Transient optical properties of coherent four-level atoms without undepleted ground-state approximation
Transmit optical properties of coherent four-level atoms without undepleted ground-state approximation

Hui-jun Li, Chao Hang, Guoxiang Huang *

Department of Physics and Institute of Theoretical Physics, East China Normal University, Shanghai 200062, China

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Abstract

We present a theoretical method for studying the transient linear and nonlinear optical property of a four-level atomic system with electromagnetically induced transparency. We start from equations of motion of density matrix that describe the response of atoms to probe and signal fields. We solve these equations by means of a method of multiple-scales. Different from previous studies, in our approach undepleted ground-state approximation is not used and significant differences from previous results are found.

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1. Introduction

Resonant interaction between light and matter is one of basic topics in nonlinear and quantum optics. In recent years, electromagnetically induced transparency (EIT) in lifetime broadened atomic systems has attracted considerable attentions [1]. The importance of EIT stems from the fact that it results in not only a large suppression of optical absorption, but also a drastic modification of dispersion property and a greatly enhanced nonlinear optical susceptibilities. There are many important applications of EIT, including ultraslow light propagation, optical information storage and retrieval, giant Kerr nonlinearity, enhanced frequency conversion, ultraslow optical solitons, and optical quantum communications and computations, etc. [1–13].

Recently, much interest has focused on the study of low-light-level photon switching via EIT-based quantum interference effect [14–22]. In order to optimize the physical property of such optical switching, it is necessary to get a sound understanding of the transient optical feature of resonant multilevel systems under various EIT configurations. Some earlier explorations on this problem for three-level \( \Lambda \)-type and \( V \)-type atoms have been made by several authors [23–25]. In a recent work, Shen et al. [26] has considered a four-level \( N \)-type EIT medium and provided general formulas for the transient evolution of the linear and nonlinear optical susceptibilities. However, in their calculation amplitude variables describing the evolution of atoms have been used and an undepleted approximation for ground state population has been assumed. Note that for a lifetime broadened medium a complete description of the system is to use density matrix [27]. In addition, for the problem of calculating nonlinear susceptibilities, the undepleted ground state approximation should be avoided because the ground state depletion is significant and such depletion gives non-negligible contribution to nonlinear susceptibilities.

In the present work, we develop an effective theoretical method for calculating the transient linear and nonlinear optical property of lifetime broadened multi-level atomic systems with EIT configurations. Instead of the amplitude variables employed in Ref. [26], we use density matrix formalism to describe the response of atoms to probe and signal fields. Of course solving the density-matrix equations is much more difficult than the amplitude-variable equations because the equation number in density-matrix formalism is much more than the...
equation number in amplitude-variable formalism. We shall introduce a powerful method of multiple-scales to solve the density-matrix equations in a systematic way. Different from the approach in Ref. [26], in our theory the undepleted ground-state approximation is not used. We show that there are significant differences between the results obtained by two different approaches and the depletion of the ground state population gives non-negligible contribution to optical nonlinear susceptibilities.

The Letter is arranged as follows. In Section 2 we present the density-matrix equations of motion for a four-level model and make an asymptotic expansion. In Section 3 we provide solutions of the asymptotic expansion up to three-order approximation based on the perturbation method of multiple-scales. In Section 4, we present the calculating result of linear and nonlinear optical susceptibilities of the system. Finally, in the last section we give a discussion and summary of our main results.

2. Model

The four-level atomic model under consideration is shown in Fig. 1. A probe field of frequency \( \omega_p \) (half Rabi frequency \( \Omega_p \)), a control field of frequency \( \omega_c \) (half Rabi frequency \( \Omega_c \)), and a signal field of frequency \( \omega_s \) (half Rabi frequency \( \Omega_s \)) together with the atomic energy states \( |j\rangle \) (\( j = 1, 2, 3, 4 \)) form a \( N \)-type configuration. The electric-field vector of the system is \( \mathbf{E} = \sum_{i=p,c,s} e_i \mathbf{E}_i \exp[i(k_i z - \omega_i t)] + \text{c.c.} \), where \( e_i \) is the unit vector of the polarization component of the electric field with envelope \( \mathbf{E}_i \) (\( i = p, c, s \)). The Rabi frequencies are defined by \( \Omega_p = (e_p \cdot \mathbf{p}_{13}) \mathbf{E}_p / \hbar, \Omega_c = (e_c \cdot \mathbf{p}_{23}) \mathbf{E}_c / \hbar, \) and \( \Omega_s = (e_s \cdot \mathbf{p}_{23}) \mathbf{E}_s / \hbar \), where \( \mathbf{p}_{ij} \) is the electric dipole moment element associated with the transition from \( |i\rangle \) to \( |j\rangle \). In interaction picture, the equations of motion of density-matrix describing atomic response to the optical fields read [27]

\[
\begin{align*}
&i \left( \frac{\partial}{\partial t} + \gamma_1 \right) \sigma_{11} - i \Gamma_{13} \sigma_{33} + \Omega_p^* \sigma_{31} - \Omega_p \sigma_{31}^* = 0, \\
&i \left( \frac{\partial}{\partial t} + \gamma_2 \right) \sigma_{22} - i(\Gamma_{23} \sigma_{33} + \Gamma_{44} \sigma_{44}) + \Omega_c^* \sigma_{32} - \Omega_c \sigma_{32}^* + \Omega_s^* \sigma_{42} - \Omega_s \sigma_{42}^* = 0, \\
&i \left( \frac{\partial}{\partial t} + \gamma_3 + \Gamma_{13} + \Gamma_{23} \right) \sigma_{33} - \Omega_p^* \sigma_{31} - \Omega_p \sigma_{31}^* - \Omega_c^* \sigma_{32} - \Omega_c \sigma_{32}^* + \Omega_s \sigma_{32}^* = 0, \\
&i \left( \frac{\partial}{\partial t} + \gamma_4 + \Gamma_{24} \right) \sigma_{44} - \Omega_p^* \sigma_{42} + \Omega_s \sigma_{42}^* = 0, \\
&i \left( \frac{\partial}{\partial t} + \gamma_{21} \sigma_{21} - \Omega_p \sigma_{31}^* + \Omega_c \sigma_{31}^* + \Omega_s \sigma_{41} = 0, \\
&i \left( \frac{\partial}{\partial t} + \gamma_{31} \right) \sigma_{31} - (\sigma_{33} - \sigma_{11}) \Omega_p + \Omega_s \sigma_{21} = 0, \\
&i \left( \frac{\partial}{\partial t} + \gamma_{41} \right) \sigma_{41} - \Omega_p \sigma_{43} + \Omega_s \sigma_{21} = 0, \\
&i \left( \frac{\partial}{\partial t} + \gamma_{32} \right) \sigma_{32} - (\sigma_{33} - \sigma_{22}) \Omega_c - \Omega_s \sigma_{42} + \Omega_p \sigma_{32}^* = 0, \\
&i \left( \frac{\partial}{\partial t} + \gamma_{42} \right) \sigma_{42} - (\sigma_{44} - \sigma_{22}) \Omega_s - \Omega_s \sigma_{43} = 0, \\
&i \left( \frac{\partial}{\partial t} + \gamma_{43} \right) \sigma_{43} - \Omega_p^* \sigma_{41} - \Omega_s^* \sigma_{42} + \Omega_c \sigma_{32}^* = 0, \\
\end{align*}
\]

where \( \sigma_{ij} \) (\( i, j = 1 \) to \( 4 \)) are density matrix elements in the interaction picture, \( \sigma_{ii} \) is the occupation probability of atom in state \( |i\rangle \), \( \Gamma_{ij} \) gives the rate per atom at which population decays spontaneously from level \( |i\rangle \) to level \( |j\rangle \), \( \gamma_j = (\Gamma_j + \Gamma_{ij})/2 \) + \( \gamma_{ij} \)/2 + \( \gamma_{ij}^{\text{coll}} \) is the damping rate of the \( \sigma_{ij} \) coherence (with \( \Gamma_j = \sum_{i<j} \Gamma_{ij} \) denoting the total rate of population out of level \( |j\rangle \)), \( \gamma_i \) is the ionization rate of state \( |i\rangle \). The quantity \( \gamma_{ij}^{\text{coll}} \) is the dipole dephasing rate due to processes (such as elastic collisions) that are not associated with the transfer of population [27]. For simplicity, we have assumed that all optical fields have exact resonances in respective energy levels.

3. Asymptotic expansion and solutions in each-order approximation

3.1. Asymptotic expansion of the density-matrix equations

We apply a perturbation method of multiple-scales [28] to solve Eqs. (1a)–(1j) in a systematic way. Different from Ref. [26], here we do not use the undepleted ground state approximation. Assuming that initially the particles of the system occupy in the ground state \( |1\rangle \), we make the following asymptotic expansion

\[
\begin{align*}
\sigma_{ij} &= \delta_{ij} \delta_{j1} + \epsilon \delta_{ij} \sigma_{ij}^{(1)} + \epsilon^2 \sigma_{ij}^{(2)} + \epsilon^3 \sigma_{ij}^{(3)} + \cdots \\
&(i, j = 1 \text{ to } 3, i \geq j), \\
\sigma_{4i} &= \delta_{i1} + \epsilon \delta_{i1} \sigma_{i1}^{(1)} + \epsilon^2 \sigma_{i1}^{(2)} + \epsilon^3 \sigma_{i1}^{(3)} + \cdots \\
&(i = 1 \text{ to } 3),
\end{align*}
\]

where \( \epsilon \) is a small parameter characterizing a small population depletion of the ground state. We assume that the control field is strong (with order of unity) and the probe and signal field are

\[
\text{Fig. 1. The energy-level diagram and excitation scheme of a lifetime broadened four-state system interacting with a probe field of half Rabi frequency } \Omega_p, \text{ a control field of half Rabi frequency } \Omega_c, \text{ and signal field of half Rabi frequency } \Omega_s.
\]
weak (with order of $\varepsilon$). To obtain a divergence-free solutions in each-order approximation, all quantities on the right-hand side of the asymptotic expansions (2a) and (2b) are considered as functions of the multi-scale variables $t_i = \varepsilon^j t$ ($i = 0, 1, 2$).

Substituting the expansions (2a) and (2b) into Eqs. (1a)–(1j), we obtain

$$i \left( \frac{\partial}{\partial t_0} + \gamma_{21(31)} \right) \sigma_{21(31)}^{(j)} + \Omega_{c} \sigma_{31(21)}^{(j)} = a^{(j)}(\beta^{(j)}),$$

$$ (j = 1 \to 3),$$

$$i \left( \frac{\partial}{\partial t_0} + \gamma_{41(14)} \right) \sigma_{41}^{(2)} = \delta^{(2)},$$

$$i \left( \frac{\partial}{\partial t_0} + \gamma_{32} \right) \sigma_{32}^{(2)} - \Omega_{c} (\sigma_{33}^{(2)} - \sigma_{22}^{(2)}) = M^{(2)},$$

$$i \left( \frac{\partial}{\partial t_0} + \gamma_1 \right) \sigma_{11}^{(2)} - i \frac{\Gamma_3}{2} \sigma_{33}^{(2)} = N^{(2)},$$

$$i \left( \frac{\partial}{\partial t_0} + \gamma_2 \right) \sigma_{22}^{(2)} - i \frac{\Gamma_3}{2} \sigma_{33}^{(2)} + \Omega_{c} \sigma_{32}^{(2)} - \Omega_{c} \sigma_{52}^{(2)} = 0,$$

$$i \left( \frac{\partial}{\partial t_0} + \gamma_3 + \Gamma_3 \right) \sigma_{33}^{(2)} - \Omega_{c} \sigma_{32}^{(2)} + \Omega_{c} \sigma_{52}^{(2)} = -N^{(2)},$$

(3a)–(3f)

The explicit expressions of $\alpha^{(j)}$, $\beta^{(j)}$, $\delta^{(2)}$, $M^{(2)}$, $N^{(2)}$ in Eqs. (3a)–(3f) are given in Appendix A. Note that here we list only those equations needed for the calculation up to third-order susceptibilities. In addition, for simplicity we have assumed $\Omega_{p}$ and $\Omega_{c}$ are real numbers and $\Gamma_{23} = \Gamma_{13} = \Gamma_{21}/2$, with $\Gamma_{3}$ and $\Gamma_{4}$ being much larger than $\gamma_{2}$ and $\gamma_{3}$.

For convenience we convert Eqs. (3a)–(3f) into the following form:

$$\hat{L}_1 \sigma_{31}^{(j)} = S_{1}^{(j)},$$

(4a)

$$\sigma_{21}^{(j)} = \frac{1}{\Omega_{c}} \left[ \frac{\beta^{(j)} - i}{\Omega_{c}} \left( \frac{\partial}{\partial t_0} + \gamma_{31} \right) \sigma_{31}^{(j)} \right],$$

(4b)

$$ (j = 1 \to 3)$$

and

$$\hat{L}_2 \sigma_{32}^{(2)} = S_{2},$$

(5a)

$$\sigma_{32}^{(2)} - \sigma_{32}^{*^{(2)}} = \frac{1}{2\Omega_{c}} \left[ \left( \frac{\partial}{\partial t_0} + \Gamma_{3} \right) \sigma_{33}^{(2)} + N^{(2)} \right],$$

(5b)

$$\sigma_{22}^{(2)} = \frac{1}{2\Omega_{c}} \left[ \left( M^{(2)} + c.c. \right) + 2\Omega_{c} \sigma_{33}^{(2)} \right. \left. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \right. \r
\[ \rho_{31}^{(3)} = -\frac{2}{\Omega} \left[ (iK_\alpha + \alpha_s - \beta_\gamma) e^{K_\beta t} - (iK_{-\alpha} + \alpha_s - \beta_\gamma) e^{K_{-\beta} t} \right] + E_0 \left( e^{i\frac{\Omega}{2} t} - e^{-i\frac{\Omega}{2} t} \right), \]

(13)

are self-Kerr and cross-Kerr nonlinear coefficients, respectively. In above expressions \( a_l, \beta_l \) \((l = p, s)\), \( E_0, E_{0p}, E_{0s}, E_{pp}, E_{ps}, \) and \( \lambda_j \) are constants. Their concrete expressions are omitted here.

4. Linear and nonlinear susceptibilities

The polarization intensity of the system is

\[ P = N_a \left[ p_{13} |\sigma_{31} e^{(k_p z - a_p t)} + p_{23} |\sigma_{32} e^{(k_p z - a_p t)} \right] + p_{24} |\sigma_{42} e^{(k_p z - a_p t)} + c.c.], \]

(14)

where \( N_a \) denotes the atomic density of the system. The expression of time-dependent optical susceptibilities induced by the probe field is given by

\[ \chi_p(t) = \frac{\kappa}{\Omega_p} \sigma_{31}(t) \approx \chi_{pp}^{(1)} \chi_p^{(3)} |e_p|^2 + \chi_{ps}^{(3)} |e_s|^2, \]

(15)

where \( \kappa = N_a |p_{13}|^2 / (\hbar \Omega) \), \( \chi_{pp}^{(3)} \) and \( \chi_{ps}^{(3)} \) denote the self-Kerr and cross-Kerr nonlinear susceptibilities, respectively. Based on the results given in the last section we obtain the explicit expression of linear and nonlinear optical susceptibilities of the probe field, expressed as

\[ \chi_{pp}^{(1)} = \kappa \rho_{31}^{(1)}, \]

(16a)

\[ \chi_{pp}^{(3)} = \frac{\kappa |p_{13}|^2}{\hbar^2} \rho_{31}^{(3)}, \]

(16b)

\[ \chi_{ps}^{(3)} = \frac{\kappa |p_{24}|^2}{\hbar^2} \rho_{31}^{(3)}, \]

(16c)

where \( \rho_{31}^{(1)} = |\sigma_{31}^{(1)}| / \sqrt{\Omega_p} \).

In Fig. 2 we have shown the transient behavior of the imaginary part of linear susceptibility \( \text{Re} \chi_{pp}^{(1)} \). The system parameters are chosen as (typical for transitions in hyperfine-split Na D lines) [26]): \( \Gamma_1 = 1.2 \times 10^8 \text{ s}^{-1}, \Gamma_2 = 2.5 \times 10^8 \text{ s}^{-1}, \gamma_2 = 3 \times 10^6 \text{ s}^{-1}, \Omega_c = 9 \times 10^8 \text{ s}^{-1}, \sigma_{31} = 2.2 \times 10^{11} \text{ cm}^{-3}, |p_{13}| = 2.1 \times 10^{-27} \text{ cm}^2. \]

We see that Im \( \chi_{pp}^{(1)} \) has an oscillation in initial time interval of the order of \( 10^{-7} \text{ s} \) and decays into nearly zero rapidly as time increases. The physical reason of nearly vanishing \( \chi_p^{(1)} \) after the transient evolution is due to the EIT-induced quantum interference effect that greatly suppresses the absorption of the probe field.

Shown in Fig. 3 is the result of the transient behavior of the self-Kerr nonlinear susceptibility of the probe field Im \( \chi_{pp}^{(3)} \) (in unit of \((\text{m/V})^2\)) with the same parameters given in Fig. 2. Curve (a) (black solid line) is the result without using undepleted ground state approximation. Curve (b) (red dashed line) is the result when the undepleted ground state approximation is used. Curve (c) (blue solid line) is the amplification of the curve (b) to 500 times in order to illustrate the transient behavior of the curve (b). We see that there is a drastic difference for Im \( \chi_{pp}^{(3)} \) between the results with and without using the undepleted ground state approximation. In Fig. 4 we have also shown the time evolution of the cross-Kerr nonlinear susceptibility of the probe field Im \( \chi_{ps}^{(3)} \) (in unit of \((\text{m/V})^2\)) with the same parameters given in Fig. 2.

Shown in Fig. 5 is the result of the population in the ground state \( |1\rangle \), i.e. \( \sigma_{11} \), as a function of time. The solid black line represents the result without using the undepleted ground state approximation. The parameters are chosen as \( \Gamma_3 = 1.2 \times 10^8 \text{ s}^{-1}, \gamma_2 = 3 \times 10^6 \text{ s}^{-1}, \Omega_c = 9 \times 10^8 \text{ s}^{-1} \) and \( \Omega_p = \Omega_c / 7 \). We see that \( \sigma_{11} \) displays an apparent transient behavior and its steady state approaches a value less than unity, due to the reason of spontaneous emission and ionization of the system. This result is quite different from that obtained under the undepleted ground state approximation [26], where \( \sigma_{11} \) is always unity (the dashed red line in Fig. 5). This shows also that the depletion of the ground state is important and can not be neglected when calculating nonlinear susceptibilities of the system.

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2 As in Ref. [26], in the present work we have studied only the case of exactly resonance, thus the optical susceptibilities have only an imaginary part (i.e. absorptive susceptibilities). A case of nearly resonance that contribute a real part of the optical susceptibilities (i.e. refractive susceptibilities) will be considered elsewhere.
We have presented a new theoretical method for studying the transient linear and nonlinear optical properties of a four-level atomic system with an EIT configuration. By means of a method of multiple-scales, we have solved the equations of motion of density matrix that describe the linear and nonlinear response of atoms to the probe and signal fields. Different from the approach given in Ref. [26], in our theory the undepleted ground-state approximation has not been employed. We have shown that the depletion of the ground state population gives significant contribution to the nonlinear optical susceptibilities of the system. Obviously, our method is quite general and can be easily applied to other multi-level systems. The results provided here may be useful for the understanding of the switching feature of EIT-based systems and have potential applications in optical information processing and transmission.

5. Summary

We have presented a new theoretical method for studying the transient linear and nonlinear optical properties of a four-level atomic system with an EIT configuration. By means of a method of multiple-scales, we have solved the equations of motion of density matrix that describe the linear and nonlinear response of atoms to the probe and signal fields. Different from the approach given in Ref. [26], in our theory the undepleted ground-state approximation has not been employed. We have shown that the depletion of the ground state population gives significant contribution to the nonlinear optical susceptibilities of the system. Obviously, our method is quite general and can be easily applied to other multi-level systems. The results provided here may be useful for the understanding of the switching feature of EIT-based systems and have potential applications in optical information processing and transmission.

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Appendix A. The coefficients in Eqs. (3)

The explicit expressions of $\alpha^{(j)}$, $\beta^{(j)}$ ($j=1, 2, 3$), $\delta^{(2)}$, $M^{(2)}$, $N^{(2)}$, in Eqs. (3) are given by

\begin{align}
\alpha^{(1)} &= 0, \\
\alpha^{(2)} &= -i \frac{\partial \sigma^{(2)}_{21}}{\partial t_1}, \\
\alpha^{(3)} &= -i \frac{\partial \sigma^{(2)}_{21}}{\partial t_1} - i \frac{\partial \sigma^{(1)}_{21}}{\partial t_2} + \Omega_p \sigma^{(2)}_{32} + \Omega_s^* \sigma^{(2)}_{41}, \\
\beta^{(1)} &= -\Omega_p, \\
\beta^{(2)} &= -i \frac{\partial \sigma^{(1)}_{31}}{\partial t_1}, \\
\beta^{(3)} &= -i \frac{\partial \sigma^{(2)}_{31}}{\partial t_1} - i \frac{\partial \sigma^{(1)}_{31}}{\partial t_2} + (\sigma^{(2)}_{33} - \sigma^{(2)}_{11}) \Omega_p, \\
\delta^{(2)} &= -\Omega_p \sigma^{(2)}_{31}, \\
M^{(2)} &= -\Omega_p \sigma^{(2)}_{31}, \\
N^{(2)} &= -\Omega_p (\sigma^{(1)}_{31} - \sigma^{(1)}_{31}).
\end{align}

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