Theory of resonances in four-wave mixing resulting from velocity-changing collisions

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(Received 28 December 1987)

A theory of four-wave mixing including effects of velocity-changing collisions is presented. Three fields with frequencies ω, ω, and $\omega+\delta$ are incident on a vapor of "two-level" atoms having upper state b and lower state a. Two of the fields are counterpropagating and the third (of frequency $\omega+\delta$) makes a small angle with one of the others. The frequency ω is a nearly resonant (inside the Doppler width) with the a-b transition frequency. The phase-conjugate signal emitted at frequency $\omega-\delta$ is calculated as a function of δ. Using a simple collision model in which collisions are phase interrupting in their effect on atomic coherence and velocity-changing in their effect on level populations, we discuss the conditions under which resonances characterized by the upper or lower radiative and collision rates can be observed. Assuming that the total $(a+b)$ state population is conserved in the absence of collisions, it is shown that velocity-changing collisions can "open" the system and lead to a resonance characterized by the lower-state width (convoluted with the residual Doppler width). With increasing pressure, the width of this induced resonance structure decreases monotonically. For sufficiently high pressure, the collisional redistribution of velocity classes is complete—the system "recloses" and the narrow resonance disappears. The interplay of the collision-induced opening, line narrowing, and reclosing of the system is discussed, as is the relationship of these narrow resonances to the so-called pressure-induced extra resonances of Bloembergen and co-workers [Indian J. Pure Appl. Phys. **16**, 151 (1978); Phys. Rev. Lett. **46**, 111 (1981)].

I. INTRODUCTION

The phenomenon of pressure-induced extra resonances observed via four-wave mixing is a subject area that has received a great deal of attention following its prediction[1] and experimental verification[2] by Bloembergen and co-workers. Both theoretical and experimental developments in pressure-induced resonances have been reviewed recently.[3]

Pressure-induced resonances refer to resonant structures that appear only in the presence of collisions. They can be observed under a wide variety of experimental configurations. To observe pressure-induced resonances via four-wave mixing in a "two-level" atom, three laser fields having frequencies ω, ω, and $\omega+\delta$ may be used. As the detuning δ is varied, the pressure-induced resonances appear as structures centered at $\delta=0$ with linewidths characterized by the spontaneous decay rates of the two levels. When one of the transition levels is the ground state, it is possible to observe very narrow resonances, whose widths are limited by transit time or residual Doppler broadening.

Most theoretical treatments of the problem have been restricted to homogeneously broadened atomic samples or to situations in which the atom-field detunings are much larger than the Doppler width. Many of the experiments were carried out for this range of detunings. In this limit atoms in all velocity subclasses essentially contribute equally to line-shape formation. If the atom-field detunings are less than the Doppler width, atoms which are Doppler shifted into resonance with the field are preferentially excited. The physical interpretation of the four-wave mixing signals differs significantly for nearly resonant fields (detuning less than the Doppler width) than for the large-detuning case.

There have been a number of experiments carried out with nearly resonant fields.[4,5] A theoretical analysis of these experiments, including effects of velocity-changing collisions and residual Doppler broadening (owing to a slight angle between two of the beams), has not been carried out to our knowledge. Lam *et al.*[5] did discuss the effect of velocity-changing collisions but did not include the residual Doppler broadening. Rothberg and Bloembergen[6] discussed the collisional narrowing of the residual Doppler broadening of the resonances (for the highly detuned case), giving the expected dependence of the resonances's width and amplitude on perturber pressure. Most other theoretical approaches neglect the residual Doppler broadening and treat collisions solely by the introduction of a number of collision rates.

It is the purpose of this paper to analyze collision-induced features that may appear in four-wave-mixing line shapes. We consider a two-level atom subjected to three fields having frequencies ω, ω, $\omega+\delta$ and calculate the signal emitted by the sample at frequency $\omega-\delta$ as a function of δ. The atom-field detuning $\Delta=\omega-\omega_0$ (ω_0 is the atomic transition frequency) is less than the Doppler width associated with the transition.

In particular, we examine in detail the conditions under which one can observe resonances characterized by the natural widths γ_a and γ_b, associated with the lower and upper transition levels, respectively. In the limit that a is the ground state, it will be seen that a resonance with width γ_a can be observed only when the system is "open" (population not conserved). Velocity-changing collisions provide a mechanism for opening the system. The total population of *each* atomic velocity class need no longer remain "closed" when collisions are present, even if the total (velocity-integrated) population does remain closed. Thus, velocity-changing collisions contribute directly to the resonance having width γ_a. The resonance having width γ_b occurs for open or closed systems, with or without collisions, for nearly resonant tuning.

Velocity-changing collisions not only lead to the appearance of the γ_a resonance, but are also responsible for the narrowing of the residual Doppler broadening. The way in which velocity-changing collisions result in opening, collisional narrowing, and the ultimate closing of the system is explored.

The calculations are carried out in lowest-order perturbation theory, using a highly simplified collision model ("strong" velocity-changing collisions for populations; homogeneous collisional decay for atomic coherence). A preliminary version of this work has appeared.[7]

In the first part of the paper we carry out a straightforward calculation of the four-wave-mixing signal, after having introduced our definitions of open and closed systems. In the second half of the paper we analyze the results, emphasizing the dependence of the γ_a resonance on the opening, narrowing, and ultimate closing produced by velocity-changing collisions.

II. CALCULATION OF THE SIGNAL

The system we consider in this paper is a collection of two-level atoms, interacting in a classical four-wave-mixing geometry with three laser beams, whose electric fields are labeled by $\mathbf{E}_f$ (forward) $\mathbf{E}_b$ (backward), and $\mathbf{E}_p$ (probe), as shown in Fig. 1. Fields $\mathbf{E}_f$ and $\mathbf{E}_b$ have the same frequency ω and are counterpropagating (wave vectors $\mathbf{k}_0$ and $-\mathbf{k}_0$, respectively), while field $\mathbf{E}_p$ has frequency $\omega+\delta$ and has a wave vector $\mathbf{k}$ which is directed along an axis at angle θ to $\mathbf{k}_0$. The $|a\rangle$ (lower) and $|b\rangle$ states are separated in energy by $\hbar\omega_0$. Both of them may be excited levels, although the interesting case, with which we are mainly concerned, is a closed system in which $|a\rangle$ is a ground state and $|b\rangle$ an excited state that can decay radiatively only via spontaneous emission to state $|a\rangle$. For reasons that will become clear in Sec. III, we adopt a slightly modified definition of a closed system. If γ_a and γ_b are, respectively, the radiative lifetimes of $|a\rangle$ and $|b\rangle$, and $\gamma_{b,a}$ is the radiative transfer rate from $|b\rangle$ to $|a\rangle$, the system is said to be closed if

$$\gamma_b=\gamma_{b,a}+\gamma_a \ . \tag{1}$$

Condition (1) implies that there is only one decay rate (which can be zero) for the sum of the populations of states $|a\rangle$ and $|b\rangle$. In other words, the total population is not conserved within an overall decay constant γ_a which may differ from zero. If, for example, states $|a\rangle$ and $|b\rangle$ have the same transit time in the laser beams, condition (1) could hold with γ_a equal to the inverse transit time. The motivation behind this definition of a closed system is discussed in Sec. III.

The atoms form a gas with a classical velocity distribution, and the internal state of a group of atoms having the velocity $\mathbf{v}$ is described by the density operator $\rho(\mathbf{v})$. In the absence of interaction with the lasers, the variation of $\rho(\mathbf{v})$ as a function of $\mathbf{v}$ is proportional to the Gaussian thermal distribution

$$W(\mathbf{v})=\frac{1}{(u\sqrt{\pi})^3}e^{-v^2/u^2} \ , \tag{2}$$

where u is the most probable atomic speed.

These atoms are immersed in a buffer gas of foreign atoms with no active structure. The active-atom density is assumed to be sufficiently low that one need consider only active-atom–foreign-gas-atom collisions. To easily account for these collisions, we make the classical set of assumptions which defines the so-called collisional and radiative impact regime. The validity conditions for the impact approximation can be stated as[8]

$$\begin{aligned}&\tau_c \ll \Omega_i^{-1}, |\omega-\omega_0|^{-1}, |\omega+\delta-\omega_0|^{-1} \ , \\ &\tau_c \ll \Gamma^{-1}, \gamma_b^{-1}, \gamma_a^{-1} \ ,\end{aligned} \tag{3}$$

where τ_c is the duration of a typical collision, Ω_i is any relevant Rabi frequency, and Γ is a macroscopic collisional rate. In this framework the evolutions under collisional and radiative interactions are decoupled and the collisions can be simply described by an additional time derivative in every equation governing a matrix element $\rho_{\alpha\beta}(\mathbf{v})$ $(\alpha=a,b;\ \beta=a,b)$ given by[9]

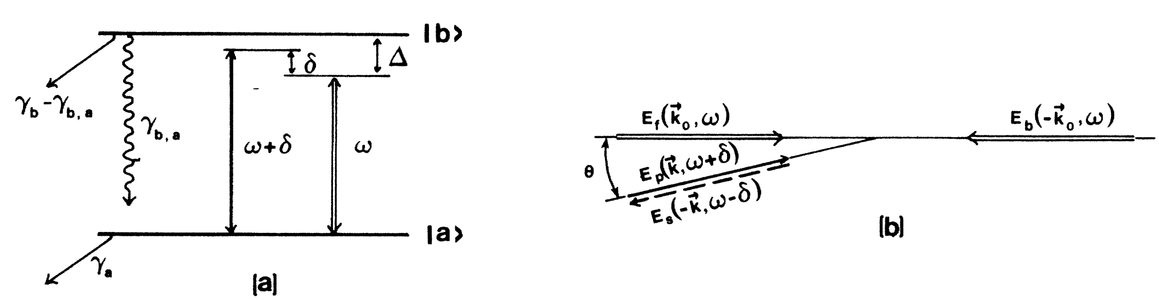

FIG. 1. (a) Level scheme for the system under consideration. For the laser frequencies ω and $(\omega+\delta)$ shown, the detuning $\Delta=(\omega-\omega_0)$ is within the Doppler width of the transition. (b) The geometry of the three input laser beams (E_f,E_b,E_p). The phase-conjugate signal, $E_s(-\mathbf{k},\omega-\delta)$, is shown as a dashed arrow.

$$\left[\frac{\partial}{\partial t}\rho_{\alpha\beta}(\mathbf{v})\right]_{\text{coll}}=-\Gamma^{\text{ph}}_{\alpha\beta}(\mathbf{v})\rho_{\alpha\beta}(\mathbf{v})(\delta_{\alpha a}\delta_{\beta b}+\delta_{\alpha b}\delta_{\beta a})-\Gamma_{\alpha\beta}(\mathbf{v})\rho_{\alpha\beta}(\mathbf{v})+\int A_{\alpha\beta}(\mathbf{v}'\to\mathbf{v})\rho_{\alpha\beta}(\mathbf{v}')d^3v' , \quad (4)$$

where $A_{\alpha\beta}(\mathbf{v}'\to\mathbf{v})$ is a collision kernel, $\Gamma_{\alpha\beta}(\mathbf{v})=\int A_{\alpha\beta}(\mathbf{v}\to\mathbf{v}')d^3\mathbf{v}'$ is a collision rate, and $\Gamma^{\text{ph}}_{\alpha\beta}$ is a (complex) decay rate associated with impact pressure broadening theories involving phase-interrupting collisions.[9] This expression, which accounts for the velocity changes of atoms during collisions, yields a coupling between different velocity classes through the collision kernel $A_{\alpha\beta}(\mathbf{v}'\to\mathbf{v})$. The kernel, which is directly related to integrals of quantum-mechanical scattering amplitudes, has no general analytical form.

The resulting equations of motion for $\rho_{\alpha\beta}(\mathbf{v})$ cannot be solved analytically unless we model the kernels in a way that permits such a solution. To carry out the illustrative calculations in this paper, we choose simple forms for the kernels as follows.

(1) For the off-diagonal element ρ_{ab}, we neglect the contributions from the second and third terms of Eq. (4). This is a very common approximation, justified when the interaction potentials are somewhat different for the two levels, which is common of atomic optical transitions.[9] We keep only a rate of destruction of ρ_{ab},

$$\left[\frac{\partial}{\partial t}\rho_{ab}(\mathbf{v})\right]_{\text{coll}}=-\Gamma^{\text{ph}}_{ab}\rho_{ab}(\mathbf{v}) , \quad (5)$$

and we furthermore assume that Γ^{ph}_{ab} is real and does not depend upon $\mathbf{v}$.

(2) For the populations ρ_α, we adopt the so-called strong collision model in which the velocity $\mathbf{v}$ of an atom is thermalized, on average, after one collision, regardless of the initial velocity $\mathbf{v}'$. The collision kernel is given by

$$A_\alpha(\mathbf{v}'\to\mathbf{v})=\Gamma_\alpha W(\mathbf{v}), \quad \alpha=a,b \quad (6)$$

with Γ_α independent of $\mathbf{v}$.

The equations of motion (in the interaction representation) for the atomic-density matrix elements, including the atom-field and collisional interactions, are

$$\left[\frac{\partial}{\partial t}+\mathbf{v}\cdot\nabla+\tilde{\gamma}_b\right]\rho_b(\mathbf{v})=\lambda_b W(\mathbf{v})+\left[i\rho_{ab}(\mathbf{v})\sum_\nu\frac{\Omega_\nu}{2}e^{-i\phi_\nu}+\text{c.c.}\right]+\Gamma_b W(\mathbf{v})\bar{\rho}_b ,$$

$$\left[\frac{\partial}{\partial t}+\mathbf{v}\cdot\nabla+\tilde{\gamma}_a\right]\rho_a(\mathbf{v})=\lambda_a W(\mathbf{v})+\gamma_{b,a}\rho_b(\mathbf{v})-\left[i\rho_{ab}\sum_\nu\frac{\Omega_\nu}{2}e^{-i\phi_\nu}+\text{c.c.}\right]+\Gamma_a W(\mathbf{v})\bar{\rho}_a , \quad (7)$$

$$\left[\frac{\partial}{\partial t}+\mathbf{v}\cdot\nabla+\tilde{\gamma}_{ab}-i\omega_o\right]\rho_{ab}(\mathbf{v})=i[\rho_b(\mathbf{v})-\rho_a(\mathbf{v})]\sum_\nu\frac{\Omega_\nu}{2}e^{-i\phi_\nu} ,$$

where the summation of ν applies to the three laser beams, $\phi_\nu=\omega_\nu t-\mathbf{k}_\nu\cdot\mathbf{R}$, $\Omega_\nu=\mu_{ab}E_\nu/\hbar$ (Rabi frequency), and λ_b,λ_a account for a possible external incoherent pumping of populations (for a closed system: $\lambda_b,\lambda_a\sim 0$ with $\lambda_a/\gamma_a\sim n_a$, unperturbed population of $|a\rangle$). In Eqs. (7) we have defined

$$\tilde{\gamma}_\alpha=\gamma_\alpha+\Gamma_\alpha, \quad \alpha=a,b , \quad (8a)$$

$$\tilde{\gamma}_{ab}=\tfrac{1}{2}(\gamma_a+\gamma_b)+\Gamma^{\text{ph}}_{ab} , \quad (8b)$$

$$\bar{\rho}_\alpha=\int d^3v\rho_\alpha(\mathbf{v}), \quad \alpha=a,b . \quad (8c)$$

Equations (7) are solved using a perturbative expansion up to third order in the field amplitudes. Among the 27 contributions to $\rho^{(3)}_{ab}$, two correspond to emission of an electromagnetic wave counterpropagating along the probe direction, with a frequency ω-δ and a wave vector $-\mathbf{k}$ (the so-called phase-conjugated emission). The phase-conjugate contribution to $\rho^{(3)}_{ab}$, obtained from a perturbative solution of Eqs. (7), is[7]

$$\rho^{(3),\text{PC}}_{ab}=\sigma_{ab}e^{i[(\omega-\delta)t+\mathbf{k}\cdot\mathbf{R}]} , \quad (9a)$$

with

$$\begin{aligned}\sigma_{ab}=-in\frac{\Omega_f\Omega_p^*\Omega_b}{8}[\tilde{\gamma}_{ab}+i(\Delta-\delta+\mathbf{k}\cdot\mathbf{v})]^{-1}\\ \times\Bigg[W(\mathbf{v})\{(1+R)[\tilde{\gamma}_b-i\delta+i(\mathbf{k}-\mathbf{k}_0)\cdot\mathbf{v}]^{-1}+(1-R)[\tilde{\gamma}_a-i\delta+i(\mathbf{k}-\mathbf{k}_0)\cdot\mathbf{v}]^{-1}\}\\ \times\{[\tilde{\gamma}_{ab}+i(\Delta-\mathbf{k}_0\cdot\mathbf{v})]^{-1}+[\tilde{\gamma}_{ab}-i(\Delta+\delta-\mathbf{k}\cdot\mathbf{v})]^{-1}\}\\ +W(\mathbf{v})\Bigg\{\tilde{\Gamma}_b(1+R)\bar{\mathcal{S}}_b[\tilde{\gamma}_b-i\delta+i(\mathbf{k}-\mathbf{k}_0)\cdot\mathbf{v}]^{-1}\\ +\tilde{\Gamma}_a\left[(1-R)\bar{\mathcal{S}}_a+R\left[\frac{1}{\Gamma_b}-\frac{1}{\Gamma_a}\right]\tilde{\Gamma}_b\bar{\mathcal{S}}_b\right][\tilde{\gamma}_a-i\delta+i(\mathbf{k}-\mathbf{k}_0)\cdot\mathbf{v}]^{-1}\Bigg\}\Bigg]\\ +(\text{same with }\mathbf{k}_0\to-\mathbf{k}_0) . \quad (9b)\end{aligned}$$

For the sake of compactness, we have introduced in Eqs. (9) the notations

$$R=\frac{\gamma_{b,a}}{\tilde{\gamma}_b-\tilde{\gamma}_a}\ , \tag{10a}$$

$$\tilde{\Gamma}_\alpha=\frac{\Gamma_\alpha}{1-\Gamma_\alpha\int\frac{W(\mathbf{v})d^3v}{\tilde{\gamma}_\alpha-i\delta+i(\mathbf{k}-\mathbf{k}_0)\cdot\mathbf{v}}}\ , \tag{10b}$$

$$\mathcal{S}_\alpha=\int\frac{W(\mathbf{v})d^3v}{\tilde{\gamma}_\alpha-i\delta+i(\mathbf{k}-\mathbf{k}_0)\cdot\mathbf{v}}\left[\frac{1}{\tilde{\gamma}_{ab}+i(\Delta-\mathbf{k}_0\cdot\mathbf{v})}+\frac{1}{\tilde{\gamma}_{ab}-i(\Delta+\delta-\mathbf{k}\cdot\mathbf{v})}\right], \tag{10c}$$

$$n=\frac{\lambda_b}{\gamma_b}-\left[\frac{\lambda_a}{\gamma_a}+\frac{\gamma_{b,a}\lambda_b}{\gamma_a\gamma_b}\right]. \tag{10d}$$

The intensity of phase-conjugate emission is proportional to the absolute square of

$$\bar{\sigma}_{ab}=\int\sigma_{ab}(\mathbf{v})d^3v\ .$$

The integration is carried out within the so-called Doppler limit defined by

$$\tilde{\gamma}_a,\tilde{\gamma}_b,\tilde{\gamma}_{ab}\ll ku,k_0u\ , \tag{11a}$$

$$|\Delta|,|\delta|\ll ku,k_0u\ . \tag{11b}$$

Condition (11a) is satisfied for pressures $\lesssim 100$ Torr. Condition (11b) is satisfied for the nearly resonant atom-field interaction of this problem—it would be violated for the large detunings $|\Delta|\gg ku$ appropriate to many experiments in pressure-induced resonances. As a result of inequalities (11), one can neglect in (9) both the part of the first term varying as $[\tilde{\gamma}_{ab}-i(\Delta+\delta-\mathbf{k}\cdot\mathbf{v})]^{-1}$ and the *entire* ($\mathbf{k}_0\to-\mathbf{k}_0$) term, since, after integration, these terms are smaller by a factor $\tilde{\gamma}_{ab}/ku$ than the other contributing terms.

To achieve the three-dimensional integration over $\mathbf{v}$ we choose the coordinate system shown in Fig. 2 with

$$\theta\ll 1\ . \tag{12}$$

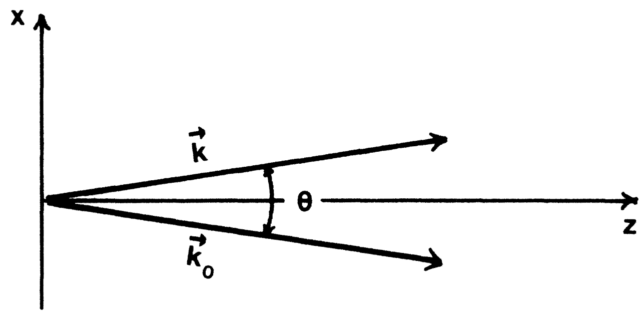

FIG. 2. Axes chosen for performing the velocity integration.

For the range of detunings δ under consideration, we can set $|\mathbf{k}|\approx|\mathbf{k}_0|\equiv K$ so that

$$\mathbf{k}\cdot\mathbf{v}=Kv_z+K\frac{\theta}{2}v_x\ ,$$

$$\mathbf{k}_0\cdot\mathbf{v}=Kv_z-K\frac{\theta}{2}v_x\ ,$$

$$(\mathbf{k}-\mathbf{k}_0)\cdot\mathbf{v}=K\theta v_x\ . \tag{13}$$

Using condition (12), an analytical integration over both v_x and v_z can be easily carried out with the result given by

$$\bar{\sigma}_{ab}=-in\frac{\Omega_f\Omega_p^*\Omega_b}{8}(\bar{\sigma}_{ab}^{\text{out}}+\bar{\sigma}_{ab}^{\text{in}})\ , \tag{14a}$$

$$\bar{\sigma}_{ab}^{\text{out}}=\frac{\sqrt{\pi}}{Ku}(\omega_{ab}^L+\omega_{ab}^{LM})\frac{i}{Ku\theta}\left[\frac{\sqrt{\pi}}{Ku\theta}\right]\times\left[(1+R)\frac{\omega_{ab}^T-\omega_b^T}{\zeta_b-\zeta_{ab}}+(1-R)\frac{\omega_{ab}^T-\omega_a^T}{\zeta_a-\zeta_{ab}}\right], \tag{14b}$$

$$\bar{\sigma}_{ab}^{\text{in}}=\left[\frac{\sqrt{\pi}}{Ku}\right]^2\omega_{ab}^{LM}(\omega_{ab}^L+\omega_{ab}^{LP})(1+R)\mathcal{D}_b\frac{\sqrt{\pi}}{Ku\theta}\omega_b^T+(1-R)\mathcal{D}_a\frac{\sqrt{\pi}}{Ku\theta}\omega_a^T+R\left[\frac{1}{\Gamma_b}-\frac{1}{\Gamma_a}\right]\mathcal{D}_a\mathcal{D}_b\ , \tag{14c}$$

$$\omega_{ab}^L=\omega\left[\frac{-\Delta+i\tilde{\gamma}_{ab}}{Ku}\right],\quad \omega_{ab}^T=\omega(\zeta_{ab})=\omega\left[\frac{-2\Delta+\delta+2i\tilde{\gamma}_{ab}}{Ku\theta}\right],$$

$$\omega_{ab}^{LM}=\omega\left[\frac{-\Delta+\delta+i\tilde{\gamma}_{ab}}{Ku}\right],\quad \omega_a^T=\omega(\zeta_a)=\omega\left[\frac{\delta+i\tilde{\gamma}_a}{Ku\theta}\right], \tag{15}$$

$$\omega_{ab}^{LP}=\omega\left[\frac{\Delta+\delta+i\tilde{\gamma}_{ab}}{Ku}\right],\quad \omega_b^T=\omega(\zeta_b)=\omega\left[\frac{\delta+i\tilde{\gamma}_b}{Ku\theta}\right],\quad \mathcal{D}_a=\frac{\Gamma_a\frac{\sqrt{\pi}}{Ku\theta}\omega_a^T}{1-\Gamma_a\frac{\sqrt{\pi}}{Ku\theta}\omega_a^T}\ ,$$

and the function $\omega(\zeta)$ is defined by[10]

$$\omega(\zeta)=e^{-\zeta^2}[1-\text{erf}(-i\zeta)], \quad \zeta \text{ arbitrary}$$
$$=\frac{i}{\pi}\int_{-\infty}^{+\infty}\frac{e^{-t^2}dt}{\zeta-t} \quad \text{only when } \text{Im}(\zeta)>0 , \qquad (16)$$

where $\text{erf}(\zeta)$ is the complex error function.

III. LINE-SHAPE ANALYSIS

The line shape (Eq. 14) may appear to have a rather complicated structure, but it simply reflects the various physical processes simultaneously occurring in the vapor. There is some question as to the best method for analyzing Eq. (14). We chose to use four (related) aspects of the problem and hope that an overall picture emerges from these components. (A) First, we consider the line shape as composed of two contributions—one from atoms have not undergone velocity-changing collisions and one from atoms that have. The former category dominates at low pressure and the latter at high pressure. In this subsection the resonance positions and widths are discussed. (B) Second, we examine the conditions under which resonances having a width characterized by the lower-state spontaneous and collision widths can be observed. It is shown that the existence of such resonances depends critically on the departure of the sum density

$$S(\mathbf{v})=\rho_a(\mathbf{v})+\rho_b(\mathbf{v}) \qquad (17)$$

from its equilibrium value. In this subsection a natural definition of a closed system emerges. Moreover, it is seen how the system opens and recloses with increasing peturber pressure, and how the reclosing is linked to the collisional narrowing of the lower-state resonance. (C) Third, we examine briefly the dependence of the resonance characterized by the upper-state spontaneous and collisional decay processes. (D) Finally, we summarize the various line-shape features and give several examples of typical line shapes at various pressures. In Sec. IV the relationship of these line shapes to the pressure-induced extra resonance of Bloembergen and co-workers[1,2] is noted.

A. Line-shape resonances and widths

In the context of the strong collision model, the line shape naturally appears as composed of two terms: σ_{ab}^{out}, associated with atoms that have not undergone velocity-changing collisions, and σ_{ab}^{in}, associated with atoms that have.

1. Atoms not having undergone velocity-changing collisions

The contribution from such atoms is dominant at low pressure when collisions are relatively infrequent (in this case $\sigma_{ab}^{\text{in}}/\sigma_{ab}^{\text{out}}$ is of order Γ_α/ku). For these atoms, the line shape results from the integration over velocity of a component $\sigma_{ab}^{\text{out}}(\mathbf{v})$ which consists of the product of three factors as follows:

$$\sigma_{ab}^{\text{out}}(\mathbf{v})\simeq\frac{1}{\tilde{\gamma}_{ab}+i(\Delta-\delta)+i\mathbf{k}\cdot\mathbf{v}}\,\frac{1}{\tilde{\gamma}_\alpha-i\delta+i(\mathbf{k}-\mathbf{k}_0)\cdot\mathbf{v}}\times\frac{1}{\tilde{\gamma}_{ab}+i\Delta-i\mathbf{k}_0\cdot\mathbf{v}}, \quad \alpha=a,b . \qquad (18)$$

The third factor represents a single-photon absorption of the forward pump beam; the second factor reflects the evolution within state $|\alpha\rangle$, that is, the probability that the grating formed by the forward pump and probe beams is not affected by velocity-changing collisions; and the first factor represents a "three-photon" process involving the absorption of one forward and one backward pump photon and the emission of a probe photon.

When integrated over velocity, the first and third factors give rise to a resonance centered at $\delta=2\Delta$ with width [full width at half maximum (FWHM)] $4\tilde{\gamma}_{ab}$. This resonance results from a velocity-selective process in which the same velocity classes of atoms are used in both the one-photon and three-photon absorption factors. The second factor in (18) gives rise to "grating resonances" centered at $\delta=0$ with widths that we write symbolically as

$$\Gamma_G(\alpha)=(2\tilde{\gamma}_\alpha)*(Ku\theta) \qquad (19)$$

representing the convolution of a Lorentzian having width $2\tilde{\gamma}_\alpha$ with a Gaussian having a characteristic residual width $(Ku\theta)$ (the FWHM of the Gaussian is 1.66 $Ku\theta$). For illustrative purposes, we may consider the case in which

$$\gamma_a \ll Ku\theta, \quad Ku\theta \ll \gamma_b \qquad (20)$$

implying that

$$\Gamma_G(b)\simeq 2\tilde{\gamma}_b=2(\gamma_b+\Gamma_b) , \qquad (21a)$$

$$\Gamma_G(a)\simeq(2\tilde{\gamma}_a)*(Ku\theta) . \qquad (21b)$$

The upper state grating has a width $2(\gamma_b+\Gamma_b)$ while the lower-state grating has a width given by the convolution of $2(\gamma_a+\Gamma_a)$ and $Ku\theta$. At low pressure and for small residual Doppler broadening, the lower-state grating resonance can be much narrower than the upper-state one. These features are shown in Fig. 3.

2. Atoms having undergone velocity-changing collisions

Any atom having undergone a velocity-changing collision is thermalized. Consequently, any correlation between the velocity classes participating in the one-photon and three-photon absorption processes is *lost*. In analogy with Eq. (18), the contribution from atoms having undergone velocity-changing collisions arises from the velocity integration (over v_z and v_z') of a term that can be written as

$$\sigma_{ab}^{\text{in}}(\mathbf{v})\simeq\frac{1}{\tilde{\gamma}_{ab}+i(\Delta-\delta)+ikv_z}\sigma_G(\delta,\alpha,v_x)\times\frac{1}{\tilde{\gamma}_{ab}+i\Delta-ik_0v_z'} , \qquad (22)$$

where v_z' and v_z are uncorrelated. The factor $\sigma_G(\delta,\alpha,v_x)$

is a term that is responsible for a collisional or "Dicke" narrowing of the grating resonances.[11] On averaging over velocity, the first and third factors give rise to very broad "resonant" structures, characterized by the full Doppler width.[12] The grating resonance associated with state $|\alpha\rangle$ has a width which starts at $\Gamma_G(\alpha)=(2\gamma_\alpha)*(Ku\theta)$ [Eq. (19)] at low pressure and monotonically decreases to a final width equal to $2\gamma_\alpha$. The condition for collisional narrowing to this final width $2\gamma_\alpha$ is

$$\frac{\Gamma_\alpha}{Ku\theta} \gg 1 \ . \tag{23}$$

If $\gamma_b \gg Ku\theta$, the excited-state grating width (for this contribution from atoms which have undergone velocity-changing collisions) is always equal to $2\gamma_b$. For the lower state, assuming $\gamma_a \ll Ku\theta$, the grating width starts at 1.66 $(Ku\theta)$ for low pressure and reduces to $2\gamma_a$ for pressures such that $\Gamma_a/Ku\theta \gg 1$.

These line-shape features are shown in Fig. 4. The contributions from atoms having undergone velocity-changing collisions become dominant at high pressure, where atoms undergo many collisions within their natural lifetimes. A quantitative condition for "high" pressure can be written

$$\frac{\Gamma_\alpha}{\gamma_\alpha} > \frac{Ku}{\bar{\gamma}_{ab}}, \quad \alpha=a,b \tag{24}$$

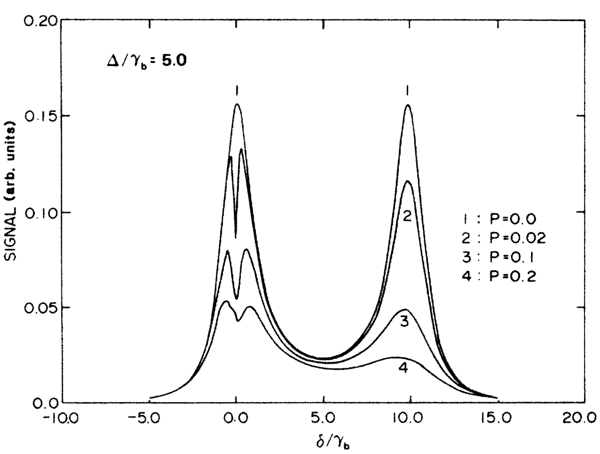

FIG. 3. Four-wave-mixing signal intensity (arbitrary units) as a function of pump-probe detuning δ. All frequencies are in units of γ_b; for the figures shown, $\Delta=5.0$, $Ku\theta=0.1$, $Ku=100$, $\gamma_a=0.01$. The collision rates are taken as $\Gamma_a=\Gamma_{ab}=4\Gamma_b$, $P=\Gamma_b/\gamma_b=0.0$, 0.02, 0.1, 0.2 corresponding to different (dimensionless) pressures. At zero pressure, there are resonances at $\delta=0$ and $\delta=2\Delta$ having widths (FWHM) equal to 2.0. As the perturber pressure increases from zero, this system "opens" and a narrow structure (corresponding to $\bar{\sigma}_{ab}^{\text{out}}$ of the text) appears whose width is $2(\gamma_a+\Gamma_a)$ convoluted with the residual Doppler width $Ku\theta$. This narrow resonance broadens with increasing pressure. At these relatively low pressures, only the contribution from $\bar{\sigma}_{ab}^{\text{out}}$ is dominant, although the marked asymmetry of the $P=0.2$ dip is due to the fact that $\bar{\sigma}_{ab}^{\text{in}}$ is beginning to make a non-negligible contribution to the line shape.

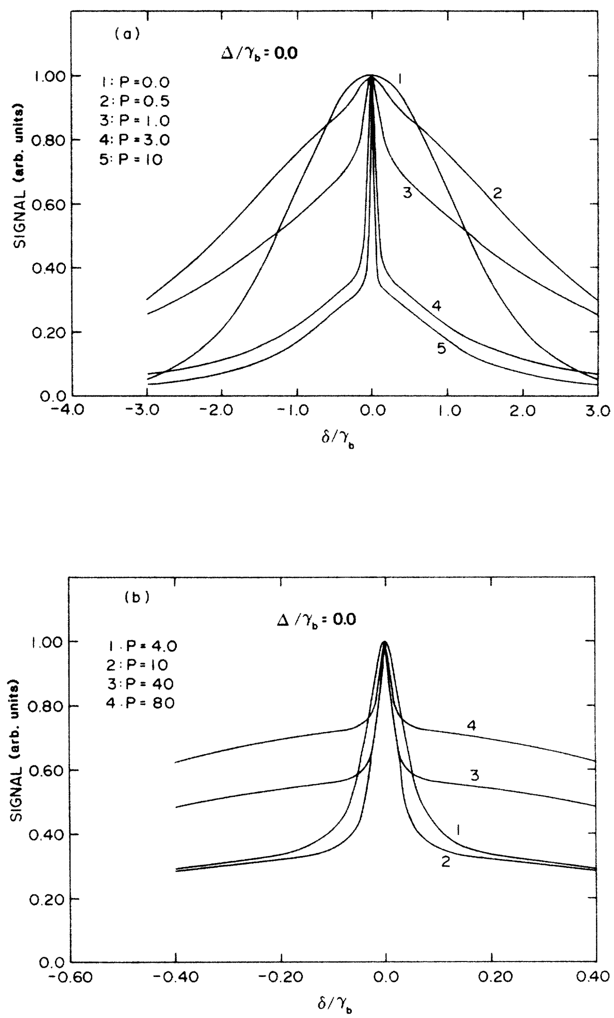

FIG. 4. Graphs of four-wave-mixing signal intensity (arbitrary units) as a function of pump-probe detuning δ. All frequencies are in units of γ_b; for the figures shown, $\gamma_a=0.01$, $\Delta=0.0$, $Ku\theta=1.0$, $Ku=100$, and $\Gamma_a=\Gamma_{ab}=4\Gamma_b$, with $P=\Gamma_b/\gamma_b=(0.001, 0.5, 1.0, 3.0, 10)$ in Fig. 4(a) and $P=(4, 10, 40, 80)$ in Fig. 5(b). These figures are intended to illustrate collisional narrowing and the "reclosing" of the system, which is why a somewhat larger value of $Ku\theta=1.0\ \gamma_b$ was chosen. At zero pressure the linewidth is the convolution of a Lorentzian having width $2\gamma_b$ with a Gaussian having width 1.66 $Ku\theta$. As the pressure increases, the system "opens" and the contribution from $\bar{\sigma}_{ab}^{\text{in}}$ begins to become important, leading to a new narrow resonance characterized by the ground-state width (convoluted with $Ku\theta$). (The $\bar{\sigma}_{ab}^{\text{out}}$ component giving rise to the narrow dip seen in Fig. 3 is negligible for the parameters chosen for these graphs.) With increasing pressure, the "broad" and narrow resonances approach their asymptotic widths $2\gamma_b$ and $2\gamma_a$, respectively, as a result of collisional narrowing. There is a range of pressures where the ratio of the narrow to broad resonance amplitude is approximately constant; at still higher pressures, the reclosing of the system leads to an asymptotic disappearance of the narrow resonance. Note that in Fig. 4(b) the scale has been expanded so that the broad resonance (of width $2\gamma_b$) appears only as an approximately constant background. All curves have been normalized to the same value at $\delta=0$; in absolute terms, the signal decreases with increasing pressure approximately as P^{-2}.

which states that the number of collisions within a radiative lifetime is large enough to redistribute velocity selected atoms excited in a range ($Ku=\pm\tilde{\gamma}_{ab}$) over the entire Doppler width Ku. The *redistribution* inequality (24) is linked to the reclosing of the system as is shown below. A schematic picture of the velocity redistribution is shown in Fig. 5.

B. Resonances characterized by lower-state radiative and collisional rates

In order to determine the conditions under which grating resonances characterized by the lower-state width are seen, it is useful to recall Eq. (7c) in which one finds that

$$\rho_{ab}(\mathbf{v})\propto[\rho_b(\mathbf{v})-\rho_a(\mathbf{v})] . \quad (25)$$

In terms of the sum density

$$S(\mathbf{v})=\rho_a(\mathbf{v})+\rho_b(\mathbf{v}) ,$$

expression (25) can be written

$$\rho_{ab}(\mathbf{v})\propto[2\rho_b(\mathbf{v})-S(\mathbf{v})] . \quad (26)$$

Up to second order in the fields, $\rho_b(\mathbf{v})$ depends only on the excited-state parameters. *Thus, any contribution to* ρ_{ab} *in third order in the fields which depends on the*

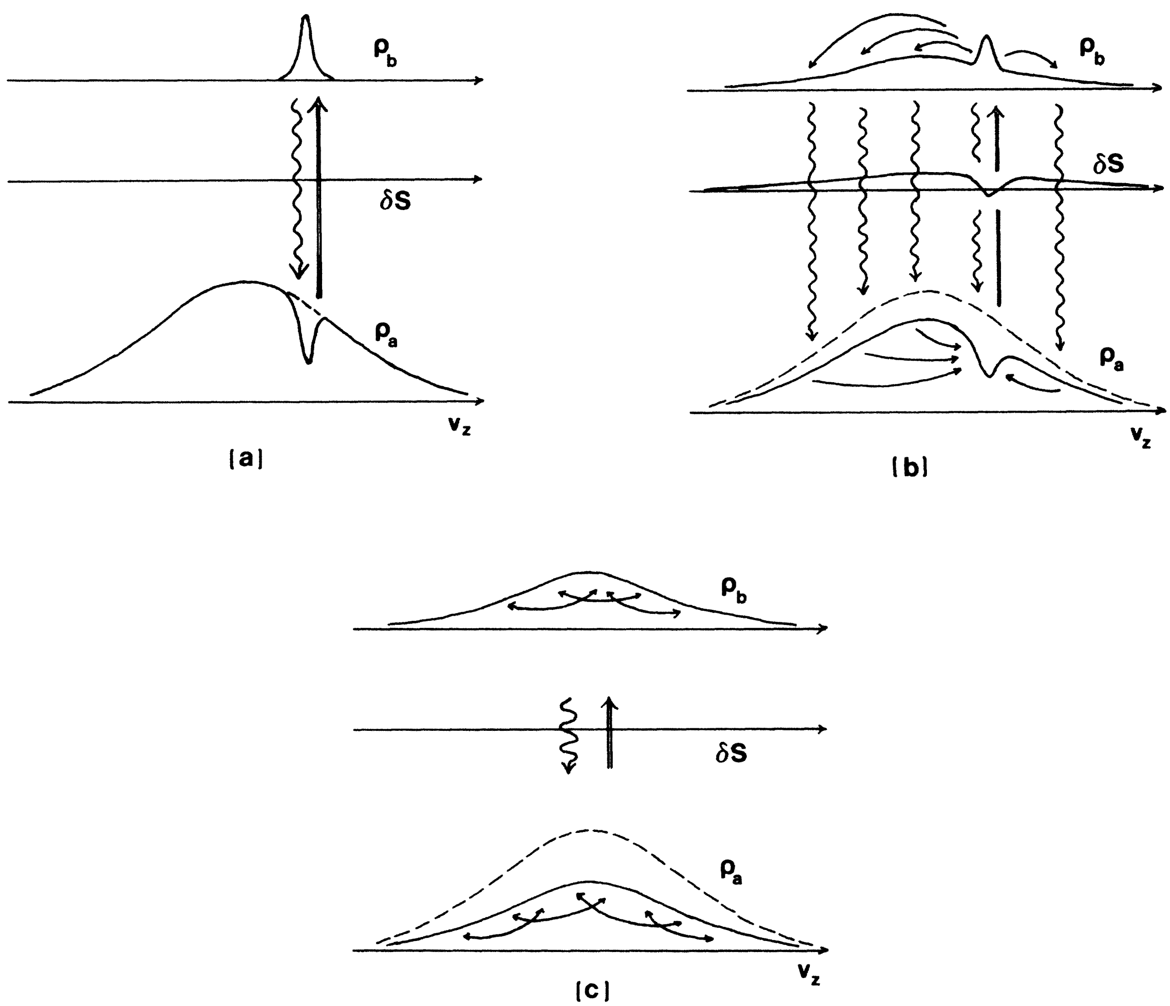

FIG. 5. Schematic representation of the opening and reclosing of the system in velocity space. The graphs shown in each of (a), (b), and (c) give the longitudinal velocity distribution for state a, $\rho_a(\mathbf{v})$, for state b, $\rho_b(\mathbf{v})$, and the deviation $\delta S(\mathbf{v})$ of the sum density $[\rho_a(\mathbf{v})+\rho_b(\mathbf{v})]$ from its equilibrium value in the absence of applied fields. (a) At zero pressure, excitation and decay occurs within a given velocity class and $\delta S(\mathbf{v})=0$, assuming that the system is "closed," as defined in the text. (b) With increasing pressure, collisions redistribute some of the velocity-selected atoms over the entire thermal width. For different collision rates in the two states, the closed nature of the system is lost, as is evidenced by a nonvanishing $\delta S(\mathbf{v})$. (c) At very high pressure, such that the redistribution in each level is complete, the system has "reclosed," and, as at zero pressure, once again finds $\delta S(v)=0$. In this limit, there is no longer any velocity selection and excitation and decay occurs over the entire thermal distribution. In each diagram, the dashed curve corresponds to the equilibrium distribution $\rho_a(\mathbf{v})$ in the absence of the fields. The upward arrows represent excitation by the fields, the downward curly arrows represent radiative decay, and the sideways arrows represent velocity redistribution.

lower-state decay rates must come from a second-order nonvanishing contribution to $S(\mathbf{v})$. The sum density serves as a measure of resonances characterized by the lower-state decay rates.

To zeroth order in the fields, the sum density calculated from Eqs. (7) is

$$S^{(0)}(\mathbf{v})=\frac{\lambda_a}{\gamma_a}+\frac{\lambda_b}{\gamma_b}\frac{\gamma_{b,a}}{\gamma_a}+\frac{\lambda_b}{\gamma_b} . \tag{27}$$

To second order in the fields, the sum density contains contributions from all combinations of two of the fields. The part of $S^{(2)}(\mathbf{v})$ responsible for phase-conjugate emission may be written

$$[S^{(2)}(\mathbf{v})]_{\mathrm{PC}}=S_{+}^{(2)}(\mathbf{v})e^{i[(\mathbf{k}-\mathbf{k}_0)\cdot\mathbf{R}-\delta t]}+\text{c.c.} \tag{28}$$

When this is substituted into Eq. (7), one finds that $S_{+}^{(2)}(\mathbf{v})$ satisfies

$$[\tilde{\gamma}_a-i\delta+i(\mathbf{k}-\mathbf{k}_0)\cdot\mathbf{v}]S_{+}^{(2)}(\mathbf{v})$$
$$=-(\gamma_d+\Gamma_d)\rho_{b+}^{(2)}+\int A_d(\mathbf{v}'\to\mathbf{v})\rho_{b+}^{(2)}(\mathbf{v}')d^3v'$$
$$+\int A_a(\mathbf{v}'\to\mathbf{v})S_{+}^{(2)}(\mathbf{v}')d^3v' , \tag{29}$$

where

$$\gamma_d=\gamma_b-\gamma_{b,a}-\gamma_a , \tag{30}$$

$$\Gamma_d=\Gamma_b-\Gamma_a , \tag{31}$$

$$A_d(\mathbf{v}'\to\mathbf{v})=A_b(\mathbf{v}'\to\mathbf{v})-A_a(\mathbf{v}'\to\mathbf{v}) , \tag{32}$$

and $\rho_{b+}^{(2)}(\mathbf{v})$ is the second-order component of $\rho_b^{(2)}(\mathbf{v})$ which contributes to the phase-conjugated emission. If $S_{+}^{(2)}(\mathbf{v})$ vanishes, the line shape to third order cannot depend on the lower-state decay rates. Thus, the existence of resonances characterized by γ_a or Γ_a depends on a nonvanishing $S_{+}^{(2)}(\mathbf{v})$.

If there are no velocity-changing collisions [$\Gamma_a=0$, $A_a(\mathbf{v}'\to\mathbf{v})=0$], $S_{+}^{(2)}(\mathbf{v})$ vanishes only if

$$\gamma_d=\gamma_b-\gamma_{b,a}-\gamma_a=0 . \tag{33}$$

Equation (33) is the condition (1) we adopted for a closed system, since it implies no deviation from the zeroth-order sum density. In the following discussion, we assume that (33) holds. In that case, $S_{+}^{(2)}(\mathbf{v})$ satisfies

$$[\tilde{\gamma}_a-i\delta+i(\mathbf{k}-\mathbf{k}_0)\cdot\mathbf{v}]S_{+}^{(2)}(\mathbf{v})-\int A_a(\mathbf{v}'-\mathbf{v})S_{+}^{(2)}(\mathbf{v}')d^3v'$$
$$=-\Gamma_d\rho_{b+}^{(2)}+\int A_d(\mathbf{v}'\to\mathbf{v})\rho_{b+}^{(2)}(\mathbf{v}')d^3v' . \tag{34}$$

It is seen that all inhomogeneous terms in Eq. (34) vanish if the collision kernels for the two states are identical. This result is independent of the specific form of the collision kernel, and is not restricted to the strong collision kernel used in this work. For identical collision kernels, $\Gamma_d=0$, $S_{+}^{(2)}(\mathbf{v})=0$, and there is no opening of the system. This result is easy to understand. For identical kernels, collisions redistribute atoms in all the velocity classes in the upper and lower levels in the same manner, on average. This implies that the total population of each velocity subclass is dynamically conserved and the system remains "closed." Thus, the existence of resonances characterized by ground-state decay rates necessarily depends on a difference between the upper- and lower-state kernels [assuming, as we do, that condition (33) or (1) holds]. This conclusion is illustrated in Fig. 6.

If $\Gamma_d\neq 0$, at low pressure the system "opens" as a result of velocity-changing collisions and narrow resonances can be seen (if they are not masked by the residual Doppler broadening, that is, if $Ku\theta<\gamma_a$). As the pressure increases to the point when condition (24) is applicable, the velocity distributions in both ground and excited states are rethermalized, $\rho_a(\mathbf{v})$, $\rho_b(\mathbf{v})\propto W(\mathbf{v})$. In this limit, it is easily seen from Eq. (34) that $S_{+}^{(2)}(\mathbf{v})=0$, i.e., the system has reclosed. In going from low to high pressure, collisional narrowing of the lower-state grating resonance can be seen.

It can be shown from Eq. (34) and the second-order solution of Eq. (7) for $\rho_b^{2+}(\mathbf{v})$ that the amplitude of that part of $S_{+}^{(2)}(\mathbf{v})$ which contributes to the resonance having width equal to [2(γ_a + collisionally narrowed residual width)] is proportional to $(Ku\theta)^2/\Gamma_a\gamma_a$. This is precisely the same factor that determines the collisionally narrowed residual width (see below). Thus, the degree of reclosing of the system is interrelated with the collisional narrowing of the system.

C. Upper-state grating resonance

At low pressure, the upper-state grating resonance has width $\Gamma_G(b)=(2\tilde{\gamma}_b)*(Ku\theta)\simeq 2\tilde{\gamma}_b$ if $Ku\theta\ll\tilde{\gamma}_b$. This is the contribution from atoms which have not experienced velocity-changing collisions. As the pressure grows, eventually reaching the limit (24), the term arising from atoms having undergone velocity-changing collisions dominates the line shape. This term has width equal to $2\gamma_b$.

D. Line-shape summary

At low pressure, the line shape is determined by atoms that have *not* undergone any collision. There is a resonance of width $4\tilde{\gamma}_{ab}$ centered at $\delta=2\Delta$ and one of width

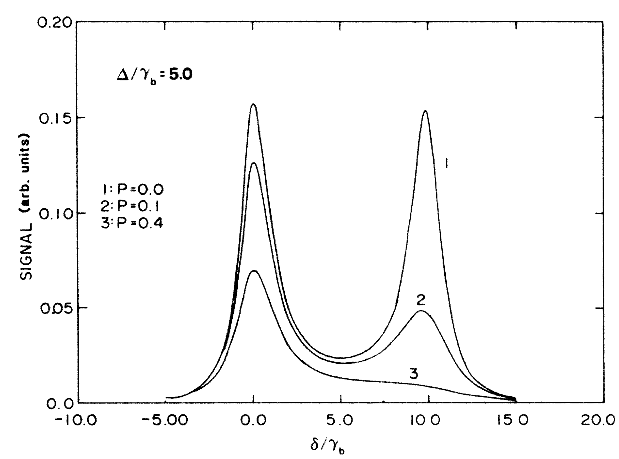

FIG. 6. Graphs similar to those of Fig. 3, but with $\Gamma_a=\Gamma_b$. In this limit of equal collision rates for the two levels, the system never "opens" and the narrow resonance is not seen.

$(2\tilde{\gamma}_b)*(Ku\theta)$ centered at $\delta=0$. If the system is "open," owing to $\Gamma_d\neq 0$, there is an additional narrow resonance centered at $\delta=0$, having width $(2\tilde{\gamma}_a)*(Ku\theta)$. These features are illustrated in Fig. 3. As the pressure grows, the no-collision terms ($\bar{\sigma}_{ab}^{\,\text{out}}$) broaden and diminish in (relative) amplitude. The major contribution begins to come from atoms that have undergone velocity-changing collisions. The resonance at $\delta=2\Delta$ vanishes and the resonances centered at $\delta=0$ have widths which reduce toward their asymptotic values of $2\gamma_a$ and $2\gamma_b$, respectively.

At "high" pressure, such that $\Gamma_{ab},\Gamma_a,\Gamma_b \gg (Ku\theta)$ and $\Gamma_a \gg \gamma_a$, $\Gamma_b \gg \gamma_b$, the line shape takes on the asymptotic form

$$\bar{\sigma}_{ab}=-i\frac{\Omega_f\Omega_p^*\Omega_b}{8}(\bar{\sigma}_{ab}^{\,\text{out}}+\bar{\sigma}_{ab}^{\,\text{in}})\ , \tag{35a}$$

$$\bar{\sigma}_{ab}^{\,\text{out}} \underset{\sim}{\rightarrow} \left[\frac{\sqrt{\pi}}{Ku}\right](\omega_{ab}^{L}+\omega_{ab}^{LM})\left[\frac{1}{2(\gamma_{ab}+\Gamma_{ab})+2i\Delta-i\delta}\,\frac{1}{\gamma_b+\Gamma_b-i\delta}\left[2+\frac{\gamma_d+\Gamma_d}{\gamma_a+\Gamma_a-i\delta}\right]\right]\ , \tag{35b}$$

$$\bar{\sigma}_{ab}^{\,\text{in}} \underset{\sim}{\rightarrow} \left[\frac{\sqrt{\pi}}{Ku}\right]^2(\omega_{ab}^{L}+\omega_{ab}^{LP})\omega_{ab}^{LM}\left\{-\frac{\gamma_d+\Gamma_d}{(\gamma_a+\Gamma_a-i\delta)(\gamma_b+\Gamma_b-i\delta)}+\frac{1}{\gamma_b+\dfrac{(Ku\theta)^2}{2\Gamma_b}-i\delta}\left[2\left[\frac{\Gamma_b}{\gamma_b+\Gamma_b-i\delta}\right.\right.\right.$$
$$\left.\left.\left.+\frac{1}{\gamma_a+\dfrac{(Ku\theta)^2}{2\Gamma_a}-i\delta}\left[\gamma_d-(1-R)\frac{(Ku\theta)^2}{2\Gamma_a\Gamma_b}\Gamma_d\right]\right]\right]\right\}\ , \tag{35c}$$

where for convenience, we recall that γ_d and Γ_d are defined by

$$\gamma_d=\gamma_b-\gamma_{b,a}-\gamma_a\ ,$$
$$\Gamma_d=\Gamma_b-\Gamma_a\ .$$

From Eq. (35) one easily verifies that the redistributed component $\bar{\sigma}_{ab}^{\,\text{in}}$ dominates as soon as

$$\frac{(\gamma_{ab}+\Gamma_{ab})\Gamma_b}{Ku\gamma_b}\gg 1\ , \tag{36}$$

which is condition (24) arrived at in Sec. III A, using simple physical arguments. The narrowing of the line shape can be seen in $\bar{\sigma}_{ab}^{\,\text{in}}$, where the widths of the dominant terms are given by $[\gamma_\alpha+(Ku\theta)^2/2\Gamma_\alpha]$, $\alpha=a,b$. Moreover, for a closed system as defined by condition (1) ($\gamma_d=0$), the amplitude of the state $|a\rangle$ (lower) resonance asymptotically approaches zero (for any finite $\gamma_a\neq 0$) as $(Ku\theta)^2/2\Gamma_a\gamma_a$. This is the same factor that determines how the resonance width approaches its asymptotic value γ_a. In other words, when the narrowing is complete, the system is also reclosed and the narrow resonance disappears.

It is interesting to note that as long as $(Ku\theta)^2/2\Gamma_a>\gamma_a$, the narrow resonance keeps a constant amplitude with respect to the γ_b peak, as can be seen in $\bar{\sigma}_{ab}^{\,\text{in}}$ rewritten as

$$\bar{\sigma}_{ab}^{\,\text{in}}\simeq\frac{1}{\gamma_b+\dfrac{(Ku\theta)^2}{2\Gamma_b}-i\delta}\left[2+\left[\frac{\Gamma_a}{\Gamma_b}-1\right]\frac{\dfrac{(Ku\theta)^2}{2\Gamma_a}}{\dfrac{(Ku\theta)^2}{2\Gamma_a}-i\delta}\right]\ .$$

For $\gamma_a \ll \gamma_b$ there is a wide range of pressures for which the ratio of the amplitudes of the resonances associated with states $|a\rangle$ and $|b\rangle$ remains constant. The ratio of amplitudes is governed by the ratio Γ_a/Γ_b. These features are shown in Fig. 4.

IV. DISCUSSION

In this paper we have examined the phase-conjugate four-wave-mixing signal that is produced when three nearly resonant (detunings within the Doppler width of the atomic transition) fields having frequencies ω, ω, and $\omega+\delta$ are incident on an ensemble of two-level atoms. The two fields having frequency ω are counterpropagating and the third field makes a small angle θ with one of these fields. We have seen that for a system which is "closed" in the absence of collisions [i.e., $(\gamma_b-\gamma_{b,a}-\gamma_a)=0$, see Fig. 1], velocity-changing collisions can play a critical role in determining the strength of the phase-conjugate emission as a function of δ. In particular, these collisions are responsible for the detailed structure of the "Rayleigh-type" resonances which appear, centered at $\delta=0$. In the absence of collisions, the $\delta=0$ resonance has a width equal to $2\gamma_b$ (convoluted with the residual Doppler width $Ku\theta$). At low pressure this width is increased owing to velocity-changing collisions. Moreover, since velocity-changing collisions "open" the system for each velocity subclass, a new resonance centered at $\delta=0$ having width $2\gamma_a$ (convoluted with $Ku\theta$) appears. If $\gamma_a \ll \gamma_b$, this new resonance can be much narrower than the resonance of width $(2\gamma_b)$ in the absence of collisions. As the pressure increases, collisional narrowing of the residual Doppler width of the resonances occurs and the resonance widths decrease monotonically towards their asymptotic values of $(2\gamma_b)$ and $(2\gamma_a)$, respectively. At the same time, the system is "reclosing" since collisions are redistributing the velocity-selected atoms over the entire Maxwellian velocity distribution. When the system is fully reclosed, the γ_a resonance disappears, just as in the absence of collisions.

It seems useful to emphasize that, although they occur simultaneously and are related to the same residual widths $(Ku\theta)^2/2\Gamma_\alpha$, the collisional narrowing and the reclosing are two different phenomena. Collisional narrowing occurs only because there is a spatial phase factor which enters in the line-shape formation. In "traditional" collisional narrowing, the phase factor is associated with an atomic coherence (optical or otherwise) and the narrowing can occur when the collisions reduce this atomic mean-free path to the point where it is less than the wavelength needed to excite this atomic coherence. In the present case, however, the relevant phase is that of the population gratings created within a *single* level by the lasers. Line shapes which are collisionally narrowed have widths which asymptotically approach their homogeneous widths, with a residual component that decreases as $(\Delta\nu)^2/\Gamma$, where $\Delta\nu$ is the relevant inhomogeneous broadening and Γ a collision rate. The population gratings discussed in this work are spatially modulated in the transverse direction. Consequently, the narrowing occurs relative to that direction, with a corresponding inhomogeneous width equal to $Ku\theta$.

In contrast to the collisional narrowing, the reclosing of the system does not depend intrinsically on the existence of a spatial phase. The reclosing is simply related to the velocity distribution within each level. The radiation fields create a nonequilibrium velocity distribution in each level and collisions tend to restore the atoms to equilibrium. The system recloses when the collision rate is much larger than the radiative transfer rate [see condition (36)]. The reclosing occurs not only for the population gratings discussed above, but also for the nonmodulated parts of ρ_a and ρ_b. Moreover, it occurs in the longitudinal as well as in the transverse directions. It can be easily shown that the sum density $S(\mathbf{v})$ which measures the degree of openess of the system, tends to zero with increasing pressure asymptotically as $[(1/\Gamma_b)-(1/\Gamma_a)]$ for all its components, modulated or not. On the other hand, for the population gratings, there is a dependence of the reclosing on $Ku\theta$ which is absent for the unmodulated component of the population. One may say that, at high pressure, there is an additional effective lifetime $(Ku\theta)^2/2\Gamma_\alpha$ for the population grating associated with state $|\alpha\rangle$, so that condition (1) defining a closed system is replaced by

$$G_d=\left[\gamma_b+\frac{(Ku\theta)^2}{2\Gamma_b}\right]-\gamma_{b,a}-\left[\gamma_a+\frac{(Ku\theta)^2}{2\Gamma_a}\right]$$

$$=\gamma_d+\frac{(Ku\theta)^2}{2\Gamma_a\Gamma_b}\Gamma_d=0\ .$$

It is precisely this factor G_d which enters the expression for the amplitude of the narrow peak in $\bar{\sigma}^{\,\mathrm{in}}_{ab}$. In summary, we see that although the amplitude of the narrow peak is determined by the factor $(Ku\theta)^2/2\Gamma_a$ [and $(Ku\theta)^2/2\Gamma_b$], and the residual width of the narrow peak is determined also by the same factor $(Ku\theta)^2/2\Gamma_a$, the origin of these two effects is different.

The collision-induced resonant structures discussed in this work differ somewhat from the pressure-induced extra resonances (PIER4) studied by Bloembergen and coworkers.[1,2] In PIER4, the atom-field detunings were always *outside* the Doppler width of the transitions. As such, velocity-changing collisions played no role in the opening or closing of the system, as defined in this paper. Moreover, there is no velocity selection in the excitation process since *all* atoms are detuned well outside the Doppler width. As a consequence, there is *no* resonance centered at $\delta=0$ in the absence of collisions for a closed system. A resonant structure centered at $\delta=0$ having width $2\tilde{\gamma}_b$ (convoluted with $Ku\theta$) appears and *grows* with increasing perturber pressure, but no resonant structure with width $2\tilde{\gamma}_a$ (convoluted with $Ku\theta$) emerges, since the system remains closed for each velocity subclass (as there is no velocity selection in the excitation process). In contrast to these results, for nearly resonant tuning (detuning within the Doppler width), resonant structures centered at $\delta=0$ *always* exist, even in the absence of collisions. The overall amplitude of these resonance structures decreases with increasing pressure, in contrast to PIER4. Moreover, again in contrast to PIER4, collisions lead to an additional resonance having width $2\gamma_a$ (convoluted with $Ku\theta$), owing to the fact that velocity-changing collisions open the system for each velocity subclass. The differences between PIER4 and four-wave mixing using nearly resonant tuning can be traced to velocity-selective excitation which is present in the latter and absent in the former.

Finally, we should like to comment on the experimental implications of our work. Several studies of the role of velocity-changing collisions on four-wave-mixing line shapes for nearly resonant tuning have been carried out.[5] Many of the features predicted in this work have been observed in those experiments (pressure-induced narrow resonant structures centered at $\delta=0$, collisional narrowing), but a quantitative comparison with theory has not yet been attempted (the theory must be extended to include effects of magnetic degeneracy or the experiments will have to be done using an atom other than sodium[5]). A systematic experimental study of the collisional modification of four-wave-mixing line shapes for nearly resonant tuning in Na is in progress,[13] and it is hoped that this study will serve as a test for the theory.

ACKNOWLEDGMENTS

This work is supported by the U.S. Office of Naval Research and the National Science Foundation Grant No. PHY8415781. M. Gorlicki wishes to thank Dr. P. Berman for his support and New York University for its hospitality during this work. The Laboratoire de Physique des Lasers is a laboratory associated with the Centre National de la Recherche Scientifique.

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