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## Theory of optical heterodyne three-level saturation spectroscopy *via* collinear non-degenerate four-wave mixing in coupled Doppler-broadened transitions

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**Résumé.** — Nous présentons une analyse théorique de la spectroscopie de saturation par hétérodynage optique à haute fréquence dans le cas d'un système couplé à trois niveaux en présence d'élargissement Doppler. Le faisceau saturant, qui est modulé en amplitude à une fréquence  $\delta$  par technique de double bande latérale avec suppression de la porteuse, est résonnant pour une des transitions. Par un processus de mélange à quatre ondes non dégénéré, cette modulation est transférée à un faisceau sonde, résonnant pour une transition couplée. Le champ saturant et le champ sonde se propagent soit dans la même direction, soit en directions opposées. La forme de raie de la modulation induite sur le faisceau sonde est calculée par un développement de la matrice densité au troisième ordre de perturbation relativement à l'amplitude des champs incidents. L'intégration sur les vitesses est effectuée dans l'approximation des grandes largeurs Doppler. Le signal est dû à la fois à des effets de saturation de la population et à des effets cohérents (Raman ou à deux photons). Ces diverses contributions apparaissent sous forme de doublets de résonances lorentziennes. Nous montrons qu'en l'absence de processus de relaxation (collisions déphasantes, ou cascades radiatives), une interférence destructive entre la saturation de la population et les processus cohérents à deux photons entraîne la disparition d'un des doublets de résonances. On en déduit que les collisions déphasantes peuvent conduire à l'existence d'un doublet de résonances supplémentaires induites par pression (PIER), qui pourraient donner des informations sur les processus collisionnels en régime d'impact. Les propriétés de ces doublets PIER sont comparées à d'autres types de signaux PIER étudiés en mélange à quatre ondes et en spectroscopie résolue dans le temps.

**Abstract.** — We present a theoretical analysis of high-frequency optical heterodyne saturation spectroscopy in Doppler-broadened coupled three-level systems. The saturating beam, which is assumed to be amplitude modulated at frequency  $\delta$  (double-sideband suppressed carrier amplitude modulation) is resonant for one of the transitions. Through non-degenerate four-wave mixing processes, this modulation is transferred to a probe beam, resonant for the coupled transition. Saturating and probe fields may be either co-propagating or counter-propagating. The lineshape of the probe modulation is analysed *via* a third-order perturbation expansion of the atomic density matrix with respect to the incident field amplitudes. The resulting signal is integrated over velocities, in the Doppler limit approximation. Both population saturation effects and coherent (Raman-type, or two-photon) processes contribute to the signal. The various contributions appear as Lorentzian-type resonance doublets. We show that, in the absence of relaxation processes (collisional dephasing, or radiative cascades), destructive interferences between population saturation and coherent two-photon processes are responsible for the disappearance of one resonance doublet. Phase-interrupting collisions are thus predicted to lead to the existence of a « pressure-induced extra-resonance » (PIER) doublet, which could yield information on collisional processes in the impact regime. The properties of the predicted PIER doublets are analysed in relation with other types of PIER signals studied in four-wave mixing, and in time-resolved saturation spectroscopy.

### 1. Introduction.

In the recent years, increased sensitivity in Doppler-free spectroscopy (e.g., saturated absorption, two-

photon or Raman spectroscopy) has been obtained thanks to the development of various high-frequency (HF) heterodyne techniques [1-5]. In particular, it has become possible to routinely reach the shot-noise limit in the detection of Doppler-free optical signals. In addition, nonlinear dispersion lineshapes may be monitored as easily as nonlinear absorption.

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In previous works, we have studied both experimentally [4] and theoretically [5] a spectroscopic technique in which a HF *amplitude modulation* (AM) is applied to the beam propagating in one arm of a ring interferometer. In this case, and when all the electromagnetic (e.m.) incident fields originate from a single laser source, the heterodyne beat observed at resonance can be interpreted as produced by a collinear nearly-degenerate four-wave mixing process.

By using two different laser sources, the advantages of HF heterodyne spectroscopy can be extended to the saturation spectroscopy of coupled three-level systems with arbitrary frequencies. We have shown recently [6] that this two-laser technique is particularly adequate to perform Doppler-free spectroscopy of weak, hardly saturable, transitions (e.g. weak UV transitions). In this paper, we present a theoretical analysis of optical heterodyne three-level saturation spectroscopy, and we discuss the various contributions to the heterodyne signal, which originate essentially in population effects, or in coherence effects (Raman-type, or two-photon type). After presenting the general framework of this study (Sect. 2), we give an expression of the heterodyne beat signal, obtained in a third-order perturbation theory, in the approximation of large Doppler widths (Sect. 3). In section 4, we discuss the interference effects occurring between population contributions and coherence

processes. In particular, we show that a new type of pressure-induced extra-resonances (PIER) should be observable.

## 2. General description.

We consider a gas medium composed of three-level atoms  $|0\rangle, |1\rangle, |2\rangle$ , for which the levels  $|0\rangle - |1\rangle$  and  $|0\rangle - |2\rangle$  are coupled by dipolar transitions (the respective dipole moments,  $\mu_1$  and  $\mu_2$ , are assumed to be real). The atomic velocity distribution  $f(\mathbf{v})$  is assumed to obey a Maxwell-Boltzmann distribution law :

$$f(\mathbf{v}) = \frac{1}{\pi^{3/2} u^3} \exp(-v^2/u^2) \quad (1)$$

( $u$  is the mean velocity).

The medium is supposed to be irradiated by two incident e.m. fields which propagate along the  $z$ -axis, a probe field  $E_1$  :

$$E_1 = \frac{1}{2} \varepsilon_1 \exp i(\omega_1 t - k_1 z) + c.c. \quad (2)$$

whose frequency is approximately resonant for the  $|0\rangle - |1\rangle$  transition, and a pump beam  $E_2$ , nearly resonant for the  $|0\rangle - |2\rangle$  transition, and whose amplitude is modulated at an audio- or radio-frequency  $\delta$  :

$$E_2 = \frac{1}{2} \varepsilon_2 \left\{ \exp i \left[ \left( \omega_2 + \frac{\delta}{2} \right) t - \varepsilon k_2 z \right] + \exp i \left[ \left( \omega_2 - \frac{\delta}{2} \right) t - \varepsilon k_2 z \right] \right\} + c.c. \quad (3)$$

In equation (3), one has  $\varepsilon = +1$  or  $\varepsilon = -1$ , whether the two fields  $E_1$  and  $E_2$  are co-propagating or counter-propagating.

[For sake of simplicity, the amplitudes  $\varepsilon_1$  and  $\varepsilon_2$  are assumed to be real].

When resonance conditions are fulfilled, an e.m. field at frequency  $\omega_1 + \delta$  (respectively  $\omega_1 - \delta$ ) is emitted along the propagation axis of the  $E_1$  field, which can be attributed to a collinear non-degenerate four-wave mixing process in which are absorbed one photon of the probe field  $E_1$  and one photon of the pump field at frequency  $\omega_2 + \delta/2$  (respectively  $\omega_2 - \delta/2$ ), and emitted one photon of the pump field at frequency  $\omega_2 - \delta/2$  (respectively  $\omega_2 + \delta/2$ ). As is shown in figure 1, this emitted field is detected *via* its heterodyne beating with the probe at frequency  $\delta$ . This field is coherently radiated by the  $(\omega_1 \pm \delta)$  components of the nonlinear polarization  $P_{NL}$ , induced in the medium by the total field  $E_1 + E_2$ . One thus has to derive the element  $\rho_{01}$ , of the density matrix,  $\rho$  :

$$P_{NL} = \mu_1 \rho_{01} + c.c. \quad (4)$$

[in equation (4), we retain only the term of interest of the induced nonlinear polarization].

The equations of motion for the density matrix can be written independently of the configuration of the atomic three-level system [folded system : V- or  $\Lambda$ -type, or cascade systems, see figure 2].

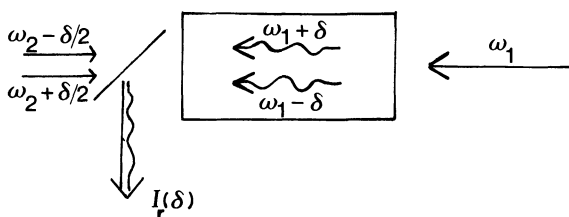


Fig. 1. — Schematics of the optical set-up (pump and probe beams are counter-propagating,  $\varepsilon = -1$ ).

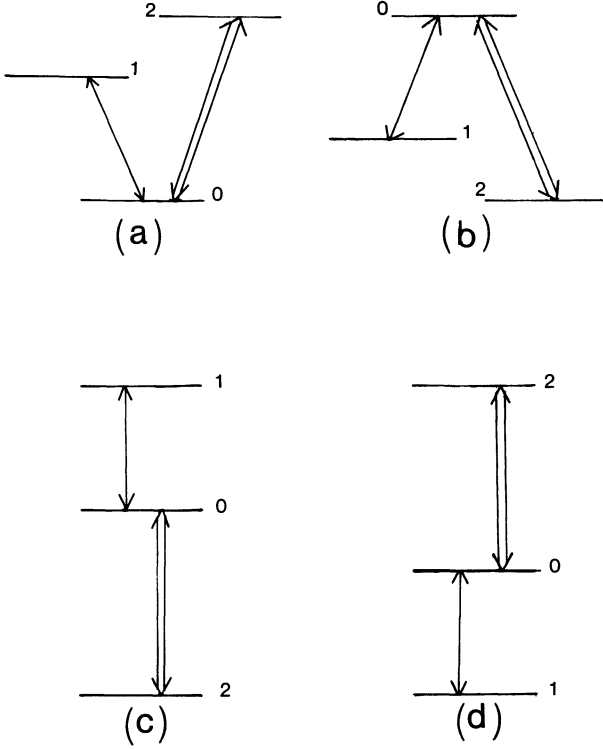


Fig. 2. — Various types of three-level atomic systems : (a) V-type folded system ; (b)  $\Lambda$ -type folded system ; (c) and (d) cascade systems.

$$\begin{aligned}
 \dot{\rho}_{00} + \Gamma_0(\rho_{00} - n_0) &= -\frac{i}{\hbar} [E_1 \mu_1(\rho_{01} - \rho_{10}) + E_2 \mu_2(\rho_{02} - \rho_{20})] + \left(\frac{d\rho_{00}}{dt}\right)_{\text{sp}} \\
 \dot{\rho}_{11} + \Gamma_1(\rho_{11} - n_1) &= -\frac{i}{\hbar} E_1 \mu_1(\rho_{10} - \rho_{01}) + \left(\frac{d\rho_{11}}{dt}\right)_{\text{sp}} \\
 \dot{\rho}_{22} + \Gamma_2(\rho_{22} - n_2) &= -\frac{i}{\hbar} E_2 \mu_2(\rho_{20} - \rho_{02}) + \left(\frac{d\rho_{22}}{dt}\right)_{\text{sp}} \\
 \dot{\rho}_{01} + (\Gamma_{01} - i\omega_{10}) \rho_{01} &= \frac{i}{\hbar} [\mu_2 E_2 \rho_{21} - \mu_1 E_1(\rho_{00} - \rho_{11})] \\
 \dot{\rho}_{02} + (\Gamma_{02} - i\omega_{20}) \rho_{02} &= \frac{i}{\hbar} [\mu_1 E_1 \rho_{12} - \mu_2 E_2(\rho_{00} - \rho_{22})] \\
 \dot{\rho}_{12} + (\Gamma_{12} - i\omega_{21}) \rho_{12} &= \frac{i}{\hbar} [-\mu_2 E_2 \rho_{10} + \mu_1 E_1 \rho_{02}].
 \end{aligned} \tag{5}$$

In equation (5),  $\dot{\rho} = \frac{d\rho}{dt}$  is the total (hydrodynamic) time-derivative of the density matrix  $\left(\frac{d}{dt} = \frac{\partial}{\partial t} + \mathbf{v} \cdot \nabla\right)$ .  $\Gamma_i$  and  $\Gamma_{ij}$  represent respectively the relaxation coefficients of the population of level  $|i\rangle$ , and of the coherence between levels  $|i\rangle$  and  $|j\rangle$  ( $\Gamma_{ij} = \Gamma_{ji}$ ).  $\hbar\omega_{ij} = \omega_i - \omega_j$  ( $i, j = |0\rangle, |1\rangle, |2\rangle$ ) is the  $i$ - $j$  transition frequency ( $\omega_{ij}$  can be positive or negative).  $n_i$  is the equilibrium population of level  $|i\rangle$ , and  $\left(\frac{d\rho_{ii}}{dt}\right)_{\text{sp}}$  represents the cascade term induced by spontaneous emission towards level  $|i\rangle$ , which can be written :

$$\left(\frac{d\rho_{ii}}{dt}\right)_{\text{sp}} = \sum_{\substack{j=0,1,2 \\ j \neq i}} \gamma_{ji} \rho_{jj}. \tag{6}$$

As an example, for a V-type folded system (see Fig. 2a) one has  $\left(\frac{d\rho_{00}}{dt}\right)_{\text{sp}} = \gamma_{10} \rho_{11} + \gamma_{20} \rho_{22}$  and

$$\left(\frac{d\rho_{11}}{dt}\right)_{\text{sp}} = \left(\frac{d\rho_{22}}{dt}\right)_{\text{sp}} = 0.$$

Following the same approach as in reference [5], one gets the expression of the reradiated field  $E_r$  :

$$E_r = -i \frac{k_1 L}{2 \varepsilon_0} \mu_1 \langle \rho_{01} \rangle + c.c. \quad (7)$$

where  $\langle \rangle$  means velocity-averaged, and  $L$  is the length of the interaction zone. The expression of the heterodyne beat signal  $I_r(\delta)$  is given by :

$$I_r(\delta) = \frac{1}{2} \varepsilon_0 c \varepsilon_1 [\varepsilon_r(+)] + \varepsilon_r^*(-)] \exp(i \delta t) + c.c. \quad (8)$$

where the amplitudes  $\varepsilon_r(\pm)$  are defined by :

$$E_r = \frac{1}{2} [\varepsilon_r(+) \exp i[(\omega_1 + \delta) t - k_1 z] + \varepsilon_r(-) \exp i[(\omega_1 - \delta) t - k_1 z]] + c.c. \quad (9)$$

### 3. Expression of the heterodyne beat signal in a perturbative approach.

In all the following, we use a lowest-order perturbation expansion (relatively to the incident field amplitudes) to calculate the components of  $\rho_{01}$  oscillating at  $\omega_1 \pm \delta$  : this implies to deal with a third-order perturbation expansion of  $\rho$  in the total field  $E_1 + E_2$ , and to retain the cross-terms proportional to  $\varepsilon_1 \varepsilon_2^2$  only, i.e. to deal with a second-order perturbation expansion relatively to the pump field  $E_2$  and a first-order expansion relatively to the probe field  $E_1$ .

To be specific, we first consider the case of a V-type folded system (cf. Fig. 2a) and we discuss lately the case of different atomic systems.

**3.1 CASE OF A V-TYPE FOLDED SYSTEM.** — For the V-type system, in the rotating-wave approximation, the density matrix element  $\rho_{01}$  evolves essentially at a positive frequency  $\omega_1$  [ $\rho_{01} = \tilde{\rho}_{01} \exp i(\omega_1 t - k_1 z)$ , where  $\tilde{\rho}_{01}(z, t)$  evolves slowly relatively to  $\omega_1$  and  $k_1$ ]. For the contribution  $\rho_{01}^{(+)}$  which evolves like  $\exp i[(\omega_1 + \delta) t - k_1 z]$ , one easily finds ( $v$  is the velocity component along  $z$ ) :

$$\begin{aligned} \rho_{01}^{(+)}(v) = & \frac{i}{8} \varepsilon_1 \varepsilon_2^2 \mu_1 \mu_2^2 \frac{\exp i[(\omega_1 + \delta) t - k_1 z]}{\Gamma_{01} + i(\omega_1 + \delta - \omega_{10} - k_1 v)} \times \\ & \times \left\{ \frac{(n_0 - n_2)}{(\Gamma_0 + i\delta)} \left( 1 - \frac{\gamma_{20}}{\Gamma_2 + i\delta} \right) \left[ \frac{1}{\Gamma_{02} + i(\omega_2 + \delta/2 - \omega_{20} - \varepsilon k_2 v)} + \frac{1}{\Gamma_{02} - i(\omega_2 - \delta/2 - \omega_{20} - \varepsilon k_2 v)} \right] + \right. \\ & \left. + \frac{1}{\Gamma_{21} + i[\omega_1 - (\omega_2 - \delta/2) - \omega_{12} - (k_1 - \varepsilon k_2) v]} \left[ \frac{(n_0 - n_2)}{\Gamma_{02} - i(\omega_2 - \delta/2 - \omega_{20} - \varepsilon k_2 v)} + \right. \right. \\ & \left. \left. + \frac{(n_0 - n_1)}{\Gamma_{01} + i(\omega_1 - \omega_{10} - k_1 v)} \right] \right\}. \quad (10) \end{aligned}$$

In this expression, two different contributions appear : (i) the first one, which is proportional to  $(\Gamma_0 + i\delta)^{-1}$ , arises from a modulation at frequency  $\delta$  of the atomic population — created at second-order — in the common level  $|0\rangle$  ; for this first term, the two resonant denominators are associated to the resonance conditions for the optical coherences created at third- and first-order respectively :  $(\omega_2 + \delta/2 - \omega_{20} - \varepsilon k_2 v) - (\omega_2 - \delta/2 - \omega_{20} - \varepsilon k_2 v) + (\omega_1 - \omega_{10} - k_1 v) = 0$  ; and  $(\omega_2 + \delta/2 - \omega_{20} - \varepsilon k_2 v) = 0$  or  $(\omega_2 - \delta/2 - \omega_{20} - \varepsilon k_2 v) = 0$ . (ii) In the second term, the population factor is replaced by a denominator associated to a Raman-type transition process between levels  $|1\rangle$  and  $|2\rangle$ , which appears at second-

order, *via* one interaction with the pump field  $E_2$  and one interaction with the probe field  $E_1$ . Inside this Raman-type contribution, the term which is proportional to  $(n_0 - n_2)$  is associated to a Raman coherence created *via* first-order optical coherences between levels  $|0\rangle$  and  $|2\rangle$ , while the term proportional to  $(n_0 - n_1)$  is associated to a Raman coherence created *via* the coupling (at first-order), by the  $E_1$  field, between levels  $|0\rangle$  and  $|1\rangle$ .

In the infinite Doppler width approximation ( $k_1 u, k_2 u \gg \Gamma$ ), the velocity integration of expression (10) is easily performed by means of residue techniques. One should notice that the Raman term yields a non-negligible contribution only for  $\varepsilon = +1$  (co-propa-

gating incident e.m. fields). Moreover, the contribution proportional to  $n_0 - n_1$ , which exists even if the medium is transparent for the pump beam ( $n_0 = n_2$ ), remains non-negligible only if  $k_2 > k_1$ . Note that the cancellation of this extra Raman contribution is well known in ordinary three-level saturation spectroscopy [7-9].

Finally, the velocity-averaged quantity  $\langle \rho_{01}^{(+)} \rangle$  can be written as :

$$\langle \rho_{01}^{(+)} \rangle = C \{ \exp i[(\omega_1 + \delta) t - k_1 z] \} \times \{ S_P(\delta) + S_R(\delta) \} \quad (11)$$

where :

$$C = \frac{i\sqrt{\pi}}{4 k_1 u} \mu_1 \mu_2^2 \varepsilon_1 \varepsilon_2^2 \quad (12)$$

and where  $S_P(\delta)$  and  $S_R(\delta)$  are associated respectively to population and Raman coherence contributions. One has

$$S_P(\delta) = \frac{(n_0 - n_2)}{\Gamma_0 + i\delta} \left[ 1 - \frac{\gamma_{20}}{\Gamma_2 + i\delta} \right] \frac{1}{\Gamma + i \left[ \Delta + \delta \left( \frac{1}{2} + \frac{k_2}{k_1} \right) \right]} \quad (13)$$

and for  $k_2 < k_1$ ,  $\varepsilon = +1$ ,

$$S_R(\delta) = \frac{(n_0 - n_2)}{\Gamma_{02} + \Gamma_{01} - \Gamma_{12} + i\delta} \left\{ \frac{1}{\Gamma' + i(\Delta + \delta/2)} - \frac{1}{\Gamma + i \left[ \Delta + \delta \left( \frac{1}{2} + \frac{k_2}{k_1} \right) \right]} \right\}. \quad (14a)$$

For  $k_2 > k_1$ , and  $\varepsilon = +1$ , one gets

$$S_R(\delta) = \frac{(n_0 - n_2)}{\Gamma_{02} + \Gamma_{01} - \Gamma_{12} + i\delta} \left\{ \frac{1}{\Gamma'' + i \left[ \Delta + \delta \left( -\frac{1}{2} + \frac{k_2}{k_1} \right) \right]} - \frac{1}{\Gamma + i \left[ \Delta + \delta \left( \frac{1}{2} + \frac{k_2}{k_1} \right) \right]} \right\} + \frac{(n_0 - n_1)}{i\delta} \left\{ \frac{1}{\Gamma'' + i \left( \Delta + \frac{\delta}{2} \right)} - \frac{1}{\Gamma'' + i \left[ \Delta + \delta \left( -\frac{1}{2} + \frac{k_2}{k_1} \right) \right]} \right\}. \quad (14b)$$

In equations (13) and (14), we have used :

$$\Delta = \frac{k_2}{k_1} (\omega_1 - \omega_{10}) - \varepsilon (\omega_2 - \omega_{20}) \quad (15)$$

$$\Gamma = \Gamma_{02} + \frac{k_2}{k_1} \Gamma_{01} \quad (16a)$$

$$\Gamma' = \left( 1 - \frac{k_2}{k_1} \right) \Gamma_{02} + \frac{k_2}{k_1} \Gamma_{12} \quad (16b)$$

$$\Gamma'' = \left( \frac{k_2}{k_1} - 1 \right) \Gamma_{01} + \Gamma_{12}. \quad (16c)$$

To determine the total signal lineshape when the frequency of one laser (e.g.  $\omega_2$ ) is tuned across resonance, while the other frequency ( $\omega_1$ ) is kept constant, one needs to take into account the emission at frequency  $\omega_1 - \delta$  by adding the quantities  $S_P^*(-\delta)$  and  $S_R^*(-\delta)$  to  $S_P(+\delta)$ , and  $S_R(+\delta)$ . Hence, one easily sees that the population contribution will appear as a doublet, whose splitting is  $\delta \left( 1 + \frac{2k_2}{k_1} \right)$ , and whose centre is located at  $\Delta = 0$  [i.e.  $\omega_2 = \omega_{20} + \varepsilon \frac{k_2}{k_1} (\omega_1 - \omega_{10})$ ]. On the other hand, the Raman contribution (for  $\varepsilon = +1$ ) appears as a pair of doublets, if  $k_2 < k_1$  [splittings  $\delta$  and  $\delta \left( 1 + \frac{2k_2}{k_1} \right)$ ],

or as a structure of three doublets if  $k_2 > k_1$  [splittings  $\delta$ ,  $\delta \left( \frac{2k_2}{k_1} - 1 \right)$  and  $\delta \left( \frac{2k_2}{k_1} + 1 \right)$ ]. All these doublets are centred on the same frequency ( $\Delta = 0$ ). A schematic of the positions of the expected resonances is presented in figure 3. All the resonances have a Lorentzian lineshape.

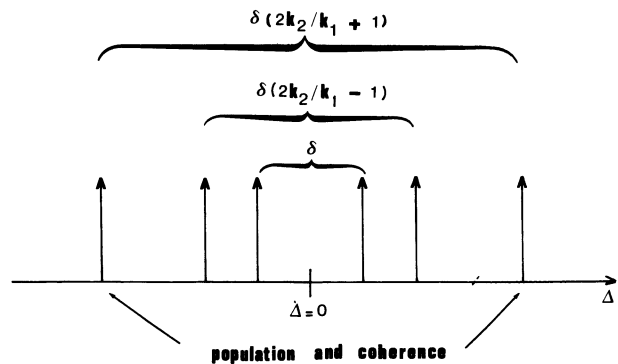


Fig. 3. — Positions of the three doublets of the heterodyne spectrum (note that the doublet with splitting  $\delta \left( \frac{2k_2}{k_1} - 1 \right)$  disappears if  $k_2 < k_1$ ).

Two points should be noted in these results :

(i) for co-propagating fields, the population doublet and one of the Raman doublets are located at the same frequencies, which implies that these two signals interfere. The effect of this interference can be dramatic, as seen in the next section ;

(ii) for vanishing  $\delta$ 's ( $\delta \ll \Gamma_{ij}$ ), the signal lineshape tends to the well-known resonance lineshapes in standard three-level saturation spectroscopy. All the doublets merge in a single resonance (at least, for weak saturation) which is a complex admixture of population and Raman-type contributions. One obvious interest of HF optical heterodyne spectroscopy is to discriminate between these two contributions in a straightforward way.

For instance, in the case  $k_2 \approx k_1$ , it is well known that the Raman contribution has a linewidth governed by  $\Gamma_{12}$  [cf. Eq. (16b)] which may be much narrower than the population contribution (in the case of ground state, or long-living states). One way of looking at these features is to monitor the transient evolution of the signal lineshape *via* time-resolved spectroscopic techniques [10-12]. The present method should allow one to monitor these narrow Raman resonances by shifting them away from the population contributions.

Another noteworthy point can be mentioned in the case  $k_2 > k_1$ . In that case, new contributions appear in three-level spectroscopy, which exist in particular for a medium transparent to the pump beam ( $n_2 = n_0 \neq n_1$ ) [7-9]. In HF optical heterodyne spectroscopy, these new contributions can be monitored independently of the relative values of  $n_2$  and  $n_0$ , since they are responsible for an additional doublet which is well resolved from the other ones (for  $\delta \gg \Gamma$ ).

**3.2 EXTENSION TO OTHER ATOMIC SYSTEMS.** — When dealing with three-level systems differing from the one of the figure 2a, minor changes in the formulae (13) to (16) have to be performed, which are relevant :

(i) to the various cascade processes induced by spontaneous emission ;

(ii) to the conditions of resonance for an atomic transition which become  $\omega_i = |\omega_{i0}|$  ;

(iii) and to the subsequent changes in the rotating wave approximation [ $\rho_{0i}$  evolves approximately like

$$\exp i(\omega_i t - \varepsilon k_i z) \quad \text{if } \omega_{i0} > 0$$

and like

$$\exp[-i(\omega_i t - \varepsilon k_i z)] \quad \text{if } \omega_{i0} < 0].$$

The heterodyne beat signal can still be written as

$$I_r(\delta) = \frac{\varepsilon_0 c \mu_1 \delta_1^2 \mu_2^2 \delta_2^2 \sqrt{\pi L}}{16 u} [S_p(\delta) + S_p^*(-\delta) + S_r(\delta) + S_r^*(-\delta)] e^{i\delta t} + c.c. \quad (17)$$

if the following changes are considered in the expression of  $S_p$  and  $S_r$  :

(a) The quantity  $\Delta$  (Eq. (15)) must be generalized to

$$\Delta = \frac{k_2}{k_1} (\omega_1 - |\omega_{10}|) - \varepsilon (\omega_2 - |\omega_{20}|). \quad (18)$$

(b) In the expression of  $S_p$  (Eq. (13)), the spontaneous emission factor  $\left[1 - \frac{\gamma_{20}}{\Gamma_2 + i\delta}\right]$  must be replaced by :

(i)  $\left[1 - \frac{\gamma_{01}}{\Gamma_1 + i\delta}\right]$  for the  $\Lambda$ -type system (cf. Fig. 2b) ;

(ii) 1 for the cascade system represented in figure 2c ;

(iii)  $\left[1 - \frac{\gamma_{20}}{\Gamma_2 + i\delta}\right] \left[1 - \frac{\gamma_{01}}{\Gamma_1 + i\delta}\right]$  for the cascade system represented in figure 2d.

If the effect of spontaneous emission is negligible, one can notice that the population contribution becomes identical for all the possible three-level systems : ( $n_0 - n_2$ ) is indeed replaced by ( $n_2 - n_0$ ) in the case of figures 2b and 2d, so that the factor ( $n_0 - n_2$ ) [or ( $n_2 - n_0$ )] always represents the population difference between the lower energy level and the upper energy level.

(c) For cascade systems (cf. Figs. 2c and 2d), the analogous of the  $S_r$  term of equation (14) corresponds to a contribution of a two-photon coherence, which is not negligible after the velocity integration only if the two incident e.m. fields are *counter-propagating* ( $\varepsilon = -1$ ).

The expression thus obtained for  $S_r$  has to be multiplied by a factor  $-1$  in the case of the atomic systems of the figures 2b and 2d.

#### 4. Pressure-induced extra-resonances in Doppler-free saturation spectroscopy.

In the previous section, we have already noted that one part of the  $S_r$  term (Raman, or two-photon, coherence contribution) is resonant for the same frequency as the  $S_p$  term (population contribution), and that their lineshapes are identical [same linewidth  $\Gamma$ , Eq. (16a)]. By adding (13) to the adequate terms of (14a) [or (14b)], one gets the following contribution :

$$\frac{n_0 - n_2}{\Gamma + i \left[ \Delta + \delta \left( \frac{1}{2} + \frac{k_2}{k_1} \right) \right]} \left[ \frac{1}{\Gamma_0 + i\delta} \left( 1 - \frac{\gamma_{20}}{\Gamma_2 + i\delta} \right) - \frac{1}{\Gamma_{01} + \Gamma_{02} - \Gamma_{12} + i\delta} \right] \quad (19)$$

which can be written in the equivalent form :

$$\frac{n_0 - n_2}{\Gamma + i \left[ \Delta + \delta \left( \frac{1}{2} + \frac{k_2}{k_1} \right) \right]} \frac{1}{\Gamma_0 + i\delta} \left[ \frac{\Gamma_{01} + \Gamma_{02} - \Gamma_{12} - \Gamma_0}{\Gamma_{01} + \Gamma_{02} - \Gamma_{12} + i\delta} - \frac{\gamma_{20}}{\Gamma_2 + i\delta} \right]. \quad (20)$$

This expression, established for the V-type configuration of figure 2a (in the case  $\varepsilon = +1$ ), may be easily generalized to any type of three-level system.

A remarkable result is that the above contribution cancels, and the interference between population saturation effects and Raman processes is *totally destructive* if :

(i) there is no phase-interrupting collision, so that

$$\Gamma_{ij} = \frac{1}{2}(\Gamma_i + \Gamma_j) \quad (21)$$

and (ii) the radiative cascade processes are negligible,

$$\gamma_{ji} \ll \Gamma_j. \quad (22)$$

[In the case of the cascade system of figure 2c, where the saturating transition lies lowest, condition (22) is not necessary.]

Thus the resonance doublet with frequency splitting  $\delta \left( 1 + \frac{2k_2}{k_1} \right)$  should appear *only in presence of specific relaxation processes* : radiative cascade processes, or dephasing collisions.

In this sense, this doublet may be considered as a type of *pressure-induced extra-resonance* (PIER). We will compare these resonances with Bloembergen's PIER [13] later on. First, let us consider the  $k_1 = k_2$  case in more detail. From equations (13)-(16), one gets

$$S_R(\delta) + S_R(\delta) = \frac{n_0 - n_2}{\Gamma + i\delta} \left[ \frac{1}{\Gamma_{01} + \Gamma_{02} + i \left( \Delta + \frac{3\delta}{2} \right)} \left( \frac{\Gamma_{01} + \Gamma_{02} - \Gamma_{12} - \Gamma_0}{\Gamma_{01} + \Gamma_{02} - \Gamma_{12} + i\delta} - \frac{\gamma_{20}}{\Gamma_2 + i\delta} \right) \right] + \frac{n_0 + n_2}{\Gamma_{01} + \Gamma_{02} - \Gamma_{12} + i\delta} \times \frac{1}{\Gamma_{12} + i \left( \Delta + \frac{\delta}{2} \right)}. \quad (23)$$

The pure Raman resonance has a linewidth  $\Gamma_{12}$ , and the doublet splitting is  $\delta$ . On the other hand, the mixed population/Raman contribution has a linewidth equal to  $\Gamma_{01} + \Gamma_{02}$  (doublet splitting  $3\delta$ ).

(i) For vanishing  $\delta$ 's ( $\delta \ll \Gamma_{ij}$ ), equation (23) yields the usual three-level saturation resonance [7, 9]. In general, its lineshape is not a pure Lorentzian. It is a Lorentzian only when conditions (21)-(22) are satisfied.

(ii) For  $\delta \gg \Gamma_{ij}$ , the  $\delta$  doublet is in quadrature with respect to the modulation of the saturating beam. Its amplitude is proportional to  $1/(i\delta)$ . The  $3\delta$  doublet is in phase, with an amplitude proportional to  $-(\Gamma_{01} + \Gamma_{02} - \Gamma_{12} - \Gamma_0 - \gamma_{20})/\delta^2$ . Note that, in this type of configuration, the contributions due to dephasing collisions and radiative cascades are of opposite sign since, in general,  $\Gamma_{01} + \Gamma_{02} \geq \Gamma_{12} + \Gamma_0$ .

## 5. Discussion.

This type of extra-resonances resorts to a larger class of pressure-induced extra-resonances, PIER. They can be compared to the ones studied by Bloembergen

*et al.* [13]. In the latter case, PIER's are observed — in presence of collisional relaxation — for *off-resonance* laser irradiation, when the difference between two incident laser frequencies matches a transition between two unpopulated excited states (or equally populated ground states). Collisional redistribution of radiation is closely related to those PIER's [14]. In comparison, the resonances discussed above present important differences :

(i) They are predicted for *resonant* irradiation, so that low-power c.w. lasers, and relatively low buffer-gas pressures (typically, a fraction of torr) could be used. They should allow one to analyse collisional processes in the impact regime (since the frequency detuning is nearly zero), in a pressure range different from the one usually explored with PIER resonances.

(ii) The predicted resonances are always *Doppler-free*, because of the velocity selection imposed by the resonant character of the interaction. In the case of off-resonance irradiation, all the velocity groups are excited, and the Doppler broadening may be elimi-

nated only in the case of a *collinear, nearly co-propagating* beam geometry, when all the incident frequencies are very close (collision-induced population, or Zeeman coherence effects [13, 15]).

It should be noted that, in the resonances analysed here, the velocity average plays a major role. In this sense, these resonances are intimately connected to the ones already studied in Doppler-free saturation spectroscopy. As noted above, the absence of PIER doublets in optical heterodyne three-level spectroscopy coincides with the presence of a single Lorentzian lineshape in standard three-level saturation spec-

troscopy. Another case of collision-induced extra signals has been pointed out long ago in time-resolved saturation spectroscopy. It has been predicted that collisional processes could be responsible for the appearance of additional coherent decaying beats in the transient evolution of saturation signals : they have been discussed in detail for coherent Raman beats [16], three-level free decay [12], three-level optical nutation [10], and two-level free decay [17]. It is clear that the above PIER's are the counterpart in the frequency domain, of these extra coherent beats in time-resolved saturation spectroscopy.

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