Polarization properties of phase-conjugate mirrors: Angular dependence and disorienting collision effects in resonant backward four-wave mixing for Doppler-broadened degenerate transitions

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We present the theory of resonant backward degenerate four-wave mixing in gaseous media for Doppler-broadened *degenerate* two-level systems. The degeneracy of the atomic levels as well as the influence of the polarization of the incident fields are taken into account within the framework of a tensorial formalism. The amplitude, line shape, and polarization of the reemitted phase-conjugate field are calculated up to the third order in the incident fields. The velocity average is performed in the approximation of large Doppler linewidths. One considers in detail the influence of depolarizing relaxation processes and the effects of pump-probe angular separation. In particular, the importance of the residual Doppler effect (shortening of the lifetime of the optically induced gratings by the atomic motion) associated with the pump-probe angle is emphasized. We show that the polarization of the reemitted fields is governed by very simple laws as soon as the residual Doppler effect overcomes the lifetimes of the atomic levels. Phase-conjugate mirrors can be simply characterized by a linear dichroism (parallel pump polarizations) or a birefringence (cross-polarized pumps). Vectorial phase conjugation is also analyzed, in the case of counter-rotating circular polarizations of the pumps. Finally, the case of two-photon transition is studied and compared with the case of a resonant transition.

I. INTRODUCTION

In recent years, increasing interest has been shown for backward degenerate four-wave mixing (DFWM)[1] in relation with its applications to phase conjugation (PC). Numerous experimental and theoretical works have been concerned with the case when the nonlinear medium consists of a resonant gas sample. These studies on resonant media are of particular interest for a good knowledge of all the physical processes involved in DFWM. Some of the properties related to the Doppler-broadened nature of these media are rather well known: it has been demonstrated that resonant DFWM is efficient only in the case of a small angular separation between the standing pump wave and the incident traveling probe wave (see Fig. 1) and, thus, the emission line shape is Doppler-free, due to velocity selection.[2-4] The saturation behavior of the PC emission has also been widely discussed[2,5] and has been demonstrated to be of dispersive origin in most cases.

Some of the polarization properties of resonant DFWM have been analyzed by several groups,[6-13] but only a few works considered the influence of level degeneracy in detail.[6,12] In particular, vectorial wave-front reversal (both phase *and* polarization conjugation) have been studied in Refs. 7 and 10. In general, most of the works in the field have been either concerned with elementary polarization selection rules[8,9,14] or based on the "scalar" model[10,11] which assumes the reemitted field to be the sum of three contributions proportional to the scalar product of two incident fields, so that

$$\vec{E}_{PC}=A(\vec{E}_0^*\cdot\vec{E}_-)\vec{E}_+ +B(\vec{E}_0^*\cdot\vec{E}_+)\vec{E}_- +C(\vec{E}_+\cdot\vec{E}_-)\vec{E}_0^* \quad (1)$$

(notations are obvious, see Fig. 1; A,B,C are constants depending upon the nonlinear susceptibility of the medium). If the scalar model seems to describe conveniently most works in solid materials, it appears to be strongly inadequate to the case of gas media. Such a model does not consider the possibility of bringing the atomic system into a linear superposition of Zeeman sublevels, and thus does not account for the contributions originating in atomic sublevel coherence. For instance, for a probe polarization orthogonal to the one of the pumps, Eq. (1) wrongly assigns resonant phase conjugation to two-photon absorption by the standing pump wave [term C in Eq. (1)] and ignores the process in which the PC emission process is induced by a Zeeman coherence grating.[11]

In this paper, we present a calculation of the PC field for a Doppler-broadened degenerate two-level system $(a,J_a\rightarrow b,J_b)$. One uses the tensorial formalism[15,16]

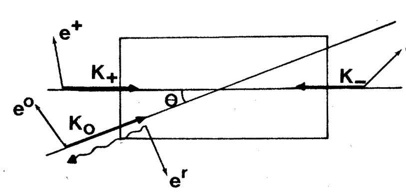

FIG. 1. Basic scheme of degenerate four-wave mixing. The standing pump wave [fields $(\vec{E}_+,\vec{E}_-)$] is along the axis defined by $\vec{K}_+=-\vec{K}_-=\vec{K}$, the probe (field $\vec{E}_0$) propagates along $\vec{K}_0$. The polarization of the fields are defined, respectively, by $\vec{e}^{\,+}$, $\vec{e}^{\,-}$, $\vec{e}^{\,0}$, and $\vec{e}^{\,r}$ for the reemitted field $\vec{E}_{PC}$.

which allows us to naturally introduce the effects of the polarizations of the incident fields. In addition, relaxation processes in a gas phase (such as disorienting collisions or dephasing collisions) and cascade effects induced by spontaneous emission are straightforwardly taken into account. The validity of our analysis is essentially limited to the case of low incident powers, so that the PC field is derived by a perturbation expansion. By comparison with previous studies, our work demonstrates the extreme importance of the pump-probe angular separation θ which severely affects the polarization properties of DFWM emission. It is shown that several regimes must be distinguished as it was already the case for a nondegenerate two-level system (see Ref. 3, which will be referred to as I in all the following). In the quasiperfect collinear regime, the Doppler effect is entirely negligible, and the problem becomes closely related to saturated absorption and polarization spectroscopy: References 6 and 8 consider only this regime for which the PC emission should depend strongly on all the details of the relaxation processes. On the other hand, for relatively large angles θ, the line shape itself is modified by the Doppler broadening and the physical processes are independent of the details of the system relaxation. An intermediate regime exists in which the line shape is weakly affected by the Doppler broadening, while the lifetime of the atomic gratings is governed by the residual Doppler effect: in this case, the emission line shape and the polarization properties of the PC mirror become uncorrelated and the latter ones are independent of the relaxation mechanisms in a gas phase. This regime is the one commonly encountered in experiments as we have shown in a previous paper.[12]

Section II is devoted to the notations and to the description of the atomic system. In Sec. III, a general expression of the reemitted field is given as an integral over the velocity distribution and is calculated through a third-order perturbation expansion of the density matrix. In Sec. IV, the velocity integration is performed for the various regimes mentioned above, depending on the pump-probe angular separation, and some general properties of the PC field polarization are discussed in Sec. V. Section VI is devoted to the discussion of several cases of practical importance; e.g., all the polarizations linear and parallel, or orthogonal, or all the polarizations circular. In all these cases, the main features of the phase-conjugate mirror can be characterized easily. One also discusses the practical framework allowing one to carry out vectorial conjugation. A more complete calculation of the influence of the angular separation θ on symmetry breaking by collisions effects is deferred to an appendix. Finally, in Sec. VII, we discuss briefly the problem of two-photon phase conjugation when the level degeneracy and polarization problems are taken into account.[17]

II. DESCRIPTION OF THE ATOMIC SYSTEM

The atomic system that we consider here is a two-level atom $a \to b$ (a is the lower level) with respective angular momentum J_a, J_b, so that each level is composed of $(2J_\alpha+1)$ sublevels $|J_\alpha, m_\alpha\rangle$ $(-J_\alpha \le m_\alpha \le J_\alpha)$. The two levels (transition frequency ω_0) are coupled by an electric dipole moment.

One uses the tensorial formalism in which atomic observables are expanded on a set of irreducible tensor operators:[15,16,18]

$$ {}_{\alpha\beta}T_Q^{(k)} = \sum_{m_\alpha, m_\beta} (-1)^{J_\beta - m_\beta} \langle J_\alpha, J_\beta, m_\alpha, -m_\beta \,|\, k, Q\rangle \times |J_\alpha, m_\alpha\rangle\langle J_\beta, m_\beta| \ , \tag{2} $$

where $\langle J_\alpha, J_\beta, m_\alpha, -m_\beta \,|\, k, Q\rangle$ are Clebsh-Gordon coefficients. In this basis, the electric dipole operator is represented by a tensor of rank 1:

$$ \vec{P} = \frac{P_{ab}}{\sqrt{3}} [{}_{ab}T^{(1)} + (-1)^{J_a - J_b}\, {}_{ba}T^{(1)}] \ , \tag{3} $$

where P_{ab} is the (real) reduced matrix element. The gas medium is composed of a collection of atoms whose translation motion is described classically. Their internal state is described by an atomic density operator $\rho(\vec{r}, \vec{v}, t)$ which is expanded on the basis of irreducible tensor operators:

$$ \rho(\vec{r}, \vec{v}, t) = \sum_{\alpha,\beta,k,Q} {}_{ab}\rho_Q^k(\vec{r}, \vec{v}, t)\, {}_{\alpha\beta}T_Q^{(k)} \ . \tag{4} $$

The relaxation of the atomic system (radiative decay, collisional broadening) is assumed to be independent of the velocity:

$$ \left[\frac{d}{dt}\, {}_{\alpha\beta}\rho_Q^k \right]_{\text{relax}} = -\Gamma_{\alpha\beta}(k)\, {}_{\alpha\beta}\rho_Q^k \ . \tag{5} $$

[In the following, we set $\Gamma_{\alpha\alpha}(k) = \Gamma_\alpha(k)$.] $\Gamma_\alpha(0)$ is the relaxation rate of the global population in level α, $\Gamma_\alpha(1)$ is the relaxation rate of the orientation (macroscopic angular momentum), and $\Gamma_\alpha(2)$ describes the relaxation of the atomic alignment ($k=2$). In the absence of laser irradiation, the population is at thermal equilibrium, and thus follows a Maxwell velocity distribution (u, mean velocity):

$$ n_\alpha(\vec{v}) = n_\alpha f(\vec{v}) \tag{6} $$

with

$$ f(\vec{v}) = \frac{e^{-v^2/u^2}}{(u\sqrt{\pi})^3} \ . \tag{7} $$

The $b \to a$ transfer induced by spontaneous emission (cascade effects) is described by the following term:[19]

$$ \left[\frac{d}{dt}\, {}_{aa}\rho_Q^k \right]_{\text{spont em}} = \Theta(b,a,k)\, {}_{bb}\rho_Q^k \tag{8} $$

with

$$ \Theta(b,a,k) = (-1)^{J_a + J_b + k + 1} \times \gamma_{ba}(2J_b+1) \begin{Bmatrix} J_b & J_b & k \\ J_a & J_a & 1 \end{Bmatrix} \ . \tag{9} $$

In Eq. (9), γ_{ba} $[\le \Gamma_b(0)]$ is the probability for spontaneous emission from $|b\rangle$ to $|a\rangle$.

We use a classical description for the incident electromagnetic (EM) fields. For the sake of simplicity, we restrict ourselves to plane waves, and we assume that propagation effects are negligible, so that the EM field $\vec{E}$ inside the medium is the sum of three incident waves:

$$\vec{E}=\frac{1}{2}(\vec{\mathscr{E}}+\vec{\mathscr{E}}^*) \tag{10}$$

with

$$\vec{\mathscr{E}}=\sum_{\mu=\pm,0}\mathscr{E}^{\mu}\vec{e}^{\,\mu}\exp(-i\Phi_\mu) \tag{11}$$

and

$$\Phi_\mu=\omega_\mu t-\vec{K}_\mu\cdot\vec{r}+\varphi_\mu\ , \tag{12}$$

where the index μ refers to the fields labeled $+,-,0$ (see Fig. 1). In Eq. (12), ω_μ is the frequency of the μ wave, $\vec{K}_\mu$ its wave vector, φ_μ its phase, and $\mathscr{E}^\mu$ its complex amplitude ($\vec{K}_+=-\vec{K}_-=\vec{K}$). The polarization vector $\vec{e}_\mu$ can be expanded on either a (real) orthonormal basis $(\vec{u}_x,\vec{u}_y,\vec{u}_z)$, or the unit complex vectors defined by

$$\vec{u}_0=\vec{u}_z,\quad \vec{u}_{\pm 1}=\mp\frac{1}{\sqrt{2}}(\vec{u}_x\pm i\vec{u}_y)\ . \tag{13}$$

They satisfy

$$\vec{u}^{\,*}_q=(-1)^q\vec{u}_{-q},\quad \vec{u}_q\cdot\vec{u}^{\,*}_{q'}=\delta_{qq'}\ . \tag{14}$$

Hence, we defined the standard components of the unit vectors $\vec{e}^{\,\mu}$ by

$$\vec{e}^{\,\mu}=\sum_q e^\mu_q\vec{u}^{\,*}_q \tag{15}$$

with

$$e^\mu_q=\vec{e}^{\,\mu}\cdot\vec{u}_q\ . \tag{16}$$

The transversality of the plane waves imposes the condition

$$\vec{e}^{\,\mu}\cdot\vec{K}_\mu=0\ . \tag{17}$$

To determine all the properties of the medium interacting with the EM fields, we need to calculate the density matrix of the system, which obeys the following master equation:[18]

$$i\hbar\dot{\rho}=[\mathscr{H}_0-\vec{P}\cdot\vec{E},\rho]+\left[\frac{d\rho}{dt}\right]_{\text{relax}}+\left[\frac{d\rho}{dt}\right]_{\text{spont em}}\ . \tag{18}$$

$\dot{\rho}$ is the total (hydrodynamic) time derivative of ρ, $\dot{\rho}=\partial\rho/\partial t+\vec{v}\cdot\vec{\nabla}\rho$. The equation of motion of ${}_{\alpha\beta}\rho^k_Q$ is thus obtained by projecting Eq. (18) in the tensorial basis [Eq. (4)].

III. EXPRESSION OF THE EMITTED FIELD

The electric field inside the medium obeys the propagation equation

$$\vec{\nabla}\times\vec{\nabla}\times\vec{E}-\frac{1}{c^2}\frac{\partial^2\vec{E}}{\partial t^2}=\frac{1}{\epsilon_0 c^2}\frac{\partial^2\vec{P}}{\partial t^2} \tag{19}$$

[where $\vec{E}$ and $\vec{P}$ are, respectively, the electric field and the macroscopic polarization at $\vec{r}$ and t (Ref. 20)]. The field of interest $\vec{E}^{\,r}(\vec{r},t)$ corresponding to the field reemitted through the four-wave mixing process is only one of the Fourier components of the total electric field, radiated by the corresponding component $\vec{P}^{\,r}(\vec{r},t)$ of the induced polarization. $\vec{E}^{\,r}$ and $\vec{P}^{\,r}$ can be written in complex notation:

$$\vec{E}^{\,r}(\vec{r},t)=\frac{1}{2}[\vec{\mathscr{E}}^{\,r}(\vec{r},t)\exp(-i\Phi_r)+\text{c.c.}]\ , \tag{20}$$

$$\vec{P}^{\,r}(\vec{r},t)=\frac{1}{2}[\vec{\mathscr{P}}^{\,r}(\vec{r},t)\exp(-i\Phi_r)+\text{c.c.}] \tag{21}$$

with $\Phi_r=\omega_r t-\vec{K}_r\cdot\vec{r}+\varphi_r$, where ω_r, $\vec{K}_r$, and φ_r are, respectively, the frequency, the wave vector, and the phase of the reemitted PC field of interest. Since we consider only degenerate four-wave mixing in the following, we assume that the dispersion relation is satisfied, so that $\omega_r=c\,|\vec{K}_r|$ (automatic phase matching of backward DFWM), and we have to determine the field exiting out of the medium [in the vacuum, where $\vec{P}(\vec{r},t)=0$]. In vacuum, the reemitted field must be a transverse plane wave. Equation (19) can be separated in an independent system of equations for the transverse component $\vec{E}^{\,r}_\perp(\vec{r},t)$ and the longitudinal one $\vec{E}^{\,r}_\parallel(\vec{r},t)$ ($\vec{E}_\parallel$ parallel to $\vec{K}_r$). In the slowly varying envelope approximation, one sees easily that $\vec{\mathscr{E}}^{\,r}_\parallel$ is everywhere proportional to the induced *local* longitudinal polarization $\vec{\mathscr{P}}^{\,r}_\parallel(\vec{r},t)$ [Eq. (19) yields $\vec{\mathscr{E}}_\parallel=-\vec{\mathscr{P}}_\parallel/\epsilon_0$]. Thus, the longitudinal field does not propagate inside the medium, in the sense that it is independent of the length of the interaction zone. In other words, its gain coefficient cancels, implying that $\vec{\mathscr{E}}^{\,r}_\parallel(\vec{r},t)$ is very small throughout the sample. On the other hand, the transverse field and the transverse polarization are coupled by the well-known propagation equation (in the slowly varying envelope approximation with $\vec{K}_r=-\vec{K}_0$):

$$(\vec{K}_0\cdot\vec{\nabla})\vec{\mathscr{E}}^{\,r}_\perp-\frac{K}{c}\frac{\partial}{\partial t}\vec{\mathscr{E}}^{\,r}_\perp=-\frac{iK^2}{2\epsilon_0}\vec{\mathscr{P}}^{\,r}_\perp \tag{22}$$

leading to ($K=|\vec{K}_0|$)

$$\vec{\mathscr{E}}^{\,r}_\perp(\vec{r},t)=\frac{iKL}{2\epsilon_0}\vec{\mathscr{P}}^{\,r}_\perp(\vec{r},t) \tag{23}$$

outside of the medium ($|\vec{r}|_\perp>L$). One assumes an optically thin sample, so that $\vec{\mathscr{P}}_\perp(\vec{r},t)$ can be considered as a constant. L is the length of the interaction zone in the medium (see I, Sec. 2).

The calculation of the emitted field is therefore reduced to the determination of the transverse component of the nonlinear polarization. The total polarization $\vec{\mathscr{P}}^{\,r}=\vec{\mathscr{P}}^{\,r}_\perp+\vec{\mathscr{P}}^{\,r}_\parallel$ is easily obtained by solving the density matrix equations in the standard tensorial basis. On the other hand, the separation between transverse and longitudinal components of $\vec{\mathscr{P}}^{\,r}$ is simpler when one deals with the coordinates in the real space than with the standard components. In the following, we give only the expression of the standard components of $\vec{\mathscr{P}}^{\,r}$ and not explicitly of $\vec{\mathscr{P}}^{\,r}_\perp$. In most applications, when the angular separation θ is small, one has $|\mathscr{P}^r_\parallel|\ll|\mathscr{P}^r_\perp|$ (as a consequence of the transverse nature of the incident fields [Eq. (17)]) and thus $\vec{\mathscr{P}}^{\,r}_\perp\simeq\vec{\mathscr{P}}^{\,r}$. In the most general case, the standard components of $\vec{\mathscr{P}}^{\,r}_\parallel$ and $\vec{\mathscr{P}}^{\,r}_\perp$ are immediately deduced from $\vec{\mathscr{P}}^{\,r}$ if the quantization axis $\vec{u}_z$ is chosen parallel to $\vec{K}_r$; such a choice for $\vec{u}_z$ is always possible, although generally not the most convenient (see Sec. V C).

The induced polarization $\vec{P}(\vec{r},t)$ is calculated through the equation

$$\vec{P}(\vec{r},t)=\mathrm{Tr}[\langle\rho\rangle\vec{P}]\ , \tag{24}$$

where $\langle\rho\rangle$ stands for the velocity-averaged density matrix. One deduces from Eqs. (3) and (5) that

$$P^{*}_{Q'}=\frac{P_{ab}}{\sqrt{3}}[\langle {}_{ab}\rho^{1}_{Q'}\rangle+(-1)^{Q'}\langle {}_{ab}\rho^{1*}_{-Q'}\rangle]\ . \tag{25}$$

The general third-order perturbation solution of Eq. (18) is given in Ref. 18, assuming the rotating-wave approximation:

$$\begin{aligned}{}^{(3)}_{ab}\rho^{1}_{Q'}=-i\frac{P^3_{ab}}{8}\frac{n}{\sqrt{3}}\sum_{\substack{k,Q,q,q',q'',\\ \lambda,\mu,\nu}}&\left[\frac{({}^{q''}_{ba}G^{1k}_{Q'Q})({}^{q}_{ba}G^{1k}_{q'Q})}{L^k_b(\mu-\nu)}+\frac{({}^{q''}_{ab}G^{1k}_{Q'Q})({}^{q}_{ab}G^{1k}_{q'Q})}{L^k_a(\mu-\nu)}\left[1-\gamma_{ba}\frac{\mathscr{A}^k_{ba}}{L^k_b(\mu-\nu)}\right]\right]\\ &\times\frac{\mathscr{E}^{\mu}\mathscr{E}^{\nu}\mathscr{E}^{\lambda}e^{\mu*}_{q'}e^{\nu}_{-q}e^{\lambda*}_{-q''}}{L(\mu-\nu+\lambda)}\left[\frac{1}{L(\mu)}+\frac{1}{L^*(\nu)}\right]e^{i(\Phi_\mu-\Phi_\nu+\Phi_\lambda)}\ .\end{aligned} \tag{26}$$

In Eq. (26), $n=n_b-n_a$ is the population inversion density. The coefficients G and $\mathscr{A}$ are given by

$${}^{q}_{\alpha\beta}G^{k'k}_{Q'Q}=(-1)^{J_a+J_\beta+Q'}[(2k+1)(2k'+1)]^{1/2}\times\begin{pmatrix}k' & 1 & k\\ Q' & q & -Q\end{pmatrix}\begin{Bmatrix}k' & 1 & k\\ J_\alpha & J_\alpha & J_\beta\end{Bmatrix}, \tag{27}$$

$$\mathscr{A}^k_{ba}=(-1)^{J_a+J_b+1}(2J_b+1)\times\begin{Bmatrix}J_b & J_b & k\\ J_a & J_a & 1\end{Bmatrix}\frac{\begin{Bmatrix}1 & 1 & k\\ J_b & J_b & J_a\end{Bmatrix}}{\begin{Bmatrix}1 & 1 & k\\ J_a & J_a & J_b\end{Bmatrix}}\ . \tag{28}$$

The denominators L, analogous to those defined in I, are given by

$$L(\mu)=\Gamma_{ab}+i(\omega_\mu-\omega_0-\vec{K}_\mu\cdot\vec{v})\ , \tag{29}$$

$$L(\mu-\nu+\lambda)=\Gamma_{ab}+i[\omega_\mu-\omega_\nu+\omega_\lambda-\omega_0-(\vec{K}_\mu-\vec{K}_\nu+\vec{K}_\lambda)\cdot\vec{v}]\ , \tag{30}$$

$$L^k_\alpha(\mu-\nu)=\Gamma_\alpha(k)+i[\omega_\mu-\omega_\nu-(\vec{K}_\mu-\vec{K}_\nu)\cdot\vec{v}]\ . \tag{31}$$

For a dipolar transition, only one optical relaxation rate $\Gamma_{ab}(k=1)$ has to be considered at third order [Eq. (26)] and we have set $\Gamma_{ab}=\Gamma_{ab}(1)$.

From Eq. (26), it is easy to get the component $\vec{P}^{\,r}$ of the induced polarization responsible for the emission. For *degenerate* four-wave mixing ($\omega_\mu=\omega_\nu=\omega_\lambda=\omega$), the phase-conjugate field is such that

$$\phi_r=(\omega t+\vec{K}_0\cdot\vec{r}+\varphi_++\varphi_--\varphi_0) \tag{32}$$

so that, in the sum over μ,ν,λ in Eq. (26), only two terms contribute, $\vec{\mathscr{P}}^{(+)}$ and $\vec{\mathscr{P}}^{(-)}$, associated, respectively, to $(\mu,\nu,\lambda)=(+,0,-)$ and $(\mu,\nu,\lambda)=(-,0,+)$. Thus

$$\vec{\mathscr{P}}^{\,r}=\vec{\mathscr{P}}^{(+)}+\vec{\mathscr{P}}^{(-)} \tag{33}$$

with $\vec{\mathscr{P}}^{(+)}$ given by

$$\mathscr{P}^{(+)*}_{Q'}=\int f(\vec{v})d^3v\sum_k\left[\frac{{}_b g^{+}_{Q'}(k)}{L^k_b(\vec{K}-\vec{K}_0)}+\frac{{}_a g^{+}_{Q'}(k)}{L^k_a(\vec{K}-\vec{K}_0)}\left[1-\gamma_{ba}\frac{\mathscr{A}^k_{ba}}{L^k_b(\vec{K}-\vec{K}_0)}\right]\right]\frac{\mathscr{C}}{L(-\vec{K}_0)}\left[\frac{1}{L(\vec{K})}+\frac{1}{L(\vec{K}_0)^*}\right] \tag{34}$$

and $\mathscr{P}^{(-)*}_{Q'}$ is deduced from (34) by exchanging $+\leftrightarrow-$ and $\vec{K}\leftrightarrow-\vec{K}$. $\vec{\mathscr{P}}^{(\pm)}$ are physically interpreted as the contribution of the diffraction of the $(\mp\vec{K})$ wave by a grating formed by waves $(\pm\vec{K})$ and $\vec{K}_0$. One has defined the following notations:

$${}_\beta g^{+}_{Q'}(k)=\sum_{Q,q,q',q''}({}^{q''}_{\beta\alpha}G^{1k}_{Q'Q})({}^{q}_{\beta\alpha}G^{1k}_{q'Q})\times e^{(+)*}_{q'}e^{0}_{-q}e^{(-)*}_{-q''}\ , \tag{35a}$$

$${}_\beta g^{-}_{Q'}(k)=\sum_{Q,q,q',q''}({}^{q''}_{\beta\alpha}G^{1k}_{Q'Q})({}^{q}_{\beta\alpha}G^{1k}_{q'Q})\times e^{(-)*}_{q'}e^{0}_{-q}e^{(+)*}_{-q''}\ . \tag{35b}$$

(One should note that these coefficients vanish for $k>2$.) Due to the degeneracy in frequency, expressions of Eqs. (29)–(31) have been simplified as

$$L(\vec{K}_\mu)=\Gamma_{ab}+i(\omega-\omega_0-\vec{K}_\mu\cdot\vec{v})\ , \tag{36}$$

$$L^k_\beta(\vec{K}_\mu-\vec{K}_\nu)=\Gamma_\beta(k)-i(\vec{K}_\mu-\vec{K}_\nu)\cdot\vec{v}\ , \tag{37}$$

and $\mathscr{C}$ is a constant defined by

$$\mathscr{C}=-\frac{i}{4}\frac{P^4_{ab}n}{3}\mathscr{E}^{+}\mathscr{E}^{-}\mathscr{E}^{0}\ . \tag{38}$$

The phase-conjugate field is then readily obtained by Eqs. (23), (33), and (34). The general expression obtained contains all the desirable information on the emission dependence (amplitude, polarization) on the angular momentum (J_a, J_b), on the incidence angle, and on the polarizations of the incident fields.

In a simple model, in which all the relaxation constants are equal, and $\gamma_{ba}=0$, expression (34) is greatly simplified by using the following relation (Appendix A):

$$\sum_k {}_b g_{Q'}^{\pm}(k) = \sum_k {}_a g_{Q'}^{\mp}(k) \ . \tag{39}$$

This implies

$$\sum_{k,\beta} {}_\beta g_{Q'}^{-}(k) = \sum_{k,\beta} {}_\beta g_{Q'}^{+}(k) = g_{Q'} \tag{40}$$

and in particular, for $J_a = J_b$,

$$\sum_k {}_\beta g_{Q'}^{\pm}(k) = \tfrac{1}{2} g_{Q'} \ . \tag{41}$$

We will see that this symmetry relation allows us to simplify some of the expressions in the next sections.

Before discussing the problem of the velocity integration of Eq. (34), let us first consider the simple case of stationary atoms. In that case, Eq. (34) simplifies because

$$L_\beta^k(\vec{K}-\vec{K}_0) = \Gamma_\beta(k) \ , \tag{42}$$

$$L(\vec{K}) = \Gamma_{ab} + i(\omega-\omega_0) = L(\omega) \ , \tag{43}$$

so that the standard components of $\vec{\mathscr{P}}^{\,r}$ are given by

$$\mathscr{P}_{Q'}^{r*} = \frac{2\Gamma_{ab}\mathscr{C}}{L(\omega)\,|L(\omega)|^2} \sum_k \left[\frac{{}_b g_{Q'}^{+}(k) + {}_b g_{Q'}^{-}(k)}{\Gamma_b(k)} + \frac{{}_a g_{Q'}^{+}(k) + {}_a g_{Q'}^{-}(k)}{\Gamma_a(k)} \times \left[1 - \gamma_{ba} \frac{\mathscr{A}_{ba}^k}{\Gamma_b(k)} \right] \right] . \tag{44}$$

In this expression, the gratings formed by waves $(\vec{K},\vec{K}_0)$ and $(-\vec{K},\vec{K}_0)$ play comparable roles, and the relative amplitude of $\mathscr{P}^{(+)}$ and $\mathscr{P}^{(-)}$ just depends on the polarization of the incident fields. Assuming that it is possible to vary the angular separation θ for a given set of incident polarizations [which means that $\vec{\mathscr{E}}_0$ is orthogonal to the $(\vec{K},\vec{K}_0)$ plane], it is thus clear that the θ dependence of the reemitted field is only due to propagation effects (conditions of transversality) since $\vec{\mathscr{P}}^{\,r}$ is independent of θ (Sec. VI A 5).

IV. ANGULAR DEPENDENCE OF THE PC FIELD FOR DOPPLER-BROADENED SYSTEMS

In all the following, we assume a Maxwellian velocity distribution $f(\vec{v})$ [Eq. (7)], and we only discuss the case of *large Doppler broadening,* i.e., $Ku \gg \Gamma_{\alpha\beta}(k)$ $(\alpha=a, b;\ \beta=a,b)$.

In Eq. (34), the velocity integration consists in the integration of products of L-type denominators multiplied by constant factors. Such an integration is entirely identical to the one discussed previously in I (Refs. 3 and 21) for a two-level system; the physical description of the angular separation dependence is still valid, the main difference being that instead of considering only a population grating, one now has to sum over various types of gratings.

If the polarizations of the pumps and probe are parallel, population gratings are formed, corresponding to a spatial modulation of the population of the Zeeman sublevels. If the polarization of the probe is orthogonal to the one of the pumps, there is no light intensity interference pattern, but the polarization of the total incident field is spatially modulated, thus producing a spatial modulation in the Zeeman atomic coherence. In the framework of the tensorial formalism, these two kinds of gratings decay differently since they originate in various combination of multipole observables with different relaxation rates. Population gratings involve the global population of the levels $(k=0)$ and longitudinal orientation $(k=1)$ and alignment $(k=2)$. On the other hand, coherence gratings correspond to transverse orientation or alignment. A second cause of grating relaxation lies in the motional washout: due to the atomic motion, an absorber runs over a grating period in a mean time of the order of Λ/u [$\lambda/2u\sin(\theta/2)$, for the $(\vec{K},\vec{K}_0)$ grating]. This introduces a *"residual Doppler broadening"* ($\approx Ku\theta$), which shortens the grating decay time.

The velocity integration of Eq. (34) will be discussed for several regimes, depending on the relative importance of the Doppler linewidth Ku, the residual Doppler broadening $Ku\theta$, and the various relaxation times $\Gamma_{\alpha\beta}(k)$: (i) the quasicollinear configuration $[Ku\theta \ll \Gamma_{ab}, \Gamma_\beta(k)]$; (ii) the orthogonal configuration $(\theta=\pi/2)$; and (iii) two intermediate regimes with $\theta \ll 1$, residual Doppler broadening either large $[Ku\theta \gg \Gamma_\beta(k), \Gamma_{ab}]$ or small compared with the optical linewidth $[\Gamma_{ab} \gg Ku\theta, \Gamma_\beta(k)]$. In all the cases with $\theta \ll 1$, only the gratings induced by waves $\vec{K}$ and $\vec{K}_0$ $(\vec{K}_0 \approx \vec{K})$ contribute efficiently to the emission. The $(-\vec{K},\vec{K}_0)$ grating yields negligible contribution because of its very large motional decay.

A. Quasicollinear configuration $(\theta \approx 0)$

For $\theta=0$ in Eq. (34), we only have to integrate over one velocity component the product of two resonant denominators [since $L_a^k(\vec{K}-\vec{K}_0)$ is independent of $\vec{v}$]. This integration yields the standard components of $\vec{\mathscr{E}}^{\,r}$ by using relation $\mathscr{E}_{Q'}^{r*} = -(iKL/2\epsilon_0)\mathscr{P}_{Q'}^{r*}$ [Eq. (23)]:

$$\mathscr{P}_{Q'}^{r*} = \mathscr{C} \frac{\sqrt{\pi}}{Ku} \frac{1}{L(\omega)} \sum_k \left[\frac{{}_b g_{Q'}^{+}(k)}{\Gamma_b(k)} + \frac{{}_a g_{Q'}^{+}(k)}{\Gamma_a(k)} \left[1 - \gamma_{ba} \frac{\mathscr{A}_{ba}^k}{\Gamma_b(k)} \right] \right] \tag{45}$$

[$L(\omega)$ is the resonant denominator introduced in Eq. (43)].

The exactly collinear configuration is not of much interest for phase conjugation and is identical to the basic scheme of polarization spectroscopy,[23] but the validity of Eq. (45) can be extended to quasicollinear configuration as long as $\theta \ll \Gamma_\alpha(k)/Ku, \Gamma_{ab}/Ku$.

It is worth comparing the reemitted intensity I^r for the two cases of stationary atoms and large Doppler broadenings, keeping the same set of polarizations and the same geometry ($\theta \simeq 0$). One finds easily that, on resonance,

$$I_{\text{Doppler}}/I_{\text{stat}} = \frac{\pi}{16}\left[\frac{\Gamma_{ab}}{Ku}\right]^2 \tag{46}$$

[one has $I_{\text{Doppler}} \propto \pi/|KuL(\omega)|^2$ and $I_{\text{stat}} \propto 16\Gamma_{ab}^2/|L(\omega)|^6$].

This result appears as quite general (at least for third order) and does not depend on the incident polarizations and the population relaxation: it involves only the ratio of the homogeneous to inhomogeneous width. This extends the results of I.

B. Orthogonal configuration ($\theta = \pi/2$)

In this case, one has to take into account the two gratings $(\vec{K},\vec{K}_0)$ and $(-\vec{K},\vec{K}_0)$ which both contribute to the PC emission. The terms to be integrated over two velocity components in Eq. (34) are terms like

$$[L(-\vec{K}_0)]^{-1}\{[L(\vec{K})]^{-1}+[L(\vec{K}_0)^*]^{-1}\}[L_\beta^k(\vec{K}-\vec{K}_0)]^{-1}$$

or like

$$[L(-\vec{K}_0)]^{-1}\{[L(\vec{K})]^{-1}+[L(\vec{K}_0)^*]^{-1}\} \times [L_\beta^k(\vec{K}-\vec{K}_0)L_\alpha^k(\vec{K}-\vec{K}_0)]^{-1}$$

(terms due to the spontaneous emission).

Assuming $\Gamma_{\alpha\beta}(k) \ll Ku$, it is easy to show [see Eq. (B5) of Appendix B] that the terms due to spontaneous emission are much smaller (by a factor of the order of γ_{ba}/Ku) than the first terms, which themselves can be rewritten as

$$[L(-\vec{K}_0)L(\vec{K})L(\vec{K}_0)^*]^{-1}\left[1+\frac{2\Gamma_{ab}-\Gamma_\beta(k)}{\Gamma_\beta(\vec{k})-i(\vec{K}-\vec{K}_0)\cdot\vec{v}}\right],$$

and in the integration, only $[L(-\vec{K}_0)L(\vec{K})L(\vec{K}_0)^*]^{-1}$ contributes notably (the other term being $[2\Gamma_{ab}-\Gamma_\beta(k)]/4Ku\sqrt{\pi}$ smaller). The integration is easily performed as in I,[3,21] and one gets

$$\mathscr{P}_{Q'}^{r*} = \frac{4\sqrt{\pi}}{(Ku)^3}\mathscr{C}g_{Q'}\frac{g(Z)}{Z}\left[e^{-Z^2}-\frac{2i}{\sqrt{\pi}}g(Z)\right] \tag{47}$$

with

$$Z = (\omega-\omega_0)/Ku \tag{48}$$

and

$$g(Z) = e^{-Z^2}\int_0^Z e^{t^2}dt\ . \tag{49}$$

The geometrical coefficient $g_{Q'}$ is defined in Eqs. (39)–(41).

The emission features are independent of the details of the relaxation processes. The polarization state of the incident fields and the angular momentum of the energy levels determine the overall efficiency, but do not affect the emission line shape which is Doppler-broadened. As could be expected, one gets the same frequency dependence as for nondegenerate two-level systems [see I, Eq. (39)].

C. Large residual Doppler broadening [$Ku\theta \gg \Gamma_{ab}, \Gamma_\beta(k)$]

The integration of Eq. (34) is analogous to the one discussed in I [Eq. (55)] and we find

$$\mathscr{P}_{Q'}^{r*} = \frac{4\sqrt{\pi}}{(Ku)^3\theta^2}\mathscr{C}g_{Q'}\frac{g(Y)-\frac{1}{2}i\sqrt{\pi}(1-e^{-Y^2})}{Y} \tag{50}$$

with

$$Y = 2(\omega-\omega_0)/Ku\theta\ . \tag{51}$$

One can remark that the contribution of the cascade effect is negligible after velocity integration (it gives a term $\gamma_{ba}/Ku\theta$ smaller than the main terms). The emission linewidth given by Eq. (50) exhibits a Doppler broadening of the order of $Ku\theta$ (see I, Sec. 4.3) and the intensity at line center is reduced by a factor of $(\Gamma/Ku\theta)^4$ by comparison with the fully collinear configuration [this factor is calculated in a simple model in which $\Gamma_{\alpha\beta}(k)=\Gamma$, $\gamma_{ba}=0$]. This reduction is both due to the partial selection of two velocity components (instead of one axial component for $\theta \approx 0$), and to the atomic thermal motion responsible for the washing out of the induced gratings (the mean lifetime of these gratings is $(Ku\theta)^{-1}$, instead of $[\Gamma_\beta(k)]^{-1}$, see I).

D. Intermediate regime: residual Doppler effect smaller than the optical linewidth [$\Gamma_{ab} \gg Ku\theta, \Gamma_\beta(k)$]

Since $\Gamma_{ab}, Ku\theta \ll Ku$, only the $(\vec{K},\vec{K}_0)$ grating contributes. The velocity integration is better performed in a basis defined by axes $\vec{K}_1=(\vec{K}+\vec{K}_0)/2$ and $\vec{K}_2=(\vec{K}_0-\vec{K})/2$ [velocity components v_1, v_2; see Eqs. (42) and (43) of I]. In the integration of Eq. (34), the main contribution comes from terms like

$$\frac{1}{[\Gamma_{ab}+i(\omega-\omega_0+K_1v_1+K_2v_2)]} \times \frac{1}{[\Gamma_{ab}+i(\omega-\omega_0+K_2v_2-K_1v_1)]}\frac{1}{[\Gamma_\beta(k)+2iK_2v_2]}\ . \tag{52}$$

Because $K_1u \approx Ku \gg \Gamma_{ab}, K_2u, \omega-\omega_0$, one easily shows [see Eqs. (59) and (60) of I] that integration of (52) over v_1 leaves us with

$$\frac{\sqrt{\pi}}{Ku}\frac{1}{\Gamma_{ab}+i(\omega-\omega_0+K_2v_2)}\frac{1}{\Gamma_\beta(k)+2iK_2v_2}\ . \tag{53}$$

In the case when $K_2u \simeq Ku\theta/2 \ll \Gamma_{ab}$, K_2v_2 can be neglected in the first denominator of (53), and one gets, after some transformations,

$$\mathscr{P}_{Q'}^{r*}=\frac{\pi}{(Ku)^2\theta}\frac{\mathscr{C}}{\Gamma_{ab}+i(\omega-\omega_0)}\sum_k\left[{}_b g_{Q'}^{+}(k)\psi(\Gamma_b(k)/Ku\theta)+{}_a g_{Q'}^{+}(k)\psi(\Gamma_a(k)/Ku\theta)-\gamma_{ba}\mathscr{A}_{ba}^{k}\,{}_a g_{Q'}^{+}(k)\frac{\psi(\Gamma_b(k)/Ku\theta)-\psi(\Gamma_a(k)/Ku\theta)}{\Gamma_a(k)-\Gamma_b(k)}\right] \tag{54}$$

with

$$\psi(x)=e^{x^2}\left[1-\frac{2}{\sqrt{\pi}}\int_0^x e^{-t^2}dt\right]. \tag{55}$$

[The (real) function $\psi(x)$ is related to the plasma dispersion function W (Ref. 24) by $\psi(x)=W(ix)$.]

Equation (54) shows clearly that the line shape is a Lorentzian [width Γ_{ab}, not affected by the residual Doppler effect], while the overall efficiency depends on the combination of atomic relaxation, spontaneous emission, and motional broadening.

In Eq. (54), two limit cases can be considered.

(i) If $\Gamma_\beta(k)\ll Ku\theta$, $\psi(0)\simeq 1$ and Eq. (54) reduces to

$$\mathscr{P}_{Q'}^{r*}=\frac{\pi}{(Ku)^2\theta}\frac{\mathscr{C}}{\Gamma_{ab}+i(\omega-\omega_0)}g_{Q'}\,. \tag{56}$$

In that case, the polarization properties are related to $g_{Q'}$ as in Eqs. (47) and (50). The motional broadening is the main limitation to the PC efficiency, so that the intensity is proportional to $(Ku\theta)^{-2}$, instead of $(Ku\theta)^{-4}$ in Eq. (50), when the line-shape Doppler broadening also contributes.

(ii) If $Ku\theta\ll\Gamma_\beta(k)$, $\psi(x)\simeq(x\sqrt{\pi})^{-1}$ and Eq. (54) is thus equivalent to Eq. (45).

V. GENERAL PROPERTIES OF THE POLARIZATION OF THE PC FIELD

A. Independence between emission line shape and polarization properties

In all the cases considered above, we have seen that the emission line shape does not depend on the incident polarizations. Reciprocally, this independence applies to the polarization of the PC wave, which is found to be independent of the frequency detuning. This property is general, independent of the incidence angle θ, as long as $\Gamma_\beta(k)\ll\Gamma_{ab}$. The emission line shape is thus exactly similar to the one obtained for nondegenerate two-level systems (see I). This property is valid at third order and, in general, fails for intense incident beams. At saturation, emission line shape and polarization properties become strongly correlated.[13]

B. Angular dependence of the polarization

When $\Gamma_{ab}\gg\Gamma_\beta(k)$, the polarization dependence can be separated in two main regimes.

(i) If $Ku\theta\lesssim\Gamma_\beta(k)$, the polarization depends on the relative value of $\Gamma_\beta(k)$ and $Ku\theta$, so that it is sensitive to the relaxation processes in the gas medium (collisions, etc.).

(ii) If $Ku\theta\gg\Gamma_\beta(k)$, the polarization state no longer depends on the relaxation processes, and is only related to the geometrical coefficients $g_{Q'}$ [Eqs. (39)–(41)].

In particular, in this regime, polarization selection rules cannot be broken by collisional effect, since the latter ones do not have enough time to operate during the grating mean lifetime.[9] A noteworthy point is that coefficients $g_{Q'}$ are identical whether one considers the grating induced by the waves $(\vec{K},\vec{K}_0)$ or waves $(-\vec{K},\vec{K}_0)$. It implies that the PC polarization remains unaffected when the polarizations of the pump waves $\vec{K}$ and $-\vec{K}$ are exchanged [for $Ku\theta\gg\Gamma_\beta(k)$]. This remarkable symmetry property allows us to predict the behavior of PC mirrors when the pumps have arbitrary linear polarization. Because of symmetry the probe polarization must be unchanged when it is directed along the bisectors of the pump polarizations. These directions represent the "neutral" axes of the PC mirror. However, in general, the amplitudes of the PC reflectivity along these two directions are not equal. This will be discussed in more detail in Sec. VI C.

C. Transversality of the reemitted PC field

As long as we are interested in a small angular separation θ, we can consider that both probe and PC wave have a polarization transverse to the pump axis $\vec{K}$. Elementary physical arguments allow one to understand that the nonlinear polarization induced in the medium is transverse. For instance, if one defines the standard basis so that $\vec{u}_0=\vec{u}_z\,||\,\vec{K}$, symmetry conditions on $3j$ symbols impose that

$$({}^{q''}_{\beta\alpha}G^{1k}_{Q'Q})({}^{q}_{\beta\alpha}G^{1k}_{q'Q})=0 \quad \text{for } q,q',q''=\pm1 \text{ and } Q'=0$$

which means that there is no longitudinal optical polarization $(Q'=0)$.

As a consequence of this transversality, Eqs. (50) and (54) [combined with Eq. (23)] yield directly the standard components of the emitted field. On the other hand, we will see in Sec. VI that for $\theta=\pi/2$, a fully longitudinal polarization can be induced in the medium, without reemission of a propagating field.

VI. POLARIZATION CHARACTERISTICS OF PC MIRRORS

In this paper, the theoretical analysis is restricted to the case of low-power incident fields, so that a third-order perturbation expansion is valid [Eq. (26)]. The emitted EM field then depends linearly on each of the incident fields, and it is possible to predict the behavior of the PC mirror for any set of incident polarizations by discussing its properties in a few remarkable cases. A complete analysis is actually given when one considers the following sets (with an arbitrary probe polarization): pumps linearly polarized, with parallel or orthogonal polarizations; pumps circularly polarized (co-rotating, or counter-rotating).

A. Cross-polarized pumps

Here we consider the case when the pump beams have linear and orthogonal polarization, and the probe beam has an arbitrary linear polarization. One assumes a small angular separation $\theta \ll 1$. For convenience, we choose the orthonormal basis so that

$$\vec{e}^{\,+}=\vec{u}_z=\vec{u}_0\ , \tag{57a}$$

$$\vec{e}^{\,-}=\vec{u}_x=\frac{1}{\sqrt{2}}(\vec{u}_{-1}-\vec{u}_{+1})\ , \tag{57b}$$

and

$$\vec{e}^{\,0}=\vec{u}_z\cos\alpha_0+\vec{u}_x\sin\alpha_0\ , \tag{58}$$

where α_0 is the $(\vec{e}^{\,0},\vec{u}_z)$ angle.

The standard components of the polarizations are easily obtained from Eqs. (13) and (16), and when replaced in Eq. (35), yield the following ${}_\beta g^{\pm}_{Q'}$ coefficients:

$${}_\beta g_0^{+}(k)=d_\beta(k)\sin\alpha_0\ , \tag{59a}$$

$${}_\beta g_0^{-}(k)=c_\beta(k)\sin\alpha_0\ , \tag{59b}$$

$${}_\beta g_{-1}^{+}(k)=-{}_\beta g_{+1}^{+}(k)=c_\beta(k)\frac{\cos\alpha_0}{\sqrt{2}}\ , \tag{60a}$$

$${}_\beta g_{-1}^{-}(k)=-{}_\beta g_{+1}^{-}(k)=d_\beta(k)\frac{\cos\alpha_0}{\sqrt{2}}\ . \tag{60b}$$

In these equations, one has introduced the following quantities:

$$c_\beta(k)=({}^{0}_{\beta\alpha}G^{1k}_{00})({}^{-1}_{\beta\alpha}G^{1k}_{10})\ , \tag{61a}$$

$$d_\beta(k)=({}^{1}_{\beta\alpha}G^{1k}_{01})^2\ . \tag{61b}$$

The definition of the $3j$ symbols used in Eq. (27) implies that

$$c_\beta(k)=\begin{cases}(-1)^{k/2}\begin{Bmatrix}1 & 1 & k\\ J_\beta & J_\beta & J_\alpha\end{Bmatrix}^2 & \text{for } k=0,2\\ 0\ \text{ for } k=1\end{cases} \tag{62a}$$

$$d_\beta(k)=\begin{cases}\frac{3}{2}\begin{Bmatrix}1 & 1 & k\\ J_\beta & J_\beta & J_\alpha\end{Bmatrix}^2 & \text{for } k=1,2\\ 0\ \text{ for } k=0\ .\end{cases} \tag{62b}$$

Equations (39)–(41), substituted into Eqs. (59) and (60), show that

$$\sum_k c_\beta(k)=\sum_k d_\alpha(k)\quad(\alpha\neq\beta)\ . \tag{63}$$

By substituting Eqs. (57)–(63) into Eq. (34), one sees that the polarization of the reemitted field verifies $\mathscr{P}^{r*}_{+1}=-\mathscr{P}^{r*}_{-1}$, and then

$$\vec{\mathscr{P}}^{\,r*}=\sum_{Q'}\mathscr{P}^{r*}_{Q'}\vec{u}_{Q'}=\mathscr{P}^{r*}_0\vec{u}_z+\mathscr{P}^{r*}_x\vec{u}_x \tag{64}$$

with

$$\mathscr{P}^r_x=\sqrt{2}\mathscr{P}^r_{-1}\ . \tag{65}$$

If we define the unitary vector $\vec{e}^{\,r}$ by

$$\vec{\mathscr{P}}^{\,r}=\mathscr{P}^r\vec{e}^{\,r} \tag{66}$$

($\mathscr{P}^r$, complex amplitude), and the angle α_r by

$$\vec{e}^{\,r}=\cos\alpha_r\,\vec{u}_z+\sin\alpha_r\,\vec{u}_x\ , \tag{67}$$

it is easy to verify that α_r is real, and, for $\theta\ll 1$, $\vec{e}^{\,r}$ is the linear polarization of the PC wave. From Eqs. (57), (58), (64), and (65), a simple relation between α_0 and α_r can be deduced:

$$\tan\alpha_0\tan\alpha_r=D\ , \tag{68}$$

where D is a quantity independent of α_0. As concerns its polarization properties, the characterization of the PC mirror will be achieved with the determination of the constant D.[12] Equation (68) can be predicted by simple arguments of conservation of the angular momentum and linearity of a third-order perturbation theory,[9] but the above calculation is required to determine D.

1. *Case of stationary atoms*

From Eq. (34), one gets for stationary atoms

$$\frac{\mathscr{P}^{(\pm)}_z}{\sin\alpha_0}=\frac{\mathscr{P}^{(\pm)}_x}{\cos\alpha_0} \tag{69}$$

so that

$$\vec{e}^{\,r}=\vec{u}_z\sin\alpha_0+\vec{u}_x\cos\alpha_0 \tag{70}$$

and thus

$$D=1\ . \tag{71}$$

This characterizes the PC mirror as the analog of a half-wave plate, whose neutral axes are along the bisectors of the pump polarizations (at least for $\theta\ll 1$; extension to an arbitrary value of θ is given in Sec. VI A 5).[12] The high symmetry of such a type of PC mirror can be understood by the equal importance of the contribution of the $(\vec{K},\vec{K}_0)$ grating and of the $(-\vec{K},\vec{K}_0)$ grating.

2. *Small residual Doppler broadening ($Ku\theta\ll\Gamma_{ab}$)*

The results of Sec. IV D apply, and yield for the induced polarization

$$\vec{\mathscr{P}}^{\,r*}=\frac{\pi}{(Ku)^2\theta}\frac{\mathscr{C}}{\Gamma_{ab}+i(\omega-\omega_0)}\times(d\,\sin\alpha_0\,\vec{u}_z+c\,\cos\alpha_0\,\vec{u}_x) \tag{72}$$

and c and d are now given by

$$d=\sum_{k=1,2}\left[d_b(k)\psi_b^k+d_a(k)\psi_a^k-\gamma_{ba}\mathscr{A}^k_{ba}d_a(k)\frac{\psi_b^k-\psi_a^k}{\Gamma_a(k)-\Gamma_b(k)}\right]\ , \tag{73a}$$

$$c=\sum_{k=0,2}\left[c_b(k)\psi_b^k+c_a(k)\psi_a^k - \gamma_{ba}\mathscr{A}_{ba}^k c_a(k)\frac{\psi_b^k-\psi_a^k}{\Gamma_a(k)-\Gamma_b(k)}\right], \quad (73b)$$

with

$$\psi_\beta^k=\psi\left[\frac{\Gamma_\beta(k)}{Ku\theta}\right]. \quad (74)$$

Equations (72) combined with Eq. (67) yield directly $D=c/d$.

Two limiting regimes can be considered in Eqs. (73).

(a) When $Ku\theta \ll \Gamma_\beta(k)$, $\psi_\beta^k \simeq Ku\theta/\Gamma_\beta(k)\sqrt{\pi}$ and coefficients c and d can be written as

$$d=\frac{Ku\theta}{\sqrt{\pi}}\sum_{k=1,2}\left[\frac{d_b(k)}{\Gamma_b(k)}+\frac{d_a(k)}{\Gamma_a(k)}\left[1-\gamma_{ba}\frac{\mathscr{A}_{ba}^k}{\Gamma_b(k)}\right]\right], \quad (75a)$$

$$c=\frac{Ku\theta}{\sqrt{\pi}}\sum_{k=0,2}\left[\frac{c_b(k)}{\Gamma_b(k)}+\frac{c_a(k)}{\Gamma_a(k)}\left[1-\gamma_{ba}\frac{\mathscr{A}_{ba}^k}{\Gamma_b(k)}\right]\right]. \quad (75b)$$

These coefficients, and thus the polarization properties, strongly depend on the details of the relaxation processes.

If the *population* grating ($k=0$) is predominant, [$\Gamma_\beta(0) \ll \Gamma_\beta(1)$, $\Gamma_\beta(2)$, i.e., $c \gg d$], only the z component of the probe is reflected, and yields a PC wave polarized along $\vec{u}_x$. This is the result predicted by the scalar theory [Eq. (1)].

As $\Gamma_\beta(k) \geq \Gamma_\beta(0)$ it is theoretically impossible to get a predominant orientation or alignment grating. However, it is worthwhile to analyze separately the effects of the various types of gratings.

For the *orientation* grating, the PC wave is polarized along $\vec{u}_z$, and only the component of the probe polarized along $\vec{u}_x$ contributes to the signal.

For the *alignment* grating, the PC wave is polarized along $(\frac{3}{2}\sin\alpha_0\,\vec{u}_z-\cos\alpha_0\,\vec{u}_x)$.

(b) When $Ku\theta$ increases, the difference existing between the contributions from various orders tends to vanish. In the limit $Ku\theta \gg \Gamma_\beta(k)$, $\psi_\beta^k \to 1$, and one has

$$d=c=\sum_{k,\beta} d_\beta(k)=\sum_{k,\beta} c_\beta(k) \quad (76)$$

so that $D=1$. The polarization dependence on the relaxation processes (collisions, etc.) disappears. However, the intermediate regime, $Ku\theta \simeq \Gamma_\beta(k)$, needs, for its description, an adequate knowledge of these processes.

The above discussion applies to most of the experimental studies. Experiments performed on various neon lines have been interpreted with the help of Eqs. (72) and (73). One has shown that in general, D takes values close to 1, but non-negligible deviations from $D=1$ are produced by a residual influence of collisional relaxation and spontaneous emission (for instance, $D \simeq 1.1$ for the 640-nm line of neon[12,13]).

3. *Large residual Doppler broadening [$Ku\theta \gg \Gamma_{ab}, \Gamma_\beta(k)$]*

The polarization characteristics remain independent of the relaxation processes (in particular $D=1$), but the emission line shape is gradually modified and undergoes a Doppler broadening which increases with the angular separation [cf. Eq. (50)].[3]

4. *Single relaxation model*

Let us consider the case when the system's relaxation can be reduced to one relaxation time per level and the spontaneous transfer is negligible.[25] Here,

$$\Gamma_\beta(k)=\gamma_\beta,\quad \gamma_{ba}=0 \quad (77)$$

so that $\psi_\beta^k=\psi_\beta$. Thus c and d can be written as [Eqs. (63)—(73)]

$$d=\psi_b\sum_k d_b(k)+\psi_a\sum_k d_a(k)\ , \quad (78a)$$

$$c=\psi_b\sum_k d_a(k)+\psi_a\sum_k d_b(k)\ . \quad (78b)$$

From these equations, one easily deduces that, for $J_a=J_b$, one has $c=d$ ($D=1$), which implies that the induced polarization $\vec{\mathscr{P}}^{\,r}$ is directed along $\sin\alpha_0\,\vec{u}_z+\cos\alpha_0\,\vec{u}_x$, independent of θ. Such a result can be extended to $J_a \neq J_b$, when $\gamma_a=\gamma_b$. In the latter example, all the gratings induced in levels a and b have the same lifetime. This allows us to interpret the prediction $D=1$ for $Ku\theta \gg \Gamma_\beta(k)$: the residual Doppler broadening becomes the main relaxation process limiting the amplitude of the optically induced gratings, and thus they can be considered as governed by a single relaxation constant [$\approx(Ku\theta)^{-1}$].

5. *Effect of the pump-probe angular separation*

When θ increases, the probe polarization may have a component along $\vec{K}$ ($\vec{u}_y$). However, this component cannot participate in the PC process because due to the conservation of angular momentum, symmetry rules forbid reemission when the three incident polarizations (pumps and probe) are orthogonal two by two. Thus we have to project $\vec{e}^{\,0}$ on the plane ($\vec{u}_x,\vec{u}_z$). As an example, let us consider the case when the ($\vec{K},\vec{K}_0$) plane is perpendicular to the pump polarization $\vec{u}_z$. The probe polarization may be projected onto $\vec{u}_z$ and $\vec{u}_{x'}=\vec{u}_x\cos\theta+\vec{u}_y\sin\theta$ (see Fig. 2). In the PC process, $\vec{u}_z$ is transformed in $\vec{u}_x$, which is decomposed in a transverse polarization $\vec{u}_{x'}\cos\theta$, and a longitudinal one (along $\vec{K}_0$) which cannot propagate. On the other hand, $\vec{u}_{x'}$ is transformed in $\vec{u}_z\cos\theta$ (since $\vec{u}_y$ does not contribute). If we assume $D=c/d=1$ [Eq. (71)], one easily sees that the PC mirror still behaves *like a $\lambda/2$ plate,* but with an efficiency reduced by $\cos^2\theta$ (in addition to the line shape and angular dependence). In particular, for $\theta=\pi/2$, the PC reflectivity cancels: an incident probe polarized along $\vec{u}_z$ generates in the medium a longitudinal nonlinear polarization which cannot radiate, and on the other hand the PC efficiency is zero for a probe polarized along $\vec{u}_y$. The latter selection rule, dictated by propagation effects, is easily extended to arbitrary incident inten-

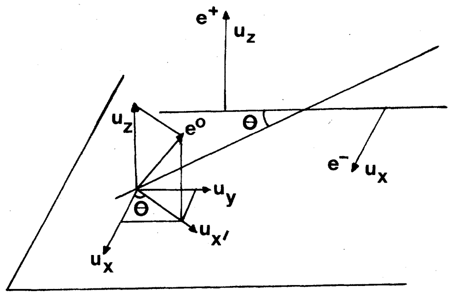

FIG. 2. Components of the probe polarization for non-negligible values of θ: there is a component along $\vec{u}_y$, for which the reemission is forbidden, because of symmetry properties.

sities (since the laws of conservation of angular momentum are not restricted to third-order theory).

6. *Probe circularly polarized*

The previous discussion allows us to predict the behavior of the PC mirror when the probe is circularly polarized. If the two efficiencies c and d are different $[Ku\theta \lesssim \Gamma_\beta(k)]$, an incident circular polarization is reflected into an elliptical one.

If $c=d$ (as is the case under broad classes of hypotheses), a circular σ^+ polarization is reflected back into a σ^- polarization. This means that, due to the reversal of the propagation axis $(\vec{K}_0 \rightarrow -\vec{K}_0)$, left (right) circularly polarized light is reflected left (right) circularly polarized. This demonstrates more completely the analogy of the PC mirror with a half-wave plate when $D=1$.

B. Parallel polarizations of the pump

We now turn to the case when the pump beams have the same linear polarization that we take as the quantization axis, $\vec{e}^{\,+}=\vec{e}^{\,-}=\vec{u}_z$. This polarization is assumed to be orthogonal to the $(\vec{K},\vec{K}_0)$ plane, so that the (linear) polarization of the probe can be decomposed like

$$\vec{e}^{\,0}=\cos\alpha_0\,\vec{u}_z+\sin\alpha_0\,\vec{u}_x \tag{79a}$$

$$=\cos\alpha_0\,\vec{u}_0+\frac{\sin\alpha_0}{\sqrt{2}}(\vec{u}_{-1}-\vec{u}_{+1}) \tag{79b}$$

($\vec{u}_x$ is now the unit vector orthogonal to $\vec{u}_z$ and $\vec{K}_0$). In that case, all the incident polarizations, and thus the PC polarization, are orthogonal to the probe wave vector $\vec{K}_0$. By reporting the standard components of the polarization vectors in Eqs. (34) and (35), one easily shows that the induced PC polarization decomposes in two components along $\vec{u}_z$ and $\vec{u}_x$:

$$\mathscr{P}_z^{(\pm)*}=\cos\alpha_0\int f(\vec{v})d^3v\frac{\mathscr{C}}{L(-\vec{K}_0)}\left[\frac{1}{L(\pm\vec{K})}+\frac{1}{L(\vec{K}_0)^*}\right]\times\sum_{k=0,2}\left[\frac{h_b(k)}{L_b^k(\pm\vec{K}-\vec{K}_0)}+\frac{h_a(k)}{L_a^k(\pm\vec{K}-\vec{K}_0)}\left[1-\gamma_{ba}\frac{\mathscr{A}_{ba}^k}{L_b^k(\pm\vec{K}-\vec{K}_0)}\right]\right], \tag{80a}$$

$$\mathscr{P}_x^{(\pm)*}=-\sin\alpha_0\int f(\vec{v})d^3v\frac{\mathscr{C}}{L(-\vec{K}_0)}\left[\frac{1}{L(\pm\vec{K})}+\frac{1}{L(\vec{K}_0)^*}\right]\times\sum_{k=1,2}\left[\frac{f_b(k)}{L_b^k(\pm\vec{K}-\vec{K}_0)}+\frac{f_a(k)}{L_a^k(\pm\vec{K}-\vec{K}_0)}\left[1-\gamma_{ba}\frac{\mathscr{A}_{ba}^k}{L_b^k(\pm\vec{K}-\vec{K}_0)}\right]\right], \tag{80b}$$

where the coefficients $f_\beta(k)$ $(k=1,2)$ and $h_\beta(k)$ $(k=0,2)$, are obtained from Eqs. (27)—(35):

$$h_\beta(k)=\left[1+\frac{k}{2}\right]\begin{Bmatrix}1 & 1 & k\\ J_\beta & J_\beta & J_\alpha\end{Bmatrix}^2, \tag{81a}$$

$$f_\beta(k)=(-1)^{k+1}\frac{3}{2}\begin{Bmatrix}1 & 1 & k\\ J_\beta & J_\beta & J_\alpha\end{Bmatrix}^2. \tag{81b}$$

Coefficients h_β characterize the PC (amplitude) efficiency for the various sublevel populations gratings (all the polarizations are parallel). On the other hand, coefficients f_β characterize the efficiency for Zeeman coherence gratings (probe polarization orthogonal to one of the pumps).

From Eqs. (39)—(41), one gets the symmetry relations

$$\sum_{k=1,2}f_b(k)=\sum_{k=1,2}f_a(k)=f/2\,, \tag{82a}$$

$$\sum_{k=0,2}h_b(k)=\sum_{k=0,2}h_a(k)=h/2\,. \tag{82b}$$

If we define the polarization $\vec{e}^{\,r}$ of the PC field like in Eq. (67), one gets,[12] from Eqs. (80), that α_r satisfies

$$\tan\alpha_r=C\tan\alpha_0\,. \tag{83}$$

In most cases of interest, C is real, i.e., $\vec{e}^{\,r}$ is a linear polarization. Like in the previous section, Eq. (83) can be predicted by simple arguments of angular momentum conservation, but the evaluation of Eqs. (80) is required to determine C.

(i) *For stationary atoms,* one finds immediately

$$C=-F_s/H_s\ , \tag{84}$$

where

$$F_s=\sum_k\left[\frac{f_b(k)}{\Gamma_b(k)}+\frac{f_a(k)}{\Gamma_a(k)}\left[1-\mathscr{A}^k_{ba}\frac{\gamma_{ba}}{\Gamma_b(k)}\right]\right] \tag{85}$$

and a similar definition for H_s.

(ii) *For Doppler-broadened systems,* one can consider the various regimes discussed at length in the previous sections. In particular, for small residual Doppler broadening ($\Gamma_\beta, Ku\theta \ll \Gamma_{ab}$), one easily gets (see Sec. IV D)

$$C=-F/H \tag{86}$$

with

$$F=\sum_{k=1,2}\left[f_b(k)\psi^k_b+f_a(k)\psi^k_a-\gamma_{ba}\mathscr{A}^k_{ba}f_a(k)\frac{\psi^k_b-\psi^k_a}{\Gamma_a(k)-\Gamma_b(k)}\right], \tag{87a}$$

$$H=\sum_{k=0,2}\left[h_b(k)\psi^k_b+h_a(k)\psi^k_a-\gamma_{ba}\mathscr{A}^k_{ba}h_a(k)\frac{\psi^k_b-\psi^k_a}{\Gamma_a(k)-\Gamma_b(k)}\right] \tag{87b}$$

[ψ^k_β is defined by Eq. (74)]. In the quasicollinear configuration [$\theta \ll \Gamma_\beta(k)/Ku$], these equations reduce to

$$F=\frac{Ku\theta}{\sqrt{\pi}}F_s,\quad H=\frac{Ku\theta}{\sqrt{\pi}}H_s\ . \tag{88}$$

The behavior is thus similar to the one obtained for stationary atoms: indeed, since the two pumps have the same polarization, the PC polarization does not depend on the relative importance of the $(\vec{K},\vec{K}_0)$ and $(-\vec{K},\vec{K}_0)$ gratings. In a quasicollinear configuration, the $(\vec{K},\vec{K}_0)$ grating is not affected by the atomic motion, and its relaxation is identical to the case when the atoms are stationary.[26]

In the opposite case, when the residual Doppler broadening overcomes the atomic decay rate [$Ku\theta \gg \Gamma_\beta(k)$], $\psi \to 1$ and thus F and H are no longer dependent on the details of the atomic relaxation: one has $F=f$ and $H=h$ [see Eqs. (82) and (87)]. According to Eqs. (81)–(83), one finds

$$C=\frac{(J-1)(J+2)}{(3J^2+3J-1)}\quad \text{for a } J\to J \text{ transition} \tag{89a}$$

$$C=\frac{-J(2J+4)}{(4J^2+8J+5)}\quad \text{for a } J\to J+1 \text{ transition}\ . \tag{89b}$$

(iii) This last result can be generalized to any value of θ in a *simple* relaxation model: one relaxation time per level, and no spontaneous emission [Eq. (77)].[25] Thus, $F=f(\psi_a+\psi_b)/2$ and $H=h(\psi_a+\psi_b)/2$, and C is given by Eqs. (89) whatever the angular separation may be. [This can be demonstrated directly from Eq. (80) by using relations (82).]

As was discussed in our previous papers,[12,13] the relation $\tan\alpha_r = C\tan\alpha_0$ implies that the PC mirror with parallel pump polarizations is a "nonreciprocal" mirror, in the sense that the polarization after two successive PC reflections differs from the incident polarization (except for $|C|=1$). When Eqs. (89) are valid, one has $|C|<1$ (except for a $J=\frac{1}{2}\to J=\frac{1}{2}$ transition; in this case $C=-1$, and a linear polarization is reflected symmetrically to $\vec{u}_z$): this means that the PC mirror has a *linear dichroism,* and that the best efficiency is obtained for incident polarizations parallel to the one of the pump beams. After one PC reflection, the polarization tends to come closer to the *preferential* direction $\vec{u}_z$ ($|\alpha_r| < |\alpha_0|$ for $|\alpha_0| < \pi/2$). Due to the dichroism of such a type of mirror, it is obvious that if the probe beam is circularly polarized, the PC reflection will be elliptically polarized.

C. Arbitrary linear polarizations of the pumps

When the two pumps are arbitrarily linearly polarized, the behavior of the PC reflection can be determined as a linear combination of the two principal configurations discussed in the above subsections (VI A and VI B). However, in numerous practical cases, the PC reemitted field is not modified if the polarization of the two pump beams are exchanged [even if only the $(\vec{K},\vec{K}_0)$ grating contributes]. Such a symmetry between the two pump polarizations occurs notably when the residual Doppler broadening is predominant [$Ku\theta \gg \Gamma_\beta(k),\gamma_{ba}$] or in the single relaxation model (see Sec. VI A 4).

Assuming these hypotheses, the symmetry of the problem implies that the bisectors of the pump polarization axes are neutral axes of the PC mirror. Indeed, let us define $(\vec{u}_z,\vec{u}_x)$ so that

$$\vec{e}^{\,+}=\cos\beta\,\vec{u}_z+\sin\beta\,\vec{u}_x\ , \tag{90a}$$

$$\vec{e}^{\,-}=\cos\beta\,\vec{u}_z-\sin\beta\,\vec{u}_x\ . \tag{90b}$$

It is easy to verify that for a probe polarized along $\vec{u}_z$, the reemitted field is along $\vec{u}_z$, with an amplitude proportional to $h\cos^2\beta+f\sin^2\beta$, while for a probe along $\vec{u}_x$, the reemission is along $\vec{u}_x$, and the amplitude proportional to $-(f\cos^2\beta+h\sin^2\beta)$ [f and h are given by Eq. (82)]. Some remarkable points can be mentioned.

(i) For $\beta=\pi/4$, the efficiency is the same along the two neutral axes: indeed, the two pumps have orthogonal polarization and the hypotheses imply that $D=1$ (see Sec. VI A). Moreover, the amplitude of the PC field can be directly shown to be proportional to c ($=d$), and it is consistent with the identity

$$c+d=f+h \tag{91}$$

when c and d take the value calculated in Eq. (76).

(ii) If $C\geq 0$ [i.e., for a $J\to J$ transition, $J\neq\frac{1}{2}$, or for a $J=0\to J=1$ transition, see Eq. (89)], there is a remarkable angle β_0 for which the reflectivity cancels along the neutral axis $\vec{u}_z$. This zero value is obtained for $\beta_0=\tan^{-1}(C^{-1/2})$. In this configuration, the PC field is always polarized along the second bisector.

(iii) In the case of a $J=\frac{1}{2}\to J=\frac{1}{2}$ transition, one has $f=h$ [see Eq. (89)], so that the neutral axes have the same efficiency whatever β may be. It means that the PC mirror thus acts as a birefringent half-wave plate for any configuration of the pump polarizations. This generalizes the behavior obtained for orthogonally polarized pumps ($\beta=\pi/4$). One should note also that the efficiency of this mirror is independent of the angle between the pump polarization axes.

D. Circular polarization for the pump beams

The case of co-rotating circular polarizations for the pumps (e.g., σ^+) is quite simple,[9] as selection rules imply that only the σ^+ component of the probe wave is reflected, and with the same σ^+ polarization. On the opposite, the configuration where the two pumps have counter-rotating circular polarizations (σ^+,σ^-) cannot be treated so simply, but presents much interest since it has been pointed out that such a PC mirror can be a way to achieve vectorial phase conjugation.[6,7,13] Indeed, selection rules show that a σ^+-polarized probe is reflected σ^-, and vice versa, as is expected for vectorial phase conjugation. Actually the reflectivity for a σ^+ probe wave generally differs from the reflectivity of a σ^- probe wave, leading to an elliptically polarized PC wave when the incident probe is linearly polarized. We have given a rather extensive discussion on the possibility of achieving vectorial conjugation in a previous paper.[13] The requirements are essentially identical to the conditions for which one finds $D=1$ in Eq. (71): an effective single relaxation model has to be assumed, due either to adequate hypotheses on the atomic relaxation, or to the Doppler shortening of the grating lifetime. A less restrictive condition can also be considered, in the case of a $J\rightarrow J$ transition: a single decay rate for each level $[\Gamma_\beta(k)=\gamma_\beta]$ is a sufficient condition for vectorial conjugation (see Sec. VI A 4). However, it should be noted that vectorial phase conjugation can be achieved only for *small* values of θ. With increasing θ, the probe polarization can get π-polarized components which are reflected with the same π polarization, but with an efficiency which is always different from the $\sigma^+\leftrightarrow\sigma^-$ efficiency. Thus vectorial PC cannot be achieved for arbitrary incidences.

VII. TWO-PHOTON RESONANCE

In this section we consider DFWM emission enhanced by a two-photon transition between degenerate levels.[3,17] We do not present the complete formalism, but we give some general and simple results deduced from a treatment analogous to the one given for a one-photon resonance.

Let us recall first some of the basic properties predicted for two-photon resonantly enhanced DFWM.[3] For a two-photon transition, the interpretation of the PC emission as a diffraction process on a grating induced by one of the pump beams and the probe beam is no longer valid: indeed, the physical process responsible for the PC emission consists in an interaction between the probe beam, and a two-photon coherence generated by absorption of one photon from each pump beam. This coherence is basically Doppler-free, and the PC emission line shape does not depend on the angular separation θ.[27] This point, which should be an advantage in comparison with one-photon DFWM in a Doppler-broadened medium, is balanced by the weakness of the PC emission, caused by the nonresonant nature of single-photon absorption or emission (see Ref. 28 of I). This explains why the observation of two-photon cw emission in gas media has not been yet reported.[28]

The cascade three-level a-b-c system is given in Fig. 3. We assume that there is a *single* relay level ($|b,J_b\rangle$). It is well known that the two-photon transition operator[29] is a sum of a scalar operator and quadrupolar (rank $k=2$) operator. Moreover, if $J_a\neq J_c$, there is no scalar contribution. These considerations imply that the PC field is proportional to

$$\vec{\mathscr{E}}^{\,r}\propto\frac{\vec{\mathscr{E}}^{\,0}}{\Gamma_{ac}(0)-i(2\omega-\omega_{ac})}+\frac{\vec{\mathscr{E}}^{\,2}}{\Gamma_{ac}(2)-i(2\omega-\omega_{ac})}\,, \tag{92}$$

where $\vec{\mathscr{E}}^{\,0}$ and $\vec{\mathscr{E}}^{\,2}$ depend on the incident polarizations and on the angular momenta.

If $\Gamma_{ac}(0)=\Gamma_{ac}(2)$, or if $J_a\neq J_c$ (leading to $\vec{\mathscr{E}}^{\,0}=\vec{0}$), the emission is a Doppler-free Lorentzian. If $\Gamma_{ac}(0)\neq\Gamma_{ac}(2)$, and if $J_a=J_c$, the emission line shape is more complicated as it is the sum of two Lorentzians, and the respective contribution of each one depends now on the atomic system (*angular momenta*, etc.) and on the *incident polarizations* (in this sense, it differs from resonant DFWM).

A. Pumps linearly and orthogonally polarized

A general symmetry rule of DFWM forbids the emission when the polarizations of the three incident fields are orthogonal two by two. Let us define the pump polarizations as being σ_x and σ_y: a π-polarized probe is not reflected. Selection rules imply also that a σ_x-polarized probe is reflected σ_y and vice versa (see Sec. VI A). Due to the equivalent role played by each of the two pump fields, the efficiency of σ_x and σ_y is identical, and the behavior of the PC mirror is strictly similar to the one described in Sec. VI A for resonant DFWM when $D=1$. The PC mirror acts as a half-wave plate, whose neutral axes are along the bisectors of the pumps polarizations. The efficiency of the PC mirror decreases with increasing θ, due to propagation effects only: in particular the analysis developed in Sec. VI A 5 still applies.

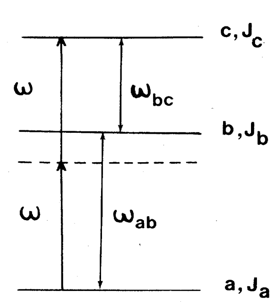

FIG. 3. Schematic of the three-level system.

B. Linear and parallel polarization of the pumps, or counter-rotating circular polarizations of the pumps

In these two cases, it is possible to define the basis $(\vec{u}_x, \vec{u}_y, \vec{u}_z)$ so that only $q=0$ components appear in the tensorial development of the two-photon coherence: $\vec{u}_z$ is chosen along the polarization of the pumps if they have an identical linear polarization (π), and $\vec{u}_z$ is chosen along the propagation axis of the pumps if they have circular counter-rotating polarizations (σ^+ and σ^-).

From the expression of ${}^{(2)}_{ac}\rho$ (${}^{(2)}_{ac}\rho$ is the two-photon coherence created at second order), given for example in Ref. 30, it can be shown that

$$\mathscr{P}^r_Q \propto \frac{\mathscr{C}^0_Q}{\Gamma_{ac}(0)-i(2\omega-\omega_{ac})}+\frac{\mathscr{C}^2_Q}{\Gamma_{ac}(2)-i(2\omega-\omega_{ac})} \quad (93)$$

with

$$\mathscr{C}^0_Q = e^{0*}_{-Q}\begin{bmatrix}1 & 1 & 0\\ q & -q & 0\end{bmatrix}\begin{bmatrix}1 & 1 & 0\\ Q & -Q & 0\end{bmatrix}\begin{Bmatrix}1 & 1 & 0\\ J_a & J_c & J_b\end{Bmatrix}^2, \quad (94a)$$

$$\mathscr{C}^2_Q = 5e^{0*}_{-Q}\begin{bmatrix}1 & 1 & 2\\ q & -q & 0\end{bmatrix}\begin{bmatrix}1 & 1 & 2\\ Q & -Q & 0\end{bmatrix}\begin{Bmatrix}1 & 1 & 2\\ J_a & J_c & J_b\end{Bmatrix}^2, \quad (94b)$$

and q is defined so that $q=0$ if the pumps are π polarized and $q=1$ if the pumps are respectively (σ^+,σ^-) polarized.

(a) *If* $J_a \neq J_c$ [or eventually, if $\Gamma_{ac}(2) \ll \Gamma_{ac}(0)$ for $J_a=J_c$] the PC mirror is dichroic in such a way that a probe polarized along $\cos\alpha_0\,\vec{u}_z+\sin\alpha_0\,\vec{u}_x$ is reflected along $\cos\alpha_0\,\vec{u}_z-\frac{1}{2}\sin\alpha_0\,\vec{u}_x$ [$C=-\frac{1}{2}$ in Eq. (83)]. Besides, the *amplitude* of the emission for a π-polarized probe is two times larger if the pumps are themselves π polarized than if they are (σ^+,σ^-) polarized. It results that for grazing incidence ($\theta \approx 0$), the *intensity* of the PC emission is 16 times larger if all the three incident beams have the same linear polarization than if the pumps are σ^+ and σ^- (the probe being necessarily σ polarized since $\theta \simeq 0$).[31] Due to the dichroism of the PC mirror, and although $\sigma^\pm$ is reflected $\sigma^\mp$, there is no way to achieve *vectorial* phase conjugation, except

(i) for $\theta \approx 0$, if the pumps are (σ^+,σ^-) polarized (as probe σ^+ and σ^- present the same reflectivity) or

(ii) for $\theta \simeq \pi/2$, and $\vec{K}_0 \| \vec{u}_z$, if the pumps have the same linear polarization π (as the probe is then necessarily σ polarized).

(b) *If* $J_a=J_c$, the PC mirror still has a dichroism between π and σ probe polarization, and this dichroism depends on the ratio of $[\Gamma_{ac}(0)-i(2\omega-\omega_{ac})]^{-1}/[\Gamma_{ac}(2)-i(2\omega-\omega_{ac})]^{-1}$. It must be noticed that the dichroism depends now on the frequency detuning $2\omega-\omega_{ac}$. At line center ($2\omega=\omega_{ac}$), the dichroism is expressed by the value of C [see Eq. (83)].

(i) For π-polarized pumps, at any angle,

$$C=\frac{[\Gamma_{ac}(0)]^{-1}\begin{Bmatrix}1 & 1 & 0\\ J_a & J_a & J_b\end{Bmatrix}^2-[\Gamma_{ac}(2)]^{-1}\begin{Bmatrix}1 & 1 & 2\\ J_a & J_a & J_b\end{Bmatrix}^2}{[\Gamma_{ac}(0)]^{-1}\begin{Bmatrix}1 & 1 & 0\\ J_a & J_a & J_b\end{Bmatrix}^2+2[\Gamma_{ac}(2)]^{-1}\begin{Bmatrix}1 & 1 & 2\\ J_a & J_a & J_b\end{Bmatrix}^2}. \quad (95)$$

(ii) For (σ^+,σ^-) pumps, and orthogonal incidence $\theta=\pi/2$, one has

$$C=\frac{[\Gamma_{ac}(0)]^{-1}\begin{Bmatrix}1 & 1 & 0\\ J_a & J_a & J_b\end{Bmatrix}^2-\frac{1}{2}[\Gamma_{ac}(2)]^{-1}\begin{Bmatrix}1 & 1 & 2\\ J_a & J_a & J_b\end{Bmatrix}^2}{[\Gamma_{ac}(0)]^{-1}\begin{Bmatrix}1 & 1 & 0\\ J_a & J_a & J_b\end{Bmatrix}^2+[\Gamma_{ac}(2)]^{-1}\begin{Bmatrix}1 & 1 & 2\\ J_a & J_a & J_b\end{Bmatrix}^2}. \quad (96)$$

The measurement of the dichroism can yield a determination of the relative importance of $\Gamma_{ac}(0)$ and $\Gamma_{ac}(2)$. It gives a way of measuring the importance of quadrupolar relaxation.[32] For $\Gamma_{ac}(0)=\Gamma_{ac}(2)$, the dichroism does not depend on the frequency detuning.

(c) *If the scalar contribution is predominant* [$J_a=J_c$, with $J_a=0$ or $\frac{1}{2}$, or with $\Gamma_{ac}(0) \ll \Gamma_{ac}(2)$], there is no longer any dichroism, and any linear polarization is reflected identically to itself. *A perfect vectorial phase conjugator is then achieved* independently of the pump polarization, either with (σ^+,σ^-) pumps or with π-polarized pumps. Moreover, the efficiency of these two kinds of PC mirror [π pumps or (σ^+,σ^-) pumps] is the same. It should be noted that all these predictions (assuming a predominant scalar contribution) are in *agreement with the scalar theory* (see the Introduction) which describes the PC field as proportional to $(\vec{E}^+\cdot\vec{E}^-)\vec{E}^{0*}$. It is also worth noting that the scalar theory is almost never valid for one-photon transition in experimental conditions, whatever the hypotheses on the relaxation processes may be (population predominant), because the motional shortening of the grating lifetime tends to equalize the contributions of each type of grating (scalar and nonscalar grating).

VIII. CONCLUSION

In this paper we have derived the general polarization properties of resonant DFWM, and we have seen how the details of the relaxation processes must be taken into account. One of the basic results of our analysis is to demonstrate that the efficiency of the various induced gratings is determined by a combination of the intrinsic atomic lifetimes $[\Gamma_\beta(k)]^{-1}$ and of the mean motional lifetime $(Ku\theta)^{-1}$. As soon as the motion-induced lifetime shortening becomes predominant [$Ku\theta \gg \Gamma_\beta(k)$], which is the case in most of the experiments, the polarization of the reemitted field obeys very simple and general laws, independent of the relaxation constants. Another noticeable result is that, in general, the scalar theory leads to strongly incorrect predictions. In particular, this is always the case when the residual Doppler effect is predominant, so that the physical processes can be described by a single relaxation time model.

An important conclusion of our work is that the most striking collision effects (such as violation of a selection rule[9]) are generally not observable in the experiments, due to their drastic reduction by the Doppler lifetime shortening (see Appendix B). However, it must be outlined that in our theory we have not taken into account elastic velocity-changing collisions. Such collisional processes are of great importance at high buffer-gas pressures, as they tend to reduce the atomic mean free path. Thus the thermal washout of the grating becomes less important, and the overall efficiency larger: this point is analogous to the Dicke narrowing effect, and was recently demonstrated by Bloembergen and co-workers.[33]

In all this work we have assumed weak incident powers in order to deal with a lowest-order perturbation theory, so that the frequency line shapes are always rather simple. It is well known that for Doppler-broadened nondegenerate two- or three-level atoms, saturation effects are responsible for complex spectral line shapes.[5] For degenerate atomic systems, the saturation processes generate still harder difficulties. In particular, experimental observations have shown that for a given set of incident polarizations, the reemitted polarization itself depends on the frequency detuning.[13] However, if the incident polarizations are only principal polarizations, and in the case of a $J=0\rightarrow J=1$ or $J=1\rightarrow J=1$ transition, theoretical solutions[34] can be obtained through the well-known theory of saturated absorption for nondegenerate two- and three-level systems. In particular, we have solved recently the case of an intense backward pump beam as well as the case of intense forward pump and probe beams.

APPENDIX A: DEMONSTRATION OF EQ. (39)

Equation (39) is a consequence of the identity

$$\sum_{k,Q} ({}^{q''}_{\beta\alpha}G^{1k}_{Q'Q})({}^{q}_{\beta\alpha}G^{1k}_{q'Q}) = \sum_{k,Q} ({}^{-q'}_{\alpha\beta}G^{1k}_{Q'Q})({}^{q}_{\alpha\beta}G^{1k}_{-q''Q}) \,. \tag{A1}$$

To demonstrate (A1), one starts from the following orthogonality relation:[35]

$$\sum_k (-1)^k(2k+1) \begin{Bmatrix} 1 & 1 & k \\ J_\alpha & J_\alpha & J_\beta \end{Bmatrix}^2 \begin{Bmatrix} 1 & 1 & k \\ 1 & 1 & k' \end{Bmatrix} = (-1)^{k'} \begin{Bmatrix} 1 & 1 & k' \\ J_\beta & J_\beta & J_\alpha \end{Bmatrix}^2 , \tag{A2}$$

and from the $3j$-$6j$ symbols relation[35]

$$\sum_{g,m_g} (2g+1) \begin{Bmatrix} j_1 & J_1 & g \\ j_2 & J_2 & f \end{Bmatrix} \begin{pmatrix} j_1 & J_1 & g \\ m_1 & M_1 & -m_g \end{pmatrix} \begin{pmatrix} j_2 & J_2 & g \\ m_2 & M_2 & m_g \end{pmatrix} = (-1)^{2f+J_1-M_1+j_2+m_2} \sum_{m_f} \begin{pmatrix} j_1 & j_2 & f \\ m_1 & m_2 & -m_f \end{pmatrix} \begin{pmatrix} J_1 & J_2 & f \\ M_1 & M_2 & m_f \end{pmatrix} \tag{A3}$$

which implies

$$(-1)^{q''+q'}(-1)^k \sum_Q \begin{pmatrix} 1 & 1 & k \\ Q' & -q' & -Q \end{pmatrix} \begin{pmatrix} 1 & 1 & k \\ -q'' & q & -Q \end{pmatrix} = \sum_{g,m} (2g+1) \begin{Bmatrix} 1 & 1 & g \\ 1 & 1 & k \end{Bmatrix} \begin{pmatrix} 1 & 1 & g \\ Q' & q'' & -m \end{pmatrix} \begin{pmatrix} 1 & 1 & g \\ -q' & -q & m \end{pmatrix} . \tag{A4}$$

Once (A4) is multiplied by $(-1)^k(2k+1)\{{}^{1}_{J_\alpha}\;{}^{1}_{J_\alpha}\;{}^{k}_{J_\beta}\}^2$, and after summation over k, the relation (A2) gives

$$\sum_{k,Q} ({}^{-q'}_{\alpha\beta}G^{1k}_{Q'Q})({}^{q}_{\alpha\beta}G^{1k}_{-q''Q}) = (-1)^{Q'+q} \sum_{g,m} 3(2g+1) \begin{pmatrix} 1 & 1 & g \\ Q' & q'' & -m \end{pmatrix} \begin{pmatrix} 1 & 1 & g \\ q' & q & -m \end{pmatrix} \begin{Bmatrix} 1 & 1 & g \\ J_\beta & J_\beta & J_\alpha \end{Bmatrix}^2 \tag{A5}$$

demonstrating the identity (A1).

APPENDIX B: EFFECT OF THE ANGULAR SEPARATION ON THE COLLISIONAL BREAKING OF SELECTION RULES

In the case of a single relaxation time model with parallel pump polarization, one knows that $C=0$ for a $J_a=0\rightarrow J_b=1$ or $J_a=1\rightarrow J_b=1$ transition [Eq. (89)]. A direct demonstration[9] of such a selection rule can be given as long as collisional processes are not considered [i.e., when one assumes $\Gamma_\beta(2)=\Gamma_\beta(1)$] (see Ref. 9). Here we discuss the value of C [Eq. (83)] for different values of the angular separation in order to see how the collisional effects become negligible.

For instance, let us consider the example of a $J_a=0\rightarrow J_b=1$ transition. As long as $Ku\theta \ll \Gamma_{ab}$, one has [see (ii) of Sec. VI B]

$$C = -\frac{F}{H} = \frac{\frac{1}{2}(\Psi_b^2-\Psi_b^1)}{\Psi_a^0+\frac{1}{3}\Psi_b^0+\frac{2}{3}\Psi_b^2-\gamma_{ba}\dfrac{\Psi_b^0-\Psi_a^0}{\Gamma_a(0)-\Gamma_b(0)}} . \tag{B1}$$

(i) *If* $Ku\theta \ll \Gamma_\beta(k)$, hence $\Psi_\beta^k \simeq Ku\theta/\Gamma_\beta(k)\sqrt{\pi}$, so that

$$C = \frac{\dfrac{1}{2}\left[\dfrac{1}{\Gamma_b(2)}-\dfrac{1}{\Gamma_b(1)}\right]}{\dfrac{1}{\Gamma_a(0)}+\dfrac{1}{3\Gamma_b(0)}+\dfrac{2}{3\Gamma_b(2)}-\dfrac{\gamma_{ba}}{\Gamma_a(0)\Gamma_b(0)}} . \tag{B2}$$

One notes that the selection rule is broken when $\Gamma_b(2)\neq\Gamma_b(1)$.

(ii) *When* $Ku\theta \gg \Gamma_\beta(k)$, $\Psi_\beta^k \simeq 1-2\Gamma_\beta(k)/\sqrt{\pi}Ku\theta$, so that

$$C=\frac{\Gamma_b(1)-\Gamma_b(2)}{2\sqrt{\pi}Ku\theta} . \tag{B3}$$

As we consider $Ku\theta \gg \Gamma_\beta(k)$, one sees that, in first approximation, now $C \simeq 0$, so that the selection rule is no longer severely violated. Equation (B3) means also that the reflectivity of a σ-polarized probe decreases as rapidly as $(Ku\theta)^{-4}$—instead of $(Ku\theta)^{-2}$ for a π polarized probe—because of the variation of the dichroism with the angular separation. One should also notice that the dichroism is now independent of cascade effects (γ_{ba}).

(iii) *For $\theta=\pi/2$,* we have mentioned in a previous paper[9] that the dichroism is given by

$$C=\frac{\Gamma_b(1)-\Gamma_b(2)}{4\sqrt{\pi}Ku} \tag{B4}$$

so that even in the presence of depolarizing collisions, there is no hope to observe any violation of the slection rule. To demonstrate Eq. (B4), one must use the identity

$$\int\int \frac{(\pi u^2)^{-1}\exp[-(v_x^2+v_z^2)/u^2]}{(\Gamma_{ab}+iKv_x)^2(\Gamma_{ab}-iKv_z)[\Gamma_\beta(k)+iK(v_x-v_z)]}dv_x dv_z=\frac{2}{(Ku)^4} \quad [\text{for } \Gamma_\beta(k),\ \Gamma_{ab} \ll Ku] . \tag{B5}$$

The case of a $J_a=1 \rightarrow J_b=1$ transition is analogous to the previous one, but involves collisional depolarization in both levels. For instance, in the case $\Gamma_\beta(k) \ll Ku\theta \ll \Gamma_{ab}$, one gets

$$C=\frac{[\Gamma_b(1)-\Gamma_b(2)]+[\Gamma_a(1)-\Gamma_a(2)]}{4\sqrt{\pi}Ku\theta} . \tag{B6}$$

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