Until now we studied the modification of absorption and dispersion properties of a (weak) probe light in three level atomic systems (of lambda, ladder or Vee type interaction scheme) due to presence of another strong (pump) light. The phenomenon of electromagnetically induced transparency (EIT) in such systems is seen to originate from a destructive interference between two distinct quantum mechanical paths leading to the excitation of the same upper level. However studies involving modification of EIT and its applications with inclusion of an additional fourth level have also been of considerable interest in recent times and are widely reported in literature. Such studies however are carried out mostly in closed-loop four-level systems of double Lambda (Λ), double ladder or diamond (◊) configuration etc. In this chapter we discuss four wave mixing (FWM) using EIT scheme in a Doppler broadened medium comprising double lambda (Λ) systems with inclusion of buffer gas. The results obtained are shown to be analogous to observed experimental results in more complex and costly setups like cold atomic ensembles or atomic beams.

5.1 Introduction

Suppression of linear absorption and modification of dispersion properties by electromagnetically induced transparency (EIT) also initiated various novel interaction schemes for enhancing nonlinear optical phenomena in absorbing media. One of these interesting nonlinear optical phenomena is the process of four-wave mixing. Harris et al. proposed using EIT to suppress absorption of the short-wavelength light generated in a four-wave mixing (FWM) scheme and showed that the FWM efficiency can be greatly enhanced [1]. Since then various groups [2-6] have studied FWM in four level systems using EIT [7]. Nonlinear optical phenomenon may also be observed at low light intensities approaching single photon energy levels [8]. Using EIT technique it is possible to explore quantum nonlinear optics and quantum information processes [9]. While studying double lambda-Λ systems one usually utilizes the continuous (CW) and pulsed lasers [10-13]. Korsunsky et. al. have studied the phase dependent nonlinear optics in double-lambda system [14]. The time dependent analysis of Ref [4,15] in forward FWM is limited by three photon destructive interference, in an ultraslow-propagation regime in an optically dense medium. Experimental studies of H. Kang et al. [16] in cold atomic
$^{87}$Rb, D1 transition (double-Λ system) using continuous-wave (cw) lasers shows slow light propagation and 10% efficiency for (backward) FWM scheme despite destructive interference of three photon and one photon excitation.

However, EIT and the FWM process in a vapor cell or solids at room temperature is affected by inhomogeneous broadening [13, 17, 18] arising due to thermal motion (in vapors) or local field fluctuations (in solids). For instance thermal motion of atoms in an atomic vapor can cause inhomogeneous broadening of distinct forms viz.(i) the transit time (or time of flight) broadening caused by movement of the atoms across the finite extent of the pump-probe beams in a transverse plane (to the z axis) [19], (ii) residual Doppler broadening owing to wave vector mismatch between pump and probe beams [20] propagating along z axis, and(iii) a small finite angular separation between pump-probe beams [21]. As we have mentioned earlier most studies of EIT and the associated nonlinear processes ignore the finite transit time (or time of flight) broadening that is of the order of $\gamma_t (\approx v_{\text{th}} k_B \approx v_{\text{th}} 2\pi / d)$ where $v_{\text{th}} = (2k_B T / m_A)^{1/2}$ is the most probable thermal velocity of an atom of mass $m_A$ at absolute temperature $T$ and $k_B$ is the Boltzmann constant. Therefore even in a room temperature atomic vapor, the transit time broadening, $\gamma_t ( > 2\pi \times 0.1 \text{ MHz})$ is much greater than the residual Doppler widths (of the order of a few KHz) induced by velocity component along z axis,

In this work we present a detailed theoretical study of EIT and the associated FWM process in a Doppler broadened medium comprising double lambda systems, including the effect of a buffer gas. The theory is more general as it includes various forms of Doppler broadening and other broadening (mentioned above) effects in presence of buffer gas in a vapor cell. In our study we consider cw laser fields (pump, coupler, probe, FWM signal), all the fields are coplanar and probe and FWM signals are weak compared with pump-coupler fields. An interaction scheme is considered in which all the applied field frequencies are on resonance. We utilize density matrix formalism and a strong collision model to incorporate dephasing and velocity changing collisions aspects of buffer gas into the theory.

5.2 Formulation:

We consider a typical four-level double Λ-type system shown in Figure 5.1. A strong laser (called pump) field given by $\vec{E}_c = \vec{E}_c \exp [i(k_c \cdot \vec{r} - \omega_c t)] + \text{c. c.}$, drives the transition $|2\rangle \leftrightarrow |3\rangle$. 

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Double lambda system
and a weak probe laser field, $\vec{E}_p = \varepsilon_p \exp[i(\vec{k}_p \cdot \vec{r} - \omega_pt)] + \text{c.c.}$ acts on the transition $|1\rangle \leftrightarrow |3\rangle$ forming a standard $\Lambda$ type configuration. The spontaneous decay rates from level $|3\rangle$ to levels $|2\rangle$ and $|1\rangle$ are $\gamma_{32}$ and $\gamma_{31}$ respectively. Another coupling (or control) laser field $\vec{E}_c = \varepsilon_c \exp[i(\vec{k}_c \cdot \vec{r} - \omega_ct)] + \text{c.c.}$, drives the transition $|2\rangle \leftrightarrow |4\rangle$ and generates a non degenerate FWM signal field $\vec{E}_e = \varepsilon_e \exp[i(\vec{k}_e \cdot \vec{r} - \omega_et)] + \text{c.c.}$, with a wave vector $\vec{k}_e$ and frequency $\omega_e$. The phase matching condition is given by $k_e^2 = k_p^2 + k_s^2$ where $k_p$, $k_c$, and $k_s$ are the wave vectors of the probe and coupling fields respectively, and $k_s$ is the wave vector of pump field. The spontaneous decay rates of the upper level $|4\rangle$ to levels $|1\rangle$ and $|2\rangle$ are $\gamma_{41}$ and $\gamma_{42}$ respectively. We assume the standard EIT condition $\Omega_p \ll \Omega_e$ and $\Omega_e \ll \Omega_s$ where $\Omega_p$ (= $\mu_{31} \cdot \varepsilon_p / \hbar$), $\Omega_e$ (= $\mu_{41} \cdot \varepsilon_e / \hbar$), $\Omega_c$ (= $\mu_{32} \cdot \varepsilon_c / \hbar$), and $\Omega_s$ (= $\mu_{42} \cdot \varepsilon_s / \hbar$), are the Rabi frequencies of the probe, FWM signal, strong pump and coupler fields.

![Diagram](image.png)

**Fig.5.1:** *EIT and FWM scheme in double ladder configuration. Here $\Omega_p$, $\Omega_e$, $\Omega_c$ and $\Omega_s$ are the Rabi frequencies of the probe, FWM signal, strong pump and coupler fields.*

### 5.2 a) Interaction Hamiltonian:

The Hamiltonian describing interaction of the fields and system under condition of near resonant excitation is obtained in the interaction picture as

$$
\mathcal{V}_{\text{int}} = -\hbar \left[ \Omega_p \exp[i(\vec{k}_p \cdot \vec{r} + \Delta_pt)] S_{31} + \Omega_c \exp[i(\vec{k}_c \cdot \vec{r} + \Delta_ct)] S_{32} \\
+ \Omega_s \exp[i(\vec{k}_s \cdot \vec{r} + \Delta_st)] S_{42} + \Omega_e \exp[i(\vec{k}_e \cdot \vec{r} + \Delta_et)] S_{41} + \text{H.c.} \right],
$$

(5.1)
where $\Delta_p = (\omega_{31} - \omega_p)$, $\Delta_c = (\omega_{32} - \omega_c)$, $\Delta_s = (\omega_{42} - \omega_s)$ and $\Delta_e = (\omega_{41} - \omega_e)$ denote detuning of the probe, pump, coupling and signal field frequencies from atomic resonance frequencies $\omega_{31}, \omega_{32}, \omega_{42}$ and $\omega_{41}$ respectively. $S_{ij} = |i\rangle\langle j|$, ($i, j = 1, 2, 3, 4$) are the atomic raising or lowering operators.

5.2 b) Density matrix formulation:

The equations describing time evolution of the slowly varying components of the density matrix elements $\tilde{\rho}_{ij}(v, t)$ can be obtained using Eqs. (5.1), (1.8b), (1.10), and (1.16) in Eq. (1.11).

Thereafter using appropriate transformations to eliminate fast oscillating (exponential) terms together with the frequency and phase matching conditions $\Delta_e - \Delta_s = \Delta_p - \Delta_c$ and $\vec{k}_e - \vec{k}_s = \vec{k}_p - \vec{k}_c$ for the FWM process, the equations of motion for the slowly varying density matrix elements $\tilde{\rho}_{ij}(v, t)$ can be obtained.

The abovementioned derivation of density matrix elements is simplified considerably if we first consider the physical process underlying EIT and FWM. Analogous to a three level $\Lambda$ case (where only a single strong field is applied) application of two strong cw (pump and coupler) fields result in rapid optical pumping of all the atomic populations into the ground state $|1\rangle$. Thus before application of a probe (or generated) field, initially all the population (to zero order in probe and signal fields), $\rho_{11}^{(0)}(v)$ is in the ground level $|1\rangle$ with a thermal, i.e., Maxwell velocity (3 dimensional) distribution given by

$$\rho_{11}^{(0)}(v) = M(v) = [\ln2/(\pi v^2)]^{3/2} \exp\left(-\ln2 \, \vec{v} \cdot \vec{v}/v^2\right).$$ (5.2)

Here $\vec{v} = \sqrt{\ln2} \, v_{th}$ and $v_{th} = \sqrt{2k_BT/m_A}$ is the most probable velocity at temperature $T$ of an atom of mass $m_A$. If the applied probe and generated signal fields are sufficiently weaker (than pump and coupler), nearly all the population continues to occupy the ground level. Under these conditions the equations of motion for the relevant slowly varying atomic variables are obtained as

$$\dot{\rho}_{21} = -\left\{i\left[(\Delta_p - \Delta_c) + (\vec{k}_p - \vec{k}_c) \cdot \vec{v}\right] + (\gamma_{21} + \Gamma_{12})\right\}\tilde{\rho}_{21} + i\Omega_c^* \tilde{\rho}_{31} + i\Omega_s^* \tilde{\rho}_{41} + \Gamma_{21} M(v) \int \tilde{\rho}_{21}(v', t) d^3v',$$ (5.3a)

$$\dot{\rho}_{31} = -\left\{i(\Delta_p + \vec{k}_p \cdot \vec{v}) + \gamma_p + \frac{\gamma_{31} + \gamma_{32}}{2}\right\}\tilde{\rho}_{31} + i\Omega_p \rho_{11}^{(0)}(v) + i\Omega_c \tilde{\rho}_{21},$$ (5.3b)
\[ \dot{\rho}_{41} = -\left\{ i(\Delta_p + \vec{k}_e \cdot \vec{v}) + \gamma_p + \frac{\gamma_1 + \gamma_2}{2} \right\} \rho_{41} + i\Omega_e \rho_{11}(v) + i\Omega_s \rho_{21}. \]  
(5.3c)

The following transformations were used in deriving the above set of equations of motion (Eq.(5.3)) for the density matrix elements:

\[ \rho_{31} = \rho_{31} e^{i(\vec{k}_p \cdot \vec{r} + \Delta_p t)}, \]  
(5.4a)

\[ \rho_{41} = \rho_{41} e^{i(\vec{k}_e \cdot \vec{r} + \Delta_e t)}, \]  
(5.4b)

\[ \rho_{21} = \rho_{21} e^{i[(\vec{k}_p - \vec{k}_e) \cdot \vec{r} + (\Delta_p - \Delta_e) t]} = \rho_{21} e^{i[(\vec{k}_e - \vec{k}_s) \cdot \vec{r} + (\Delta_e - \Delta_s) t]}. \]  
(5.4c)

The above density matrix equations for slowly varying atomic variables are written in plane-wave approximation for the probe (FWM signal) and pump (coupler) fields. In typical experimental situation a co-propagating geometry is utilized in which all the applied and generated fields are propagating along z direction. Thus the Doppler shift terms in Eq.(5.3) can henceforth be expressed as \( \vec{k}_m \cdot \vec{v} = k_m v_z, \) \( m = p, e \) and \( (\vec{k}_p - \vec{k}_e) \cdot \vec{v} = (k_p - k_e) v_z \).

5.2 c) **Inclusion of transit time broadening:**

The plane-wave approximation is valid and is utilized in most studies of the EIT and slow light performed using sufficiently large, well collimated probe beam diameter. For sufficiently large laser beam diameters \([20]\) the transit time broadening effects arising from finite extent of the laser beam intensity in the transverse plane can be ignored. Eqs.(5.3) can be further generalized to include the finite beam width (in a transverse plane to propagation axis- z direction) of the probe and generated (signal) beam by replacing \( \Omega_p \) and \( \Omega_e \) with space-time dependent probe and signal amplitude \( \Omega_p(\vec{r}, t) \) and \( \Omega_e(\vec{r}, t) \). Consequently including space dependence in the slowly varying quantities \( \rho_{ij}(\vec{r}, t, \nu) \), introducing the Fourier transformation of a function as

\[ f(\vec{r}, t, \nu) = \int_{-\infty}^{+\infty} \frac{d^2 q}{2\pi^2} e^{i\vec{q} \cdot \vec{r}} \int_{-\infty}^{+\infty} \frac{d\omega}{2\pi} e^{-i\omega t} f(q, \omega, \nu), \]  
(5.5)

and using equations (1.10) and (5.5) in Eq (5.3) we get

\[ \left\{ i(\Delta_p - \omega) + i(k_p v_z + \vec{q} \cdot \vec{v}) + \gamma_p + \frac{\gamma_1 + \gamma_2}{2} \right\} \tilde{\rho}_{31}(q, \omega, \nu) = i\Omega_p \rho_{11}^{(0)}(v) + i\Omega_s \rho_{21}(q, \omega, \nu), \]  
(5.6a)
\[ \{i(\Delta_e - \omega) + i(k_e v_z + q_{\perp} \vec{v}_{\perp}) + \gamma_p + \frac{\gamma_1 + \gamma_2}{2}\} \tilde{\rho}_{41}(q_{\perp}, \omega, v) = i\Omega_e \rho_{11}^{(0)}(v) + i\Omega_s \tilde{\rho}_{21}(q_{\perp}, \omega, v), \]  
(5.6b)

\[ \{i[(\Delta_p - \Delta_e) - \omega] + i(k_p - k_e) v_z + i q_{\perp} \vec{v}_{\perp} + (\gamma_2 + \Gamma_1)\} \tilde{\rho}_{21}(q_{\perp}, \omega, v) = i\Omega_c^{*} \tilde{\rho}_{31}(q_{\perp}, \omega, v) + i\Omega_s^{*} \tilde{\rho}_{41}(q_{\perp}, \omega, v) + \Gamma_2 \cdot M(v) \int \tilde{\rho}_{21}(q_{\perp}, \omega, v') d^3v', \]  
(5.6c)

We now proceed to obtain, from the set of Eqs.(5.6), the one-photon coherences from which the characteristics of the probe and signal fields can be determined. For arbitrarily strong pump and coupler fields, we solve the above set of density-matrix equations to leading order in the (Rabi) amplitude of a weak probe, \(\Omega_p\) and generated FWM field, \(\Omega_e\). This is done by first replacing \(\Omega_p\) and \(\Omega_e\) by \(\lambda_p \Omega_p\) and \(\lambda_e \Omega_e\) (where \(\lambda_p\) and \(\lambda_e\) are perturbation expansion parameters) and using the following expansion for the density matrix elements in Eq.(5.6):

\[ \tilde{\rho}_{ij} = \lambda_e \tilde{\rho}_{ij}^{(\Omega_e)} + \lambda_p \tilde{\rho}_{ij}^{(\Omega_p)} + \ldots \ldots \]  
(5.7)

Here \(\tilde{\rho}_{ij}^{(\Omega_e)}\) and \(\tilde{\rho}_{ij}^{(\Omega_p)}\) are the one-photon coherences to first order in the Rabi amplitude \(\Omega_p\) and \(\Omega_e\) respectively, of the probe and signal fields. Using the above procedure we find that the relevant first order density matrix equations are:

\[ A_{41} \tilde{\rho}_{41}^{(\Omega_p)}(q_{\perp}, \omega, v) = i\Omega_s \tilde{\rho}_{21}^{(\Omega_p)}(q_{\perp}, \omega, v), \]  
(5.8a)

\[ A_{31} \tilde{\rho}_{31}^{(\Omega_p)}(q_{\perp}, \omega, v) = i\Omega_p M(v) + i\Omega_c \tilde{\rho}_{41}^{(\Omega_p)}(q_{\perp}, \omega, v), \]  
(5.8b)

\[ A_{21} \tilde{\rho}_{21}^{(\Omega_p)}(q_{\perp}, \omega, v) = i\Omega_c^{*} \tilde{\rho}_{31}^{(\Omega_p)}(q_{\perp}, \omega, v) + i\Omega_s^{*} \tilde{\rho}_{41}^{(\Omega_p)}(q_{\perp}, \omega, v) + \Gamma_2 \cdot M(v) \int \tilde{\rho}_{21}^{(\Omega_p)}(q_{\perp}, \omega, v') d^3v', \]  
(5.8c)

\[ A_{41} \tilde{\rho}_{41}^{(\Omega_e)}(q_{\perp}, \omega, v) = i\Omega_e M(v) + i\Omega_s \tilde{\rho}_{21}^{(\Omega_e)}(q_{\perp}, \omega, v), \]  
(5.8d)

\[ A_{31} \tilde{\rho}_{31}^{(\Omega_e)}(q_{\perp}, \omega, v) = i\Omega_c \tilde{\rho}_{21}^{(\Omega_e)}(q_{\perp}, \omega, v), \]  
(5.8e)

\[ A_{21} \tilde{\rho}_{21}^{(\Omega_e)}(q_{\perp}, \omega, v) = i\Omega_c^{*} \tilde{\rho}_{31}^{(\Omega_e)}(q_{\perp}, \omega, v) + i\Omega_s^{*} \tilde{\rho}_{41}^{(\Omega_e)}(q_{\perp}, \omega, v) + \Gamma_2 \cdot M(v) \int \tilde{\rho}_{21}^{(\Omega_e)}(q_{\perp}, \omega, v') d^3v'. \]  
(5.8f)

Here
Double lambda system

\[ A_{41}(q_\perp, \omega, v) = \left\{ i(\Delta_e - \omega) + i(k_e v_z + q_\perp \vec{v}_\perp) + \left( \gamma_p + \frac{\gamma_{41} + \gamma_{42}}{2} \right) \right\}, \quad (5.9a) \]

\[ A_{31}(q_\perp, \omega, v) = \left\{ i(\Delta_p - \omega) + i(k_p v_z + q_\perp \vec{v}_\perp) + \left( \gamma_p + \frac{\gamma_{31} + \gamma_{32}}{2} \right) \right\}, \quad (5.9b) \]

\[ A_{21}(q_\perp, \omega, v) = \left\{ i[(\Delta_p - \Delta_e) - \omega] + i(k_p - k_e) v_z + i q_\perp \vec{v}_\perp \right\} + \left( \gamma_{21} + \Gamma_{21} \right). \quad (5.9c) \]

Eq. (5.8) can be solved for the coherences, \( \rho_{jk}^s \) in the interaction picture that are related to their counterparts in the Schrodinger picture, \( \rho_{jk}^s \) through the relation \( \rho_{jk}^s = \rho_{jk}^s e^{i\omega t} \). Thus the slowly varying, velocity averaged coherences (to first order in Rabi amplitudes of the probe and signal) in the Schrodinger picture, henceforth denoted \( I_{jk}^{(Qm)} \) for brevity (by dropping the superscript, s) can be obtained as

\[ I_{31}^{(Qp)}(q_\perp, \omega) = e^{-i(k_p, \vec{r} - \omega t)} \int \rho_{31}^{(Qp)}(q_\perp, \omega, v) d^3v \]

\[ = i\Omega_p \left[ \int \frac{(A_{21} A_{41} + |\Omega_2|^2) M(v) d^3v}{A_{31} A_{41} \xi} - \Gamma_{21} |\Omega_2|^2 \right] \frac{\left\{ \int \frac{M(v) d^3v}{A_{31} \xi} \right\}^2}{\left\{ 1 - \Gamma_{21} \int \frac{M(v) d^3v}{\xi} \right\}} \], (5.10a) \]

\[ I_{41}^{(Qe)}(q_\perp, \omega) = e^{-i(k_e, \vec{r} - \omega t)} \int \rho_{41}^{(Qe)}(q_\perp, \omega, v) d^3v \]

\[ = i\Omega_e \left[ \int \frac{(A_{21} A_{31} + |\Omega_3|^2) M(v) d^3v}{A_{31} A_{41} \xi} - \Gamma_{21} |\Omega_3|^2 \right] \frac{\left\{ \int \frac{M(v) d^3v}{A_{31} \xi} \right\}^2}{\left\{ 1 - \Gamma_{21} \int \frac{M(v) d^3v}{\xi} \right\}} \], (5.10b) \]

\[ I_{31}^{(Qe)}(q_\perp, \omega) = e^{-i(k_e, \vec{r} - \omega t)} \int \rho_{31}^{(Qe)}(q_\perp, \omega, v) d^3v \]

\[ = -i\Omega_e \Omega_3 \Omega_e \left[ \int \frac{M(v) d^3v}{A_{31} A_{41} \xi} + \Gamma_{21} \left\{ \int \frac{M(v) d^3v}{A_{31} \xi} \right\} \frac{\left\{ \int \frac{M(v) d^3v}{A_{41} \xi} \right\}}{\left\{ 1 - \Gamma_{21} \int \frac{M(v) d^3v}{\xi} \right\}} \right], (5.11a) \]

\[ I_{41}^{(Qp)}(q_\perp, \omega) = e^{-i(k_e, \vec{r} - \omega t)} \int \rho_{41}^{(Qp)}(q_\perp, \omega, v) d^3v \]

\[ = -i\Omega_p \Omega_3 \Omega_e \left[ \int \frac{M(v) d^3v}{A_{31} A_{41} \xi} + \Gamma_{21} \left\{ \int \frac{M(v) d^3v}{A_{31} \xi} \right\} \frac{\left\{ \int \frac{M(v) d^3v}{A_{41} \xi} \right\}}{\left\{ 1 - \Gamma_{21} \int \frac{M(v) d^3v}{\xi} \right\}} \right]. (5.11b) \]
where
\[ \zeta(q_\perp, \omega, v) = A_{21}(q_\perp, \omega, v) + |\Omega_c|^2/A_{31}(q_\perp, \omega, v) + |\Omega_s|^2/A_{41}(q_\perp, \omega, v). \tag{5.12} \]

An inspection of Eq.(5.10) shows that these coherences are directly proportional to either the probe or signal field Rabi frequency. Thus these are proportional to the linear polarization and hence the real and imaginary parts of Eq.(5.10) describe dispersion and absorption of probe and signal fields in the inhomogeneously broadened double lambda system. On the other hand Eq.(5.11) reveals that these coherences are proportional to the product of the Rabi frequencies of the other three fields, that is, these yield the nonlinear polarization arising due to the frequency mixing of the other three fields.

**5.3 Propagation of the signal and probe waves:**
To calculate the efficiency of the generated FWM signal we consider the propagation of the weak signal and probe waves through an extended medium composed of double (Λ) systems. In chapter 3 we have developed a formalism for propagation of a probe pulse through an EIT medium comprising of three level Λ systems. Although that formalism can be applied here, for the sake of simplicity and clarity, in the present case we consider the applied probe (and therefore the generated signal) pulses to be of very large duration in time, i.e., the probe (and the signal) can be approximated as nearly monochromatic, cw fields. Consequently for this case we can use steady state solutions for the coherences obtained by setting \( \omega = 0 \) in Eqs.(5.10) and (5.11). Propagation of a field of the form given by Eq.(1.3) is described by the Maxwell’s wave equation
\[ (\nabla^2 - \frac{1}{c^2} \frac{\partial^2}{\partial t^2})E_a = \frac{4\pi}{c^2} \frac{\partial^2}{\partial t^2} (P_{a}^{L} + P_{a}^{NL}) , \quad (a = e, p) \tag{5.13} \]
where \( P_{a}^{L} \) and \( P_{a}^{NL} \) respectively, are the macroscopic linear and nonlinear polarizations associated with and aligned along the direction of the field \( \vec{E}_a \). In general the macroscopic polarization \( P \), i.e., the ensemble average of the induced dipole moment per unit volume in a medium of number density \( N \) is given by
\[ P = N \text{ Tr}(\mu < \rho >) = \sum_{i,j,k,l=1}^{4} \mu_{ijk} < \rho_{kij} > = \mu_{13} < \rho_{31} > + \mu_{14} < \rho_{41} > + \text{c.c.}, \tag{5.14} \]
Here angular brackets $\langle \rangle$ denote averaging over thermal velocities $\langle \rho_{ij} \rangle = \int \rho_{ij}(\nu) dv$. The induced macroscopic linear and non-linear polarization is in the medium is proportional to velocity averaged coherence (off diagonal elements) $I_{\Omega e}^{(j)}, I_{\Omega p}^{(j)}$ where $j = 3$ and 4. Thus we can write the polarization in the form (using the notation of Eqs.(5.9) and (5.10)):

$$P_{41} = N\mu_{14} \left( I_{\Omega e}^{(3)} + I_{\Omega p}^{(4)} \right) e^{i(k_{\nu}x - \omega_{e}t)} + \text{c.c.},$$  \hspace{1cm} (5.15a) \\

$$P_{31} = N\mu_{13} \left( I_{\Omega p}^{(3)} + I_{\Omega e}^{(4)} \right) e^{i(k_{p}x - \omega_{p}t)} + \text{c.c.},$$  \hspace{1cm} (5.15b) \\

Substituting Eqs. (5.15a) and (5.15b) along with Eq. (1.3) in Eq. (5.13) and assuming the validity of the slowly-varying amplitude approximation and neglecting diffraction of the beams in the transverse plane, we get the following coupled waves equations for signal and probe field amplitudes:

$$\frac{\partial E_{e}}{\partial z} = \frac{\alpha_{e}}{2} E_{e} + \frac{\beta_{e}}{2} E_{p} ,$$  \hspace{1cm} (5.16a) \\

$$\frac{\partial E_{p}}{\partial z} = \frac{\alpha_{p}}{2} E_{p} + \frac{\beta_{p}}{2} E_{e} .$$  \hspace{1cm} (5.16b) \\

The second term $\beta_{j}$, ($j = s, p$) on the right-hand side of Eqs. (5.16a) and (5.16b) essentially represents mutual coupling between the probe field and the FWM field while the first terms $\alpha_{j}$ ($j = s, p$) describe absorption and dispersion properties of the atomic medium. These are given by

$$\alpha_{e} = i \frac{8\pi^{2}N|\mu_{14}|^{2}}{\lambda_{e}\hbar\gamma_{D}} \left( \frac{I_{\Omega e}^{(3)}}{\Omega_{e}/\gamma_{D}} \right),$$  \hspace{1cm} (5.17a) \\

$$\alpha_{p} = i \frac{8\pi^{2}N|\mu_{13}|^{2}}{\lambda_{p}\hbar\gamma_{D}} \left( \frac{I_{\Omega p}^{(3)}}{\Omega_{p}/\gamma_{D}} \right),$$  \hspace{1cm} (5.17b) \\

$$\beta_{e} = i \frac{8\pi^{2}N|\mu_{13}||\mu_{14}|}{\lambda_{e}\hbar\gamma_{D}} \left( \frac{I_{\Omega p}^{(3)}}{\Omega_{p}/\gamma_{D}} \right),$$  \hspace{1cm} (5.17c)
\[
\beta_p = \frac{8\pi^2 N}{\lambda_p} \frac{\mu_{13}}{\mu_{14}} \frac{|I_{1\Omega_e}(\gamma_e/\gamma_D)}{I_{31}(\Omega_e/\gamma_D)} \cdot \text{(5.17d)}
\]

It should be noted that since \(\alpha_m \propto i l_{1j}^{(\Omega_m)}\), (\(m = e, p\) and \(j = 3, 4\)) the \textit{real} and \textit{imaginary} parts of \(\alpha_e (\alpha_p)\) respectively, describe absorption and dispersion properties of the generated signal (probe) field in the medium. The dispersion vanishes at line center since the imaginary part of \(\alpha_e (\alpha_p)\) is zero at exact resonance. Thus it can be shown and verified numerically that the coefficients \(\alpha_m\) and \(\beta_m\), \((m = e, p)\) given by Eqs. (5.17) are purely real at line center. For this particular case in which all fields are on resonance, together with the boundary condition at the input \(\varepsilon_p(z = 0) = \varepsilon_p(0)\) and \(\varepsilon_e(z = 0) = 0\), we can solve the coupled Eqs.(5.16) to obtain the field amplitudes for the signal, \(\varepsilon_e(z)\) and the probe wave, \(\varepsilon_p(z)\) in the medium as

\[
\frac{\varepsilon_e(z)}{\varepsilon_p(0)} = \frac{\beta_e}{2\Phi} \exp\left\{\left[\frac{\alpha_p + \alpha_e}{2}\right]z\right\} \sinh(\Phi z) \cdot \text{(5.18a)}
\]

\[
\frac{\varepsilon_p(z)}{\varepsilon_p(0)} = \exp\left\{\left[\frac{\alpha_p + \alpha_e}{2}\right]z\right\} \left(\cosh(\Phi z) + \frac{(\alpha_p - \alpha_e)}{4\Phi} \sinh(\Phi z)\right) \cdot \text{(5.18b)}
\]

where \(\Phi = \sqrt{\frac{\beta_e \beta_p}{4 + (\alpha_p/4 - \alpha_e/4)^2}}\cdot \text{(5.18c)}\)

It is also evident that Eq.(5.16b) (and similarly (5.16a)) can be obtained from the probe pulse propagation equation (3.41) developed in Chapter 3 in the steady state limit (time derivatives are zero), assuming negligible (diffraction) variation in probe pulse envelope in the transverse plane compared with that in the z direction \((i \frac{c}{2\omega_p} q_z^2 \ll \frac{\partial}{\partial z})\) and for nonlinear polarization component generated at wave vector \(k_{nl} = k_p\) (or \(k_e\)).

5.4 \textit{Results and discussion:}

We now present numerical results for EIT and propagation effects of the probe and generated FWM signal by applying the theory to a rubidium atomic vapor. The double lambda system can be realized, for example, by excitation of the \(5S_{1/2} \rightarrow 5P_{1/2}\) transition with energy level separation wave length \(\lambda = 795\) nm in a \(^{87}\text{Rb}\) atom using (strong) pump, coupler and (weak) probe laser
Double lambda system

fields. The tuning the field frequencies to the combination of the upper level 5P_{1/2} and hyperfine substructure of the ground state 5S_{1/2} makes the system a four-level double lambda system [7, 8]. In our numerical calculations both drive fields (pump and coupler) are fixed on resonance with their respective transitions. The dephasing rate of the two-photon coherence excited between the two (unlinked) lower states |1> and |2> is around \(\gamma_{21} = 2\pi \times 200\) Hz at an atomic density, \(N = 2 \times 10^{12}\) atoms/cm\(^3\) of the \(^{87}\text{Rb}\) vapor [22]. For a Ne buffer gas (of pressure \(p\) Torr) in the vapor cell, the velocity changing collision parameter is \(\Gamma_{21}/2\pi = p \times 2\) MHz and the collisional dephasing parameter is given by, \(\gamma_p = 2.7 \times \Gamma_{21}\). All parameters are expressed in units of the Doppler width \(\gamma_D/2\pi = 270\) MHz. An optimum value of the velocity changing collision parameter, \(\Gamma_{21} = 0.085\gamma_D\) (which corresponds to a fixed Ne buffer gas pressure, \(p = 11.5\) Torr) is used in all numerical calculations.

The EIT phenomenon for both the probe and generated signal field is illustrated in Fig.5.2 by plotting the absorption characteristics as a function of the two photon detuning \(\delta/\gamma_D\). The pump Rabi frequency is fixed at a value \(\Omega_c/\gamma_D = 0.03\) and the coupler Rabi frequency \((\Omega_s/\gamma_D)\) values are varied through 0, 0.01, 0.03 and 0.05. Curves a and b in Fig.5.2 (i) are the probe absorption profiles (given by the imaginary part of the coherence, \(I_{31}^{(a_p)}\gamma_D/\Omega_p\) ) in the absence of the coupler field i.e., \(\Omega_s/\gamma_D = 0\). Thus curves a and b display respectively, the effect of transit time broadening and buffer gas on the typical absorption profiles of a probe beam in an isolated three level \(\Lambda\) system. From the absorption profile in the absence of the buffer gas (curve a) it is clear that substantial absorption occurs due to the transit time broadening. In contrast, the probe absorption profile in the presence of (11.5 Torr) Ne buffer gas (curve b) shows that velocity changing collisions give rise to perfect EIT at line center by eliminating the transit time broadening. At higher detuning values absorption is lower due to the presence of large collisional dephasing \(\gamma_p\). Nonlinear generation cannot occur in the absence of the coupler field, \(\Omega_s/\gamma_D = 0\).

In Fig. 2(i) [2(ii)] the curves c, d, and e [c', d' and e'] depict the probe (signal) absorption profiles in the presence of (11.5 Torr) Ne buffer gas as the value of the coupler Rabi frequency, \(\Omega_s/\gamma_D\) is varied through 0.01, 0.03 and 0.05. Curves c, d, and e (in Fig.5.2 (i)) reveal that the on-resonance probe absorption increases (or probe EIT diminishes) as one increases the strength of the coupling field amplitude \(\Omega_s/\gamma_D\). Simultaneously curves c', d' and e' (in Fig.5.2(ii)) which
Double lambda system
depict the signal absorption profiles (obtained from imaginary part of \( \Gamma_{41}^{(\Omega_e)} \frac{\gamma_D}{\Omega_e} \)) also show a similar but reverse trend. That is, initially for weak coupling field amplitudes \( \Omega_s/\gamma_D \), signal absorption is very high. As the coupling field amplitude increases, the line center (\( \Delta_e = \Delta_p = 0 \)) signal absorption decreases (or EIT increases), become equal to probe absorption when both the strong driving fields are equal, \( \Omega_c = \Omega_s \) and continues to decrease with further increase in coupling field amplitude. This decrease in EIT of the probe field can be attributed to creation of an additional (third) absorption channel when the coupler field is applied on the \( |2\rangle \rightarrow |4\rangle \) transition, due to which the destructive interference that causes EIT is incomplete.

**Fig.5.2:** EIT of (i) probe and (ii) generated FWM signal as a function of two photon detuning \( \delta/\gamma_D \) for a fixed pump Rabi frequency \( (\Omega_e/\gamma_D) = 0.03 \) and various coupling field Rabi frequencies \( (\Omega_s/\gamma_D) \). (i) Curves \( a \) and \( b \) distinguish the effect of buffer gas collisions on typical EIT characteristics of an isolated lambda system (obtained when coupler field, \( \Omega_s/\gamma_D = 0 \)). Curve \( a \) is the absorption profile in the absence of the buffer gas (\( \Gamma_{21} = 0 \)) whereas curve \( b \) is that in the presence of a Ne buffer gas (of pressure \( p = 11.5 \) Torr) corresponding to a velocity changing collision parameter \( \Gamma_{21} = 0.085 \gamma_D \). Curves \( c, d \) and \( e \) in (i) are the probe absorption profiles and \( c', d' \) and \( e' \) in (ii) are those for the signal field in
the presence of a buffer gas of pressure $p = 11.5$ Torr ($\Gamma_{21} = 0.085\gamma_D$). The coupler Rabi frequencies ($\Omega_s/\gamma_D$) values for $c, c'$ is 0.01, $d, d'$ is 0.03 and $e, e'$ is 0.05. The buffer gas dephasing parameter for Ne is, $\gamma_p = 2.7\Gamma_{21}$.

It is well known that EIT in an (isolated) three level $\Lambda$ system such as, for instance, that formed by the transitions $|1\rangle \rightarrow |3\rangle \rightarrow |2\rangle$ is caused by destructive interference between a one-photon (direct) probe absorption path $|1\rangle \rightarrow |3\rangle$ and another (indirect) three-photon path created via the transitions $|1\rangle \rightarrow |3\rangle \rightarrow |2\rangle \rightarrow |3\rangle$. Now application of a third (coupler) field on the transition $|2\rangle \rightarrow |4\rangle$ (which generates a signal field on the transition $|1\rangle \rightarrow |4\rangle$) in turn creates an additional three-photon $|1\rangle \rightarrow |4\rangle \rightarrow |2\rangle \rightarrow |3\rangle$ path for excitation of the level $|3\rangle$, the strength of which depends upon the coupler field intensity. This can be seen from the $|\Omega_s|^2$ dependent second term in the numerator (of the first term) of Eq.(5.10a). Similar inference can be drawn for the adjacent $\Lambda$ subsystem formed by the generated signal and the coupler field which through the action of the pump field $\Omega_c/\gamma_D$ (see the $|\Omega_c|^2$ dependent second term in the numerator (of the first term) of Eq.(5.10b)) affects the EIT of the signal field. It is clear from the above Figures that when the coupler and pump amplitudes (or intensities) become equal, the minimum EIT possible in each lambda subsystem is only 50% of that for an isolated lambda system.

Furthermore, in Fig.5.3 line center (i.e., on-resonance, $\Delta_p = \Delta_e = 0$) absorption values for probe (curves $p$) and generated FWM signal (curves $e$) are plotted as a function of the coupler field Rabi frequency $\Omega_s/\gamma_D$ for two distinct values of pump field Rabi frequencies, $\Omega_c/\gamma_D = 0.02$ (solid curves) and $\Omega_c/\gamma_D = 0.03$ (dashed curves). Also shown in this figure are the gain factor, obtained from the imaginary part of the nonlinear coherence, $l_{31}^{(\Omega_p)}(\Omega_D/\Omega_p)$ [Eq.(5.11a)] or $l_{41}^{(\Omega_p)}(\Omega_D/\Omega_p)$ [Eq.(5.11b)] as a function of the coupler amplitude $\Omega_s/\gamma_D$ for a fixed pump amplitudes $\Omega_c/\gamma_D$. Since the same parameters $|\mu_{13}| \cong |\mu_{14}|$, $\lambda_e \cong \lambda_p$ and $N$ appear in the expressions for absorption and gain coefficients defined by Eq.(5.17), the absorption $\alpha_p$, $\alpha_e$ and gain coefficients $\beta$ (= $\beta_p = \beta_e$ ) as defined in Eqs.(5.17) can be obtained from the line center absorption and gain factor values depicted on y-axis of Fig.5.3 by multiplying by a factor $\eta (= -\frac{8\pi^2N|\mu_{13}|^2}{\lambda_p\hbar\gamma_D})$. From Fig. 5.3(i) we find that the contributions from gain factor (for nonlinear
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generation $\beta$) and absorption (loss $\alpha$) to the FWM signal (and probe) field are opposite in sign and the magnitudes of these are unequal ($\alpha \neq \beta$) for the pump and coupler fields, $\Omega_c/\gamma_D = \Omega_m/\gamma_D = 0.02$ and 0.03. This is the effect of transit time broadening ($\vec{q}\cdot\vec{r}$) due to which even when the absorption loss in both lambda subsystems becomes equal, $\alpha (= \alpha_p = \alpha_e)$ when $\Omega_c = \Omega_s$, the gain factor (nonlinear generation coefficient $\beta$) is different from the absorption coefficient. Therefore transit time broadening ($\vec{q}\cdot\vec{r}$) has major influence on absorption (loss $\alpha$) and gain factor (generation coefficient $\beta$) of both probe and generated FWM signals.

![Figure 5.3](image)

**Fig.5.3:** (i) and (ii). The effect of buffer gas on absorption and gain factor values at line center (i.e., on-resonance, $\Delta_p = \Delta_e = 0$) for probe (curves $p$) and generated FWM signal (curves $e$) plotted as a function of the coupler field Rabi frequency $\Omega_s/\gamma_D$ for two distinct values of pump field Rabi frequencies, $\Omega_c/\gamma_D = 0.02$ (solid curves) and $\Omega_c/\gamma_D = 0.03$ (dashed curves). The gain factor is obtained from the imaginary part of the nonlinear coherence, $I_{31}^{(n_e)} \gamma_D/\Omega_p$ [Eq.(5.11a)] and is identical in value to that obtained from $I_{41}^{(n_p)} \gamma_D/\Omega_p$ [Eq.(5.11b)]. Since the parameters $|\mu_{13}| \cong |\mu_{14}|$ and $\gamma_e \cong \gamma_p$ the absorption coefficients $\alpha_p$, $\alpha_e$ and nonlinear gain parameter $\beta (= \beta_p = \beta_e)$ as defined in Eqs.(5.17) are obtained.
respectively, by multiplying the line center absorption and gain factor values depicted on y-axis by a factor \( \eta = \frac{8\pi^2 N |\mu_{13}|^2}{\omega_p \hbar \gamma_D} \).

To overcome transit time broadening effect and to prevent the attenuation of probe and generated FWM signal fields in the medium a buffer gas is added to the system. Fig.5.3(ii) shows buffer gas effect on gain factor (nonlinear generation \( \beta \)) and absorption (loss coefficient \( \alpha \)) of probe and FWM signal for pump–coupler fields, \( \Omega_c/\gamma_D = \Omega_m/\gamma_D = 0.02 \), and 0.03. Since other parameters appearing in the expressions Eq.(5.17) for \( \alpha_e \), \( \beta_e \) (and \( \alpha_p \), \( \beta_p \)) are the same, we find that \( \alpha (= \alpha_p = \alpha_e) \) and \( \beta (= \beta_p = \beta_e) \) are nearly equal when \( \Omega_c/\gamma_D = \Omega_m/\gamma_D = 0.03 \) in presence of buffer gas. Thus it is expected that a balance (steady-state) will be established in the situation where the propagation loss determined by absorption coefficient \( \alpha \) is compensated for by the effective (nonlinear) gain given by \( \beta \) in the medium. Whereas Fig.5.3 (i) shows that in absence of buffer gas the balance will not be established, as the propagation loss determined by absorption coefficient \( \alpha \) do not compensate (nonlinear) gain \( \beta \) in the medium.

![Graph showing variation of relative intensity of probe and generated signal as a function of dimensionless parameter \( \alpha z \)](image)

\[ \Omega_c/\gamma_D = \Omega_s/\gamma_D = 0.03 \]

\[ \Gamma_{21}/\gamma_D = 0.085 \]

\[ \Gamma_{21}/\gamma_D = 0 \]

**Fig.5.4:** Variation of the relative intensity of the probe (curves \( p \)) and generated signal (curves \( e \)) as a function of the dimensionless parameter \( \alpha z \) where \( z \) is the propagation distance and...
the value of the absorption coefficient $\alpha$ is evaluated at line center (on-resonance). The pump and coupler Rabi frequencies are, $\Omega_c/\gamma_D = \Omega_s/\gamma_D = 0.03$. For this value of driving field the probe and signal propagate unattenuated through the vapor with matched intensities for $\alpha z \geq 5$ in the presence of buffer gas, $I_{21} = 0.085\gamma_D$ (dashed curves) whereas both the fields are attenuated in the absence of the buffer gas, $I_{21} = 0$ (solid curves). The buffer gas collisional dephasing rate is, $\gamma_p = 2.7\Gamma_{21}$.

We now illustrate how this competition between the absorption $\alpha$ and nonlinear gain $\beta$ affects the propagation characteristics of the probe and signal fields by plotting in Fig.5.4 the relative signal (probe) intensity $\left| \frac{\varepsilon_2(z)}{\varepsilon_p(0)} \right|^2 \left( \frac{\varepsilon_p(z)}{\varepsilon_p(0)} \right)^2$ (using Eq.(5.18)) as a function of $\alpha z$. The absorption coefficient-length product, $\alpha z$ can be varied either by changing the atomic density or length of the vapor cell.

We observe from Fig 5.4 that initially (at the input interface) the probe has maximum value and the signal field is zero. As the fields propagate further in the medium, generation of the signal field via FWM takes place due to which the signal intensity grows at the expense of the probe fields so that the intensity of the probe decreases. However when the generated signal becomes sufficiently strong and comparable to the probe field strength, the reverse process also can occur in which the signal, coupler and the pump via FWM regenerate the probe field. In the course of propagation in the vapor, eventually these two process must reach a balance i.e., a matching of intensity (or amplitude) of the probe and the signal fields should occur. However as seen in Fig.5.2, finite absorption loss ($\alpha$) causes attenuation of the fields as they propagate in the vapor. It is observed from Fig.5.4 that in the absence of the buffer gas (solid curves p and e) matching of the probe and signal field intensities does not occur and the fields are eventually attenuated for $\alpha z \geq 8$.

On the other hand in the presence of a buffer gas (dashed curves p and e) we find that beyond a certain propagation distance characterized by the value of $\alpha z \geq 5$, the generated signal and probe intensities (or amplitudes) are matched and the probe and signal fields propagate through the medium without further dissipation. As seen in Fig. 5.3 the condition $\alpha = \beta$ is satisfied at these values of coupler and pump field Rabi frequencies $\Omega_c/\gamma_D = \Omega_s/\gamma_D = 0.03$. Hence
the propagation loss $\alpha$ is compensated for by the nonlinear gain $\beta$ and the probe and signal fields propagate without any loss in the vapor. We further note that under these matched and no loss conditions the probe and signal amplitude are, $\varepsilon_e(z)/\varepsilon_p(0) = \varepsilon_p(z)/\varepsilon_p(0) = 1/2$ i.e., the signal (and probe) intensity relative to the incident probe intensity is 25% each. The mismatch and attenuation of the signal and probe in the absence of the buffer gas is due to fact that absorption loss $\alpha$ caused by transit time broadening is much larger than the nonlinear gain $\beta$ as observed in Fig 5.3.

5.5 Conclusion:

To summarize, we have studied the buffer gas collisions effect on EIT associated FWM signal generation in an inhomogeneous (Doppler) broadened medium. A strong collision model is considered for velocity changing collisions. Furthermore, under optimal condition of FWM generation the maximum transparency achieved for either the probe or the generated FWM field is only 50% of the one obtains in the case of an isolated 3 level ($\Lambda$) system. The forward FWM scheme is more efficient in a inhomogeneous (Doppler) broadened medium than cooled (homogenous medium) system of Ref [16] provided taking care of major effect of transit time broadening and consideration of buffer gas collisions. The maximum FWM efficiency is obtained with matched coupling fields and buffer gas collisions; at higher propagation lengths the conversion efficiency reaches nearly 25%. Our results are consisted with the earlier experimental studies of Ref [16] (homogenous medium) in forward four mixing scheme.
5.5 References

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