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Theoretical Studies of Resonance Enhance Stimulated Raman Scattering (RESRS) of Frequency Doubled Alexandrite Laser Wavelengths in Cesium Vapor

(NASA-CR-176464) THEORETICAL STUDIES OF RESONANCE ENHANCE STIMULATED RAMAN SCATTERING (RESRS) OF FREQUENCY DOUBLED ALEXANDRITE LASER WAVELENGTHS IN CESIUM VAPOR Semiannual Report (Brown Univ.) 10 p G3/36 N86-17731 Unclas 05240

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Introduction

It is well known that the presence of a real atomic level which is nearly resonant with the pump field can greatly enhance the Raman emission cross-section. In order to accurately calculate the Raman gain in systems where resonance enhancement plays a dominant role, expressions for the pump and signal susceptibilities must be derived. These expressions should be valid for arbitrary field strengths in order to allow for pump and signal saturation. In addition, the theory should allow for arbitrary longitudinal and transverse relaxation rates. This latter point is extremely vital for three level atomic systems such as the alkali earth metals since they do not have population reservoirs and can have widely varying spontaneous lifetimes on the three pertinent transitions. Moreover, the dephasing rates are strong functions of electronic states and are therefore also different for the three coupled pairs of levels. These considerations are not as important when molecular systems are concerned since the large reservoir of rotational states serve to produce essentially equal longitudinal recovery rates for the population of the three levels.

The most general solution to date is that of Temkin and Panock[1,2] who have solved the semiclassical rotating wave limit of a three level system under the constraint of a single longitudinal and transverse relaxation rate for the entire system of levels.

We have for the first time solved the three level system with three arbitrary longitudinal and transverse relaxation rates. There is no need for setting either pair of rates equal and the expressions are valid for arbitrarily strong fields. The next step will be to velocity integrate these expressions for the cases of copropagating and counterpropagating pump and signal fields in order to study the cesium system pumped by the doubled alexandrite laser emissions.

Density Matrix Solutions for the Susceptibilities of a Three-Level System with Arbitrary Relaxation Rates and Field Strengths

The density matrix formalism is fully equivalent to the Schrodinger wave function however it is more applicable to the statistical case, in which the wave function is not known exactly. In this case the elements of the density matrix are taken to be ensemble averages of possible configurations of the system. As a result every wave function can be expressed as a unique density matrix, but not all density matrices can be expressed as wave functions, reflecting the fact that the wave function contains only quantum uncertainties where as the density matrix contains statistical as well as quantum uncertainties.

For a given wave function

$$|\Psi> = \sum C_n |n>$$

the density matrix is written as

$$\rho_{ij} = \overline{C_i^* C_j}$$

where the bar denotes the ensemble average. The time dependence of the elements is given by

$$\rho_{ij} = \frac{i}{h}[\rho,H]_{ij}$$

where H is the total Hamiltonian of the system used to describe the three and four level atoms pictured below

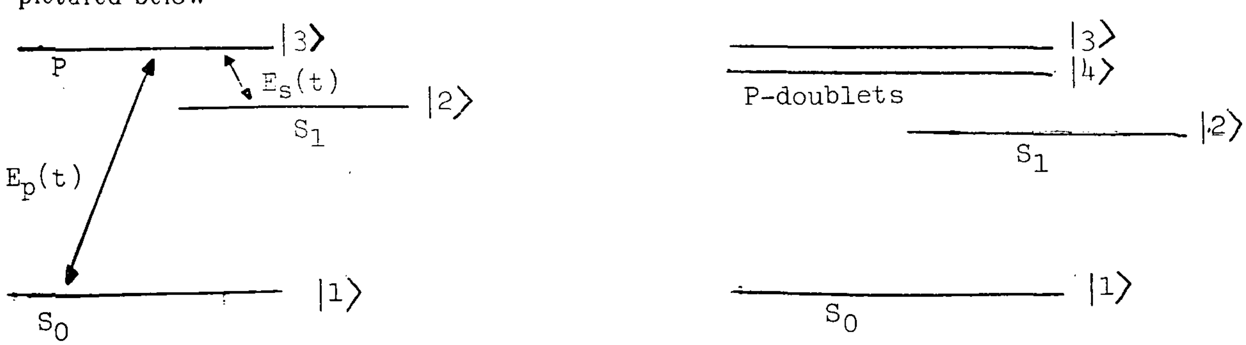

Figure 1

Both configurations approximate electronic states in an alkali atom. The lower states $|1>$ and $|2>$ are s-states with principle quantum numbers n and n+1. In the four level atom $|3>$ and $|4>$ are p-states with j=1/2 and 3/2 respectively and principle quantum number n. In the three level atom the splitting of the upper p-level has been neglected to simplify the calculations and results.

The Hamiltonian in the dipole approximation is given by:

$$H = H_0 - \vec{\mu}\cdot\vec{E}$$

where H_0 is the unperturbed atomic Hamiltonian and $-\vec{\mu}\cdot\vec{E}$ is the atom field Hamiltonian in the dipole approximation. We will assume that the dipole matrix element μ_{12} is zero because both are s-states.

The time evolution of the density matrix elements for the three level system are therefore described by

$$\frac{d\rho_{11}}{dt} = \frac{-i}{h}(\rho_{13} - \rho_{31})\vec{\mu}_{13}\cdot\vec{E}(t) - \gamma_{11}(\rho_{11} - \rho_{11}^{0})$$

$$\frac{d\rho_{22}}{dt} = \frac{-i}{h}(\rho_{23} - \rho_{32})\mu_{23}\cdot\vec{E}(t) - \gamma_{22}(\rho_{22} - \rho_{22}^{0})$$

$$\frac{d\rho_{22}}{dt} = \frac{i}{h}\left[(\rho_{13} - \rho_{31})\vec{\mu}_{12} + (\rho_{32} - \rho_{23})\vec{\mu}_{23}\right]\cdot\vec{E}(t)\gamma_{33}(\rho_{33} - \rho_{33}^{0})$$

$$\dot{\rho}_{13} = \frac{i}{h}\left[(\rho_{33} - \rho_{11})\mu_{13} - \mu_{23}\rho_{12}\right]\cdot\vec{E}(t) - i(\omega_{31} - i\gamma_{13})\rho_{13}$$

$$\dot{\rho}_{32} = \frac{i}{h}\left[(\rho_{33} - \rho_{22})\mu_{23} - \mu_{13}\rho_{12}\right]\cdot\vec{E}(t) + i(\omega_{32} - i\gamma_{32})\rho_{32}$$

$$\dot{\rho}_{12} = \frac{i}{h}\left[\mu_{13}\rho_{32} - \mu_{32}\rho_{13}\right]\cdot\vec{E}(t) - i(\omega_{21} - i\gamma_{12})\rho_{12}$$

where $\omega_{ij} = (E_i - E_j)/h$, $E(t) = E_p\cos(\omega_p t + \psi_p) + E_s\cos(\omega_s t + \psi_s)$ and γ_{ij} are phenomenological relaxation and dephasing rates, and ρ_{31}, ρ_{23} and ρ_{12} are complex conjugates of ρ_{13}, ρ_{32} and ρ_{12} respectively. The equations describing the four level system are given by:

$$\dot{\rho}_{11} = \frac{i}{h}\left[\mu_{13}(\rho_{31} - \rho_{13}) + \mu_{14}(\rho_{41} - \rho_{14})\right]\cdot\vec{E}(t) - \gamma_{11}\rho_{11}$$

$$\dot{\rho}_{22} = \frac{i}{h}\left[\mu_{23}(\rho_{32} - \rho_{23}) + \mu_{24}(\rho_{42} - \rho_{24})\right]\cdot E(t) - \gamma_{22}\rho_{22}$$

$$\dot{\rho}_{33} = \frac{i}{h}\left[\mu_{13}(\rho_{13} - \rho_{31}) + \mu_{23}(\rho_{23} - \rho_{32}) + \mu_{24}(\rho_{42} - \rho_{24})\right]\cdot E(t) - \gamma_{33}\rho_{33}$$

$$\dot{\rho}_{44} = \frac{i}{h}\left[\mu_{10}(\rho_{14} - \rho_{41}) + \mu_{24}(\rho_{24} - \rho_{42}) + \mu_{34}(\rho_{34} - \rho_{43})\right]\cdot E(t) - \gamma_{44}\rho_{44}$$

$$\dot{\rho}_{12} = \frac{i}{h}\left[\mu_{13}\rho_{32} - \mu_{23}\rho_{33} + \mu_{14}\rho_{42} - \mu_{42}\rho_{34}\right]\cdot E(t) - i(\omega_{21} - i\gamma_{12})\rho_{12}$$

$$\dot{\rho}_{13} = \frac{i}{h}\left[\mu_{13}(\rho_{33} - \rho_{11}) + \mu_{14}\rho_{43} - \mu_{23}\rho_{12} - \mu_{43}\rho_{14}\right]\cdot E(t) - i(\omega_{31} - i\gamma_{13})\rho_{13}$$

$$\dot{\rho}_{14} = \frac{i}{h}\left[\mu_{14}(\rho_{44} - \rho_{11}) + \mu_{13}\rho_{34} - \mu_{24}\rho_{12} - \mu_{34}\rho_{13}\right]\cdot E(t) - i(\omega_{41} - i\gamma_{14})\rho_{14}$$

$$\dot{\rho}_{23} = \frac{i}{h}\left[\mu_{23}(\rho_{33} - \rho_{22}) + \mu_{24}\rho_{42} - \mu_{13}\rho_{21} - \mu_{24}\rho_{42}\right]\cdot E(t) - i(\omega_{32} - i\gamma_{23})\rho_{23}$$

$$\dot{\rho}_{24} = \frac{i}{h}\left[\mu_{24}(\rho_{44} - \rho_{22}) + \mu_{23}\rho_{34} - \mu_{14}\rho_{21} - \mu_{34}\rho_{23}\right]\cdot E(t) - i(\omega_{42} - i\gamma_{24})\rho_{24}$$

$$\dot{\rho}_{34} = \frac{i}{h}\left[\mu_{34}(\rho_{44} - \rho_{33}) + \mu_{23}\rho_{14} - \mu_{23}\rho_{24} - \mu_{14}\rho_{31} - \mu_{24}\rho_{32}\right]\cdot E_{(t)} - i(\omega_{43} - i\gamma_{34})\rho_{34}$$

It is obvious that these equations with their complex conjugates are much to complicated to solve. The remainder of this paper will be devoted to the three level system.

Steady State Solutions

To obtain the steady state solutions to the density matrix equation we first take out the explicit oscillations of the off diagonal elements

$$\rho_{13} = \Lambda e^{i\omega_p t} \qquad \rho_{32} = \lambda e^{-i\omega_s t}$$

$$\rho_{12} = D e^{i(\omega_p - \omega_s)t}$$

and then neglect all terms which oscillate faster than ω_p or ω_s in the off diagonal element equations and all DC terms in the diagonal element equations. This is the rotating wave approximation which therefore disallows the possibility of Bloch-Seigert shifts in the resonances. If we then set all time derivatives equal to zero and define the following:

$$\beta_s = \frac{\mu_{23}E_s e^{i\psi_s}}{2h} \qquad \beta_p = \frac{\mu_{13}E_p e^{i\psi_p}}{2h}$$

$$L_s = \omega_s - \omega_{32} + i/\tau_1 = \delta_s + i/\tau_1$$
$$L_p = \omega_p - \omega_{31} - i/\tau_2 = \delta_p - i/\tau_2$$
$$L_{sp} = \omega_s - \omega_p + \omega_{21} + i/\tau_3 = \delta_s - \delta_p + i/\tau_3$$

the equations become

$$L_s\lambda = \beta_s^*\Delta_{32} - \beta_p^*D$$
$$L_p\Lambda = -\beta_p\Delta_{13} - \beta_s D$$
$$L_{sp}D = -\beta_p\lambda + \beta_s^*\Lambda$$

and

$$0 = 2\mathrm{Im}\beta_s\lambda - 4\mathrm{Im}\beta_p^*\Lambda + \frac{1}{3}\left(\frac{2}{T_1} + \frac{1}{T_3}\right)(\Delta_{13} - \Delta_{13}^{o}) + \frac{1}{3}\left(\frac{1}{T_1} - \frac{1}{T_3}\right)(\Delta_{32} - \Delta_{32}^{o})$$

$$0 = 2\mathrm{Im}\beta_p^*\Lambda - 4\mathrm{Im}\beta_s\lambda + \frac{1}{3}\left(\frac{1}{T_3} + \frac{2}{T_2}\right)(\Delta_{32} - \Delta_{32}^{o}) + \frac{1}{3}\left(\frac{1}{T_2} - \frac{1}{T_3}\right)(\Delta_{32} - \Delta_{32}^{o})$$

We can now eliminate D from the first three equations and solve for λ and Λ

$$\lambda = \frac{\beta_s^*}{L_s}\left\{\Delta_{32} + |\beta_p|^2 R\left[\frac{\Delta_{13}}{L_p} + \frac{\Delta_{32}}{L_s}\right]\right\}$$

$$\Lambda = -\frac{\beta_p}{L_p}\left\{\Delta_{13} - |\beta_s|^2 R\left[\frac{\Delta_{13}}{L_p} + \frac{\Delta_{32}}{L_s}\right]\right\}$$

where

$$R = \frac{L_s L_p}{L_s\beta_s^2 - L_p\beta_p^2 + L_s L_p L_{sp}} .$$

In terms of two real quantities, R_1 and R_2, we have that:

$$R = R_1 - iR_2 = \frac{(\delta_p^2 + \tau_2^{-2})(\delta_s^2 + \tau_1^{-2})(A - iB)}{A^2 + B^2}$$

$$A = \delta_s(\delta_p^2 + \tau_2^{-2})(\delta_s^2 + \tau_1^{-2} - \beta_p^2) - \delta_p(\delta_s^2 + \tau_1^{-2})(\delta_p^2 + \tau_2^{-2} - \beta_s^2)$$

$$B = \tau_3^{-1}(\delta_s^2 + \tau_1^{-2})(\delta_p^2 + \tau_2^{-2}) + \beta_p^2\tau_1^{-1}(\delta_p^2 + \tau_2^{-2})(\delta_s^2 + \tau_1^{-2}) + \beta_s^2\tau_2^{-1}(\delta_s^2 + \tau_1^{-2})$$

This then yeilds

$$\mathrm{Im}\beta_s\lambda = \frac{-|\beta_s|^2}{|L_s|^2}\left[\tau_1^{-1} + \frac{|\beta_p|^2}{|L_s|^2}\alpha_3\right]\Delta_{32} - \frac{|\beta_p|^2|\beta_s|^2}{|L_s|^2|L_p|^2}\alpha_2\Delta_{13}$$

and

$$\mathrm{Im}\beta_p^*\Lambda = \frac{-|\beta_s|^2}{|L_p|^2}\left[\tau_2^{-1} - \frac{|\beta_p|^2}{|L_p|^2}\alpha_1\right]\Delta_{13} - \frac{|\beta_p|^2|\beta_s|^2}{|L_s|^2|L_p|^2}\alpha_2\Delta_{32}$$

where

$$\begin{aligned}
\alpha_1 &= 2\tau_2^{-1}\delta_p R_1 - (\delta_p^2 - \tau_2^{-2})R_2 \\
\alpha_2 &= (\delta_p\tau_1^{-1} - \delta_s\tau_0^{-1})R_1 + (\tau_1^{-1}\tau_2^{-1} + \delta_p\delta_s)R_2 \\
\alpha_3 &= 2\tau_1^{-1}\delta_s R_1 - (\delta_s^2 - \tau_1^{-2})R_2
\end{aligned}$$

Substituting these into the last two density matrix equations then gives

$$\frac{1}{3}\left(\frac{2}{T_1} + \frac{1}{T_3}\right)\Delta_{13}^0 + \frac{1}{3}\left(\frac{1}{T_1} - \frac{1}{T_3}\right)\Delta_{32}^0$$

$$= -\left\{\frac{2|\beta_s|^2}{|L_s|^2}\left[\tau_2^{-1} + 2|\beta_p|^2\left(\frac{-\alpha_2}{|L_p|^2} + \frac{\alpha_3}{|L_s|^2}\right)\right] - \frac{1}{3}\left(\frac{1}{T_1} - \frac{1}{T_3}\right)\right\}\Delta_{32}$$

$$+ \left\{\frac{4|\beta_p|^2}{|L_p|^2}\left[\tau_2^{-1} - |\beta_s|^2\left(\frac{\alpha_1}{|L_p|^2} + \frac{\alpha_2}{2|L_s|^2}\right)\right] + \frac{1}{3}\left(\frac{2}{T_1} + \frac{1}{T_3}\right)\right\}\Delta_{13}$$

$$\frac{1}{3}\left(\frac{1}{T_3}+\frac{2}{T_2}\right)\Delta_{32}^0+\frac{1}{3}\left(\frac{1}{T_2}-\frac{1}{T_3}\right)\Delta_{13}^0$$

$$=+\left\{\frac{4\,|\,\beta_s\,|^2}{|\,L_s\,|^2}\left[\tau_1^{-1}+|\,\beta_p\,|^2\left(\frac{-\alpha_2}{2\,|\,L_p\,|^2}+\frac{\alpha_3}{|\,L_s\,|^2}\right)\right]+\frac{1}{3}\left(\frac{1}{T_3}-\frac{2}{T_2}\right)\right\}\Delta_{32}$$

$$-\left\{\frac{2\,|\,\beta_p\,|^2}{|\,L_p\,|^2}\left[\tau_2^{-1}-|\,\beta_s\,|^2\left(\frac{\alpha_1}{|\,L_p\,|^2}+\frac{2\alpha_2}{|\,L_s\,|^2}\right)\right]-\frac{1}{3}\left(\frac{1}{T_2}+\frac{1}{T_3}\right)\right\}\Delta_{13}$$

Then by making the following substitutions and definitions

$$\Gamma_1=\frac{1}{3}\left(\frac{2}{T_1}+\frac{1}{T_3}\right)+\frac{4\,|\,\beta_p\,|^2}{\delta_p^2+\tau_2^{-3}}\left\{\tau_2^{-1}-|\,\beta_s^2\,|^2\left[\frac{\alpha_1}{\delta_p^2+\tau_2^{-2}}+\frac{\alpha_2}{2(\delta_s^2+\tau_1^{-2})}\right]\right\}$$

$$\Gamma_2=\frac{2\,|\,\beta_s\,|^2}{\delta_s^2+\tau_1^{-2}}\left\{\tau_1^{-1}+2\,|\,\beta_p\,|^2\left[\frac{-\alpha_2}{\delta_p^2+\tau_2^{-2}}+\frac{\alpha_3}{2(\delta_s^2+\tau_1^{-2})}\right]\right\}-\frac{1}{3}\left(\frac{1}{T_1}-\frac{1}{T_3}\right)$$

$$\Gamma_3=\frac{2\,|\,\beta_p\,|^2}{\delta_p^2+\tau_2^{-2}}\left\{\tau_2^{-1}-|\,\beta_s\,|^2\left[\frac{\alpha_1}{\delta_p^2+\tau_2^{-2}}+\frac{2\alpha_2}{\delta_s^2+\tau_1^{-2}}\right]\right\}-\frac{1}{3}\left(\frac{1}{T_2}-\frac{1}{T_3}\right)$$

$$\Gamma_4=\frac{1}{3}\left(\frac{1}{T_3}+\frac{2}{T_2}\right)+\frac{4\,|\,\beta_p\,|^2}{\delta_s^2+\tau_1^{-2}}\left\{\tau_1^{-1}+|\,\beta_p\,|^2\left[\frac{-\alpha_2}{2(\delta_p^2+\tau_2^{-2})}+\frac{\alpha_3}{\delta_s^2+\tau_1^{-2}}\right]\right\}$$

The equations become

$$\frac{1}{3}\left(\frac{2}{T_1}+\frac{1}{T_3}\right)\Delta_{13}^0+\frac{1}{3}\left(\frac{1}{T_1}-\frac{1}{T_3}\right)\Delta_{32}^0=\Gamma_1\Delta_{13}-\Gamma_2\Delta_{32}$$

$$\frac{1}{3}\left(\frac{1}{T_3}+\frac{2}{T_2}\right)\Delta_{32}^0+\frac{1}{3}\left(\frac{1}{T_2}-\frac{1}{T_3}\right)\Delta_{13}^0=-\Gamma_3\Delta_{13}+\Gamma_4\Delta_{32}$$

with the solutions

$$\Delta_{13}=\frac{\Gamma_4\left[\frac{1}{3}\left(\frac{2}{T_1}+\frac{1}{T_3}\right)\Delta_{13}^0+\frac{1}{3}\left(\frac{1}{T_1}-\frac{1}{T_3}\right)\Delta_{32}^0\right]+\Gamma_2\left[\frac{1}{3}\left(\frac{1}{T_3}+\frac{2}{T_2}\right)\Delta_{32}^0+\frac{1}{3}\left(\frac{1}{T_2}-\frac{1}{T_3}\right)\Delta_{13}^0\right]}{\Gamma_1\Gamma_4-\Gamma_2\Gamma_3}$$

$$\Delta_{32}=\frac{\Gamma_1\left[\frac{1}{3}\left(\frac{1}{T_3}+\frac{2}{T_2}\right)\Delta_{32}^0+\frac{1}{3}\left(\frac{1}{T_2}-\frac{1}{T_3}\right)\Delta_{13}^0\right]+\Gamma_3\left[\frac{1}{3}\left(\frac{2}{T_1}+\frac{1}{T_3}\right)\Delta_{13}^0+\frac{1}{3}\left(\frac{1}{T_1}-\frac{1}{T_3}\right)\Delta_{32}^0\right]}{\Gamma_1\Gamma_4-\Gamma_2\Gamma_3}$$

The polarization of the system can now be determined from the expectation value of the dipole operators by:

$$\langle P \rangle = \mathrm{Tr}(\mu\rho)$$
$$= 2\mathrm{Re}(\mu_{31}\rho_{13}) + \alpha\mathrm{Re}(\mu_{32}\rho_{23})$$

which, after substituting for ρ_{13} and ρ_{32} becomes

$$P = 2\mathrm{Re}\left[\frac{-|\mu_{13}|^2}{2hL_s^*}\left\{\Delta_{13} - |\beta_s|^2 R\left[\frac{\Delta_{13}}{L_p} + \frac{\Delta_{32}}{L_s}\right]\right\}E_p e^{i(\omega_p t+\psi_p)}\right]$$
$$+ 2\mathrm{Re}\left[\frac{-|\mu_{23}|^2}{2hL_s^*}\left\{\Delta_{32} + |\beta_p|^2 R^*\left[\frac{\Delta_{13}}{L_p^*} + \frac{\Delta_{32}}{L_s^*}\right]\right\}E_s e^{i(\omega_s t+\psi_s)}\right]$$

By comparing this equation with

$$P + \mathrm{Re}\left(\chi_p E_p e^{i(\omega_p t+\psi_p)}\right) + \mathrm{Re}\left(\chi_s E_s e^{i(\omega_s t+\psi_s)}\right)$$

the complex susceptibilities are found to be given by:

$$\chi_p = \frac{-|\mu_{13}|^2}{hL_p}\left\{\Delta_{13} - |\beta_s|^2 R\left[\frac{\Delta_{13}}{L_p} + \frac{\Delta_{32}}{L_s}\right]\right\}$$

and

$$\chi_s = \frac{+|\mu_{23}|^2}{hL_s^*}\left\{\Delta_{32} + |\beta_p|^2 R^*\left[\frac{\Delta_{13}}{L_p} + \frac{\Delta_{32}}{L_s}\right]\right\}$$

In their complex forms

$\chi_p = \chi_p' + i\chi_p' \quad \chi_s = \chi_s' + i\chi_s'$

$$\chi_p'' = \frac{|\mu_{13}|^2}{h(\delta_p^2+\tau_2^{-2})}\left\{\Delta_{13}\left[\tau_2^{-1} - \frac{|\beta_s|^2\alpha_1}{(\delta_p^2 + \tau_2^{-2})}\right] + \Delta_{32}\frac{|\beta_s|^2\alpha_2}{(\delta_s^2 + \tau_1^{-2})}\right\}$$

$$\chi_s'' = \frac{|\mu_{32}|^2}{h(\delta_s^2+\tau_1^{-2})}\left\{\Delta_{32}\left[\tau_1^{-1} + \frac{|\beta_p|^2\alpha_3}{(\delta_s^2 + \tau_1^{-2})}\right] + \Delta_{13}\frac{|\beta_p|^2\alpha_2}{(\delta_p^2 + \tau_2^{-2})}\right\}$$

$$\chi_p' = \frac{|\mu_{13}|^2}{h(\delta_p^2 + \tau_2^{-2})}\left\{\Delta_{13}\left[\delta_p - \frac{|\beta_s|^2\gamma_1}{(\delta_p^2 + \tau_2^{-2})}\right] - \Delta_{32}\frac{|\beta_s|^2\gamma_2}{(\delta_s^2 + \tau_1^{-2})}\right\}$$

$$\chi_s' = \frac{|\mu_{32}|^2}{h(\delta_s^2 + \tau_1^{-2})}\left\{\Delta_{32}\left[\delta_s - \frac{|\beta_p|^2\gamma_3}{(\delta_s^2 + \tau_1^{-2})}\right] - \Delta_{13}\frac{|\beta_p|^2\gamma_2}{(\delta_p^2 + \tau_2^{-2})}\right\}$$

where

$$\gamma_1 = (\delta_p^2 - \tau_2^{-2})R_1 + 2\tau_2^{-1}\delta_p R_2$$

$$\gamma_2 = (\delta_p\delta_s + \tau_1^{-1}\tau_2^{-1})R_1 + (\delta_s\tau_2^{-1} - \delta_p\tau_1^{-1})R_2$$

$$\gamma_3 = (\delta_p^2 - \tau_1^{-2})R_1 - 2\tau_1^{-1}\delta_s R_2$$

These susceptibilities are the first results which allow for completely arbitrary relaxation and dephasing rates as well as arbitrarily strong pump and signal fields.

References

(1) R. L. Panock and R. J. Temkin, ***Interaction of two laser fields with a three level molecular system***, IEEE Journal of Quantum Electronics, QE-13(6), p. 425-434, 1977.

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