. (51) and (52) to be

\[ a_{1,1-1} = \frac{\gamma_2 - i\Delta_p}{\gamma_m + \gamma_0/2 - i\Delta_p} + 4 \left| V_{21}^{p1} \right|^2 \frac{\Gamma_{21} - i\Delta_p}{\left( \Gamma_{21} + i(\Delta_s + \Delta_p) \right) \left( \Gamma_{21} - i(\Delta_s - \Delta_p) \right)}, \tag{51} \]

\[ a_{1,1-1}^{0,0} = 4V_{12}^{p1}V_{21}^{p2} \frac{\Gamma_{21} + i(\Delta_s - \Delta_p) / 2}{\left( \Gamma_{21} + i\Delta_s \right) \left( \Gamma_{21} - i\Delta_p \right)}. \tag{52} \]

where for convenience we have introduced the pump detuning from resonance \( \Delta_p \equiv \Delta - \delta = \omega_{p1} - \omega_1 \) and pump-probe detuning \( \Delta_p \equiv w - \delta = \omega_{p1} - \omega_1 \).

We are interested in the shape of the narrow dip centered at the pump frequency and associated with the CPO effect. This structure has a characteristic bandwidth of the order of \( \gamma_m \) which is much less than the natural bandwidth of the transition from the excited to the ground state as the level \( |m\rangle \) is assumed to be metastable. Thus, in Eqs. (51) and (52), we use the approximation

\[ \gamma_m, \Delta_p \ll \Gamma_{21}, \gamma_2 \tag{53} \]
and obtain  
\[ \frac{a_{6,-1}^6}{a_{1,-1}^1} \approx 4 \frac{V_{12}^1 V_{21}^{P1}}{\gamma_2 \Gamma_{21} (1 + \Delta_p^2 / \Gamma_{21}^2)} \frac{1 + \kappa_{\text{TLS}}}{S} \frac{\gamma_m + \gamma_0/2 - i \Delta_p}{(W - i \Delta_p)^{\gamma_m}}, \]
where \( W \) is the bandwidth of the CPO dip 
\[ W = \gamma_m (1 + \kappa) \quad (55) \]
and the parameter \( S = (1 + \Delta_p^2 / 2 \Gamma_{21}^2 - i \Delta_s / 2 \Gamma_{21}) \).
Combining Eqs. (47), (48), and (53), we obtain the medium response at the probe frequency in the region of the CPO resonance 
\[ \sigma_{21}^{0,-1} = -i V_{21}^{P1} \frac{1}{1 + \kappa \Gamma_{21} + i (\Delta_s - \Delta_p)} (1 - XS), \]
where the first term in the brackets determines the saturation of the system due to the pump and the second term \( X \) includes the coherent interaction between pump and probe fields 
\[ X = 1 - \frac{1}{1 + \kappa_{\text{TLS}}} \frac{\gamma_m - i \Delta_p \kappa_{\text{TLS}}}{W - i \Delta_p}. \]

Further, for simplicity we consider the case of small pump detunings and assume that \( S \approx 1 \).

**Bandwidth** The bandwidth of the resonance is determined by the function \( W \) [see Eq. (55)]. In the case of the “classical” CPO, for which \( \gamma_2 \approx \gamma_o \approx \gamma_m \), we have \( \kappa \gg \kappa_{\text{TLS}} \), so that in the denominator of Eq. (53), we can omit \( \kappa_{\text{TLS}} \) and obtain the same result as in [12] 
\[ W = \gamma_m (1 + \kappa). \]
Thus the minimal bandwidth of the CPO resonance is determined by \( \gamma_m \) and this resonance experiences power broadening. In the opposite case for which \( \gamma_2 \gg \gamma_o \approx \gamma_m, \kappa \) and \( \kappa_{\text{TLS}} \) are of the same order. In this case, the bandwidth experiences fast saturation 
\[ W = \gamma_m + \gamma_0 \frac{\kappa_{\text{TLS}}}{2} + \kappa_{\text{TLS}}^{-1} \]
and will not exceed \( \gamma_m + \frac{\gamma_0}{2} \).

**Depth** The depth is determined from Eq. (55) by putting \( \Delta_p = 0 \). We obtain \( \alpha (\Delta_p = 0) / \alpha_0 = 1/(1 + \kappa)^2 \).

**Amplitude** From Eq. (55), we can determine the amplitude of the CPO dip. The dip itself comes from the multiplier \( (\gamma_m - i \Delta_p \kappa_{\text{TLS}})/(W - i \Delta_p) \) in Eq. (57). Its amplitude \( \alpha_0 \) is given by the difference between Eq. (56) with and without this term at point \( \Delta_p = 0 \) 
\[ \alpha_0 = \frac{\kappa \gamma_o / 2 \gamma_m}{(1 + \kappa)^2} \frac{2 \gamma_m + 1 + \kappa}{\gamma_0}. \]
It follows from Eq. (60) that, in the high saturation regime, the amplitude of the dip decreases as \( 1 / \sqrt{V_{21}^2} \).

The function \( \alpha_0 \) has an extremum at the point 
\[ \kappa_c = \frac{\gamma_0 / 2 \gamma_m + 1}{4} \left( \sqrt{1 + 8 (\gamma_0 / 2 \gamma_m + 1)^{-1}} - 1 \right). \]

**For the case in which \( \gamma_o \gg \gamma_m \)** it is easy to show that the maximum amplitude of the dip \( \alpha_0 = 0.25 \) is obtained at the point \( \kappa_c = 1 \) [12], but in the general situation there is no simple expression so that a graphical representation is required (see Fig. 2). It can be seen from Fig. 2 that with decreasing \( \gamma_o / \gamma_m \) the maximal achievable dip amplitude decreases.

**IV. NUMERICAL RESULTS**

In this section we numerically analyze the absorption spectrum of a weak probe field at the frequency \( \omega_{pl} \) that scans the TLS (Fig 1). The TLS is driven by two pump fields that are symmetrically detuned from resonance \( (\Delta = 0) \). Also we consider the case when there can be an additional weak field whose detuning is symmetrically displaced from resonance with respect to the first probe [see Eq. (13)]. In this case, we demonstrate strong parametric interaction between the weak fields in the presence of the pumps. As we mentioned in the previous sections, the branching ratio of \( \gamma_2 \), the decay rate from the excited to the ground state, to \( \gamma_o \), the decay rate from the excited to the metastable state, plays an important role in the properties of the CPO. We therefore analyze two cases numerically: \( \gamma_2 \approx \gamma_o \approx \gamma_m \) and \( \gamma_2 \gg \gamma_o \approx \gamma_m \).

**A. Case 1 \( \gamma_2 \approx \gamma_o \approx \gamma_m \)**

This range of values for the relaxation constants has been considered in our previous work on CPO [12,13]. However, here two strong pump fields are applied to the same transition. So, instead one CPO resonance in the probe spectrum, we obtain two CPO dips centered at the pump frequencies \( w = \pm \delta \) (Fig. 3). According to Eq. (60), increasing the pump Rabi frequencies leads to considerable power broadening of the CPO resonances [Fig. 3(b)]. The bandwidth of the CPO resonances estimated from the numerical simulation is well described by Eq. (55) which was derived for the case in which there is only one strong field [16]. The behavior of each of
these two resonances appears to be independent - while the probe field is in the region of the CPO resonance created by the first pump, it does not experience any coherent effects that come from the second pump field. The situation became quite different when we add a second symmetrically displaced weak field [Fig. 3 (red dashed lines)]. In the presence of the second weak field, FWM occurs. Due to the effect of the FWM, the absorption of two photons from the pump fields at the frequencies \(\omega_{s1}\) and \(\omega_{s2}\) leads to the generation of two photons at the probe frequencies \(\omega_{p1}\) and \(\omega_{p2}\). As a result, the dips in Fig. 3(a) get deeper and there is even amplification of the probe field is in the region of the CPO resonance created by the first pump, it does not experience any coherent effects that come from the second pump field. The situation became quite different when we add a second symmetrically displaced weak field [Fig. 3 (red dashed lines)]. In the presence of the second weak field, FWM occurs. Due to the effect of the FWM, the absorption of two photons from the pump fields at the frequencies \(\omega_{s1}\) and \(\omega_{s2}\) leads to the generation of two photons at the probe frequencies \(\omega_{p1}\) and \(\omega_{p2}\). As a result, the dips in Fig. 3(a) get deeper and there is even amplification of the probe frequencies comparable with the transverse relaxation rate \(\Gamma_{21}\) and the existence of narrowband CPO resonances. In Figs. 3(c) and (d), the existence of the CPO resonances at the subharmonic frequencies \(w = \pm (2n + 1) \delta\) where \(n = 0, 1, 2, 3, \ldots\) is clearly seen. For example the resonance that is located at the \(w/\Gamma_{21} = 0.15\) (\(n = 1\)) on the Fig. 4(c) corresponds to the process of the three-wave mixing between pump fields \(\omega_{sub,0.15} = 2\omega_{s1} - \omega_{s2}\). In this case, as in the previous one, there is strong parametric interaction between the weak symmetrically displaced fields [Fig. 4(b) and (d)]. In addition, the pump Rabi frequencies are of the same order as the transverse relaxation rate. This leads to parametric interaction between the weak fields not only in the CPO region, as in the previous case, but in a broad spectral range, as shown in Fig. 4(d) [17].

**V. CONCLUSION**

In summary, we have demonstrated the appearance of narrowband CPO responses at frequencies shifted from the pump frequency. The new CPO resonances appear at the frequencies \(\omega_{sub} = \omega_0 \pm (2n + 1) \delta\) due to multiphoton mixing between components of the bichromatic pump. We expect that these spectral features will al-
low spectral filtration of the signal field from the strong pump in experiments with slow light, and biphoton generation based on the effect of CPO. Also, in the region of the CPO dip there is effective parametric interaction between probe fields for both the cases considered in Sec. IV. The subharmonic resonances that appear when the condition $\gamma_2 \gg \gamma_0 \approx \gamma_m$ is satisfied can be investigated in atomic TLS systems where the decay via dipole-forbidden transitions are taken into account, and in certain NV centers in diamond [18]. In addition, the predicted subharmonic CPO resonances may have contributions to the resonances observed in the recent experiment [19] where the was driven by a bichromatic pump.

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