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# Theory of quantum beat and polarization interference in four-wave mixing

J. Erland and I. Balslev
*Fysisk Institut, Odense Universitet, DK-5230 Odense M, Denmark*
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We analyze theoretically the quantum beat and the polarization interference phenomena observed as oscillations in pulsed four-wave mixing. We show that resolution of the signal with respect to the delay between the incoming pulses as well as the detection frequency provides valuable information on the nature of the oscillations. Most importantly, it is very simple to distinguish between polarization interference from two independent optical transitions and quantum beats from a three-level system. In case of polarization interference, the phase of the oscillations shifts $\pi$ when the detected frequency passes any of the resonances. In case of quantum beat, the phase has little dependence on the detected frequency near the resonance.

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## I. INTRODUCTION

In the exploration of exciton kinetics and dynamics in solids the nonlinear optical phenomena studied by ultrafast spectroscopy are very essential [1–5]. Accurate information has been gained by studying the four-wave mixing (FWM) signal as a function of the delay $\tau$ between the two incident pulses. If the solid contains atomic systems with two closely lying transitions, one may observe oscillations in the delay time domain with the beat frequency of the pair of transitions. Such oscillations have been observed in GaAs quantum wells and assigned to the interference between heavy-hole and light-hole excitons [6–8], between free and bound excitons [9,10], between excitons and biexcitons [11,12], and between Landau split magnetoexcitons [13].

The solid may contain two types of independent, but coherently excited two-level systems. Or it may contain a three-level system with two closely lying transition frequencies. The two oscillatory phemomena are called polarization interference and quantum beat, respectively.

As did Koch *et al.* [14], we address the question of how to distinguish experimentally between polarization interference and quantum beat. In Ref. [14] a method based on the true time resolution of the FWM signal by means of a third laser beam was developed. An alternative method to be considered in this paper is the frequency-domain counterpart, namely the dependence on the detected frequency studied experimentally by Lyssenko *et al.* [15].

We consider here a thin sample illuminated by two pulsed laser beams propagating nearly normal to the sample. The atomic system in the sample is characterized by two distinct dipole-allowed optical transitions with nearly the same transition frequency. This can be realized in the following model systems:

(a) Two independent two-level systems with nearly equal transition frequencies. In the present paper this shall be called a II system, as the double I symbolizes the structure of the transitions.

(b) A V-type three-level system with two allowed transitions between a common ground state and two closely lying excited states.

(c) A $\Lambda$-type three-level system with two allowed transitions between a higher state and two closely lying lower states.

(d) A cascade system with almost equal frequencies of lower and upper transition. Let us use the name E type for this three-level system since the letter E symbolizes the level diagram.

These four situations are shown in Figs. 1(a)–(d).

Let the applied electric field of the beams consist of two pulses, one delayed $\tau$ with respect to the other. The fields of the two pulses are given by

$$E_1 = E_p(t)\exp[i(\mathbf{k_1}\cdot\mathbf{r} - \omega_p t)],$$
$$E_2 = E_p(t-\tau)\exp[i(\mathbf{k_2}\cdot\mathbf{r} - \omega_p t)], \quad (1)$$

where $\mathbf{r}$ is the space coordinate and $t$ is the time. For simplicity we let the two pulses have a common temporal envelope function $E_p(t)$ (centered about $t = 0$) and a common carrier frequency $\omega_p$. The two pulses have different wave vectors $\mathbf{k_1}, \mathbf{k_2}$ and arrival times. We assume a near-resonant condition, relatively short pulses, and small-area pulses. Thus $\omega_p$ is close to the transition frequencies, and

$$|\omega_i - \omega_j|, \gamma_{i,j} \ll \{1/[\text{width of } E_p(t)]\} \ll \omega_{i,j},$$
$$\int M_{i,j} E_p dt/\hbar \ll 1, \quad (2)$$

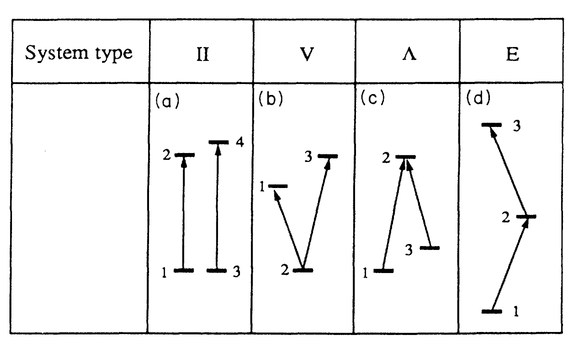


FIG. 1. Level diagrams of types II, V, $\Lambda$, and E, considered in the text.

 

where $\omega_i$, $\gamma_i$, and $M_i$ are frequency, linewidth, and dipole moment, respectively, of the $i$th transition. We detect the signal diffracted in the direction $2\mathbf{k}_2 - \mathbf{k}_1$. This means that we detect the waves generated by the third-order polarization $P^{(3)}(t,\tau)$ proportional to $E_2E_2E_1^*$. In order to gain maximum information it is useful to perform a spectral selection of the diffracted signal. The frequency of the detected signal will be denoted by $\omega$. Thus the optical intensity detected is proportional to $[\widetilde{P}^{(3)}(\omega,\tau) + \text{c.c.}]^2$, where $\widetilde{P}^{(3)}(\omega,\tau)$ is the Fourier transform of $P^{(3)}(t,\tau)$ with respect to $t$.

## II. THIRD-ORDER RESPONSE OF THREE-LEVEL SYSTEMS

In the present section we set up the density matrix equations of motions for a general three-level system with the levels 1, 2, and 3, see Fig. 1. The 1-2 and the 2-3 transitions are dipole allowed with transition dipole moments $M_{21}$ and $M_{32}$. We introduce the complex frequencies $\Omega$ as follows:

$$\Omega_{21} = \omega_2-\omega_1-i\gamma_{21},\ \Omega_{32} = \omega_3-\omega_2-i\gamma_{32},\ \Omega_{31} = \omega_3-\omega_1, \tag{3}$$

where $\hbar\omega_i$ are energies of the levels and the quantities $\gamma_{ij}$ describe the dephasing rates. In the following we shall calculate the response of an E-type system, but the final results can be applied to V- and $\Lambda$-type systems by a suitable sign change of the frequencies involved.

We introduce the occupation densities, i.e., the diagonal density matrix elements $n_1$, $n_2$, and $n_3$. The transition amplitudes, i.e., the off-diagonal density matrix elements, will be denoted as $s_{21}$, $s_{32}$, and $s_{31}$.

In setting up the optical Bloch equations we neglect longitudinal relaxation, i.e., relaxation of the occupation densities towards equillibrium. Then [16]

$$\dot{s}_{21} + i\Omega_{21}s_{21} = (i/\hbar)[M_{21}(n_1-n_2) - M_{32}s_{31}]E, \tag{4}$$

$$\dot{s}_{32} + i\Omega_{21}s_{32} = (i/\hbar)[M_{32}(n_2-n_3) + M_{21}s_{31}]E, \tag{5}$$

$$\dot{s}_{31} + i\Omega_{31}s_{31} = (i/\hbar)[M_{32}s_{21} - M_{21}s_{32}]E, \tag{6}$$

$$\dot{n}_1 = (i/\hbar)M_{21}[s_{21} - s_{21}^*]E, \tag{7}$$

$$\dot{n}_3 = -(i/\hbar)M_{32}[s_{32} - s_{32}^*]E, \tag{8}$$

$$n_2 = N - n_1 - n_3, \tag{9}$$

where $N$ is the density of three-level systems. We solve the above equations using Greens-function techniques similar to those employed by Yajima and Taira [17].

Let us first assume that level 2 is initially occupied. Then $n_2(t=0) = N$, and $n_1(t=0) = n_3(t=0) = 0$. The initial transition amplitudes $s_{ij}$ are also considered to be zero. The first-order transition amplitudes $s_{21}^{(1)}$ and $s_{32}^{(1)}$ are calculated using Eqs. (4) and (5) with the density matrices at their initial values. Next the electric field and the first-order matrices produce second-order quantities $n_1^{(2)}$, $n_3^{(2)}$, $n_2^{(2)}$, and $s_{31}^{(2)}$ via Eqs. (6)–(9). Finally these second-order quantities and the electric field create the third-order transition amplitudes $s_{21}^{(3)}$ and $s_{32}^{(3)}$ via Eqs. (4) and (5). The final expression for the third-order polarization can be written as

$$\begin{aligned} P^{(3)}(t) &= M_{21}s_{21}^{(3)} + M_{32}s_{32}^{(3)} \\ &= \int_{-\infty}^{t} dt' \int_{-\infty}^{t'} dt'' \int_{-\infty}^{t''} dt''' E(t')E(t'') \\ &\quad \times [A(t,t',t'',t''')E^*(t''') + B(t,t',t'',t''')E(t''')], \end{aligned} \tag{10}$$

where $A$ and $B$ contain appropriate Greens functions. When the electric field is considered as a sum $E(t) = E_1(t) + E_2(t)$, then the above expression multiplies 16-fold. Therefore there is a great need for neglecting irrelevant terms at an early stage. We consider only terms selected by the wave vector conservation, we neglect terms that vanish for vanishing pulse overlap and retain only resonant terms. This leads to the following selection: In V- and $\Lambda$-type systems the only relevant term is $E_2(t')E_2(t'')E_1^*(t''')$. In E-type systems this term as well as $E_1^*(t')E_2(t'')E_2(t''')$ are relevant, the latter giving a response via $s_{31}^{(2)}$.

The resulting expression then becomes

$$\begin{aligned} P^{(3)}(t) &= M_{21}s_{21}^{(3)} + M_{32}s_{32}^{(3)} \\ &= \int_{-\infty}^{t} dt' \int_{-\infty}^{t'} dt'' \int_{-\infty}^{t''} dt''' \\ &\quad \times [A(t,t',t'',t''')E_2(t')E_2(t'')E_1^*(t''') \\ &\quad + B(t,t',t'',t''')E_1^*(t')E_2(t'')E_2(t''')]. \end{aligned} \tag{11}$$

Our result for $A$ is

$$\begin{aligned} A &= iNM_{21}^2 e^{-i\Omega_{21}(t-t')} \\ &\quad \times [2M_{21}^2 e^{i\Omega_{21}^*(t''-t''')} + M_{32}^2 e^{i\Omega_{32}^*(t''-t''')}] \\ &\quad - iNM_{32}^2 e^{-i\Omega_{32}(t-t')} \\ &\quad \times [2M_{32}^2 e^{i\Omega_{32}^*(t''-t''')} + M_{21}^2 e^{i\Omega_{21}^*(t''-t''')}]. \end{aligned} \tag{12}$$

$B$ is given by

$$\begin{aligned} B &= iNM_{21}^2 M_{32}^2 e^{-i\Omega_{31}(t'-t'')} \\ &\quad \times [e^{-i\Omega_{21}(t-t')} + e^{-i\Omega_{32}(t-t')}] \\ &\quad \times [e^{-i\Omega_{21}^*(t''-t''')} + e^{-i\Omega_{32}^*(t''-t''')}]. \end{aligned} \tag{13}$$

Inserting a pair of pulses given by (1) we find

$$\begin{aligned} P^{(3)}(t,\tau) &\propto A(t,\tau,\tau,0)\Theta(t-\tau)\Theta(\tau) \\ &\quad + B(t,0,\tau,\tau)\Theta(t)\Theta(-\tau), \end{aligned} \tag{14}$$

where $\Theta$ is the unit step function. Various integrations involving the envelope function $E_p(t)$ of (1) are not written explicitly. It is seen that $A$ and $B$ describe the signal for positive and negative delays, respectively.

In case of frequency selection of the diffracted signal used by Lyssenko *et al.* [15], it is relevant to calculate the Fourier transform of $P^{(3)}(t,\tau)$ and insert the above expressions for $A$ and $B$. For positive delay we get

$$\begin{aligned} \widetilde{P}^{(3)}(\omega,\tau>0) \propto & \frac{2M_{21}^4 e^{i\Omega_{21}^*\tau} + M_{21}^2M_{32}^2 e^{i\Omega_{32}^*\tau}}{\Omega_{21}-\omega} \\ & - \frac{2M_{32}^4 e^{i\Omega_{32}^*\tau} + M_{21}^2M_{32}^2 e^{i\Omega_{21}^*\tau}}{\Omega_{32}-\omega}. \end{aligned} \tag{15}$$

The signal for negative delay is given by

$$\tilde{P}^{(3)}(\omega,\tau<0)\propto 2M_{21}^2M_{32}^2\left[\frac{1}{\Omega_{21}-\omega}+\frac{1}{\Omega_{32}-\omega}\right]e^{-i\Omega_{31}\tau}. \tag{16}$$

The above results apply to E-type systems with level 2 initially occupied. Only slight modifications are necessary for treating V-type or Λ-type systems (also with level 2 initially occupied): There is no signal for negative delay ($B$ is zero), the definitions of $\mathrm{Re}(\Omega_{21})$ for the V-type system and of $\mathrm{Re}(\Omega_{32})$ for the Λ-type systems should reverse sign, and the minus sign in (15) should be changed to a plus sign.

We now turn to the situation in which the level 1 or 3 is initially occupied. The calculations proceed along the same lines. We shall not give the analytical results here because they hold little interest in relation to oscillations. The signal in this case is nonoscillatory. The reason for this is that quantum beat requires that *two* transitions with almost equal transition frequency be coherently driven by the first pulse. This condition is not fulfilled when level 1 or 3 is initially occupied.

Results for II systems can easily be derived from the above calculations. Considering (16) with $M_{32}=0$ we get the contribution from a single two-level system. Two such systems [see Fig. 1(a)] give the third-order polarization expressed as

$$\tilde{P}^{(3)}(\omega,\tau>0)\propto\left[\frac{2M_{21}^4e^{i\Omega_{21}^*\tau}}{\Omega_{21}-\omega}+\frac{2M_{43}^4e^{i\Omega_{43}^*\tau}}{\Omega_{43}-\omega}\right]. \tag{17}$$

The above results all refer to experiments based on frequency selection of the diffracted signal. In case of the real-time resolution performed by Koch *et al.* [14], it is relevant to consider the result given in Eq. (14) directly (without Fourier transform). For V-type systems in which $B$ is zero we get

$$\begin{aligned}P^{(3)}&(t,\tau>0)\\&\propto\Theta(t-\tau)[2M_{21}^4e^{-i\omega_{21}(t-2\tau)}+2M_{32}^4e^{-i\omega_{32}(t-2\tau)}]\\&\quad+M_{21}^2M_{32}^2\Theta(t-\tau)[e^{-i\omega_{21}(t-\tau)+i\omega_{32}\tau}\\&\qquad+e^{-i\omega_{32}(t-\tau)+i\omega_{12}\tau}].\end{aligned} \tag{18}$$

For II-type systems we get

$$\begin{aligned}P^{(3)}&(t,\tau>0)\\&\propto\theta(t-\tau)[M_{21}^4e^{-i\omega_{21}(t-2\tau)}+M_{43}^4e^{-i\omega_{43}(t-2\tau)}].\end{aligned} \tag{19}$$

## III. DISCUSSION

Here we shall apply the above results to the case where the dephasing rates are somewhat smaller than the frequency difference, i.e., no spectral overlap of the two transitions.

The square of the real part of the third-order polarization $P^{(3)}$ is the detected signal $I^{(3)}$. We show in Fig. 2 the dependence of $I^{(3)}$ on $\tau$ and $\omega$ near one of the resonances for a II system as given in (17). In Fig. 3 we show the result for a three-level system with level 2 initially occupied, as given in (15). Note that the dependence of the signal on the detector $\omega$ is characterized by Lorenzians about each resonance. The dependence on $\tau$ is an exponential decay with superimposed oscillations.

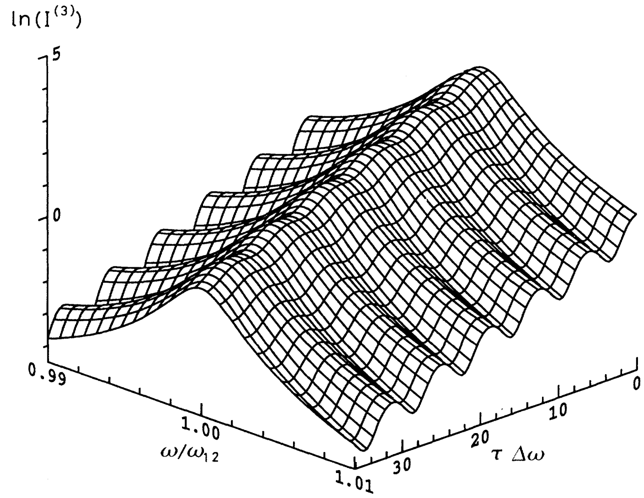


FIG. 2. $(\omega,\tau)$ dependence of the diffracted signal near the upper resonance of a II-type system. The parameters are $\Delta\omega=\omega_{12}-\omega_{34}=0.04\,\omega_{12}$ and $\gamma_{12}=\gamma_{34}=0.002\,\omega_{12}$.

It is appropriate to express any of the results (15), (16), or (17) as

$$I^{(3)}=I_{\mathrm{ave}}[1+I_m\sin(\Delta\omega\tau+\phi)], \tag{20}$$

where $\Delta\omega$ is the difference between the two transition frequencies. $I_{\mathrm{ave}}$, $I_m$, and $\phi$ can be expressed as analytically functions of $\omega$ and/or $\tau$. Note that $\phi$ depends only on $\omega$. Instead of giving all the analytical results explicitly we show in Fig. 4 an overview of the characteristics of the systems considered.

As seen in Figs. 2–4, there is a distinct difference between three-level systems exhibiting quantum beats and II systems exhibiting polarization interference:

(i) Quantum beat is characterized by a small or no change of $\phi$ and $I_m$ when passing through the resonance. The reason for this is that the beating between the two terms with different frequency denominator is unimpor-

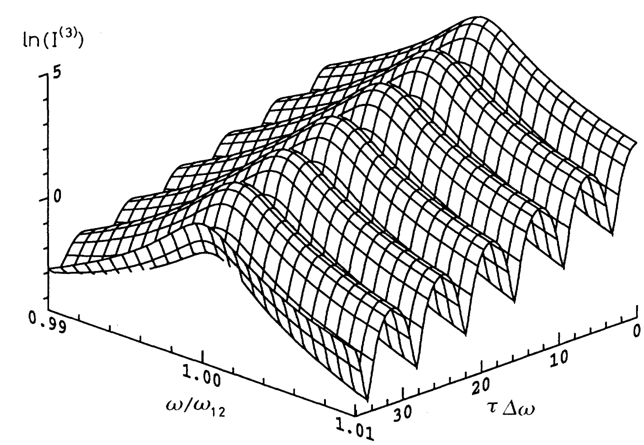


FIG. 3. $(\omega,\tau)$ dependence of the diffracted signal near the upper resonance of a V-type system. The units on the axes are arbitrary. $\Delta\omega=(\omega_{12}-\omega_{23})=0.04\,\omega_{12}$ and $\gamma_{12}=\gamma_{23}=0.002\,\omega_{12}$.

tant. A detailed analysis of (15) shows that *between* the resonances $\phi$ changes from $-\pi/2$ to $\pi/2$ and back. But this happens where the FWM signal is small.

(ii) In contrast to this, the polarization interference must include two terms with different frequency denominators (17). Therefore this case has the following characteristics: $\phi$ changes from $-\pi/2$ to $\pi/2$ (or visa versa) and $|I_m|$ goes through a minimum when the detector frequency passes any of the resonances.

As for the real-time behavior, Eq. (18) indicates a complex quantum beat behavior. Taking the square of (18) and considering components with frequency $\Delta\omega = \omega_{21} - \omega_{32}$, it can be shown that the beat contains terms with phases $\Delta\omega(t-2\tau)$, $\Delta\omega(t-\tau)$, $\Delta\omega t$, and $\Delta\omega\tau$. This situation for quantum beat is somewhat more complicated than claimed in [14]. As for polarization interference it is seen from (19) that one obtains a single beat term and that this has the phase $\Delta\omega(t-2\tau)$, which is also found in [14].

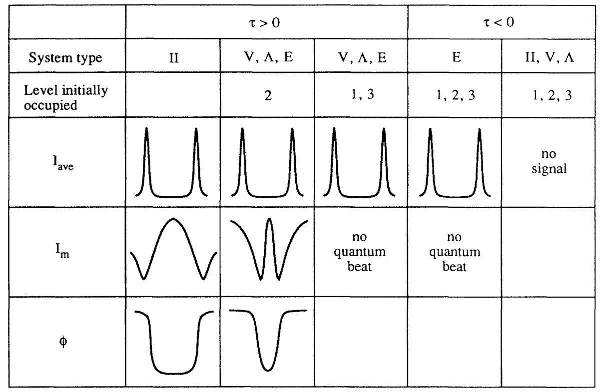


FIG. 4. Characteristics of the diffracted signal in terms of the functions $I_{\text{ave}}, I_m$, and $\phi$ for different systems, sign of $\tau$, and level initially occupied. The curves show the functions of the detected frequency.

## IV. CONCLUSION AND OUTLOOK

There is a detailed agreement between the above mentioned theoretical results for II and V systems and the experiments by Lyssenko *et al.* [15]. Here the II system is the $I_1, I_2$ transitions in CdSe, and the V-type three-level system is the light-hole and heavy-hole excition transitions in GaAs quantum wells.

This agreement encourages further theoretical studies of related systems. The most obvious is to consider the exciton-biexciton system. A proper model is then a four-level system: the ground state, the exciton state, the biexciton state, and the state formed by two free excitons. Preliminary considerations of these systems give oscillations also for negative delay. Another interesting aspect of the exciton-biexciton system is the case of spectrally narrow incident pulses at the two-photon transition. The $(\omega, \tau)$ dependence of the diffracted signal in this case is yet to be explored.

Our results also call for further experimental studies of $\Lambda$ and V systems, in order to confirm the peculiar dependence of the phase $\phi$ in the region between the resonances where the signal is rather small (see Fig. 4).

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