

## Line shapes of saturated absorption spectroscopy in optically thick media

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(Received 26 January 1998)

The line shapes of saturation spectroscopy in optically thick two-level media are calculated for atomic and molecular transitions. Line narrowing and distortion effects due to propagation are presented. For Doppler-broadened media it is shown that, due to multiphoton processes, the natural linewidth limits the ultimate narrowing that can be reached. Thus, up to  $\alpha_0 L = 20$ , only when the rate-equation approximation is valid subhomogeneous linewidths are expected. [S1050-2947(98)07708-7]

PACS number(s): 32.70.Jz, 32.80.Bx, 32.90.+a

### I. INTRODUCTION

Pump-probe spectroscopy of atoms and molecules has been studied for many years [1] and the line shape of Doppler free resonances investigated for optically thin [2–7] and for strongly absorbing media [7–15]. Basov *et al.* [7] and Rautian *et al.* [8] predicted that power broadened linewidths would show line narrowing in saturation spectroscopy in optically thick media. An increase in the propagation depth, which is equivalent to increasing the overall linear absorption of the cell, would produce narrower linewidths with high contrast transmission signal. Svanberg *et al.* [9] demonstrated this, performing saturated absorption experiments in an optically thick cell containing Na vapor and showed high contrast along with line narrowing below the natural spontaneous emission rate of the transition. Both conditions have been emphasized as advantageous for laser stabilization purposes. Experiments on infrared transitions in SF<sub>6</sub> have also shown line narrowing [12], but just to the point of eliminating the power broadening of the resonance.

The basic phenomenon behind power broadened narrowing is the attenuation of the pump beam due to propagation, which implies the saturation of a narrower class of atomic velocities. Another source of line narrowing acting on the probe transmission is the strong absorption occurring on the wings of the saturated resonance line. These effects, intuitively, might justify the subhomogeneous linewidths observed. Using the rate-equations approximation in their calculations Svanberg *et al.* [10] obtained subnatural linewidths. In their experiments in Na vapor, subnatural line narrowing did appear but associated with crossover resonances. The strictly two-level transitions observed in [10] never reached subnatural width. Recent experiments by Caiyan *et al.* [13] with two Na cells in tandem configuration do report strongly subhomogeneous linewidths, again for crossover resonances between hyperfine lines. The major difficulty in testing the two-level predictions in Na is the presence of optical pumping cycles due to its hyperfine levels. It is important to emphasize the difference between subnatural linewidth in an optical two-level transition and subnatural linewidth in a coupled three-level system. This last case may show “subnatural” lines whose widths are in fact associated to the sublevels coherence relaxation rates. These relaxations manifest themselves in Raman resonances for two copropagating

different light beams. Thus the corresponding narrow lines can only be used for precise comparison between two optical frequencies and not for absolute optical calibration purposes. Similar narrow lines also occur due to population pulsation resonance in open two-level systems [16].

As will be shown here, calculations for an optically thick medium of two-level atoms, taking into account multiphoton processes in the cross saturation of the probe transmission, do not give subnatural linewidths. An exception is when dephasing collisions dominate the transition dipole relaxation rate. In this particular case contributions from multiphoton processes are negligible, the rate equation is a good approximation and a linewidth below the homogeneous width can be attained. Baklanov and Chebotayev [2] and Haroche and Hartmann [5] showed, for the case where population decay is dominated by collisions, that the maximum percentage of transparency induced in a thin sample by a strong counterpropagating pump beam, for an infinitely Doppler-broadened transition, is 62% of the linear absorption as opposed to 100% predicted by the rate-equation approximation. Their results will be discussed here for the case of optically thick condition.

Preliminary account of the calculations have been presented earlier along with experiments done on a collision broadened molecular transition of SF<sub>6</sub> with radiation from a CO<sub>2</sub> laser [12]. In both experiments and calculations it was shown that the linewidth could not be less than the homogeneous width. The results are extended to calculations where spontaneous emission and phase interrupting collisions are the dominant mechanism of decay, restricted to two-level media and single sample cell. The line shapes were calculated as the spectral power added or subtracted to the probe beam by the induced polarization [5]. The probe has constant incident intensity and is always weak. Thus, no possibility is allowed for the generation of instabilities [17].

Optically thick media with multiple resonances were studied recently by many authors. The shift and the resolution of lines in Na vapor have been investigated by Zhankui *et al.* [11] who analyzed their data using the rate-equations approximation. Solutions using Maxwell-Bloch equations for optically thick three-level media have been reported in the literature by Schmidt-Iglesias *et al.* [14] who also studied theoretically the related problem of spectrally resolving independent pairs of nearby resonance lines [15]. The impor-

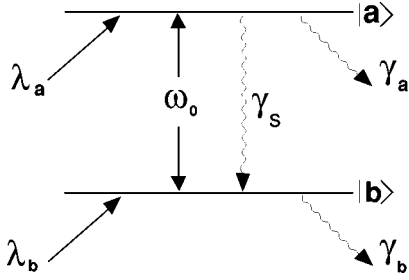


FIG. 1. Diagram of the two-level atom or molecule. The levels have pumping rates  $\lambda_a$  and  $\lambda_b$  and relaxation rates  $\gamma_a$  and  $\gamma_b$  to the nonresonant levels. The spontaneous emission rate from  $a$  to  $b$  is  $\gamma_s$ .

tant case of coupled three-level systems in an optical pumping configuration including crossover resonances, which is relevant to explain the narrow lines of the Na experiment in [10,13], is also left out of this work.

The restriction of the calculations to the case of homogeneously broadened transition brings in the line shapes with absorption and amplification features. The amplification of a probe in a two-level medium pumped by a strong near resonant field, observed for an atomic beam by Wu *et al.* [18] in the thin sample condition, can be enhanced in thick medium. The same possibility exists for wave mixing generation by related parametric processes, which have been discussed for thin media by Boyd *et al.* [19]. The work herein will not explore the details of gain generation when the homogeneous linewidth of the power broadened homogeneous linewidth is bigger than the Doppler, despite its recent interest and discussion in the literature [20]

## II. THE TWO-LEVEL SYSTEMS

The two-level systems composing the medium are represented in Fig. 1. The transition is assumed nondegenerate, with dipole matrix element  $\mu$  and central frequency  $\omega_0$ . For the upper level the relaxation is specified by two explicit coefficients:  $\gamma_s$ , the spontaneous emission rate of the transition, i.e., from level  $a$  towards level  $b$ , and  $\gamma_a$ , the decay rate towards the reservoir consisting of all other nonresonant levels. The decay rate for the lower level is  $\gamma_b$ . Dephasing collision relaxation on the transition dipole is given by the rate  $\gamma_{ph}$ . Three distinct cases will be discussed according to the relative values of the relaxation rates present: first, *relaxation dominated by the spontaneous emission, as in very low pressure atomic vapors*;  $\gamma_s \gg \gamma_a = \gamma_b \approx \gamma_{ph}$  then, *the infrared molecular transitions dominated by hard collisions where*  $\gamma_a = \gamma_b \gg \gamma_s \approx \gamma_{ph}$  and third the *transitions dominated by phase interrupting collisions* in which  $\gamma_{ph} \gg \gamma_a = \gamma_b \approx \gamma_s$ . In all cases  $\gamma_a = \gamma_b$  so that the two-level systems are closed, with the sum of the populations remaining constant. These are commonly found cases that exclude the species where optical pumping cycles occur. The relaxation rate for the transition coherence is

$$\gamma_{ab} = (\gamma_a + \gamma_b + \gamma_s)/2 + \gamma_{ph}. \quad (1)$$

The total field in the medium is given by the superposition of two plane waves

$$E(z,t) = \mathcal{E}_F(z) \cos(\omega_F t - k_F z) + \mathcal{E}_P(z) \cos(\omega_P t - k_P z), \quad (2)$$

where  $\mathcal{E}_F(z)$  and  $\mathcal{E}_P(z)$  are the amplitudes,  $\omega_F$  and  $\omega_P$  the angular frequencies, and  $k_F$  and  $k_P$  the wave vectors of pump and probe fields, respectively. The fields can be frequency tuned but their amplitudes before entering the absorbing medium are considered to have flat intensity spectra.

The density matrix equations for the two-level system are solved in the rotating-wave approximation [5]. The total optical polarization is obtained from the density matrix, which describes the medium under the action of the two fields:

$$P(z,t) = \int_{-\infty}^{\infty} \text{Tr}[\rho(z,t,v_z)\mu] dv_z, \quad (3)$$

where the terms of  $P(z,t)$  oscillating as  $\exp\{\pm(k_i z - \omega_i t)\}$  gives  $P_i(z,t)$  ( $i=F$  or  $P$ ). The general inhomogeneously Doppler-broadened case, with width  $\Delta\omega_D = k\mu\sqrt{\ln 2}$  [half-width at half maximum (HWHM)] can only be integrated numerically. Throughout the medium the intensity  $I_i \propto \mathcal{E}_i^2$  will vary as

$$\frac{dI_i}{dz} = -\alpha_i I_i = -\left\langle E_i(z,t) \frac{dP_i(z,t)}{dt} \right\rangle, \quad (4)$$

where  $\alpha_i$  is the beam absorption coefficient and  $\langle \rangle$  denotes time averaging.  $P_i(z,t)$  is the nonlinear polarization with spatial wave vector  $k_i$ .

Including only the strong pump field in the equation we get the nonlinear absorption coefficient  $\alpha_F$  of the pump beam [1]. In order to determine the intensity  $I_F(z)$  it is necessary to integrate its absorption along the cell between 0 and  $z$  for copropagating beams and between  $L$  and  $z$  for the counter-propagating beam configuration.

At each location the probe absorption coefficient  $\alpha_P$  was calculated by solving the equations in the presence of the strong pump and probe field to first order. It is the cross saturation of the pump on the probe absorption that contains the physics of interest here.

The line shapes for the saturated homogeneously broadened transitions depend on the Bennett hole width,

$$\Gamma_s^B = \gamma_{ab} \sqrt{1+S}, \quad (5)$$

where  $S = I_F(z)/I_{\text{sat}}$  is the dimensionless pump intensity and  $I_{\text{sat}}$  the saturation intensity defined as

$$I_{\text{sat}} = \hbar \gamma \gamma_{ab} c n \epsilon_0 / 2 \mu^2, \quad (6)$$

where  $n$  is the nonresonant refraction index,  $\epsilon_0$  the electric vacuum permeability, and

$$\gamma = 2(\gamma_a + \gamma_s) \gamma_b / (\gamma_a + \gamma_b) \quad (7)$$

is an effective relaxation rate. The power broadened linewidth is

$$\Gamma_s = \frac{\gamma_{ab}}{2} [1 + \sqrt{1+S}] \quad (8)$$

and the low and high saturation regimes occur for  $S \ll 1$  and  $S \gg 1$ , respectively.

The curves calculated for the transmitted probe intensity were normalized to incident intensity at  $z=0$ , and written as  $I_P/I_{P0}$ .

For the purpose of comparison of the signal linewidths as a function of the sample optical thickness, the values of the relaxation rates were always chosen such that  $\gamma_{ab}$  had the same value for different relaxation mechanisms. Thus, for the thin cell with weak pump power all the calculated signals show equal linewidths,  $2\gamma_{ab}$ .

### III. HOMOGENEOUSLY BROADENED TRANSITION

The homogeneously broadened cases occur for transversal pump-probe experiments with atomic beam [18], in very high pressure molecular gases, and within cold atoms media [21]. Velocity integration is unnecessary and the pump-probe signal in an optically thin sample does not depend on the propagation direction of the beams [16]. The geometric axial alignment of the probe with respect to the pump is irrelevant as long as the volumes being pumped and probed are the same.

In an optically thick cell the co-propagating pump-probe arrangement is not obviously equivalent to the counterpropagating one. The thick medium causes both the pump and the probe beam intensity to be attenuated through the cell. Thus the population difference and the level coherence created by the pump field depend on the position along the cell.

Physically, the total probe absorption is the integration (addition) of the accumulated induced absorptions from a stack of thin cells, covering the whole medium. As stated above, in a homogeneously broadened medium, the contribution from each thin cell is the same for both copropagating or counterpropagating beams, as long as the beam intensities are preserved. However, due to their non-negligible absorption the pump intensity and the probe intensity are different at each location of the thick cell, when one changes from copropagating to counterpropagating arrangement. For a weak probe though the pump is not disturbed. The induced probe change signal is always the integration of a signal on a probe that is attenuated from its entrance into the cell to a specific slice, then changed by the pump at that specific slice, and then again attenuated through its way out of the cell. For instance, in a region of strong pump, the probe line shape is produced at a certain slice near the cell entrance of the pump on a copropagating probe that will be attenuated afterwards, when it proceeds through the cell, or is created on the same slice, near the cell entrance of the pump, but on a probe that has already been attenuated because it came from the other side, in a counterpropagating alignment. The overall integration, calculated in first-order perturbation on the probe beam amplitude, ends up the same, showing that the thick medium condition does not break the directional symmetry because the probe is not disturbing the pump. Conversely, the inhomogeneously Doppler broadened transition already shows the directional asymmetry in the thin cell condition.

In contrast with the Doppler broadened case, the output probe intensity for a homogeneous broadening condition has been numerically calculated considering the pump beam fixed at resonance,  $\omega_F = \omega_0$ , and high input pump intensity,  $I_F = 100I_{\text{sat}}$ . This is the same pump power used to calculate narrow lines in the Doppler-broadened condition of the next

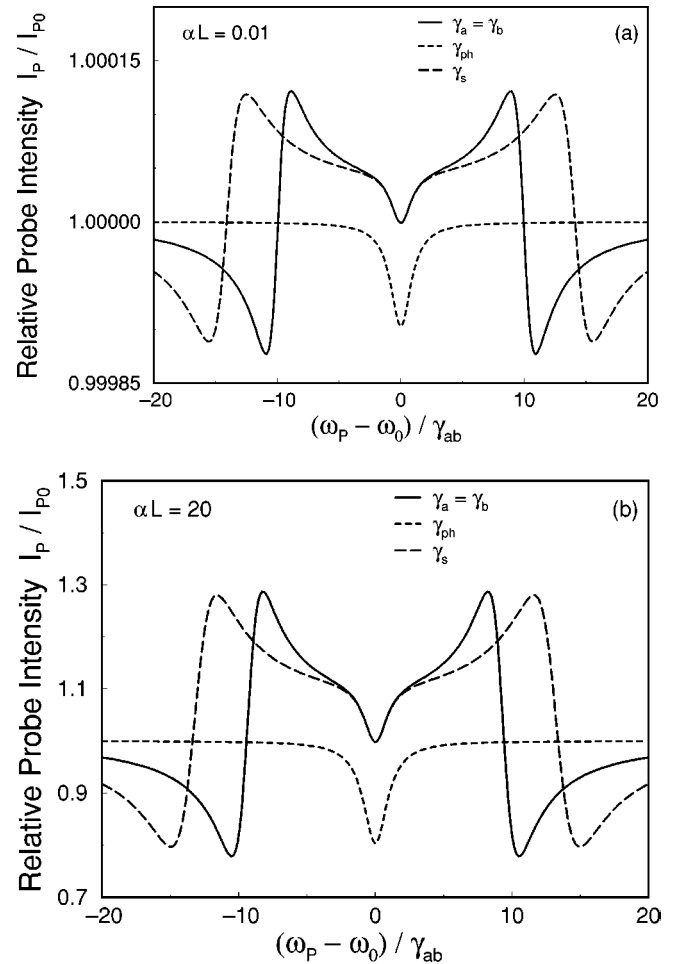


FIG. 2. Line shapes of saturated absorption in the homogeneously broadened medium for high saturation intensity,  $I_F = 100I_{\text{sat}}$ . (a) For an optically thin cell,  $\alpha_0L = 0.01$  and (b) for an optically thick cell,  $\alpha_0L = 20$ .  $I_P/I_{P0}$  is the output probe intensity normalized to its incident intensity. The short dashed line corresponds to relaxation dominated by phase interrupting collisions  $\gamma_{ab} = \gamma_{\text{ph}}$  (when the rate-equation approximation is valid). The long dashed and the solid lines correspond to spontaneous emission,  $\gamma_{ab} = \gamma_s$ , and to hard collisions,  $\gamma_{ab} = \gamma_a = \gamma_b$ , as the main relaxation mechanisms, respectively.

sections. The result is shown in Fig. 2(a) for the thin cell, with optical thicknesses  $\alpha_0L = 0.01$  and in Fig. 2(b) for the thick cell, with  $\alpha_0L = 20$ . In both cases copropagating and counterpropagating configurations give the same line shapes.

For transitions with relaxation dominated by dephasing collisions power broadened Lorentzian lines result. However, the coherent processes under high pump power induce extra features to the line, including the possibility of gain without population inversion. These line shapes have been predicted and were observed in atomic beam experiments [18]. The occurrence of amplification, when  $I_F/I_{\text{sat}} \gg 1$  in Fig. 2(a), for strong pumping near and on resonance of a two-level thin medium has been discussed by various authors [1,5] and can be associated with the dynamic Stark splitting of the transition as well as the stimulated Raman and Rayleigh emission processes [5]. This line shape has also been studied by Boyd *et al.* [19] to describe parametric gain in four-wave mixing on two-level systems. The line shape of Fig. 2(b) shows that significant gain, on the order of 30% at

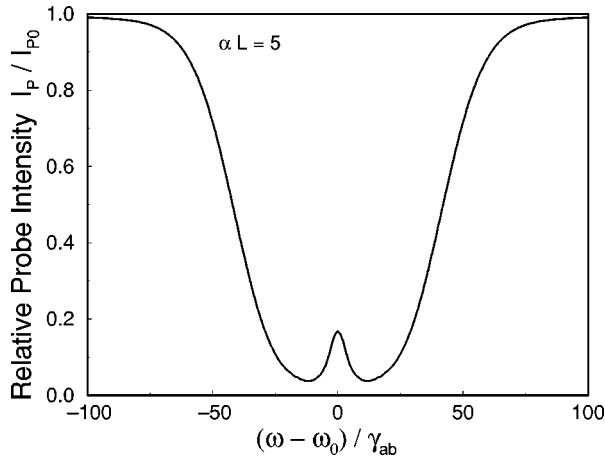


FIG. 3. Counterpropagating saturated absorption signal in a Doppler-broadened medium. The levels' populations relaxation rates were taken equal  $\gamma_a = \gamma_b$  and  $\gamma_s = \gamma_{ph} = 0$ , as in the hard collision model for molecules. For these curves we used  $\Delta\omega_D = 50\gamma$ ,  $I_F = 100I_{sat}$ , and  $\alpha_0 L = 5$ .

$\pm 10$  linewidths probe detuning, appear in a thick cell, with  $\alpha_0 L = 20$  pumped by  $I_F/I_{sat} = 100$ , while the gain was less than 0.015% for the same pump conditions in the thin cell case of Fig. 2(a). Thus the pump-probe line shapes in optically thick homogeneously broadened media do not depend on the relative beams direction unless the probe beam goes off the weak intensity condition. The main goal of this work is the line narrowing in Doppler-broadened media and so further discussion of the line shapes for the homogeneously broadened media will be treated elsewhere.

#### IV. COUNTERPROPAGATING BEAMS IN DOPPLER-BROADENED TRANSITIONS

The inhomogeneously broadened case also presents strong effects due to coherent processes. Those process play a key role in determining the signals line shapes and particularly their ultimate linewidths. They are responsible for breaking the symmetry between copropagating and counterpropagating beam configuration [1]. Such asymmetry is due to different Doppler selection of resonance in stimulated light scattering from a two-level atom in a strong field, as explained by Baklanov and Chebotayev [4]. In the rate equation limit all these coherent effects are washed out. The thick medium condition within the weak probe approximation just enhances the asymmetry existing for thin cells, preserving the essential features of the line shapes.

The Doppler-broadened transition, common in atomic vapors, was calculated considering a single laser producing the pump and the probe laser beams. Thus  $\omega_F = \omega_P = \omega$  is the frequency tuned through the resonance at  $\omega_0$ . The Doppler width is assumed always dominant ( $\Delta\omega_D \gg \gamma_{ab}, \Gamma_S$ ) and the two incident beams have constant intensity tunable over the Doppler line. With high incident intensity the strong beam initially propagates losing power at a constant rate. After its pump intensity reaches a value below the medium saturation intensity the usual exponential beam absorption takes place. Spectrally, a sharp edged profile, due to saturation contrast enhancement on the slope of the Doppler width appears with nearly the inhomogeneous linewidth,  $\Delta\omega_D$ .

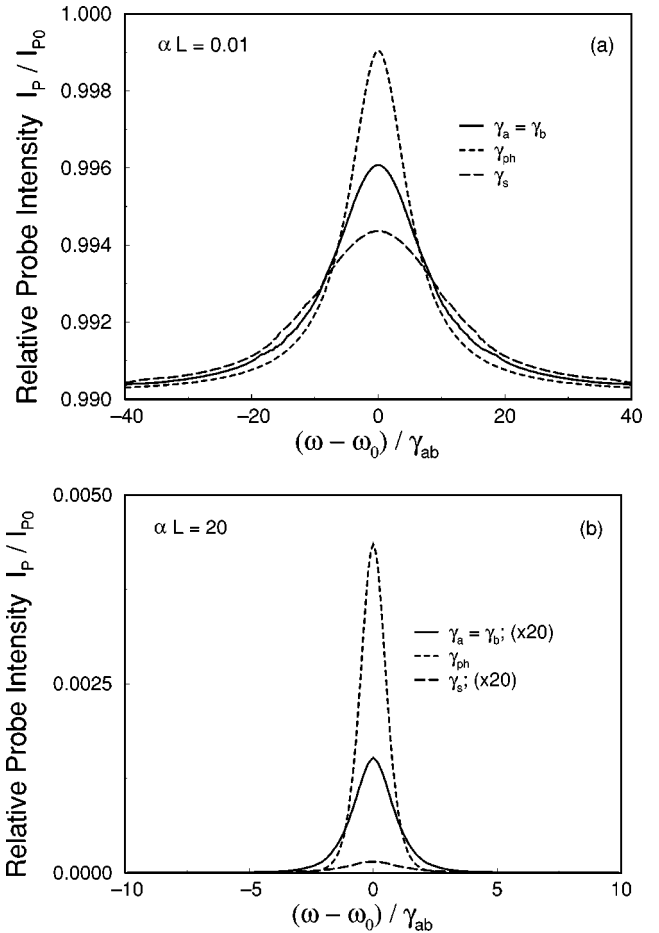


FIG. 4. Probe beam saturation line shapes calculated in the Doppler limit,  $\Delta\omega_{Doppler}/\gamma_{ab} \rightarrow \infty$ , (a) for an optically thin cell and (b) for a thick cell. The three limits of relaxation conditions, the optical thickness and the pump beam intensity are as in Fig. 2. Two vertical scales in (b) were expanded with respect to (a).

The probe beam spectrum, shown in Fig. 3, was calculated for the condition  $\Delta\omega_D = 50\gamma$  ( $\gamma_a = \gamma_b = \gamma$  and  $\gamma_s = \gamma_{ph} = 0$ ) and for a cell depth  $\alpha_0 L = 5$ . For the high input pump intensity used in the calculations,  $I_F = 100I_{sat}$ , a wide power broadened probe signal line would have resulted for the case of a thin cell. At the other extreme case, that is, for a thick cell with  $\alpha_0 L = 20$ , the results show a very narrow and very small sub-Doppler signal.

Because the intensity of the pump beam decreases as it propagates through the medium, the probe signal, which would have a power broadened linewidth in a thin cell, will have its line narrowed as the sample becomes thicker, other parameters being kept unchanged. This narrowing from power-broadened width down to the homogeneous width is intuitive and has been observed experimentally for atoms [10] and molecules [12]. Crossing the natural width barrier by further propagation narrowing was discussed previously based on results of rate-equation calculation [10].

Figure 4 shows the line shapes of the saturated absorption in the case of very strong pump input and in the Doppler limit ( $\Delta\omega_D/\gamma_{ab} \rightarrow \infty$ ). Three spectra are given in Fig. 4(a) for the optically thin condition ( $\alpha_0 L = 0.01$ ), corresponding to the limits stated in Sec. II. For comparison purposes the values chosen for the different  $\gamma_i$  was always such that  $\gamma_{ab}$

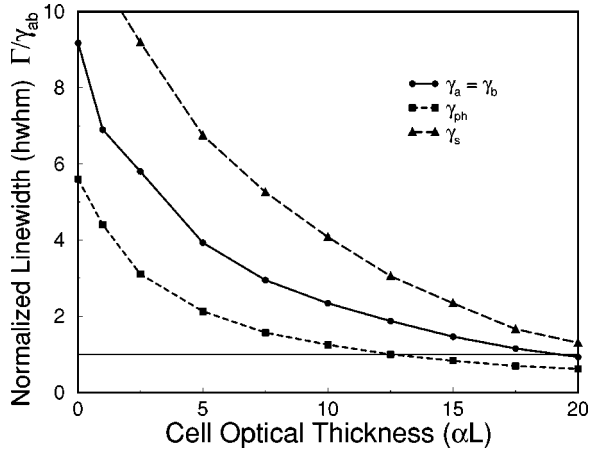


FIG. 5. Linewidths of saturated absorption signal calculated for the Doppler-broadened medium in a counterpropagating configuration as a function of the cell optical thickness. The three limits of relaxation conditions and pump beam intensity are as in Fig. 2. Notice that only for the transitions with relaxation dominated by phase interrupting collisions does the width ratio become less than unit, i.e., the line becomes narrower than the homogeneous line  $\gamma_{ab}$ .

had the same value. Therefore all lines in the low pump power optically thin cell have the same width. One can see in Fig. 4(a) that for optically thin sample conditions, three different saturation broadenings are present.

For the case of phase interrupting collisions multiphoton processes vanish, and the result is the same as obtained using the rate-equation approximation. This limit of fast relaxation of the optical polarization corresponds to a physical situation where the slow population rates drive the system behavior and coherencies washout fast. The increase in transparency, i.e., the decrease in absorption at line center induced by the pump, is the highest and the power broadening is the weakest as shown in Fig. 4(a). For higher input pump intensities, the transmission evolves to full transparency at resonance because the populations of the two levels of the transition are equalized by the pump beam.

The spectra for hard collision and spontaneous emission relaxation are also shown in Fig. 4(a). For the thin cell condition [1] the saturated absorption coefficients are such that at resonance only 62% and 45% of full transparency can be reached respectively. Thus, for both cases the center portion of the resonance is less transparent than the one resulting from rate-equation approximation. At the same time the power-broadened linewidths are larger as shown in the heavy and long dashed lines of Fig. 4(a). These effects of different transparency peaks and linewidths are preserved in the optically thick cell limit.

Figure 4(b) contains the line shapes for the thick media condition. The narrowest and highest line is for the phase interrupting collision case. Notice that the induced transparency at resonance condition is two orders of magnitude higher than the induced transparency for the cases where the relaxation is dominated by hard collisions or by spontaneous emission.

The power-broadened line narrows with increasing cell length, all other parameters remain unchanged. This can be seen in Fig. 5 where the calculated linewidths corresponding

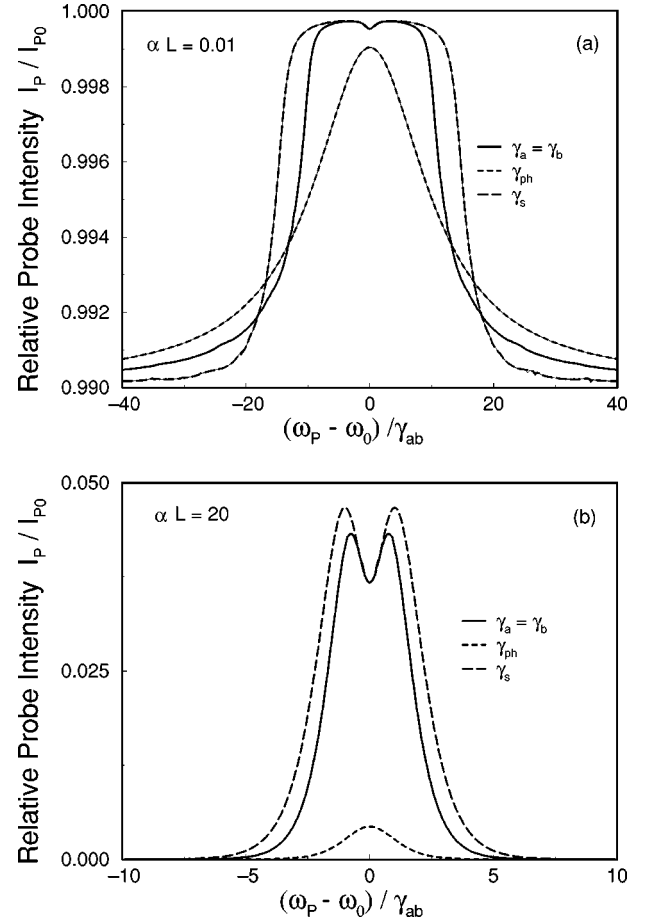


FIG. 6. Line shape of saturated absorption spectra for copropagating beams calculated for the Doppler-broadened medium. The pump beam frequency is fixed, equal to the transition frequency  $\omega_0$ , and the probe frequency was tuned. Other conditions are as in Fig. 4.

to spectra like that of Fig. 4 were plotted for  $\alpha_0 L$  varying between 0 and 20. The hard collision case, was experimentally studied using an SF<sub>6</sub> resonance and radiation from a CO<sub>2</sub> laser [12] and gives a linewidth reaching the limit  $\gamma_{ab} = \gamma$  for  $\alpha_0 L = 20$ . To our knowledge, the spontaneous emission atomic transition case for a pure two-level atom has never been reported experimentally. Its calculated line narrowing as seen in the dashed line of Fig. 5 is larger than the natural linewidth ( $\gamma_s/2$ ). The contrasts of the signals [9,10,12] in the three cases follow the inverse of the linewidths [22]. Therefore, the two-level atomic case will have the least narrowing and the least contrast in its signal.

## V. COPROPAGATING BEAMS

The ideal arrangement for copropagating pump-probe spectroscopy in Doppler-broadened media [3,4] is the non-degenerate case, where the pump frequency  $\omega_F$  is different from the probe frequency  $\omega_P$ . Spectral lines result from the tuning of  $\omega_P$ , keeping  $\omega_F$  fixed. Figure 6(a) shows the calculated line shapes for resonant pump frequency,  $\omega_F = \omega_0$ , and for high input pump intensity  $I_F = 100I_{\text{sat}}$  in an optically thin cell. Now, opposite to what happened with counterpropagating beams described in the previous section, the solution for the case of dominant dephasing collision given by

the short dashed line is the one that has the least induced transparency. Its linewidth is also less broadened than the others. In fact this line remains Lorentzian and its width should be equal to the corresponding one of Fig. 4(a), that is, for the counterpropagating condition. The only difference observed is a factor of two in frequency scale because in the counterpropagating spectra the pump frequency was also tuned along with the probe, both equal to  $\omega$ . Figure 6(a) also shows the known fact of how in the copropagating pump-probe condition, as opposed to the counterpropagating case, the multiphoton processes distort the line shape from its low saturation Lorentzian shape. These peculiar line shapes with sharp edges and a flat top with a dip have been calculated many years ago by Baklanov and Chebotayev [4], in the optically thin medium condition.

For thick medium,  $\alpha_0 L = 20$ , the calculated line shapes behave according to Fig. 6(b). Again, for transitions dominated by phase interrupting collisions, the resulting line is similar to the one for thin cells. Its width is narrowest as in the counterpropagating case (observe always the factor of two in frequency scale as explained above). For the other two cases the optically thick cell enhances the on-resonance dips of the line spectra. The induced transparency is much more significant for the atomic transitions dominated by spontaneous emission than the ones dominated by phase interrupting collisions. The small extra percentage of induced transparency for the thin cell condition once again leads to one order of magnitude difference for the thick medium condition. The signal contrast in the copropagating case was not systematically studied here. In particular, the conditions of finite Doppler width where probe amplification has been predicted [5] in thin cell deserve further detailed investigation for optically dense media [20].

## VI. CONCLUSIONS

To summarize our results, the semiclassical density matrix calculation for pump-probe energy transfer in a resonant two-level medium was done for optically thick medium conditions, determining the associated spectral line shapes with their propagation narrowing effects. Numerical integration of the probe beam has been done for homogeneously broadened

and Doppler-broadened gas media under conditions of relaxation rates describing atomic media with pure radiative decay, molecular media with homogeneous width dominated by hard collisions, and atomic or molecular media in the presence of a buffer gas, where phase interrupting collisions rates are the fastest and the rate-equation approximation is valid.

The homogeneously broadened thick media shows line shapes symmetrical with respect to copropagating versus counterpropagating configurations. The non-rate-equation approximation cases, when strong pumping is on resonance, show line shapes with an enhancement on both the absorption and the amplification portions of the probe line. The effects are the same known from thin media experiments [18]. The detuned pump condition, when the induced transparency [23] may turn into an important probe amplification [18,5] for thin cell, might give further enhancement in the gain effect for the optically thick medium. The strength attainable for such Raman-type gain in thick media was recently discussed by Brown *et al.* [20] in the limit of no pump absorption and with the power-broadened homogeneous linewidth close to the Doppler width.

In the Doppler broadened case, line narrowing due to propagation was calculated and only the case when the transition decay is dominated by phase interrupting collisions, where the rate-equation approximation is valid, lead to “sub-natural,” or better, subhomogeneous (less than  $\gamma_{ab}$ ) linewidth in the optically thick media. Though not shown in this article, signal contrast for counterpropagating arrangement was also calculated, and verified to be overestimated when the rate-equation approximation is used. Copropagating configuration, on the other hand, when the pump is set fixed on resonance and the probe is tuned, gives much higher induced transparency for the radiative relaxation limit.

## ACKNOWLEDGMENTS

The work was partially supported by Brazilian Agencies: Conselho Nacional de Pesquisa e Desenvolvimento (CNPq), Financiadora de Estudos e Projetos (FINEP), and Fundação de Amparo a Pesquisa do Estado de Pernambuco (FACEPE).

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