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Saturation in degenerate four-wave mixing: theory for a two-level atom

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Résumé. — Nous présentons dans cet article une théorie des effets de saturation dans le mélange dégénéré à quatre ondes utilisant la méthode de l'atome habillé. Nous étudions essentiellement le cas d'une raie élargie par effet Doppler, mais certains résultats peuvent être appliqués au cas du mélange non dégénéré pour des atomes immobiles. Nous montrons qu'il existe une relation étroite entre ces deux problèmes : la génération dans le milieu est essentiellement produite quand le décalage en fréquence entre deux faisceaux incidents est égal à la fréquence de Rabi. Dans le cas du mélange dégénéré, cette condition est réalisée pour deux classes de vitesse grâce au déplacement Doppler. Nous calculons analytiquement les formes de raie pour des systèmes à deux niveaux en présence d'une onde pompe intense dans le cas de la saturation avant (pompe intense parallèle à l'onde sonde) et arrière (pompe intense et sonde se contre-propageant). Nous étudions également les effets des collisions et l'influence de l'intensité de l'onde pompe faible quand cette intensité devient telle que la théorie des perturbations ne s'applique plus.

Abstract. — We present in this paper a theory of the saturation effects in degenerate four-wave mixing which makes use of the dressed-atom method. We mainly study the case of a Doppler-broadened line, but several results can be applied to the case of nearly degenerate four-wave mixing for a Doppler-free medium. We show that a close relationship does exist between the two problems: the generated beam is mainly created when the detuning between two incident beams is equal to the Rabi frequency. In the degenerate case, this condition is fulfilled by two velocity groups because of the Doppler shift. We analytically calculate the lineshapes for two-level atoms in the case of forward saturation (the intense pump beam propagates in the same direction than the probe beam) and in the case of backward saturation (the intense pump beam and the probe beam propagate in opposite directions). We also study collisional effects and the influence of the intensity of the weak pump beam when perturbation theory can no longer be applied.

Degenerate four-wave mixing has been a subject of considerable interest during the recent years. The possible application of this method to optical phase conjugation [1, 2] is one of the main reasons for this interest. In this context, atomic vapours have been studied for several reasons. First, it has been shown that reflectivities much larger than 100% can be obtained with Na vapour [3-6]. This permits to study several kinds of lasers closed by a sodium vapour phase-conjugate mirror [6-8]. A second interest comes from the application to high resolution spectroscopy. In particular the Doppler-free heterodyne spectroscopy introduced by Ducloy, Bloch and their co-workers [9-11] has been extremely successful. A third reason for the study of atomic vapours comes from the puzzling effect of a saturating laser beam. First theories [12-14] for a Doppler broadened transition were made using perturbation theory and predicted that the intensity of the phase conjugate emission has a Lorentzian dependence with the frequency of the incoming beams. However, it was experimentally shown by Liao et al. [15] that at moderate intensity of the pump beams, a dip appears at resonance. This dip was clearly related to an effect of pump beam saturation. There has been a considerable misunderstanding concerning this dip since a dip was also predicted in the case of homogeneously broadened media [16-18]. The salient importance of the inhomogeneous width was demonstrated by Bloch et al. [19] who showed that the phase-conjugate intensity was different according to the relative direction of propagation of probe and saturating pump beams (the two pump beams having different intensities). This anisotropy effect cannot be interpreted using an...
homogeneous broadening theory and shows the need for a theory that takes into account the Doppler width. The first step in that direction was done by Bloch and Ducloy [20] who derived a complete semiclassical treatment of the effect of one saturating beam on the four-wave mixing lineshape. Their theory was valid for a three-level atom and correctly predicted the anisotropy between the cases of forward and backward saturation (forward corresponds to the situation where the probe beam propagates in the same direction as the saturation pump beam, backward to the opposite situation). This theory was later on extended to the case of two-level atoms [21] and the same conclusion holds. In the meantime, we have proposed another model [22] to understand these lineshapes. This model makes use of the dressed-atom approach [23] which was previously successfully applied to the study of several effects of saturation in the radiofrequency [24, 25] and optical ranges [26-28] and in non-linear optics [29]. The main advantage of the dressed atom model [22] is that it clarifies the underlying physics. In the case of backward saturation which was considered in [22], we have shown that the Doppler effect tunes two velocity groups to a four-wave mixing process which is fully resonant in the atomic frame. In particular, we explain why the phase-conjugate intensity decreases by several order of magnitude when the Rabi frequency $Q_1$ associated to the saturating pump beam becomes larger than the Doppler-width [21, 22]. It should also be stressed that the dressed-atom method permits to have simple calculation in the secular limit ($\Omega_1 \gg \Gamma$, $\Gamma$ being the natural width of the transition).

The aim of this paper is to detail and to extend to other situations the dressed-atom method introduced in [22]. After having introduced (§ 1) our model and our notations, we show that it permits to calculate the intensities of the Rabi sidebands in non degenerate four-wave mixing for an homogeneously broadened medium (§ 2), problem which has been first considered by Harter and Boyd [30, 31]. We show afterwards (§ 3) how to calculate the dispersion and absorption lineshapes for a Doppler-broadened system and we compare our results with those of Baklanov and Chebotayev [32] and Haroche and Hartmann [33]. Following an idea closely related to [34], we deduce from the corresponding formulae the lineshapes in degenerate four-wave mixing both in the cases of backward and forward saturation (§ 4). We study afterwards the effect of collisional damping in the case of backward saturation (§ 5) and we show that the situation is different from the one encountered in the case of the Bloembergen pressure-induced extra-resonances [35-40] (see also [29, 41, 42]) where collisions increase the four-wave mixing generation. Finally, we consider the case where the weak pump beam can also saturate the medium (§ 6) and we show how the lineshape is modified.

1. Description of the model and preliminary calculations

1.1 Atoms and fields. — We consider an atomic vapour where the atoms can be described by two-level systems. The ground state is labelled $|\rangle$ and the excited state $|\rangle$. The resonance frequency is $\omega_0 = (E_+ - E_-)/\hbar$. We consider a closed system: when an atom is excited in the upper level $|\rangle$, it decays towards a lower level $|\rangle$ by emitting a spontaneous photon. The radiative lifetime is $\Gamma^{-1}$. The atoms interact with electromagnetic waves of same frequency $\omega_1$ and same polarizations $\epsilon$. The intense pump beam is $E_1$, the weak incident beams are $E_2$ and $E_3$. These beams propagate in directions $K_1$, $K_2$ and $K_3$. We consider the four-wave mixing generation in directions $(2K_1 - K_2)$ and $(K_1 + K_2 - K_3)$. In the last case, we use the usual phase-conjugate geometry [1, 2]: $K_2 = -K_1$ and we analyse both the backward saturation case $K_3 \approx K_2$ and the forward saturation case $K_3 \approx K_1$ (Fig. 1).

![Fig. 1. Geometry for backward and forward saturation.](image_url)

The electric dipole moment is calculated in the atomic frame. The frequencies of the incident beams are Doppler-shifted. In the two cases considered in figure 1, we have, for an atom of velocity $v_\parallel$ along the $z$ axis, $\omega_1 = \omega_1(1 + v_\parallel/c)$ and $\omega_2 = \omega_2(1 - v_\parallel/c)$. The problem which is degenerate in the laboratory frame appears as non degenerate in the atomic frame. This point makes the whole difference between the theories that assume an homogeneous broadening and those which take into account the Doppler effect.
1.2 HAMILTONIAN. — We make a quantum mechanical description of the intense beam \( E_1 \) and a classical description of the weak beam \( E_2 \). (For the moment, we only consider two beams). We first consider the Hamiltonian \( H_0 \) of the atoms dressed by the photons of the intense beam \( E_1 \) and we shall treat afterwards the coupling with \( E_2 \) as a perturbation.

At the rotating wave approximation, the Hamiltonian of the dressed-atom [23-25] in the laboratory frame is:

\[
H_0 = \omega_0 S_z + \hbar \omega_1 a^+ a - \frac{\hbar d}{\hbar} [S_+ a e^{i \phi} + S_- a^+ e^{-i \phi}] .
\]

(1)

The atomic operators are expressed as function of « pseudo-spin » operators \( S_z, S_+ \) and \( S_- a \) and \( a^+ \) are the annihilation and creation operators of photons in the \( (k_1, \phi) \) mode of the electromagnetic field. \( d \) is the matrix element of the electric dipole moment between levels \( |-\rangle \) and \( |+\rangle \). \( \omega_1 \) is a coupling constant.

We proceed now by making a unitary transformation \( T \) in order to obtain the Hamiltonian in the atomic rest frame. We consider

\[
T = \exp ia^+ (k_1 \cdot r)
\]

(2)

\( T \) depends on time because \( r \) has to be replaced by the atomic motion

\[
r = r_0 + vr .
\]

(3)

The new Hamiltonian is:

\[
H_T = T H_0 T^+ + i\hbar \frac{\partial T}{\partial t} T^+ .
\]

(4)

Using (1), (2) and (3), we obtain:

\[
H_T = \omega_0 S_z + \hbar \omega_1 a^+ a - \frac{\hbar d}{\hbar} [S_+ a + S_- a^+] .
\]

(5)

We note that the frequency of the quantized field is now \( \omega_1 = \omega_k - k_1 \cdot v \) instead of \( \omega_k \). The eigenstate of the field is a Glauber coherent state [43, 44]. In the laboratory frame, this state is labelled \( | \alpha \rangle \) where \( \alpha \) is a complex number \( \alpha = | \alpha | e^{i \phi} \). At time \( t \), the state of the field is:

\[
| \alpha(t) \rangle = e^{-|\alpha|^2 / 2} \sum_n \frac{|\alpha|^n}{\sqrt{n!}} e^{-i(n\omega_1 - \phi)} | n \rangle .
\]

(6)

In the atomic rest frame, the state of the field is transformed into:

\[
| \alpha_T(t) \rangle = T | \alpha(t) \rangle
\]

\[
| \alpha_T(t) \rangle = e^{-|\alpha|^2 / 2} \sum_n \frac{|\alpha|^n}{\sqrt{n!}} e^{-i(n\omega_1 - k_1 \cdot r - \phi)} | n \rangle
\]

(7)

where \( r \) is given by (3).

The mean value of \( a^+ a \) in the Glauber state is \( \bar{n} = | \alpha |^2 \). The Rabi nutation frequency \( \Omega_1 \) is equal to [23]:

\[
\Omega_1 = -2 \frac{\hbar d}{\hbar} \sqrt{\bar{n}} .
\]

(8)

The eigenstates \( | i, n \rangle \) of \( H_T \) are:

\[
| 1, n \rangle = \cos \varphi | +, n \rangle + \sin \varphi | -, n + 1 \rangle
\]

(9a)

\[
| 2, n \rangle = -\sin \varphi | +, n \rangle + \cos \varphi | -, n + 1 \rangle
\]

(9b)

where

\[
\tan 2\varphi = \frac{\Omega_1}{\omega_0 - \omega_1} .
\]

(10)

The eigenvalues \( E_{1,n} \) and \( E_{2,n} \) of \| 1, n \rangle \) and \| 2, n \rangle \) are:

\[
E_{1,n} = \left( n + \frac{1}{2} \right) \hbar \omega_1 + \frac{\hbar \Omega_1}{2}
\]

(11a)

\[
E_{2,n} = \left( n + \frac{1}{2} \right) \hbar \omega_1 - \frac{\hbar \Omega_1}{2}
\]

(11b)

where

\[
\Omega_1 = \left[ \Omega_1^2 + (\omega_0 - \omega_1)^2 \right]^{1/2}
\]

(12)

The energy levels of the dressed-atom thus consist of a ladder of doublets separated by \( \hbar \Omega_1 \). The distance between two levels of the doublet is \( \hbar \Omega \) (Fig. 2). In the following, we shall always assume that \( \Omega \ll \omega_1 \).

Fig. 2. — Energy levels of the dressed-atoms. The energy levels consist of a ladder of doublets separated by \( \hbar \Omega_1 \). The distance between two levels \| 1, n \rangle \) and \| 2, n \rangle \) of the same doublet is \( \hbar \Omega \).

1.3 RADIATIVE RELAXATION. — We now include the relaxation due to spontaneous emission. The master equation for the density matrix \( \sigma \) of the dressed-atom is:

\[
\frac{d\sigma}{dt} = \frac{1}{i\hbar} [H_T, \sigma] + \left\{ \frac{d\sigma}{dt} \right\}_{rel} .
\]

(13)
The matrix elements of $r$ are labelled by $\sigma_{ij}^p$:

$$\sigma_{ij}^p = \langle i, n \mid \sigma \mid j, n - p \rangle.$$  \hfill (14)

The relaxation terms have been derived by Cohen-Tannoudji and Reynaud [26]:

$$\begin{align*}
\left\{ \frac{d}{dt} \sigma_{11n}^p \right\}_{rel} &= - \Gamma \cos^4 \phi \sigma_{11n}^p + \Gamma \sin^4 \phi \sigma_{22n}^p \\
\left\{ \frac{d}{dt} \sigma_{22n}^p \right\}_{rel} &= - \Gamma \sin^4 \phi \sigma_{22n}^p + \Gamma \cos^4 \phi \sigma_{11n}^p \\
\left\{ \frac{d}{dt} \sigma_{12n}^p \right\}_{rel} &= - L \sigma_{12n}^p
\end{align*}$$

(15a, 15b, 15c)

$\Gamma^{-1}$ is the radiative lifetime of the upper level and $L$ is equal to:

$$L = \frac{\Gamma}{2} (1 + 2 \sin^2 \phi \cos^2 \phi).$$  \hfill (16)

If we assume that the initial state of the system is $| - > \otimes | \sigma_r(0) \rangle$ and if we make the secular approximation ($\Gamma \ll \Omega$), we obtain for the stationary solution of (13):

$$\begin{align*}
\Delta_p^s &= \sigma_{22n}^p - \sigma_{11n}^p = p(n) \cos^4 \phi - \sin^4 \phi \\
&\quad \times \left[ \cos^4 \phi + \sin^4 \phi \times e^{-i(p \omega_1 - k \cdot r_0 - \theta_1)} \right] \\
= p(n) & e^{-i(p \omega_1 - k \cdot r_0 - \theta_1)} \sigma_{22n}^p + \sigma_{11n}^p = 0
\end{align*}$$

(17a, 17b, 17c)

In stationary regime, we look for solutions of the type:

$$\begin{align*}
\frac{d}{dt} \Delta_p^s &= - \left[ ip \omega_1 + \Gamma (\cos^4 \phi + \sin^4 \phi) \right] \Delta_p^s \\
&\quad - i \Omega_2 \cos^2 \phi \left[ e^{i(\omega_2 t - k \cdot r_0 - \theta_2)} \sigma_{12n}^p - e^{-i(\omega_2 t - k \cdot r_0 - \theta_2)} \sigma_{21n}^p \right] \\
&\quad + \Gamma (\cos^4 \phi - \sin^4 \phi) p(n) e^{-i(p \omega_1 - k \cdot r_0 - \theta_1)} \\
&\quad + \Gamma (\cos^4 \phi - \sin^4 \phi) p(n) e^{-i(p \omega_1 - k \cdot r_0 - \theta_1)} \sigma_{21n}^p + \sigma_{12n}^p = 0
\end{align*}$$

(20a, 20b, 20c)

In stationary regime, we look for solutions of the type:

$$\begin{align*}
\Delta_p^s &= \Delta p(n) e^{-i(p \omega_1 - k \cdot r_0 - \theta_1)} \\
\sigma_{12n}^p &= \sigma_{12} p(n) e^{-i(p \omega_1 - k \cdot r_0 - \theta_1)} e^{-i(\omega_2 t - k \cdot r_0 - \theta_2)} \\
\sigma_{21n}^p &= \sigma_{21} p(n) e^{-i(p \omega_1 - k \cdot r_0 - \theta_1)} e^{-i(\omega_2 t - k \cdot r_0 - \theta_2)}
\end{align*}$$

(21a, 21b, 21c)

We obtain:

$$\begin{align*}
\Delta &= \frac{\cos^4 \phi - \sin^4 \phi}{\cos^4 \phi + \sin^4 \phi} \times \frac{1}{1 + \frac{\Omega_2^2 \cos^4 \phi}{\Gamma (\cos^4 \phi + \sin^4 \phi) \frac{L}{L^2 + (\omega_1 - \omega_2 + \Omega)^2}}}
\end{align*}$$

(22a)

where $p(n) = e^{-|n|^2/2} |n|^2/n!$. We remark that the coherences between levels of the same multiplet are equal to 0 at the secular approximation.

1.4 INTRODUCTION OF THE WEAK FIELD. — We now introduce the weak field $E_2$. Its magnitude is assumed to be much smaller than $|E_1|$. It implies that the energy levels (11) remain almost unchanged when $E_2$ is applied. In particular, we emphasize that our treatment cannot describe the standing wave situation where $|E_1| = |E_2|$. We treat $E_2$ as a classical field. At the rotating wave approximation, the Hamiltonian becomes:

$$H = H_T + V$$

(18)

where

$$V = \frac{\hbar \Omega_2}{2} \left[ e^{-i(\omega_2 t - k \cdot r_0 - \theta_2)} S_+ + e^{i(\omega_2 t - k \cdot r_0 - \theta_2)} S_- \right]$$

(19)

where $\omega_2$ is the frequency of wave $E_2$ in the atomic rest frame and $\Omega_2 = -d |E_2|/\hbar$.

In the following, we note that the main effect on the four-wave mixing efficiency comes from the velocity groups for which $\omega_2 = \omega_1 \pm \Omega$ (this point clearly appears on the first example [22] that we have considered). It is thus important to solve the density matrix equation $\sigma$ for $\omega_2 \sim \omega_1 \pm \Omega$. We first consider $\omega_2 \sim \omega_1 + \Omega$. In that case, the problem appears to be similar to a set of two-level systems ($\langle 2, n \rangle$ and $\langle 1, n + 1 \rangle$) interacting with a monochromatic wave of frequency $\omega_2$. The master equation can thus be exactly solved if we neglect the non secular contributions and if we assume that $p(n) \sim p(n - 1)$ ($p(n)$ is a slow function of $n$). In these conditions, the density matrix equation becomes:
A similar system can be established for \( \omega_2 \approx \omega_1 - \Omega \) and the solutions analogous to (22) are obtained by replacing \((\omega_1 - \omega_2 + \Omega)\) by \((\omega_2 - \omega_1 + \Omega)\) and \(\omega_2 \cos^2 \varphi\) by \(-\omega_2 \sin^2 \varphi\). The time dependence associated with \(\sigma_{12}^{(2a)}\) is now \(e^{-i(\varphi + 1)\omega_2 t} e^{i\omega_2 t}\) and reciprocally, we have now \(e^{-i(\varphi - 1)\omega_2 t} e^{-i\omega_2 t}\) for \(\sigma_{21}^{(2a)}\).

1.5 ELECTRIC DIPOLE MOMENT. — The mean value \(< D >\) of the component of the electric dipole moment along the polarization axis \(\varepsilon\) is:

\[
<D> = \text{Tr} \sigma(D \cdot \varepsilon) \quad (23a)
\]

\[
<D> = \sum_{n,p} \sum_{l,j} \sigma_{nm}^{(l)} \langle j, n - p | D \cdot \varepsilon | i, n \rangle . \quad (23b)
\]

Using the wavefunctions (9) and the matrix element \(< + | D \cdot \varepsilon | - > = d\) (which is assumed to be real), we first deduce that \(p\) must be equal to 1 or -1 and we then transform (23b) into

\[
<D> = D_\pm + \text{c.c.} \quad (24a)
\]

\[
D_\pm = \sum_n (-d \sin \varphi \cos \varphi A_\pm^n - d \sin^2 \varphi \sigma_{12}^{(1a)} + d \cos^2 \varphi \sigma_{12}^{(1b)}) . \quad (24b)
\]

Using (21), (22) and (24b), we obtain the value \(D_\pm\) of \(D_\pm\) when \(\omega_2 \sim \omega_1 + \Omega\).

\[
D_\pm = \frac{d(\cos^4 \varphi - \sin^4 \varphi)}{(\cos^4 \varphi + \sin^4 \varphi) + \frac{Q_2^2}{L} \cos^4 \varphi \frac{L}{L^2 + (\omega_1 - \omega_2 + \Omega)^2}} \times \exp[-i\omega_2 t] \times
\]

\[
\times \left[ - \sin \varphi \cos \varphi e^{i(k_1 r + \theta_1)} - \frac{i\omega_2 \cos^4 \varphi e^{i(k_1 r - k_2 r + \theta_2)}}{2[L + i(\omega_1 - \omega_2 + \Omega)]} - \frac{i\omega_2 \cos^2 \varphi \sin^2 \varphi}{2[L - i(\omega_1 - \omega_2 + \Omega)]} e^{i(2k_1 - k_2) r + \theta_1 - \theta_2} \right] . \quad (25)
\]

We obtain a similar formula for the dipole moment \(D_\pm^r\) around the second resonance \(\omega_2 \sim \omega_1 - \Omega\):

\[
D_\pm^r = \frac{d(\cos^4 \varphi - \sin^4 \varphi)}{(\cos^4 \varphi + \sin^4 \varphi) + \frac{Q_2^2}{L^2} \cos^4 \varphi \frac{L}{L^2 + (\omega_1 - \omega_2 - \Omega)^2}} \times \exp[-i\omega_2 t] \times
\]

\[
\times \left[ - \sin \varphi \cos \varphi e^{i(k_1 r + \theta_1)} + \frac{i\omega_2 \sin^4 \varphi}{2[L + i(\omega_1 - \omega_2 - \Omega)]} e^{i(k_1 r - k_2 r + \theta_2)} + \frac{i\omega_2 \sin^2 \varphi \cos^2 \varphi}{2[L - i(\omega_1 - \omega_2 - \Omega)]} e^{i(2k_1 - k_2) r + \theta_1 - \theta_2} \right] . \quad (26)
\]

1.6 PERTURBATIVE EXPANSION. — In the following, we shall often use a perturbative expansion (in power of \(Q_2/r\)) rather than the exact expressions (25) and (26) of \(D_\pm\). At second order in \(\Omega_2/r\), we obtain when adding the contribution of (25) and (26)

\[
D_\pm = D_\pm^{(0)} + D_\pm^{(1a)} + D_\pm^{(1b)} + D_\pm^{(2)} \quad (27a)
\]

\[
D_\pm = \left( D_\pm^{(0)} + D_\pm^{(1a)} + D_\pm^{(1b)} + D_\pm^{(2)} \right) e^{-i\omega_2 t} \quad (27b)
\]

where the upper indice corresponds to the power in \(\Omega_2/r\). Some care should be taken with the 0th order term \(D_\pm^{(0)}\) which is obviously not resonant for \(\omega_2 = \omega_1 \pm \Omega\) and which must be taken only once. The two first order terms \(D_\pm^{(1a)}\) and \(D_\pm^{(1b)}\) have been separated according to their different phase dependence. Finally, we recall that the following expressions of \(D_\pm^{(1a)}, D_\pm^{(1b)}\) and \(D_\pm^{(2)}\) are valid around the resonances \(\omega_1 - \omega_2 \sim \pm \Omega\) when \(\Omega \gg r\).
We show now how these expressions can be applied to several four-wave mixing problems.

2. Rabi sidebands in non degenerate four wave mixing in a Doppler-free medium.

Our paper is mainly devoted to the study of degenerate four-wave mixing in the Doppler limit (ku > \Omega). However the preceding theory can also be used to interpret what occurs in non degenerate four-wave mixing in a Doppler-free medium. This situation has been extensively investigated by Harter and Boyd [30, 31] who have shown that a strong enhancement of the four-wave mixing signal occurs when \( W_1 - W_2 = \pm \Omega \). As we shall see thereafter, a close link does exist between these two problems.

The formulae (26) (27) and (28) have been written for the degenerate case in the laboratory frame. If we want to study non-degenerate four-wave mixing in a Doppler-free medium, we have to use the preceding formulae expressed in the atomic frame: we change \((W_1 t - k_1 \cdot r)\) into \((W_1 t - k_1 \cdot r)\) and \((W_2 t - k_2 \cdot r)\) into \((W_2 t - k_2 \cdot r)\). In this section \( \omega_1 \) and \( \omega_2 \) are two independent frequencies coming from two different sources.

We first consider the case of backward saturation (Fig. 1a). The intense pump beam \( E_1 \) has a frequency \( \omega_1 \) which is kept fixed. The weak beam is divided into two beams \( E_2 \) and \( E_3 \) of wavevectors \( k_2 \) and \( k_3 \) almost parallel (\( k_2 \sim k_3 \)). Then \( \Omega_2 \) becomes

\[
\Omega_2 = \frac{\Omega_1^2}{L} \exp\left(i(k_1 t + \theta_1) - (k_2 t - \theta_2 + \theta_3)\right) \times \left\{ \frac{L \sin^4 \varphi}{L^2 + (\omega_1 - \omega_2 - \Omega)^2} + \frac{L \cos^4 \varphi}{L^2 + (\omega_1 + \omega_2 + \Omega)^2} \right\}.
\]

When we insert this value in (28d), we obtain a component \( D^{(2b)} \) of the electric dipole moment \( D^{(2)} \) which radiate in the \( (k_1 + k_2 - k_3) \) direction, the generated beam being the phase conjugate of the \( E_3 \) beam.

Usually, we take opposite pump beams \( (k_1 + k_2) \approx 0 \) and the generation of the fourth beam is almost phase-matched. When \( \omega_1 \) is kept fixed and \( \omega_2 \) varies, two resonances located on the Rabi sidebands

\[
\omega_1 - \omega_2 = \pm \Omega = \pm \left[ \Omega_1^2 + (\omega_0 - \omega_1)^2 \right]^{1/2}
\]

should be observed. For thin optical media, the half-width of these resonances is:

\[
L = \frac{\Gamma}{2} \left[ \frac{\Omega_1^2}{2 \Omega^2} \right]
\]

and the ratio of their intensities

\[
(tan^4 \varphi)^2 = \left[ \frac{\Omega}{\Omega + (\omega_0 - \omega_1)} \right]^4.
\]

It can also be noticed that when the intense beam is resonant \( (\omega_1 = \omega_0) \), the Rabi sidebands disappear \( (cos^4 \varphi = tan^4 \varphi) \) because the levels of the dressed atom are almost equally populated at the secular approximation \( (L \ll \Omega) \).

Other four-wave mixing processes in Doppler-free media can be described using formulae (26)-(28).
When \( E_1 \) and \( E_2 \) propagate in almost parallel directions, a beam is generated in the forward direction \((2k_1 - k_2)\). (This process is associated with the absorption of two photons of beam \( E_1 \) and amplification of one photon of beam \( E_2 \)). In the case where phase-matching problems can be omitted, the properties of the resonances are deduced from (28c) in the perturbative limit \((Q_2 \ll T)\). We note that, contrary to the preceding situation, the two Rabi sidebands have the same intensity. Other features (width, cancellation when \( \omega_1 = \omega_0 \)) are similar.

For higher values of \( \Omega_2 \) \((\Gamma < \Omega_2 \ll \Omega_1)\), the particular properties of each resonance can be deduced from (25) and (26).

### 3. Mean value of the dipole moment in the Doppler limit.

We now consider the case of Doppler-broadened systems. We assume that \( \Omega_1 \ll ku \) and \( \Omega_2 < \Gamma \). The first condition implies that the wave functions of the dressed-atom vary on a frequency range small compared to the Doppler width. The second condition permits to apply the formulae (27) and (28) valid at low intensity of the weak field. The same assumptions will be made in sections 4 and 5.

#### 3.1 AVERAGING OVER THE VELOCITY DISTRIBUTION.

The intense pump beam \( E_1 \) propagates in the \(-k\) direction. Because of the Doppler shift, the actual frequency in the atomic rest frame is \( \omega_1 = \omega_1(1 + v_1/c) \). We shall consider the cases where the weak field \( E_2 \) (which has the same frequency \( \omega_0 \) in the laboratory frame) propagates either in an almost parallel direction \((k_2 \sim -k)\) or in the opposite direction \((k_2 = k)\). In the first case, we have \( \omega_2 \approx \omega_1 \) while in the second case \( \omega_2 \approx \omega_1 = -2\omega_0 v_1/c \).

The dependence upon \( v_1 \) in formula (28) can be replaced by a dependence upon \( \varphi \) because of the definition of \( \tan 2 \varphi \). If we replace \( \omega_1 \) by \( \omega_1(1 + v_1/c) \) and we call \( \delta \) the actual detuning from the resonance \( \delta = \omega_0 - \omega_1 \), we have:

\[
\tan 2 \varphi = \frac{\Omega_1}{\delta - kv_2}.
\]  

We shall now integrate formulae (28) giving the electric dipole moment \( \tilde{D}_+(\delta, v_2) \) over the velocity distribution.

\[
\tilde{D}_+^{(1)}(\delta, v_2) \propto \frac{\Omega_2}{4\sqrt{\pi} ku} \frac{d e^{i(k_2r + \theta_2)}}{d s} \frac{1}{1 + s^2} \frac{1}{1 + s^4} \times
\]

\[
\left[ \frac{s^4}{(1 + s^2)^2} \right] \exp \left( \frac{-s^2}{4\delta - \frac{1 - s^2}{2} - \frac{1}{s}} \right) \]

\[
\exp \left( \frac{-s^2}{4\delta - \frac{1 - s^2}{2} + s} \right)\]  

where

\[
e(s) = \frac{\Gamma}{\Omega_1} \left[ \frac{1}{2} + \frac{s^2}{(1 + s^2)^2} \right]
\]
and

\[ \bar{\delta} = \frac{\delta}{\Omega_1} = \frac{\omega_0 - \omega_L}{\Omega_1}. \]  

(42)

Using \( s' = -1/s \) in the first part of (40), we transform (40) into :

\[ \tilde{D}^{(1a)}(\bar{\delta}) = \frac{\Omega_2}{2\sqrt{\pi} \kappa u} \int_0^{+\infty} ds \frac{1-s^2}{s} \frac{s^4}{1+s^4} \frac{1}{\left(1-4\bar{\delta}s - 3s^2 + 2i\varepsilon(s)s\right)}. \]  

(43)

Since the two poles of \( (1 - 4\bar{\delta}s - 3s^2 + 2i\varepsilon(s)s) \) are in the upper half plane, it is easy to integrate (43) using a contour integration in the lower half plane. We obtain using the residue theorem :

\[ \tilde{D}^{(1a)}(\bar{\delta}) = -\frac{\sqrt{\pi}}{2} \frac{\Omega_2}{k_u} e^{i(k_0+\theta_0)} \left[ \frac{2\sqrt{2}}{64\Delta^2 + 48\bar{\delta}^2 + 25} \right]. \]  

(44)

Formula (44) describes the saturated dispersion and saturated absorption lineshapes at secular limit \( (\Omega_1 \gg \Gamma) \). The predicted behaviour is in agreement with preceding calculations performed with the classical Bloch equations [32, 33]. For instance (44) shows that at resonance \( \bar{\delta} = 0 \), the absorption of the weak beam is equal to 0.6 times its usual value obtained for large detunings or in absence of the pump beam. This result is related to the fact that at resonance the weak beam interacts with velocity groups which are not fully saturated because the levels of the dressed-atom are shifted by the dynamic Stark effect [48].

3.4 Dispersion and Absorption of the Intense Pump Beam. — In contrast with the previous situation, the modification of the strong beam due to its interaction with the weak beam has not been extensively studied. The reason for that is obvious when the mere purpose is saturation or dispersion spectroscopy. However, we will show that the knowledge of the dispersion and absorption of the pump beam is an important step to calculate four-wave mixing signals in the case of backward saturation.

The components of the dipole moment, the phase of which vary like the phase of \( E_1 \), are \( D^{(0)} \) and \( D^{(2)} \) (28a and 28d). However we only keep here the second term (28d) which depends on the weak beam. In the infinite Doppler width limit, we obtain using (28d), (38), (41) and (42) :

\[ \tilde{D}^{(2a)}(\bar{\delta}) = \frac{\Omega_2^2}{2\sqrt{\pi} \Gamma k u} \int_0^{+\infty} ds \frac{1-s^2}{s} \frac{s^4}{1+s^4} \left[ \frac{\varepsilon(s)}{\varepsilon^2(s)} + \frac{1}{2} \left( \frac{\bar{\delta}^2 - \frac{1-s^2}{2s} - \frac{1}{s}}{2} \right) + \frac{\varepsilon(s)}{\varepsilon^2(s)} \right]. \]  

(45)

A simple method to integrate (45) is to replace the resonance curves \( \varepsilon(s)/[\varepsilon^2(s) + f^2(s)] \) by Dirac \( \delta \) functions \( \pi \delta[f(s)] \). Such an approach is valid when the width of the resonance curve is much smaller than the variation of the other functions, i.e. when \( \Gamma \ll \Omega_1 \) (secular limit). The roots \( s_1 \) and \( s_2 \) for the first and second denominators of (45) are :

\[ s_1 = \sqrt{4\bar{\delta}^2 + 3 - 2\bar{\delta}} \]  

\[ s_2 = \frac{1}{3}\sqrt{4\bar{\delta}^2 + 3} - \frac{2}{3}\bar{\delta} = \frac{s_1}{3}. \]  

(46a)  

(46b)

The relation \( s_2 = s_1/3 \) permits to gather the two terms of (45) and leads to :

\[ \tilde{D}^{(2a)}(\bar{\delta}) = 2\sqrt{\pi} \frac{\Omega_2^2}{\Gamma k u} e^{i(k_0+\theta_1)} \left( 8\bar{\delta}^2 + 3 \right) \frac{64\bar{\delta}^4 + 48\bar{\delta}^2 + 65}{(64\bar{\delta}^4 + 48\bar{\delta}^2 + 25)^2}. \]  

(47)

It can be noticed that \( \tilde{D}^{(2a)} \) has the same phase than \( E_1 \). It means that, at second order in \( \Omega_2/\Gamma \), the dispersion of \( E_1 \) is modified but not its absorption.
4. Degenerate four-wave mixing in the Doppler limit.

4.1 Survey of the Method. — We have treated the case of \((2k_1 - k_2)\) emission in a preceding section (§ 3.2). We consider here the cases of backward and forward saturation (Fig. 1). For both cases, we will deduce the amplitude of the phase-conjugate beam from the formulae of § 3.3 and 3.4. Following an idea of Ducloy and Bloch [34], we consider one of the incident beam \(E_i\) as a superposition of two beams almost parallel \(E_j\) and \(E_3\) which propagate in the \(k_j\) and \(k_3\) directions. As in (29), we replace \(\Omega_2^2\) by a combination of \(\Omega_2\) and \(\Omega_3\) similar to the one already written in (29). The phase conjugate emission comes from a component of the electric dipole moment linear in \(\Omega_3\) and whose phase is conjugate of the phase of beam \(E_3\). Starting from the dipole \(D_+\), the phase conjugate emission is generated by \(D_+^{(c)}\) such as

\[
D_+^{(c)} = \frac{1}{2} \frac{\partial D_+}{\partial \Omega_3} e^{(k_3 \cdot r + \theta_3)} .
\]

We now apply this formula to the two situations of figure 1.

4.2 Backward Saturation. — We consider the case where the intense pump beam propagates in a direction opposite to the two weak beams (pump \(E_2\) and probe \(E_3\) (Fig. 1a)). The conjugate beam is generated in a direction opposite to the probe beam. To find the lineshape, we use (47) and (48) with \(j = 2\). Using \(k_1 + k_2 = 0\), we find that the amplitude of the field generated in the \(-k_3\) direction is proportional to:

\[
\tilde{E}_3^{(c)}(\delta) = 2\sqrt{\pi} \frac{\Omega_2 \Omega_3}{k_2} \frac{(8 \delta^2 + 3)}{(64 \delta^4 + 8 \delta^2 + 25)^2} \cdot \frac{\delta^3}{(64 \delta^4 + 8 \delta^2 + 25)^2} \times \left[ -2(64 \delta^2 + 3) - 3(152 \delta^2 + 25) - 16 \delta_0(64 \delta^4 + 80 \delta^2 + 5) \right].
\]

As in the preceding case, we note that the phase conjugate emission is equal to 0 at resonance. In fact, \(\delta\) comes from the derivation \(d\tilde{E}_3^{(c)} / d\Omega_2\) or \(d\tilde{E}_3^{(c)} / d\delta\). On the other hand, we observe that \(\tilde{E}_3^{(c)}\) is complex while \(\tilde{E}_3^{(o)}\) is real. This behaviour is similar to the one obtained by Bloch and Ducloy [20] in the case of three-level atoms. We now compare (49), (50) and the formula obtained for atoms at rest [16-18].

4.4 Discussion. — In theories [16-18] assuming motionless atoms, the dipole moment \(D_+^{(o)}\) which generates the phase conjugate emission is equal to:

\[
D_+^{(o)}(\delta) = \frac{\Omega_2 \Omega_3}{\Omega_1^2} \frac{d}{d\delta} \left( e^{(k_3 \cdot r + \theta_3)} \frac{\delta^3}{(64 \delta^4 + 8 \delta^2 + 25)^2} \right) ,
\]

At the secular approximation, the intensity of the generated beam \(\sim |D_+^{(o)}|^2\) only depends on \(\delta = \delta / \Omega_1\). When \(\Omega_1\) increases, the peak intensity remains constant but the width increases proportionally to \(\Omega_1\). We also note that in the wing of the line (\(\delta \gg 1\)), the intensity decreases like \(\delta^{-2}\).

In order to interpret this lineshape, let us come back to (45) which is at the origin of (49). The fact that we have replaced the resonance curves by Dirac \(\delta\) functions shows that only two velocity groups (which correspond to the roo\(\delta s s_1\) and \(s_2\)) contribute to the phase conjugate emission. For these two velocity groups the four-wave mixing process is fully resonant in the energy diagram of the dressed-atom. It explains why the contribution of the other velocity groups which only contribute through non resonant terms can be neglected. The two velocity groups are determined by the equation \(\omega_1 - \omega_2 = \pm \Omega\). This demonstrates that a close link does exist with the enhancement due to the Rabi sidebands in non degenerate four-wave mixing (see § 2). The Doppler shift indeed selects the velocity groups for which a non degenerate emission on the Rabi sidebands occurs in the atomic frame. Since we are able to determine the contribution of each resonant velocity group, it is obviously possible to extent the formula for situations where the Boltzmann factor \(e^{-\Delta}\) cannot be replaced by 1, i.e. when \(\delta \sim k_2\). We have indeed developed such an approach. The corresponding results will be published together with the experimental observations [49, 50]. The fact that we add the amplitude corresponding to the two velocity groups explains why the signal is equal to 0 at resonance (\(\delta = 0\)). For this situation, the two resonant velocity groups have a contribution equal in magnitude but opposite in sign.

4.3 Forward Saturation. — We now consider the case (Fig. 1b) where the probe beam is almost parallel to the intense pump beam \((k_3 \approx k_1\) Fig. 1a). To find the lineshape, we use (44) and (48) with \(j = 1\). Using \(k_1 + k_2 = 0\); we find:

\[
\tilde{E}_3^{(c)}(\delta) = -\frac{\sqrt{\pi} \Omega_2 \Omega_3}{k_2} \frac{d}{d\delta} \left( e^{(k_3 \cdot r + \theta_3)} \frac{\delta^3}{(64 \delta^4 + 8 \delta^2 + 25)^2} \right) \times \left[ -2(64 \delta^2 + 3) - 3(152 \delta^2 + 25) - 16 \delta_0(64 \delta^4 + 80 \delta^2 + 5) \right].
\]
the square of the dipole moment. Comparing (49), (50) and (51), we have :

\[ |\tilde{y}_{+}^{(be)}|^2 \sim \frac{\Omega_{1}^4}{\Gamma^2(ku)^2} |\tilde{B}_{+}^{(oo)}|^2 \]  

\[ |\tilde{y}_{+}^{(be)}|^2 \sim \frac{\Omega_{1}^2}{\Gamma^2} |\tilde{Y}_{+}^{(ce)}|^2 . \]  

For a purely radiative system, a motionless atom model can be considered when \( \Omega_{1} > ku \) \(^{(1)}\). In these conditions, we predict a huge variation of the phase conjugate reflectivity from the Doppler limit case toward the motionless atom case. This factor \( \Omega_{1}^4/\Gamma^2(ku)^2 \), which was first noticed in [22], can be of the order of \( 10^{-4}\text{-}10^{-5} \). It explains the behaviour obtained in a computational method (see [21], Fig. 3) where the peak intensity of the phase conjugate emission decreases by several orders of magnitude when \( \Omega_{1}/ku \) varies from 1 to 10. Contrary to genuine intuition, the Doppler inhomogeneous width does not lead to a decrease but to a strong increase of the phase conjugate emission. The reason comes from the fact that the Doppler effect tunes the four-wave mixing process to be fully resonant for certain velocity groups. On the other hand, in theories assuming motionless atoms, the four-wave mixing process is always non-resonant.

The ratio of the intensities for backward and forward saturations (53) is similar to the one obtained by Bloch and Ducloy [20] for a three-level atom. At the secular limit, the phase-conjugate emission is stronger in the case of backward saturation.

The general aspect of the lineshapes obtained in these various situations (Fig. 3) is the same. The phase-conjugate emission is equal to 0 at resonance, peaks for a detuning of the order of the Rabi frequency and decreases for larger detunings. However a careful examination shows differences. For instance, around \( \delta \approx 0 \), we have a \( \delta^2 \) variation in the Doppler limit and a \( \delta^6 \) variation for motionless atoms. Similarly for large detunings, the phase conjugate emission decreases like \( \delta^{-2} \) in the Doppler limit and like \( \delta^{-6} \) for motionless atoms. (This last result should be treated with some care because for large detunings it may be useful to keep the Boltzmann factor in the Doppler limit [49, 50].) In conclusion, we believe that the differences, both in order of magnitude and in variation with \( \delta \), should now prevent wrong or doubtful interpretation of the experimental results. We can also add that the phase conjugate emission is not always equal to 0 at resonance. We have shown that for a three-level atom, the intensity differs from 0 in the Doppler limit for a particular choice of polarization [51].

Finally, we observe that if we assume \( \delta \gg \Gamma \) and we make \( \Omega_{1} \to 0 \), we find that the formula obtained in the case of backward saturation tends towards the usual perturbation formula [14] while this is not true in the case of forward saturation. It comes from the fact that in backward saturation, the secular approximation can be applied when \( |\delta + \Omega_{1}^2|^{1/2} \gg \Gamma \) because in the atomic frame, the probe beam and the saturating beam have different frequencies. On the other hand, in forward saturation, these two beams have the same frequency and the condition is always \( \Omega_{1} \gg \Gamma \).

5. Effect of collisional damping in the case of backward saturation.

The knowledge of the effect of collisional damping in four-wave mixing is important for several reasons. First, experiments are often performed in a range of pressure where the collisional broadening is larger than the natural width. Second, the effect of collisions is often less obvious than one would imagine. For example, the well known Bloembergen resonance [35-40] (PIER 4) are created by collisions. In the degenerate case their magnitude increases with the...
buffer gas pressure. These resonances have been observed and studied theoretically for detunings from the resonance much larger than the Doppler width. We propose to investigate here what occurs for detunings smaller than the Doppler width (2).

In the presence of collisions, the relaxation terms (15) have to be modified. They become \[ \begin{align*}
\frac{\text{d}}{\text{d}t} \sigma_{11}^n \bigg|_{\text{rel}} &= - \Gamma \cos^4 \varphi \sigma_{11}^n + \Gamma \sin^4 \varphi \sigma_{22}^n - 2 \gamma \sin^2 \varphi \cos^2 \varphi (\sigma_{11}^n - \sigma_{22}^n) \\
\frac{\text{d}}{\text{d}t} \sigma_{22}^n \bigg|_{\text{rel}} &= - \Gamma \sin^4 \varphi \sigma_{22}^n + \Gamma \cos^4 \varphi \sigma_{11}^n + 2 \gamma \sin^2 \varphi \cos^2 \varphi (\sigma_{11}^n - \sigma_{22}^n) \\
\frac{\text{d}}{\text{d}t} \sigma_{12}^n \bigg|_{\text{rel}} &= -(L + \gamma (\cos^4 \varphi + \sin^4 \varphi)) \sigma_{12}^n
\end{align*} \]

where \( \gamma \) is the relaxation rate of the atomic coherence due to collisions (\( \gamma \) is proportional to the buffer gas pressure). We have taken \( \gamma \) real for sake of simplicity. The collisions tend to equalize the population between levels of the dressed atom.

When we solve the Bloch equations with the relaxation terms [54, (28d)] becomes:

\[ B^{(2)}_+ = \frac{\Omega_1^2}{\Gamma} \times \]
\[ \left[ \frac{\cos^4 \varphi - \sin^4 \varphi}{(\cos^4 \varphi + \sin^4 \varphi)^2} \times \frac{L' \sin^4 \varphi}{L'^2 + (\omega_1 - \omega_2 - \Omega)^2} + \frac{L' \cos^4 \varphi}{L'^2 + (\omega_1 - \omega_2 + \Omega)^2} \right] \]

with

\[ R = 2 \gamma/\Gamma \]
\[ L' = L + \gamma (\cos^4 \varphi + \sin^4 \varphi) \]

\[ (55) \]

If the homogeneous width is much smaller than the inhomogeneous Doppler width, we can proceed as in (§ 3) and use formula (36) to average over the velocity distribution. The dipole moment which generates the phase-conjugate emission in the case of backward saturation is given by a formula which generalizes (49):

\[ B^{(0)}_+ (\delta) = 2\sqrt{\pi} \frac{\Omega_2 \Omega_3}{\Gamma k u} \times \]
\[ \delta \left[ \frac{w_0 (w_0^2 + 56) + 36 R (w_0^2 + 20) + 324 R^2 w_0}{(w_0^2 + 16) + 20 R w_0 + 36 R^2} \right] \]

with

\[ \delta = 8 \delta^2 + 3 \]

\[ (58) \]

When there is no collisional broadening \( R = 0 \) and (57) is identical to (49). We now consider the situation of a large collisional damping \( R \gg 1 \). In order to obtain simple formulae, we also assume that \( |\delta| \gg 1 \), i.e. \( \delta \gg \Omega_1 \) (but \( |\delta| \ll \kappa u \) since we have assumed an infinite Doppler width). In these conditions, (57) becomes:

\[ B^{(0)}_+ (\delta) \approx 4 \sqrt{\pi} \frac{\Omega_2 \Omega_3}{\Gamma k u} e^{-(k_0 r + \theta_0)} \left[ \frac{\delta^3}{[4 \delta^2 + R]^2} \right] \]

\[ (59) \]

The maximum of emission is obtained for \( \delta = \frac{\sqrt{3} R}{2} \) and the intensity at the maximum decreases like \( R \). When a buffer gas is added, we thus expect to observe a decrease in the phase-conjugate emission. Equation (59) is only an approximate formula, nevertheless when we compute exactly (57), we obtain the same behaviour (see Fig. 4).

Fig. 4.—Effect of collisional damping on the lineshape in the case of backward saturation for a Doppler broadened line. Position of the maximum (continuous line) and intensity at the maximum (dashed line) versus collisional damping \( R = 2 \gamma/\Gamma \). The curves correspond to the simplified formula (59) and the points to the exact calculation (formula (57)). (Note that the curves are plotted on a log-log scale.)

The effect of a buffer gas is thus strongly different from what is obtained in the case of Bloembergen resonances for detunings much larger than the Doppler width. To make the difference apparent, we can use formula (51) valid for a Doppler-free medium. The relaxation rate \( \Gamma_0 \) of the coherence is equal to \( \Gamma_0 = \Gamma + \gamma \). When \( \Gamma \gg 1 \), we have \( \Gamma_0 \approx \gamma \) and (51) becomes:

\[ B^{(0)}_+ (\delta) = \frac{\Omega_2 \Omega_3}{\Omega_1^2} \times \]
\[ \left[ \frac{R \delta^3}{\delta^2 + R^2} \right] \]

\[ (60) \]
If we compare (59) and (60), we note that the maximum of phase-conjugate emission occurs for similar values of \( \delta \sim \sqrt{R} \) and that the peak intensity also decreases like \( R \). However, there is a dramatic difference between the wings of the two curves. When \( \delta \gg R \), the intensity does not depend upon \( R \) in the Doppler limit while it increases like \( R^2 \) for the case of motionless atoms. The factor \( R^2 = y^2/\Gamma^2 \) is precisely the enhancement factor due to collisions which is observed in PIER 4 experiments in the degenerate case [36].

This study shows that the pressure-induced effects for two-level atoms are very different inside and outside the Doppler width.

6. Saturation due to the weak pump beam in the case of backward saturation.

Up to now, we have only considered the case where perturbation theory can be applied for the weak beam (\( \Omega_2 < \Gamma \)). However, as we mention in § 1, the theory can easily be extended to situations where \( \Omega_2 \gtrsim \Gamma \) provided that \( \Omega_2 \ll \Omega_1 \). \(^{(1)}\)

Using (25), (26), (36) and (48), we predict that the intensity of the phase-conjugate emission in the case of backward saturation (Fig. 1a) should be:

\[
\frac{\delta y_{\text{sat}}}{\delta} = \frac{1}{2} \frac{\partial \tilde{D}_+}{\partial \Omega_2} \Omega_3 \cos(\theta_2 - \theta_1) \quad (61)
\]

with

\[
\tilde{D}_+(\delta) = \frac{1}{u \sqrt{\pi}} \int_{-\infty}^{+\infty} \mathrm{d} v_2 D_+(\delta, v_2)
\]

and

\[
D_+(\delta, v_2) = - d \cos^4 \varphi - \sin^4 \varphi \cos \varphi \sin \varphi \times 
\]

\[
\times \left[ 1 - \frac{\Omega_2^2 L}{\Gamma \cos^4 \varphi + \sin^4 \varphi} \left( \omega_1 - \omega_2 + \Omega \right)^2 + L^2 + \Omega_2^2 \frac{L}{\Gamma \cos^4 \varphi + \sin^4 \varphi} \right] - 
\]

\[
\times \frac{\Omega_2^2 L}{\Gamma \cos^4 \varphi + \sin^4 \varphi} \left( \omega_1 - \omega_2 - \Omega \right)^2 + L^2 \times \frac{\sin^4 \varphi}{\cos^4 \varphi + \sin^4 \varphi}
\]

(62)

Because \( \Gamma, \Omega_2 \ll \Omega_1 \ll ku \), we can replace the resonance curves by Dirac \( \delta \) functions using the same procedure as in § 3.4.

\[
D_+(\delta, v_2) = - d \cos^4 \varphi - \sin^4 \varphi \cos \varphi \sin \varphi \times 
\]

\[
\times \left[ 1 - \frac{\pi \Omega_2^2 L}{\Gamma \cos^4 \varphi + \sin^4 \varphi} \left( \omega_1 - \omega_2 + \Omega \right)^2 + L^2 + \Omega_2^2 \frac{L}{\Gamma \cos^4 \varphi + \sin^4 \varphi} \right]^{1/2} \delta(\omega_1 - \omega_2 - \Omega)
\]

(63)

The integration (62) is then very easy to perform, however it does not lead to a simple analytical formula. We have plotted the intensity of the phase conjugate emission (\( \sim | \tilde{D}_+^{(1)} |^2 \)) for several values of \( \Omega_2/\Gamma \) on figure 5. We observe that the general shape of the curve remains unchanged. In particular we always have a phase conjugate emission equal to 0 at resonance. We have plotted the peak intensity of the phase conjugate emission versus \( \Omega_2/\Gamma \) on figure 6. We note that it attains a maximum when \( \Omega_2/\Gamma \sim 2.5 \) and then it decreases toward its asymptotic value.

Finally, we note that the preceding formalism can be easily adapted to calculate high-order non-linear optical susceptibilities [53]. For example, a process which involves the absorption of two photons of beam \( E_2 \) and the emission of two photons of beam \( E_3 \) should have an amplitude proportional to \( \frac{\partial^3 \tilde{D}_+}{\partial \Omega_2^2} \Omega_3^2 \) where \( \tilde{D}_+ \) is given by (62) and (64).

7. Conclusion.

In this paper we have presented a new formalism, based on the dressed-atom theory, to understand and calculate several effects in degenerate four-wave mixing in Doppler broadened media. The main advantage of the formalism is that it permits simple physical

\(^{(1)}\) Since this paper has been completed, we have received a reprint of M. Ducloy et al. [52] who studied a similar problem for three-level atoms using Bloch equations. Their results are similar to ours.
interpretations to be obtained. It also permits to simplify the calculations at the secular limit ($\Omega_1 > \Gamma$). We have indeed obtained analytical formulae, in that limit, for almost all the problems that we have considered in this paper.

There are several extensions to this work which will be presented in other publications together with the experimental results. We have studied the effect of pump beam polarization in the case of a three-level atom and show that the lineshape in degenerate four-wave mixing strongly depends on the relative polarization of the two beams which propagate in the same direction. We have also considered the case of nearly degenerate four-wave mixing and show that narrow natural width limited resonances are predicted and observed. These resonances are located at frequencies corresponding to transitions between the energy levels of the dressed-atom [51, 56] and show that the dressed-atom is strongly connected with the understanding of saturation in four-wave mixing.

References

[40] Bloembergen, N., Ann. Physique Fr. (to be published).
[50] Pinard, M., Verkerk, P. and Grynberg, G., to be published.