Nonlinear Pump Induced Effects in a Nondegenarated Three level System

by

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Abstract

Using a density matrix approach we calculated the nonlinear susceptibility of a nondegenerated three level system. This is a semiclassical approach were the atomic system is quantized and the electromagnetic field is treated classically. The nonlinear susceptibility shows similar behavior as a function of pump intensity as the linear absorption and refraction in the electromagnetically induced transparency effect. The physics studied here can easily be applied to Erbium doped amplifiers were the nonlinear susceptibility seen by the signal field is modified by the presence of the pump laser.

Introduction

The increasingly demanding application of Erbium-doped fiber amplifiers (EDFA) in fiber optic communication systems (including solitons), requires a deep understanding of the effects of the pump laser on the optical properties of the amplifier at the signal level. Amplification provided by EDFAs is produced by optically pumping an absorption band to provide signal gain in the emission band. Promoting carriers to upper energy levels modifies the optical properties of the amplifier. There has been few studies on the pump induce refractive index change at the signal level in EDFAs [1-3] -- all focused on the pump-induced effects on the linear index of refraction, and none on the nonlinear index of refraction n_2.

The modification of the optical properties of a material in the presence of a resonant pump field is widely familiar. In the linear regime a transition in which the lower level is more heavily populated than the upper level exhibits absorption of radiation whose frequency is close to the energy gap of the material. For low power the level population of the lower state is changed little, and the absorption coefficient is independent of the radiation intensity. Associated with the absorption profile, there is a dispersion profile (variation of the refractive index of the medium with frequency), this two are related through deep principles of causality.

As the radiation field intensity is progressively increased, a point is reached where the excitation process begins to dominated the decay process and a significant population begins to build in the upper state with the consequent reduction in the absorption, this regime is called saturation of the absorption. If the intensity is sufficiently high, a point can be reached such that the population in the upper level will approach the population in lower level, the absorption will approach zero and the absorption is said to be bleached. However, if some additional excitation process of the upper state is brought into play, then it becomes possible to have more atoms in the upper level than the lower state. This is the key point of electromagnetically induced transparency (EIT)[4]. If under the correct conditions a second laser is applied to couple the upper level to third transition, then the absorption can be switched off in a situation were the population of the lower level remain essentially unchanged. Secondly the effect occurs for any intensity level of the radiation field coupling the two lower levels.

The same process is predicted here for the nonlinear regime. It was shown that in the nonlinear regime a connection of nonlinear absorption with the nonlinear refraction could be accomplished through a dispersion formula similar to the Kramers-Krönig integral based on the degenerate two-photon absorption spectrum [5]. We will show that in our model the nonlinear refraction can be switched off for any value of the intensity of the signal field.

Semiclassical Theory for $\chi^{(3)}$ in a Quasi-Three Level System

The most general system is shown in Fig. 1.0. We will assume that there is no connection between level $|4\rangle$ and $|2\rangle$, and the total electric field in the system is a linear combination of the pump, probe, and signal field (*p*, *pr*, *s*).

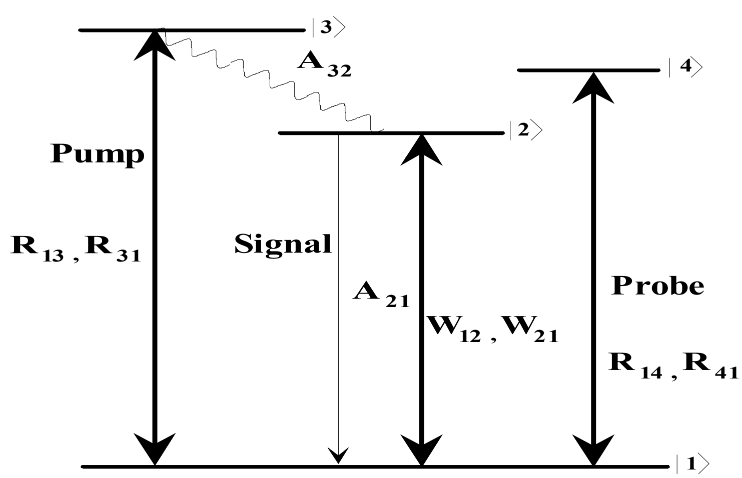

Figure 1.0 Energy State in the quasi-three level system for the study of the nonlinear susceptibility $\chi^{(3)}$ when there is a pump laser present in the system.

Let the transverse electric field in the system be

$$E(z,t) = \left[\hat{e}_p E_p \exp(-i\omega_p t + ik_p z) + \hat{e}_{pr} E_{pr} \exp(-i\omega_{pr} t + ik_{pr} z)\right] + \hat{e}_s E_s \exp\left(-i\omega_s t + ik_s z\right) + cc \tag{1}$$

In Eq. (1), E_j, ω_j, and k_j (j=p, pr, s) are the electric field, angular frequency, and wave vector for the pump, probe, and signal light respectively. The total Hamiltonian of the system is given by

$$\begin{aligned} H = \sum_{n=1}^{4} \hbar\omega_n |n\rangle\langle n| - \mu_p \cdot E_p \left(|3\rangle\langle 1| + |1\rangle\langle 3|\right) - \mu_{pr} \cdot E_{pr} \left(|4\rangle\langle 1| + |1\rangle\langle 4|\right) \\ - \mu_s \cdot E_s \left(|2\rangle\langle 1| + |1\rangle\langle 2|\right) \end{aligned} \tag{2}$$

where $\varepsilon_n = \hbar\omega_n$ is the eigenenergy of state $|n\rangle$ (n=1,2,3,4). In the absence of a driving field, the density matrix tends to evolved in time as $\exp(-i\omega_{nm}t)$. Thus, the term of the electric field that oscillated with this frequency is a more effective driving term. Therefore, we can drop the positive frequency driving field (this is called the rotating wave approximation [6]). Using Eq. (2) to calculate the equation of motion of the density operators we find after some algebra, that

$$\begin{aligned} \frac{\partial \rho_{11}}{\partial t} &= A_{21}\rho_{22} - R_{13}(\rho_{11} - \rho_{33}) - R_{14}(\rho_{11} - \rho_{44}) - i\Omega_s(\rho_{12} - \rho_{21}) \\ &- i\Omega_p(\rho_{13} - \rho_{31}) - i\Omega_{pr}(\rho_{14} - \rho_{41}) \\ \frac{\partial \rho_{22}}{\partial t} &= A_{32}\rho_{33} - A_{21}\rho_{22} + i\Omega_s(\rho_{21} - \rho_{12}) \\ \frac{\partial \rho_{33}}{\partial t} &= R_{13}(\rho_{11} - \rho_{33}) - A_{32}\rho_{33} + i\Omega_p(\rho_{13} - \rho_{31}) \\ \frac{\partial \rho_{44}}{\partial t} &= R_{14}(\rho_{11} - \rho_{44}) + i\Omega_{pr}(\rho_{14} - \rho_{41}) \end{aligned}$$

$$\begin{aligned}
\frac{\partial \rho_{12}}{\partial t} &= -(\gamma_{12} + i\Delta_{12})\rho_{12} - i\Omega_s(\rho_{22} - \rho_{11}) \\
\frac{\partial \rho_{13}}{\partial t} &= -(\gamma_{13} + i\Delta_{13})\rho_{13} - i\Omega_p(\rho_{33} - \rho_{11}) \\
\frac{\partial \rho_{14}}{\partial t} &= -(\gamma_{14} + i\Delta_{14})\rho_{14} - i\Omega_{pr}(\rho_{44} - \rho_{11})
\end{aligned} \tag{3}$$

In Eq. (3) γ_{nm} represent the dipole diphase relaxation time, Ω_n are the Rabi frequencies for each of the fields and are equal to $\mu_n E_n/\hbar$, and the $\Delta_{nm}=\omega_j - \omega_{nm}$ are the detuning factor from the resonance transition frequencies. In addition, the R_{nm} are the pumping rates for the pump and the probe fields. We have to assume that the probe fields can promote considerable amount of carriers to state $|4\rangle$. The A's are the corresponding relaxation rate for each transition.

The rate equation for the system is the time evolution of the populations of the different levels. They are express in terms of : The pumping rates R_{mn}, the transition probabilities W_{mn}, and the relaxation rates A_{mn}. Where m,n=1,2,3,4 corresponding to the different energy levels. Particles leaving n-level per unit time will contribute negatively to the time evolution of that level (depopulation).

$$\begin{aligned}
\frac{dN_1}{dt} &= R_{13}N_1 + R_{31}N_3 - W_{12}N_1 + W_{21}N_2 + A_{21}N_2 - R_{14}N_1 + R_{41}N_4 \\
\frac{dN_2}{dt} &= W_{12}N_1 - W_{21}N_2 + A_{32}N_3 - A_{21}N_2 \\
\frac{dN_3}{dt} &= R_{13}N_1 - R_{31}N_3 - A_{32}N_3 \\
\frac{dN_4}{dt} &= R_{14}N_1 - R_{41}N_4 \\
N &= N_1 + N_2 + N_3 + N_4
\end{aligned} \tag{4}$$

Solving the above set of equations for the steady state case. Assuming that the relaxation rate is $A_{32}>>A_{21}$, the non-radioactive decay rate A_{32} is dominant over the pumping rate $R=R_{13}=R_{31}$, and that $R^{'}=R_{14}=R_{41}$, we can obtain from Eq. (4)

$$
\begin{aligned}
N_1 &= \frac{(\tau W_{21}+1)}{(\tau W_{21}+1)(k+1)+R\tau+W_{12}\tau} \\
N_2 &= \frac{R\tau+W_{12}\tau}{(\tau W_{21}+1)(k+1)+R\tau+W_{12}\tau} \\
N_3 &= 0 \\
N_4 &= \frac{k(\tau W_{21}+1)}{(\tau W_{21}+1)(k+1)+R\tau+W_{12}\tau}
\end{aligned}
\tag{5}
$$

in Eq. (5) we used $A_{21}=\tau^{-1}$ which is the fluorescence life time, and $k=R'\tau_1$. The population in level $|3\rangle$ is zero because of the predominant non-radioactive decay to level $|2\rangle$. Using the following definitions for the transitions and pumping rates

$$
W_{12}\tau=\frac{1}{1+\eta}\frac{I_s}{I_s^{sat}},\quad W_{21}\tau=\frac{\eta}{1+\eta}\frac{I_s}{I_s^{sat}},\quad R\tau=\frac{I_p}{I_p^{th}},\quad R'\tau_1=\frac{I_{pr}}{I_{pr}^{th}},\quad I_i^{sat,th}=\frac{\hbar\omega_i}{\left[\frac{\sigma_i^{peak}}{1+\delta_i}\right]\tau_i}
$$

$$
\delta_i=\frac{2(\omega_i-\omega_{i1})}{\Delta\omega_{i1}},\quad \sigma_i^{peack}=\frac{\lambda_i^2}{2\pi n^2\tau_i\Delta\omega_{i1}}\; for\, i=2,3,4 \quad \rho_{ij}=\sigma_{ij}\exp(-i\omega_j t)\; for\; j=p,pr,s
$$

in the above relations the $I_{s,p,pr}$ correspond to the signal, pump, and probe intensities, $\Delta\omega_{ij}$ to the FWHM of the homogenously transition such that $\gamma_{12}=\Delta\omega_{12}/2$ is the inverse of the dipole relaxation time. There is thresholds pump power where the system is fully inverted, and a probe and signal saturation power where the system is totally bleached denote by *th,* and *sat.* The ω_{ij} denotes the frequency difference between level *i* and *j*, and we allow for some detuning factor in the transitions.

Solving Eq. (3) for the steady state case we get after some algebra for the non-diagonal elements of the density matrix

$$\begin{aligned}
\sigma_{12} &= \Omega_s \frac{(\sigma_{11} - \sigma_{22})}{\gamma_{12} - i\Delta_{12}} \\
\sigma_{13} &= \Omega_p \frac{(\sigma_{11} - \sigma_{33})}{\gamma_{13} - i\Delta_{13}} \\
\sigma_{14} &= \Omega_{pr} \frac{(\sigma_{11} - \sigma_{44})}{\gamma_{14} - i\Delta_{14}}
\end{aligned} \tag{6}$$

As we pointed [6] in the rotating wave approximation the term with the negative exponent in the electric field is a better driving field, then the total polarization of the system can be expressed as

$$P = \varepsilon_o \{\chi_p \exp(-i\omega_p t) + \chi_s \exp(-i\omega_s t) + \chi_{pr} \exp(-i\omega_{pr} t)\} = Ntr(\mu_{12}\rho_{12} + \mu_{13}\rho_{13} + \mu_{14}\rho_{14})$$

using the above expression we get for the susceptibility for each of the fields

$$\begin{aligned}
\chi_s &= \frac{N\mu_{12}^2}{\hbar} \frac{(\rho_{22} - \rho_{11})}{(\Delta_{12} - i\gamma_{12})} \\
\chi_p &= \frac{N\mu_{13}^2}{\hbar} \frac{(\rho_{33} - \rho_{11})}{(\Delta_{13} - i\gamma_{13})} \\
\chi_{pr} &= \frac{N\mu_{14}^2}{\hbar} \frac{(\rho_{44} - \rho_{11})}{(\Delta_{14} - i\gamma_{14})}
\end{aligned} \tag{7}$$

Where N is the density of the rare earth atoms in the system. Using the rate equations to calculated the population difference, we get for the total susceptibility at the signal field

$$\chi_s(\omega_s) = \frac{nNc\sigma_s^{peak}}{\omega_s} \frac{(\delta_s + i)}{1 + \delta_s{}^2} \frac{\left(\frac{I_p}{I_p^{th}} - 1\right)}{\left(\frac{I_s}{I_s^{sat}} + 1\right)\left(\frac{I_p}{I_p^{th}} + \frac{I_{pr}}{I_{pr}^{th}} + 1\right)} \tag{8}$$

In the Eq. (8) n is the index of refraction, and we have defined the dipole relaxation time to be equal to twice the FWHM of the homogeneously broadened transition, or $\gamma_{12}=\Delta\omega_{12}/2$. The detuning normalized factor is defined as

$$\delta_j = \frac{2\cdot(\omega_j - \omega_{mn})}{\Delta\omega_{mn}} \tag{9}$$

If the intensity of the probe beam in Eq. (8) is neglected, we recover the same result of Desurvire [7]. In that case, there is a resonance enhancement of the refractive index due to the presence of the pump field. On the other hand, if there is no pump field and no probe field, then the susceptibility is the same as the case of a two level system, which is described in [6]. Eq. (8) is the total signal susceptibility. To get the nonlinear susceptibility, we expand Eq. (8) in powers of I_s and retain the linear terms in I_s, that gives us

$$\chi_s(\omega_s) = \frac{nNc\sigma_s^{peak}}{\omega_s}\frac{(\delta_s + i)}{(\delta_s^2+1)}\frac{\left(\frac{I_p}{I_p^{th}}-1\right)}{\left(\frac{I_p}{I_p^{th}}+\frac{I_{pr}}{I_{pr}^{th}}+1\right)}\left(1-\frac{I_s}{I_s^{sat}}+....\right) \tag{10}$$

Using the expression $\chi = \chi^{(1)} + 3\chi^{(3)}\left|\ E^2\right| + ...$, we get for the real part of the third order nonlinear susceptibility

$$\chi_s^{(3)}(\omega_s) = \frac{n^2\varepsilon_o Nc^2\sigma_s^{peak}}{\omega_s \bar{I}_s^{sat}}\frac{\delta_s}{(\delta_s^2+1)}\frac{\left(1-\frac{I_p}{I_p^{th}}\right)}{\left(\frac{I_p}{I_p^{th}}+1\right)^2} \tag{11}$$

The linear susceptibility at the probe field is given by

$$\chi_{pr}(\omega_{pr}) = \frac{nNc\sigma_{pr}^{peak}}{\omega_s} \frac{(\delta_{pr} + i)}{1+\delta_{pr}^2} \frac{\left(\frac{I_p}{I_p^{th}} - 1\right)}{\left(\frac{I_p}{I_p^{th}} + \frac{I_{pr}}{I_{pr}^{th}} + 1\right)} \tag{12}$$

Finally expanding Eq. (12) in term of powers of I_{pr}, and retaining only the linear terms in I_{pr}, we obtain for the nonlinear susceptibility at the probe field frequency as a function of pump power I_p

$$\chi_{pr}^{(3)} = \frac{n^2\varepsilon_o Nc^2\sigma_{pr}^{peak}}{\omega_s \bar{I}_{pr}^{sat}} \frac{\delta_{pr}}{\delta_{pr}^2 + 1} \frac{\left(\frac{I_p}{I_p^{th}} + 2\right)}{\left(\frac{I_p}{I_p^{th}} + 1\right)^2} \tag{13}$$

In Fig. 2 we can see that the nonlinear susceptibility of the signal field becomes zero when the intensity of the pump is equal to the threshold pump intensity for population inversion. This is a nonlinear case of electromagnetically induce transparency [33]. The reason for the vanishing of the nonlinear susceptibility is simple, there is a long lived upper energy level. As the intensity of the pump increases the system will reach a point where the population in the lower level is the same as in the upper level, and the system is bleached. At this point there is a reduction of the absorption with a consequent increase in the refraction through the KK relation. Classically in a two level system this will be the end of the story, but if another energy level comes into play then there is a possibility to invert the population, and the system will show optical gain, which is the case for the laser. However if a second laser is turned on so as to couple the level of the first transition to a third level, then the absorption experienced in the first transition can be switched off. Therefore changing the refractive index. Because the nonlinear refractive index follows the same KK dispersion relations then the same argument can also be applied to this case.

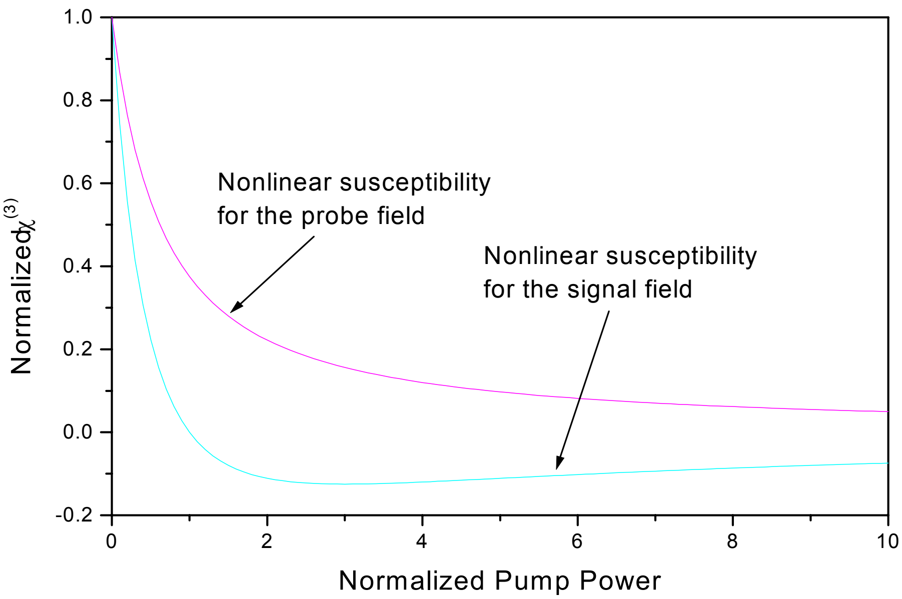

Figure 2. Decrease of the nonlinear susceptibility due to the presence of a pump field, in the case of the probe and the signal field.

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