DIAGRAMMATIC DENSITY MATRIX EVALUATION
OF TRANSIENT FOUR-WAVE MIXING IN NONLINEAR MEDIUMS

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Abstract
A Feynman diagrammatic representations are introduced to density matrix perturbation solutions for analyzing transient four-wave mixing technique. Using this approach, an analytical expression of the density matrix is derived for nonlinear mediums.

Keywords : Feynman diagrams; Four-wave mixing; Density matrix.

1. Introduction
Many applications of the diagrammatic density matrix perturbation technique used to analyze and describe third order nonlinear optical processes have been investigated [1-4]. A diagrammatic density matrix perturbation technique has been applied by Fujimoto and Yee [4] to develop a unified theoretical description of transient four-wave mixing processes with emphasis on their applications to the measurements of transient phenomena. The interest of the diagrams is that they provide a physical interpretation for the density matrix terms that they present. The effect of varying experimental parameters for the measurement of transient behavior can often be determined directly from the diagrams.

On the other hand, the four-wave mixing is one of the most interesting investigated nonlinear optical effects. Payne et al. [5] have studied the effects of four-wave mixing on four photon resonance excitation. This technique is used to study many cases as the nonlinear interaction of light in gas and vacuum [6], the third-order optical nonlinearity and the nonlinear absorption in molecules [7] and it is theoretically investigated in a fiber loop mirror [8]. It is to be noted that four-wave mixing is restricted only to the steady-state case. So, to analyze the transient behavior of nonlinear optical processes, the transient four-wave mixing, which is realized by introducing the time as an additional variable, must be considered.

Starting from the work of Fujimoto and Yee [4], we introduce in this paper a Feynman diagrammatic representations for the density matrix perturbation solutions such that each term is uniquely represented by a diagram. In this work, the diagrammatic formalism allows the easy identification perturbation terms which will contribute to a given process. It has been used for the exact calculation of the density matrix perturbation terms. So, using density matrix perturbation theory, we can determine the effects of both population and dephasing relaxation [9].

We give in Section 2 a brief description of the three pulse scattering technique and the diagrammatic formalism. The third order matrix density perturbation terms are given also as integral forms by analyzing transient four-wave mixing technique. The analytical expression of the density matrix is derived explicitly in Section 3. The summary of this theoretical framework is exposed in Section 4.

2. Third-order density matrix perturbation terms
To describe transient third-order nonlinear optical processes and their applications to the measurements of transient phenomena, we analyze the three pulse scattering transient four-wave mixing. Let us consider three incident pulses $\hat{E}_1$, $\hat{E}_2$ and $\hat{E}_p$ interacting with a two level system in a crossed beam geometry (Fig. 1). In this technique $\hat{E}_1$ and $\hat{E}_2$ are the pump beams which may be written as $E_1(\omega) e^{ik_1 r} + c.c.$ and $E_2(\omega) e^{ik_2 r} + c.c.$, respectively, where the two laser pulses are separated by a variable time delay $\tau_p$. $E_p$ is the probe beam which is written as $E_p(\omega + \Delta \omega) e^{ik_p r} + c.c.$, respectively, where the two laser pulses are separated by a variable time delay $\tau_p$. $E_p$ is the probe beam which is written as $E_p(\omega + \Delta \omega) e^{ik_p r} + c.c.$, respectively, where the two laser pulses are separated by a variable time delay $\tau_p$. $E_p$ is the probe beam which is written as $E_p(\omega + \Delta \omega) e^{ik_p r} + c.c.$, with $\tau$ is the variable time delay between the probe beam and the pump beams and c.c. is the complex conjugate. The fields $\hat{E}_1$ and $\hat{E}_2$ interact with the system to produce an excited grating which scatters the field $\hat{E}_p$ into $\hat{k}_1 = \hat{k}_p + \hat{k}_1$ and $\hat{k}_4 = \hat{k}_p + \hat{k}_1 - \hat{k}_2$ directions.
The nonlinear process is described by contributions of third-order density perturbation terms of the form $\rho_{ng}^{(3)}(t)$ into $\vec{k}_3$ and $\vec{k}_4$ directions. Following the works presented in Refs. [3] and [10], we assume that they are 48 possible third order density matrix terms by considering the system evolves from the ground state. The antiresonant perturbation terms are negligible and the terms involving two consecutive absorption or emission interactions by either the ket or bra components of the density matrix are negligible. Eight of these terms into $\vec{k}_3$ direction are given by the diagrams A-D and A'-D' in the case of a semiconductor where $n=c$ (conduction band) and $g=v$ (valence band) (Fig. 2) while an additional 16 terms may be obtained by considering all permutations of the field interactions. The remaining 24 terms are complex conjugates and are represented by diagrams which are mirror images of those given by the first 24 terms. Thus, up to a permutation of field interactions, the eight diagrams of Fig. 2 and their mirror images represent all possible topologies of the class of 48 third order density matrix perturbation terms which are consistent with the above assumptions.

In the eight third-order density perturbation terms, $g$ is the ground state and $n$ is the excited state. For analyzing the three pulse scattering, we interest to diagrammatic representation for $\rho_{ng}^{(3)}(t)$ term into $\vec{k}_3$ direction. This representation describes a process by considering the fields $\vec{E}_p$ and $\vec{E}_1^*$ produce an intermediate excited state $\rho_{nn}^{(2)}$ which has a spatial periodicity of $\vec{k}_p - \vec{k}_1$. This periodic excited state then scatters the field $\vec{E}_2$ into $\vec{k}_3$ direction (Fig. 2a).

We give here the algebraic expressions of these terms which correspond to diagrams A-D. The density matrix $\rho_{ng}^{(3)}(t)$ terms for each diagram (A-D) into $\vec{k}_3$ direction can be noted by a general form which is [11]

$$
\rho_{ng}^{(3)}(t) = \int_{-\infty}^{t} \int_{-\infty}^{t} \int_{-\infty}^{t} e^{-i\gamma(t_1-t_2)\vec{E}(t_1-t_2)} e^{-i\gamma(t_1-t_3)\vec{E}(t_1-t_3)} e^{-i\gamma(t_1-t_4)\vec{E}(t_1-t_4)} e^{-i\gamma(t_1-t_5)\vec{E}(t_1-t_5)} \rho_{nn}^{(2)}(t_1) \, dt_1 \, dt_2 \, dt_3 \, dt_4 \, dt_5,
$$

(1)

with $\omega_{1,2,3} = \omega_{ij} = \omega_i - \omega_j$ are the oscillation frequencies and $\Gamma_{1,2,3} = \Gamma_{ij}$ are the relaxation rates of the $|i\rangle < |j\rangle$ density matrix state. $\Gamma_{ij}$ is the population relaxation rate $T_{1}^{-1}$ if the state is diagonal or the dephasing relaxation rate $T_{2}^{-1}$ if the state is off diagonal. The other parameters are defined as

$\mu_i \geq < n / g >$, or, $< g / \mu / n >$,

with $\mu$ is the electric dipole.

$\varepsilon_i = \pm 1$,

$\tau_i = \tau_p, \tau_0$,
with \( i = 1, 2 \) and 3.
\[
\omega_1 = \omega_{eg}, \omega_{gn}, \quad \omega_2 = 0, \quad \omega_3 = -\omega_1, \omega_1,
\]
\[
\Gamma_1 = \Gamma_3 = \Gamma_{eg} \Gamma_{ge}
\]
and \( \Gamma_2 = \Gamma_{ea} \Gamma_{ge} \).

\[
\begin{align*}
\rho_{eg}^{(3)} & \quad \text{in the } \vec{k}_p + \vec{k}_2 - \vec{k}_1 \text{ direction} \\
& \quad \text{which describe the three pulse scattering.}
\end{align*}
\]

\( (a) \) : The field \( \vec{E}_p \) is scattered by the excited grating caused by the fields \( \vec{E}_g \) and \( \vec{E}_e \).

\( (b) \) : \( \vec{E}_p \) is scattered by the excited grating caused by the fields \( \vec{E}_g \) and \( \vec{E}_e \).

\( (A) \)

\( (B) \)

\( (A') \)

\( (B') \)

\( (C) \)

\( (D) \)

\( (C') \)

\( (D') \)

\textbf{Figure 2:} Diagrammatic representations for the density matrix perturbation terms \( \rho_{eg}^{(3)} \) in the \( \vec{k}_p + \vec{k}_2 - \vec{k}_1 \) direction which describe the three pulse scattering. (a) : The field \( \vec{E}_p \) is scattered by the excited grating caused by the fields \( \vec{E}_g \) and \( \vec{E}_e \). (b) : \( \vec{E}_p \) is scattered by the excited grating caused by the fields \( \vec{E}_g \) and \( \vec{E}_e \).
So, by using the following variables
\[ t_1 = t_3 - t_3, \]
\[ t_2 = t_2 - t_2, \]
and
\[ t_1 = t_1 - t_1, \]
\[ \rho_{eg}^{(3)} = \langle \mu_1 < \mu_2 < \mu_3 > e^{(-i\omega_3 - \Gamma_3) t_3} e^{(-i\omega_2 - \Gamma_2) t_2} e^{(-i\omega_1 - \Gamma_1) t_1} e^{i\omega_1 + i\omega_2 + i\omega_3} \times \int_{-\infty}^{t_1} e^{i\omega_3 - i\omega_2 - i\omega_1} e^{i\Gamma_1 t_1} d\tau_1 \int_{-\infty}^{t_2} e^{i\omega_2 - i\omega_1} e^{i\Gamma_2 t_2} d\tau_2 \int_{-\infty}^{t_3} e^{i\omega_1} e^{i\Gamma_3 t_3} d\tau_3. \]  

Habitually, for the evaluation of diagrammatic density matrix terms it was necessary to resort to the numerical methods because it is difficult to find an explicit form. Fortunately, we can explicit the integrals in Eq. (2) if the integral formula of error function \( \text{erf}(z) \) is used.

3. Analytical expression of diagrammatic density matrix

As usual, it is difficult to find an analytical expression of \( \rho_{eg}^{(3)}(t) \). In this section, we give in the case of a resonant system a novel analytical expression of diagrammatic density matrix perturbation \( \rho_{eg}^{(3)}(t) \) which can be written as

\[ \rho_{eg}^{(3)}(t) = e^{-\alpha t} \int_{-\infty}^{t} e^{-\alpha x^2} dx \int_{-\infty}^{\infty} e^{-\alpha x^2} dx \int_{-\infty}^{\infty} e^{-\alpha x^2} dx, \]

where \( \alpha = \Gamma_1 + i\omega_1 \), and

\[ \text{Re} = \langle \mu_1 < \mu_2 < \mu_3 > t_3^{-1} e^{i\omega_3 + i\omega_2 + i\omega_1} \times \]

with

\[ \alpha = (\Gamma_3 - \Gamma_1) t_3 + (\Gamma_2 - \Gamma_1) t_2 + (\Gamma_1 + \frac{t_1^2}{4} - \Gamma_1 t_1 - \Gamma_2 t_2 - \Gamma_3 t_3), \]

and

\[ \beta = (\omega_1 - \omega_2) t_3 + (\omega_2 - \omega_3) t_2 + (\omega_3 t_1), \]

where

\[ \Gamma_2 = \Gamma_{nn} = \Gamma_{gg} = 1/T_2, \]

and

\[ \Gamma_1 = \Gamma_3 = 1/T_1. \]

\[ \rho_{eg}^{(3)}(t) = \langle \mu_1 < \mu_2 < \mu_3 > t_3^{-1} e^{i\omega_3 + i\omega_2 + i\omega_1} \times \]

\[ 2(2\preceq p + \preceq n + \preceq m) \preceq b_{p+1} \preceq 2^{1/2} S_p(t') + C^{2p+1} b^{2(p+1)} T_p(t'), \]

where the different coefficients used in this equation are given by

\[ S_p(t') = \sum_{k=0}^{p} A(k) t^{p-k-1} e^{-t'} - \Gamma (p+1/2)(1+\text{erf}t'), \]

\[ H_p(t') = \sum_{k=0}^{p} \tilde{A}(k) t^{p-k-2k} + \frac{(2p)!}{2^p} \tilde{g} e^{-t'}, \]

and if we consider that the amplitude of excitation field is a Gaussian

\[ E(t) = e^{-t^2 / t_c^2}, \]

with \( t_c \) is the coherent time of laser, the general expression for the density matrix terms become

\[ a = \frac{1}{2} (\Gamma_3 - 2\Gamma_2 + \Gamma_1) t_x + \frac{(\tau_2 - \tau_3)}{t_c}, \]

and

\[ b = \frac{1}{2} (\Gamma_2 - 2\Gamma_1) t_x + \frac{(\tau_2 - \tau_3)}{t_c}. \]

To get Eq. (3), we have used the following parameters

\( x_1 = t_1 / t_c - \Gamma_1 t_x / 2, \)
\( x_2 = t_2 / t_c - (\Gamma_2 - \Gamma_1) t_x / 2, \)
\( x_3 = t_3 / t_c - (\Gamma_3 - \Gamma_2) t_x / 2, \)
\( t_1 = t_x t_1 + \Gamma_1 t_x^2 / 2, \)
\( t_2 = t_x x_2 + (\Gamma_2 - \Gamma_1) t_x e^2 / 2, \)

and

\( t_3 = t_x x_3 + (\Gamma_3 - \Gamma_2) t_x e^2 / 2. \)

With the help of the \( \text{erf} \) function development defined by [12]

\[ \text{erf}(t) = \frac{2}{\sqrt{\pi}} \sum_{n=0}^{\infty} \left( \frac{(-1)^n}{n! (2n+1)} \right) t^{2n+1}, \]

the known identity [12]

\[ \int_{-\infty}^{\infty} e^{-x^2} dx = \sqrt{\pi}, \]

and after performing the tedious integrations of Eq. (3), the final result can be written as

\[ \text{erf}(t) = \left( \frac{1}{\sqrt{\pi}} \int_{-\infty}^{\infty} e^{-x^2} dx \right)^{2n+1}, \]

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\[ S_p(t') = \left( -\frac{1}{4} \right) e^{-t'/\gamma} \sum_{k=0}^{\infty} A(k) \sum_{p=0}^{k-1} \left( C_p^{2k-2\alpha} \left( \frac{a}{2} \right)^{2(\beta-\gamma)-1} F_\alpha(t') + C_{p+1}^{2k-2\alpha} \left( \frac{a}{2} \right)^{2(\beta-\gamma)-1} P_\alpha(t') \right) \]

\[ \Gamma(p + \frac{1}{2}) \left( 1 + \text{erf} t' \right) - \sum_{n=0}^{\infty} \left( \frac{(-1)^n}{n!(2n+1)} \right) \sum_{p=0}^{k-1} C_{2k}^{2k-2\alpha} d^{2k-2\alpha} G_p(t') + C_{2k+1}^{2k+1} \left( 2^{\alpha+1} H_p(t') \right) \]

\[ T_p(t') = \left( -\frac{1}{4} \right) e^{-t'/\gamma} \sum_{k=0}^{\infty} \tilde{A}(k) \sum_{p=0}^{k-1} \left( C_p^{2k-2\alpha} \left( \frac{a}{2} \right)^{2(\beta-\gamma)-1} F_\alpha(t') + \sum_{p=0}^{k-1} C_{2k+1}^{2k+1} \left( \frac{a}{2} \right)^{2(\beta-\gamma)-1} P_\alpha(t') \right) + \right. \]

\[ \sqrt{\pi} e^{-t'/\gamma} \left( \frac{(2p)!}{2^p} \right)^{2(\gamma-1)/2} \left[ 1 + \text{erf} \left( \sqrt{2} \left( t' + \alpha \right) \right) \right], \]

\[ F_\alpha(t') = \sum_{q=0}^{k-1} B(q) e^{-(t'+\alpha/2)^2} e^{-2(t'+\alpha/2)^2} - \sqrt{2\pi} \left( \frac{2\pi - 1}{4} \right)^{2(\gamma-1)/2} \left[ 1 + \text{erf} \left( \sqrt{2} \left( t' + \alpha / 2 \right) \right) \right], \]

\[ \Gamma(p' + \frac{1}{2}) \left( 1 + \text{erf} t' \right) - \sum_{n=0}^{\infty} \left( \frac{(-1)^n}{n!(2n+1)} \right) \sum_{p'=0}^{k-1} C_{2k}^{2k-2\alpha} d^{2k-2\alpha} G_p(t') + C_{2k+1}^{2k+1} \left( 2^{\alpha+1} H_p(t') \right) \]

\[ T_p(t') = \left( -\frac{1}{4} \right) e^{-t'/\gamma} \sum_{k=0}^{\infty} \tilde{A}(k) \sum_{p=0}^{k-1} \left( C_p^{2k-2\alpha} \left( \frac{a}{2} \right)^{2(\beta-\gamma)-1} F_\alpha(t') + \sum_{p=0}^{k-1} C_{2k+1}^{2k+1} \left( \frac{a}{2} \right)^{2(\beta-\gamma)-1} P_\alpha(t') \right) + \right. \]

\[ \sqrt{\pi} e^{-t'/\gamma} \left( \frac{(2p)!}{2^p} \right)^{2(\gamma-1)/2} \left[ 1 + \text{erf} \left( \sqrt{2} \left( t' + \alpha \right) \right) \right], \]

\[ F_\alpha(t') = \sum_{q=0}^{k-1} B(q) e^{-(t'+\alpha/2)^2} e^{-2(t'+\alpha/2)^2} - \sqrt{2\pi} \left( \frac{2\pi - 1}{4} \right)^{2(\gamma-1)/2} \left[ 1 + \text{erf} \left( \sqrt{2} \left( t' + \alpha / 2 \right) \right) \right], \]

\[ P_p(t') = \sum_{q=0}^{k-1} \tilde{B}(q) e^{-(t'+\alpha/2)^2} e^{-2(t'+\alpha/2)^2}, \]

\[ A(k) = 2^{2k} \prod_{i=1}^{k} (p - i / 2), \]

\[ \tilde{A}(k) = \prod_{i=0}^{k-1} (p - i), \]

\[ B(q) = 4^{k-1} \prod_{i=1}^{k} (s - i / 2), \]

\[ \tilde{B}(q) = \prod_{i=0}^{k-1} (s - i), \]

Eq. (4) provides the third order density matrix perturbation terms into \( \hat{k}_3 \) direction in the case of transient four-wave mixing by considering the fields are on resonance. The other terms into \( \hat{k}_4 \) direction can be obtained in the same manner. Consequently, by using this analytical formulae, one can perform numerical calculations to illustrate the experimentally measured scattering probe signal described by

\[ S(\tau, \tau_p) \propto \int_{-\infty}^{\infty} P^{(3)} |^2 dt, \]

where the nonlinear optical polarization \( P^{(3)} \) is given by [4]

\[ P^{(3)} = \langle g | \mu | n \rangle \rho^{(3)}_{\text{ng}} \pm c.c. \]

4. Conclusion

In summary, we have studied the density matrix perturbation solutions by introducing a Feynman diagrammatic representations. An analytical expression of the density matrix is expressed in the case of two level system by analyzing transient four-wave mixing technique. The use of diagrammatic perturbation technique was shown to facilitate the rapid identification and calculation of density matrix perturbation solutions.

References