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Nonlinear Effects in Optical Pumping of Atoms by a High-Intensity Multimode Gas Laser. General Theory

Martial Ducloy

Laboratoire de Spectroscopie Hertzienne de l'Ecole Normale Supérieure, Associé au Centre National de la Recherche Scientifique, 75231 Paris Cedex 05, France

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The optical pumping of atoms by a multimode gas laser, in the presence of a magnetic field, is studied theoretically. The atoms are described by their density matrix, which is expanded on an irreducible tensorial set. The atomic relaxation is assumed to be isotropic. In order to avoid the usual perturbation theory, we use the so-called "Broad-Line approximation" (BLA). BLA is valid when the width of the "Bennett hole" created by one laser mode in the velocity distribution of the excited atoms is comparable to the spacing between modes; under these conditions, the atomic response does not depend on the velocity and the multimode laser irradiation is equivalent to a broad-line excitation. For the atomic system, BLA leads to a set of coupled equations which is valid at arbitrary intensities of the laser field and which depends on this field through a unique parameter, γ. This parameter can be understood as a laser-induced transition probability. In this article, we analyze the nonlinear effects which can be deduced from this theory, for *any* value of the angular momenta involved in the laser transition: Hanle-effect broadening, alignment coupling, saturation resonances on the populations are shown. The exact calculations, valid at arbitrary laser intensities for $J=1 \leftrightarrow J=0$, $J=1 \leftrightarrow J=1$, and $J=1 \leftrightarrow J=2$ transitions, and the corresponding experimental results will be presented in forthcoming papers.

I. INTRODUCTION

The development of gas lasers has opened a new field of experiments in atomic physics. In particular, the study of the fluorescent light from atomic levels optically pumped with a laser beam and submitted to a magnetic field has allowed the measurement of relaxation times, collision cross sections,[1,2] Landé g factors,[3-5] etc. Recently, these experiments have been extended to some molecular levels.[6,7]

The theoretical interpretation of these experiments has been developed by Dumont and Decomps.[8] The laser field was described classically and its effects on the atoms were calculated by a perturbation development up to the second order. Tsukakoshi and Shimoda[2] extended a similar treatment up to the fourth order, taking into account the first nonlinear effects. Recently, Dumont[4,5] has developed a theoretical study up to the fourth order, taking into account the atomic relaxation processes by means of a more sophisticated model: disorienting collisions, velocity diffusion by collisions and by trapping of fluorescence lines, etc. This theory gives a good explanation for the behavior of certain nonlinear effects observed on the levels populations, particularly the effect of mode locking.[4] However, this theory is not well adjusted to the study of the "Hanle-effect" shape (magnetic depolarization of the levels), and it fails at high laser intensities: One must take into account the higher orders and, furthermore, it is not sure that the perturbation development is always convergent. In recent publications,[9] several authors have studied the monomode laser operation by a method which is not based upon a perturbation development. They have shown that, at high intensities, the exact solution is different from the one obtained by perturbation theory.

The aim of this paper is to study the optical pumping of atomic levels by a multimode gas laser through a nonperturbative method. The equations of motion of the atomic system, which are set-up in Sec. II, are simplified by means of some approximations. These approximations and their physical meaning are analyzed in Sec. III. Applications to a linearly polarized laser (Sec. IV) and to a general polarization (Sec. V) are considered. The solution for *any* value of the angular momenta of the levels is studied using a perturbation expansion. In Sec. VI, we analyze the experimental conditions which ensure the validity of the approximations, and subsequently we discuss the limits of the results deduced from this theory. In forthcoming papers, exact calculations (valid at all orders in the laser field) and experimental results for small J values ($J=0$, 1, 2) will be presented.

II. EQUATIONS OF MOTION

We consider a gas of atoms having two levels, b and a, of energies W_b and W_a (Fig. 1). Every level has a Zeeman structure completely determined by its angular momentum J_β and its Landé factor g_β ($\beta = a$ or b). The atoms are excited by a discharge which populates all the levels; they are irradiated by a laser beam resonant for the b-a transition and submitted to a magnetic field. The system is studied by means of the intensity and the polarization of the fluorescent light emitted from the b and a levels. The fluorescence from level β can be expressed as a function of the different components of the density matrix of β, ${}_\beta\rho$. This function depends on the polarization of the fluorescence line (see Refs. 8 and 10). As our detection setup does not resolve the spectral shape of the lines, we have only to determine the velocity-averaged density matrix,

$$ {}_\beta\bar{\rho}(r,t) = \int_\beta \rho(v,r,t)\,dv\,. \tag{I.1}$$

A. Basic Assumptions

Most of the starting hypotheses are the same as those of Dumont.[4,5] They will be shortly recalled with the same notations as those of Ref. 4. The atomic system will be described by its density matrix ρ, which consists of four submatrices:

$$\rho = \begin{pmatrix} {}_b\rho & {}_{ba}\rho \\ {}_{ab}\rho & {}_a\rho \end{pmatrix}. \tag{I.2}$$

${}_{ab}\rho$ and ${}_{ba}\rho$ represent the "optical coherences" between a and b. They evolve at optical frequencies. ${}_\beta\rho$ represents the sublevels' populations and the Zeeman coherence for the β level. As Dumont,[4] we, too, describe the density-matrix evolution by a Bolzmann-type equation:

$$\dot{\rho} = \left(\frac{\partial}{\partial t} + v\frac{\partial}{\partial r}\right)\rho(v,r,t) = -i[\mathcal{H}(v,r,t),\rho(v,r,t)] + \Lambda(v) + \left(\frac{d}{dt}\rho(v,r,t)\right)_{\text{relax}} + \left(\frac{d}{dt}\rho(v,r,t)\right)_{tr}. \tag{I.3}$$

v and r are, respectively, the projections of the velocity and of the position of the atom on the laser axis,[11] $\mathcal{H}$ is the atomic Hamiltonian, and Λ represents the excitation of the atoms by the discharge. $[(d/dt)\rho]_{tr}$ represents the coupling by spontaneous emission from b to a[12] and $[(d/dt)\rho]_{\text{relax}}$ contains all the relaxation processes.

1. *Density Matrix* ρ

Owing to the symmetry of the various interactions, it is more convenient to develop the density matrix on a set of irreducible tensors,[13,14]

$$ {}_{\alpha\beta}\rho(v,r,t) = \sum_{k,q} {}_{\alpha\beta}\rho_q^k(v,r,t)\,{}_{\alpha\beta}T_q^{(k)}, \tag{I.4}$$

with

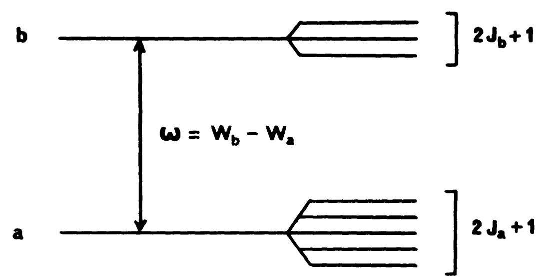

FIG. 1. Diagram of the laser transition.

$$ {}_{\alpha\beta}\rho_q^k = \mathrm{Tr}(\rho\,{}_{\alpha\beta}T_q^{(k)+}) \tag{I.5}$$

2. *Excitation Matrix* $\Lambda(v)$

The discharge is assumed to be isotropic and homogeneous; it introduces only a global population in the atomic levels. The velocity distribution is assumed to be Maxwellian, independent of the internal state:

$$\begin{aligned} \Lambda(v) &= W_M(v)(\lambda_a\,{}_aT_0^{(0)} + \lambda_b\,{}_bT_0^{(0)}), \\ W_M(v) &= (u\sqrt{\pi})^{-1}e^{-v^2/u^2}. \end{aligned} \tag{I.6}$$

u is the mean quadratic velocity.

3. *Hamiltonian* $\mathcal{H}$

$\mathcal{H}$ consists of three parts:

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_z + R(r,t). \tag{I.7}$$

$\mathcal{H}_0$ is the free-atom Hamiltonian:

$$\mathcal{H}_0 = \sum_{\beta=a,b} W_\beta(2J_\beta+1)^{1/2}\,{}_\beta T_0^{(0)}. \tag{I.8}$$

$\mathcal{H}_z$ is the Zeeman Hamiltonian:

$$\mathcal{H}_z = \sum_{\beta=a,b}\omega_\beta\left(\frac{J_\beta(J_\beta+1)(2J_\beta+1)}{3}\right)^{1/2}{}_\beta T_0^{(1)}. \tag{I.9}$$

The quantization axis is assumed to be parallel to the magnetic field H. $\omega_\beta = -(g_\beta\mu_B H)$ is the β Zeeman splitting (μ_B = Bohr magneton). $R(r,t)$ is the Hamiltonian of interaction with the laser beam:

$$R(r,t) = -\vec{\mathrm{P}}\cdot\vec{\mathrm{E}}(r,t) = -\sum_q(-)^q P_q E_{-q}. \tag{I.10}$$

$\vec{\mathrm{P}}$ is the electric dipole operator and $\vec{\mathrm{E}}$ is the laser electric field. P_q and E_q are the standard components of $\vec{\mathrm{P}}$ and $\vec{\mathrm{E}}$:

$$P_q = (P_{ab}/\sqrt{3})[{}_{ab}T_q^{(1)} + (-)^{J_a-J_b}\,{}_{ba}T_q^{(1)}]. \tag{I.11}$$

(The reduced matrix element P_{ab} is made real by means of a convenient choice of the relative phase of states a and b.) The laser consists of several modes,

$$\vec{E}(r,t)=\sum_{\mu}\left[\vec{\mathcal{E}}^{\mu}e^{-i(\omega_\mu t-k_\mu r)}+\vec{\mathcal{E}}^{\mu *}e^{i(\omega_\mu t-k_\mu r)}\right]. \quad (I.12)$$

We assume the modes to have the same polarization, $\vec{e}$[15]:

$$\vec{\mathcal{E}}^{\mu}=\mathcal{E}^{\mu}\vec{e}=|\mathcal{E}^{\mu}|e^{i\phi_\mu}\vec{e}, \quad (I.13)$$

$$\vec{E}=\mathcal{E}\vec{e}+\mathcal{E}^{*}\vec{e}^{*}, \quad (I.14)$$

with

$$\mathcal{E}=\sum_{\mu=1}^{N}\mathcal{E}^{\mu}e^{-i(\omega_\mu t-k_\mu r)}. \quad (I.15)$$

$|\mathcal{E}^{\mu}|$ and ϕ_μ are the amplitude and phase of the μ mode. N is the number of modes.

Lastly, we assume the laser beam to be a traveling wave: k_μ is always positive. The standing-wave pumping, which is considered in (Ref. 26), does not introduce any fundamental change of the theoretical results, in comparison with the running-wave pumping.

4. *Relaxation Processes*

They have been analyzed by Dumont in a detailed manner.[4] Following him, we assume the relaxation to be isotropic[13] and we use the "strong-collision" model[16] for velocity changes due to collisions or trapping of fluorescence lines (no speed memory).

$$\left(\frac{d}{dt}\,{}_{\beta}\rho_q^k(v,r,t)\right)_{\rm relax}=-\Gamma'_{\beta}(k)\,{}_{\beta}\rho_q^k(v,r,t)+\gamma'_{\beta}(k)W_M(v)\,{}_{\beta}\bar{\rho}_q^k(r,t), \quad (I.16)$$

where ${}_{\beta}\bar{\rho}$ is defined by (I.1) and $W_M(v)$ by (I.6). $\Gamma'_{\beta}(k)$ is the k-tensor relaxation rate due either to the destruction of the tensorial quantity or to the velocity change of the atom. $\gamma'_{\beta}(k)$ is the restoration rate from any other velocity. The relaxation rates of the velocity-averaged density matrix, ${}_{\beta}\bar{\rho}$, are given by

$$\Gamma_{\beta}(k)=\Gamma'_{\beta}(k)-\gamma'_{\beta}(k). \quad (I.17)$$

In Eq. (I.16) any coupling between different positions is ignored. This approximation is valid if the trapping of the fluorescence lines is either inexistent or complete, i.e., if the mean free path of a fluorescence photon is either large or small compared to the laser beam diameter. If the mean free path is of the order of the beam diameter, the coupling between distant positions cannot be ignored.[17] This case will be considerεd in a forthcoming paper.

For the optical coherence, we shall neglect the velocity changes:

$$\left(\frac{d}{dt}\,{}_{ab}\rho_q^k(v,r,t)\right)_{\rm relax}=-\Gamma_{ab}(k)\,{}_{ab}\rho_q^k(v,r,t). \quad (I.18)$$

Indeed, on the one hand, the velocity changes due to fluorescence trapping do not restore optical coherence; on the other hand, Berman and Lamb[18] have shown that there is no velocity-changing collision for the optical coherence if the scattering potentials are different in the a and b levels (a highly likely situation): in this case, it is not possible to define a classical trajectory for ${}_{ab}\rho$, because the velocity changes are different in the a and b states.

$\Gamma_{ab}(k)$ possibly has an imaginary part which corresponds to a frequency shift of the optical transition. In general, this shift is much smaller than the collision broadening[19,20] and we shall neglect it. These last two assumptions are not very important for the velocity-averaged density matrix in levels a and b: the existence of ${}_{ab}\rho$ velocity changes or of a collision-induced frequency shift would have little weight on the final results.

5. *Transfer by Spontaneous Emission*

The transfer by spontaneous emission from b to a has the spherical symmetry. It is expressed by means of a $6J$ coefficient[12]:

$$\left(\frac{d}{dt}\,{}_{a}\rho_q^k\right)_{tr}=\Theta(b,a,k)\,{}_{b}\rho_Q^k=(-)^{J_a+J_b+k+1}\gamma_{ba}(2J_b+1)\begin{Bmatrix}k & J_b & J_b\\ 1 & J_a & J_a\end{Bmatrix}\,{}_{b}\rho_q^k. \quad (I.19)$$

γ_{ba} is the $b\to a$ transition probability.

B. Density-Matrix Equations

With the help of the previous assumptions, we can write (I.3) as a set of equations coupling the ρ tensorial components. We use the well-known rotating-wave approximation: in ${}_{a}\rho$ and ${}_{b}\rho$ the optical frequencies' components are neglected, and in ${}_{ab}\rho$ only the optical frequencies' components are kept.

After some algegra, the following equations are obtained:

$${}_{a}\dot{\rho}_Q^k=-[iQ\omega_a+\Gamma'_a(k)]\,{}_{a}\rho_Q^k+\gamma'_a(k)W_M(v)\,{}_{a}\bar{\rho}_Q^k+\Theta(b,a,k)\,{}_{b}\rho_Q^k+iP_{ab}\sum_{k'Q'q}(-)^{k+k'}\,{}^{q}_{ab}G_{Q'Q}^{k'k}\left[e_{-q}\mathcal{E}\,{}_{ab}\rho_{Q'}^{k'}+(-)^{k+k'+Q}e_q^{*}\mathcal{E}^{*}\,{}_{ab}\rho_{-Q'}^{k'*}\right], \quad (I.20a)$$

$${}_{b}\dot{\rho}_Q^k=-[iQ\omega_b+\Gamma'_b(k)]\,{}_{b}\rho_Q^k+\gamma'_b(k)W_M(v)\,{}_{b}\bar{\rho}_Q^k+iP_{ab}\sum_{k'Q'q}{}^{q}_{ba}G_{Q'Q}^{k'k}\left[e_{-q}\mathcal{E}\,{}_{ab}\rho_{Q'}^{k'}+(-)^{k+k'+Q}e_q^{*}\mathcal{E}^{*}\,{}_{ab}\rho_{-Q'}^{k'*}\right], \quad (I.20b)$$

$$ {}_{ab}\dot{\rho}^{k'}_{Q'} = -\left[i\left(\frac{\omega_a+\omega_b}{2}Q'-\omega\right)+\Gamma_{ab}(k')\right]{}_{ab}\rho^{k'}_{Q'} - i\frac{\omega_a-\omega_b}{2}\left(Q'\frac{J_a(J_a+1)-J_b(J_b+1)}{k'(k'+1)}{}_{ab}\rho^{k'}_{Q'}+a(k')\,{}_{ab}\rho^{k'-1}_{Q'}+a(k'+1)\,{}_{ab}\rho^{k'+1}_{Q'}\right) $$

$$ +iP_{ab}\mathcal{E}^*\sum_{k''Q''q'} e^*_{-q'}\left[{}^{q'}_{ba}G^{k'k''}_{Q'Q''}\,{}_b\rho^{k''}_{Q''}+(-)^{k'+k''}\,{}^{q'}_{ab}G^{k'k''}_{Q'Q''}\,{}_a\rho^{k''}_{Q''}\right]+iP_{ab}\mathcal{E}^*e^*_{Q'}\delta_{k'1}\frac{nW_M(v)}{\sqrt{3}}, \tag{I.20c} $$

where $\omega = W_b - W_a$ is the atomic frequency and where two geometrical factors are defined by means of the $3J$ and $6J$ coefficients:

$$ {}^{q}_{\beta\alpha}G^{k'k}_{Q'Q} = (-)^{J_\alpha+J_\beta+Q'}[(2k+1)(2k'+1)]^{1/2}\begin{pmatrix}k' & 1 & k\\ Q' & q & -Q\end{pmatrix}\begin{Bmatrix}k' & 1 & k\\ J_\beta & J_\beta & J_\alpha\end{Bmatrix}, \tag{I.21} $$

$$ a(k') = \frac{1}{k'}\left(\frac{[(J_a+J_b+1)^2-k'^2][k'^2-(J_a-J_b)^2][k'^2-Q'^2]}{4k'^2-1}\right)^{1/2}. \tag{I.22} $$

Let us point out that, in Eqs. (I.20), ρ represents the laser-induced modifications of the density matrix. To obtain the total density matrix, we must add the discharge-induced a and b populations when the laser is off, $\lambda_\alpha/\Gamma_\alpha(0)$. Indeed, the total density matrix is

$$ \rho_t = \begin{pmatrix} {}_b\rho + \dfrac{\lambda_b}{\Gamma_b(0)}\,{}_bT^{(0)}_0 W_M(v) & {}_{ba}\rho \\ {}_{ab}\rho & {}_a\rho + \dfrac{\lambda_a}{\Gamma_a(0)}\,{}_aT^{(0)}_0 W_M(v) \end{pmatrix}. \tag{I.23} $$

That is the reason why the discharge term λ_β does not appear in Eqs. (I.20a) and (I.20b). But in Eq. (I.20c) there is a source term proportional to the laser-off populations inversion:

$$ n = \frac{\lambda_b}{\Gamma_b(0)(2J_b+1)^{1/2}} - \frac{\lambda_a}{\Gamma_a(0)(2J_a+1)^{1/2}}. \tag{I.24} $$

In (I.20c), the different tensorial orders are coupled when ω_a and ω_b are different (anomalous Zeeman effect). This coupling disappears either if the Landé factors are equal or if the laser transition is a $J=1$–$J=0$ transition. We shall neglect this coupling in the following. The validity of this approximation will be considered later.

To solve Eqs. (I.20), Dumont[4] (as have other authors[2]) has used an iteration procedure: at first order in the electric field, optical coherence (represented by ${}^{(1)}_{ab}\rho^{k'}_{Q'}$) appears which is due to the n source term in Eq. (I.20c). This optical coherence is put into Eqs. (I.20a) and (I.20b) and causes tensorial quantities to appear in levels a and b at the second order. These quantities have the following form[21]:

$$ {}^{(2)}_{\beta}\rho^k_Q(v,r,t) = \sum_{\mu,\nu}{}^{(2)}_{\beta}\rho^k_Q(v,\nu,\mu)\mathcal{E}^{\nu*}\mathcal{E}^{\mu} \times\exp\{i[(\omega_\nu-\omega_\mu)t-(k_\nu-k_\mu)r]\}. \tag{I.25} $$

These solutions are modulated at all beat frequencies, $\omega_\nu-\omega_\mu$. If we neglect small frequency shifts, the modes frequencies are equidistant:

$$ \Delta\omega = c\Delta k = \pi(c/L), \tag{I.26} $$

where c is the light velocity and L is the cavity length.

We can write (I.25) following the general form:

$$ {}_\beta\rho^k_Q(v,r,t) = \sum_p {}_\beta\rho^k_Q(v,p)e^{ip(\Delta\omega t-\Delta k r)}, \tag{I.27} $$

where p is an integer. It is easy to show that the higher-order terms will appear in the same way. Equation (I.27) will be taken as a starting equation to find the stationary solution of (I.20) by a method without iteration.

C. Stationary Operation

The Fourier expansion (I.27) is put into (I.20c):

$$ {}_{ab}\dot{\rho}^{k'}_{Q'} = [i(\omega-Q'\omega_Z)-\Gamma_{ab}(k')]\,{}_{ab}\rho^{k'}_{Q'} + iP_{ab}\sum_{\mu,p'}\mathcal{E}^{\mu*}\,{}_{ab}\zeta^{k'}_{Q'}(v,p') \times\exp i[(\omega_\mu+p'\Delta\omega)t-(k_\mu+p'\Delta k)r], \tag{I.28} $$

with

$$ \omega_Z = \tfrac{1}{2}(\omega_a+\omega_b) \tag{I.29} $$

and

$$ {}_{ab}\zeta^{k'}_{Q'}(v,p') = e^*_{\mathcal{Q}'}\frac{nW_M(v)}{\sqrt{3}}\delta_{k'1}\delta_{p'0} + \sum_{k''Q''q'} e^*_{-q'}\left[{}^{q'}_{ba}G^{k'k''}_{Q'Q''}\,{}_b\rho^{k''}_{Q''}(v,p') + (-)^{k'+k''}\,{}^{q'}_{ab}G^{k'k''}_{Q'Q''}\,{}_a\rho^{k''}_{Q''}(v,p')\right]. \tag{I.30} $$

The stationary solution is immediately obtained:

$$ {}_{ab}\rho^{k'}_{Q'} = iP_{ab}\sum_{\mu,p'}\frac{\mathcal{E}^{\mu*}\,{}_{ab}\zeta^{k'}_{Q'}(v,p')\exp\{i[(\omega_\mu+p'\Delta\omega)t-(k_\mu+p'\Delta k)r]\}}{\Gamma_{ab}(k')+i[\omega_\mu+p'\Delta\omega-\omega+Q'\omega_Z-(k_\mu+p'\Delta k)v]}. \tag{I.31} $$

In multiplying (I.31) by $\mathcal{E}$ [to obtain the source term of (I.20)], the following expression appears:

$$\mathcal{E}\,{}_{ab}\rho_{Q'}^{k'} = iP_{ab}\sum_{\mu,\nu,p'}\frac{\mathcal{E}^{\mu*}\mathcal{E}^{\nu}\,{}_{ab}\zeta_{Q'}^{k'}(v,p')\exp i[(\omega_\mu-\omega_\nu+p'\Delta\omega)t-(k_\mu-k_\nu+p'\Delta k)r]}{\Gamma_{ab}(k')+i[\omega_\mu+p'\Delta\omega-\omega+Q'\omega_z-(k_\mu+p'\Delta k)v]}. \quad \text{(I.32)}$$

Or, in another way,

$$\mathcal{E}\,{}_{ab}\rho_{Q'}^{k'} = i\,(P_{ab})^{-1}\sum_{s,p'}{}^{s}_{p'}S_{Q'}^{k'}(v,\omega_z)\,{}_{ab}\zeta_{Q'}^{k'}(v,p')\times\exp[i(p'+s)(\Delta\omega t-\Delta kr)], \quad \text{(I.33)}$$

where [with the approximation $k_\mu+p'\Delta k\simeq\bar{k}$ (Ref. 22)]

$$ {}^{s}_{p'}S_{Q'}^{k'}(v,\omega_z) = \sum_\mu\frac{P_{ab}^2\,\mathcal{E}^{\mu*}\mathcal{E}^{\mu-s}}{\Gamma_{ab}(k')+i(\omega_\mu+p'\Delta\omega-\omega+Q'\omega_z-\bar{k}v)}. \quad \text{(I.34)}$$

$\mathcal{E}^{\mu-s}$ is the field amplitude corresponding to the frequency $\omega_\nu=\omega_\mu-s\Delta\omega$. The sum on μ is from $s+1$ to N if s is positive, or from 1 to $N+s$ if s is negative.

$P_{ab}^2[\Gamma_{ab}(k')+i(\omega_\mu+p'\Delta\omega-\omega+Q'\omega_z-\bar{k}v)]^{-1}$ is the atomic cross section associated to the absorption or stimulated emission of a photon. Subsequently, ${}^{s}_{p'}S_{Q'}^{k'}$ can be understood as the probability that a $p'\Delta\omega$-modulated atomic quantity so interacts with a laser photon as to become modulated at frequency $(p'+s)\Delta\omega$. ${}^{s}_{p'}S_{Q'}^{k'}$ characterizes the linear response of atoms to the laser irradiation.

Finally, we bring Eq. (I.33) and its complex conjugate in (I.20) and, with the help of the Fourier expansion (I.27), we obtain the following relations between the Fourier coefficients, for the stationary operation:

$$[\Gamma_a'(k)+i(Q\omega_a+p\Delta\omega)]\,{}_a\rho_Q^k(v,p)=\gamma_a'(k)W_M(v)\,{}_a\bar{\rho}_Q^k(p)+\Theta(b,a,k)\,{}_b\rho_Q^k(v,p) - \sum_{k'Q'qs}(-)^{k+k'}\,{}^{q}_{ab}G_{Q'Q}^{k'k}\,[e_{-q}\,{}^{s}_{(p-s)}S_{Q'}^{k'}(v,\omega_z)\,{}_{ab}\zeta_{Q'}^{k'}(v,p-s) + (-)^{k+k'+Q+1}e_q^*\,{}^{s}_{(-p-s)}S_{-Q'}^{k'}(v,\omega_z)^*\,{}_{ab}\zeta_{-Q}^{k'}(v,-p-s)^*], \quad \text{(I.35a)}$$

$$[\Gamma_b'(k)+i(Q\omega_b+p\Delta\omega)]\,{}_b\rho_Q^k(v,p)=\gamma_b'(k)W_M(v)\,{}_b\bar{\rho}_Q^k(p) - \sum_{k'Q'qs}{}^{q}_{ba}G_{Q'Q}^{k'k}\,[e_{-q}\,{}^{s}_{(p-s)}S_{Q'}^{k'}(v,\omega_z)\,{}_{ab}\zeta_{Q'}^{k'}(v,p-s) + (-)^{k+k'+Q+1}e_q^*\,{}^{s}_{(-p-s)}S_{-Q'}^{k'}(v,\omega_z)^*\,{}_{ab}\zeta_{-Q'}^{k'}(v,-p-s)^*]. \quad \text{(I.35b)}$$

The Doppler effect at frequency $p\Delta\omega$ has been neglected.[22] s is an integer defined by $\omega_\mu-\omega_\nu=s\Delta\omega$. If the laser has N modes, the sum on s is from $-N+1$ to $N-1$.

Up to now, we have not done any hypotheses on the number of modes N, the Doppler width $\Delta\nu=\bar{k}u$, the relative importance of the mode splitting $\Delta\omega$, and of the various relaxation rates, $\Gamma_{ab}(k')$ and $\Gamma_\beta(k)$. The application range of (I.35) is very large: the laser is either monomode or multimode, the transition may be visible or infrared, the pressure range is not limited, and we can consider atomic or molecular transitions.

III. BROAD-LINE APPROXIMATION

In this section, our purpose is to simplify the expression of ${}^{s}_{p'}S_{Q'}^{k}$ [see (I.34)]. We shall do some approximations that we shall call "broad-line approximation" (BLA). The validity of these approximations will be examined in a detailed manner, in the last section. We shall see that they are fairly well fulfilled for the visible or near-infrared transitions of neutral atoms, in the usual pressure range (1–5 Torr).

We assume the laser to be phase-locked (the free-running modes case will be considered in paragraph Sec. III C). All the modes have the same phase ϕ, that we shall take equal to zero: $\mathcal{E}^\mu$ is real and positive.

A. Broad-Line Approximation

1. *Approximate Expression for S by Means of an Integral*

The optical coherence relaxation rate is assumed to be greater than the mode splitting, and the laser oscillates on a great number of modes:

$$\Gamma_{ab}(k')>\Delta\omega\,, \quad \text{(I.36)}$$

$$N\gg 1\,. \quad \text{(I.37)}$$

We also assume that $\mathcal{E}^\mu$ is a slowly varying function of μ: $(\mathcal{E}^{\mu+1}-\mathcal{E}^\mu)/\mathcal{E}^\mu\ll 1$. In this case, the right-hand side of (I.34) is slowly varying with μ and S can be written as an integral:

$$
{}^{s}_{p'}S^{k'}_{Q'}(v,\omega_Z) = \int_{-N/2+s}^{N/2} \frac{P^2_{ab}\,\mathcal{E}^{\mu}\mathcal{E}^{\mu-s}dx_\mu}{\Gamma_{ab}(k')+i[\overline{\omega}-\omega+(x_\mu+p')\Delta\omega+Q'\omega_z-\overline{k}v]}, \tag{I.38}
$$

where s is assumed to be positive. $\overline{\omega}$ is the laser mean frequency, $\overline{\omega}=N^{-1}\sum_\mu \omega_\mu$, and x_μ is equal to $(\omega_\mu-\overline{\omega})/\Delta\omega$. For negative s, the integral limits are $-N/2$ and $s+N/2$.

2. *Elimination of the Dependence on the Magnetic Field and on the Modulation Order*

In general, the experimental values of the magnetic field are weak enough to verify the following condition:

$$
N\Delta\omega \gg \omega_Z . \tag{I.39a}
$$

Owing to the resonant factor of the left-hand side of (I.35), in weak field, the modulations at the $p\Delta\omega$ frequency are off resonance for large-p values and we assume

$$
N\Delta\omega \gg p'\Delta\omega,\ \ s\Delta\omega . \tag{I.39b}
$$

As $\mathcal{E}^\mu$ is slowly varying with μ, (I.38) is replaced by

$$
{}^{s}_{p'}S^{k'}_{Q'} = \int_{-N/2}^{+N/2} \frac{(P_{ab}\mathcal{E}^{\mu})^2 dx_\mu}{\Gamma_{ab}(k')+i[\overline{\omega}-\omega+x_\mu\Delta\omega-\overline{k}v]} . \tag{I.40}
$$

3. *Elimination of the Velocity Dependence*

The laser width is assumed to be much greater than the Doppler width:

$$
N\Delta\omega \gg \Delta\nu = \overline{k}u . \tag{I.41}
$$

The previous equation becomes

$$
{}^{s}_{p'}S^{k'}_{Q'} = \gamma - i\delta = \int_{-N/2}^{+N/2} \frac{(P_{ab}\mathcal{E}^{\mu})^2 dx_\mu}{\Gamma_{ab}(k')+i[\overline{\omega}-\omega+x_\mu\Delta\omega]} . \tag{I.42}
$$

If the laser is centered on the atomic frequency ($\overline{\omega}=\omega$) and if the modes nearly have the same intensity, then

$$
(\mathcal{E}^{\mu})^2 \simeq \overline{\mathcal{E}}^2 = I/N , \tag{I.43a}
$$

with

$$
I = \sum_\mu |\mathcal{E}^\mu|^2 . \tag{I.43b}
$$

We obtain

$$
\delta = 0 ,\qquad \gamma = 2\frac{P^2_{ab}I}{N\Delta\omega}\tan^{-1}\left(\frac{N\Delta\omega}{2\Gamma_{ab}(k')}\right) . \tag{I.44}
$$

If

$$
N\Delta\omega \gg \Gamma_{ab}(k') , \tag{I.45}
$$

then

$$
\gamma = \pi\frac{P^2_{ab}I}{N\Delta\omega} ; \tag{I.46}
$$

γ is independent of the relaxation rate, $\Gamma_{ab}(k')$. On the other hand, if

$$
N\Delta\omega \ll \Gamma_{ab}(k') , \tag{I.47}
$$

then

$$
\gamma = \frac{P^2_{ab}I}{\Gamma_{ab}(k')} . \tag{I.48}
$$

Condition (I.45) is much more likely than (I.47).

B. Equations of Motion in the Broad-Line Approximation

We assume ${}^{s}_{p'}S^{k'}_{Q'}(v,\omega_Z)$ to be equal to γ, which is a laser-induced transition probability which is independent of the magnetic field and the atomic velocity. Subsequently, the solution of (I.35) has, obviously, the following form:

$$
{}_{\beta}\rho^{k}_{Q}(v,p) = {}_{\beta}\overline{\rho}^{k}_{Q}(p) W_M(v) . \tag{I.49}
$$

The internal and external variables are independent. The equations of motion become[23]

$$
[\Gamma_a(k)+i(Q\omega_a+p\Delta\omega)]\,{}_{a}\overline{\rho}^{k}_{q}(p) = \Theta(b,a,k)\,{}_{b}\overline{\rho}^{k}_{Q}(p) - \gamma\sum_{k'Q'qs}(-)^{k+k'}\,{}^{q}_{ab}G^{k'k}_{Q'Q}[e_{-q}\,{}_{ab}\overline{\zeta}^{k'}_{Q'}(p-s)+(-)^{k+k'+Q+1}e^{*}_{q}\,{}_{ab}\overline{\zeta}^{k'*}_{-Q'}(-p-s)] , \tag{I.50a}
$$

$$
[\Gamma_b(k)+i(Q\omega_b+p\Delta\omega)]\,{}_{b}\overline{\rho}^{k}_{Q}(p) = -\gamma\sum_{k'Q'qs}{}^{q}_{ba}G^{k'k}_{Q'Q}[e_{-q}\,{}_{ab}\overline{\zeta}^{k'}_{Q'}(p-s)+(-)^{k+k'+Q+1}e^{*}_{q}\,{}_{ab}\overline{\zeta}^{k'*}_{-Q'}(-p-s)] , \tag{I.50b}
$$

where

$$
{}_{ab}\overline{\zeta}^{k'}_{Q'}(p') = e^{*}_{Q'}\frac{n}{\sqrt{3}}\delta_{k'1}\delta_{p'0} + \sum_{k''Q''q'} e^{*}_{-q'}[{}^{q'}_{ba}G^{k'k''}_{Q'Q''}\,{}_{b}\overline{\rho}^{k''}_{Q''}(p') + (-)^{k'+k''}\,{}^{q'}_{ab}G^{k'k''}_{Q'Q''}\,{}_{a}\overline{\rho}^{k''}_{Q''}(p')] . \tag{I.51}
$$

In these equations, the relaxation rates are the velocity-averaged ones, $\Gamma_\beta(k)$ [see (I.17)]. Introducing the following notations[24]:

$$
{}^{qq'}_{\beta}H^{kk''}_{QQ''} = \sum_{k'Q'} {}^{q}_{\beta\alpha}G^{k'k}_{Q'Q}\;{}^{-q'}_{\beta\alpha}G^{k'k''}_{Q'Q''} , \tag{I.52}
$$

$$ {}^{qq'}_{\beta\alpha}H^{kk''}_{QQ''} = \sum_{k'Q'} (-)^{k'+k''+1}\, {}^{q}_{\beta\alpha}G^{k'k}_{Q'Q}\, {}^{-q'}_{\alpha\beta}G^{k'k''}_{Q'Q''} \,. \tag{I.53} $$

We finally obtain[25]

$$ \begin{aligned} [\Gamma_a(k)+i(Q\omega_a+p\Delta\omega)]\, {}_a\bar\rho^{k}_{Q}(p) = {} & \Theta(b,a,k)\, {}_b\bar\rho^{k}_{Q}(p) + (-)^{k}\frac{2n\gamma}{\sqrt3}\sum_{q'q} e_{-q}e^{*}_{q'}\, {}^{q}_{ab}G^{1k}_{q'Q} + 2\gamma \sum_{\substack{k''Q''\\ qq's}} (-)^{k+k''} e_{-q}e^{*}_{q'}\, {}^{qq'}_{ab}H^{kk''}_{QQ''}\, {}_b\bar\rho^{k''}_{Q''}(p-s) \\ & -\gamma \sum_{\substack{k''Q''\\ qq's}} (-)^{k+k''}[e_{-q}e^{*}_{q'} + (-)^{k+k''+Q+Q''} e_{-q'}e^{*}_{q}]\, {}^{qq'}_{a}H^{kk''}_{QQ''}\, {}_a\bar\rho^{k''}_{Q''}(p-s)\,, \end{aligned} \tag{I.54a} $$

$$ \begin{aligned} [\Gamma_b(k)+i(Q\omega_b+p\Delta\omega)]\, {}_b\bar\rho^{k}_{Q}(p) = {} & -\frac{2n\gamma}{\sqrt3}\sum_{qq'} e_{-q}e^{*}_{q'}\, {}^{q}_{ba}G^{1k}_{q'Q} + 2\gamma \sum_{\substack{k''Q''\\ qq's}} e_{-q}e^{*}_{q'}\, {}^{qq'}_{ba}H^{kk''}_{QQ''}\, {}_a\bar\rho^{k''}_{Q''}(p-s) \\ & -\gamma \sum_{k''Q''qq's} [e_{-q}e^{*}_{q'} + (-)^{k+k''+Q+Q''} e_{-q'}e^{*}_{q}]\, {}^{qq'}_{b}H^{kk''}_{QQ''}\, {}_b\bar\rho^{k''}_{Q''}(p-s)\,. \end{aligned} \tag{I.54b} $$

This system of equations couples the density-matrix components inside levels a and b. The terms that are due to the evolution of the atoms in the magnetic field and to the relaxation, have been placed in the left-hand side of these equations. The right-hand side gets together the contributions of the spontaneous emission and of the interaction with the laser field. Let us analyze these last contributions.

(1) The $\bar\rho_a$ term represents the effect of the laser on the atoms in the lower level: this is the effect of a photon absorption. As it can be seen in (I.54), the angular part of the absorption process is decoupled from the absorption probability γ. This property is also obtained in the optical pumping with an ordinary light source.[29] The analogy between the two situations is studied in Sec. III D. The angular coefficients could be obtained from considerations on the conservation of the total angular momentum of the system atom plus laser photon, during the absorption process. The photon, in a given spin state, is absorbed by an atom in level a, having a given angular momentum, and brings this atom into the upper level. This transition of an atom from level a to level b changes the global angular properties of the atomic gas in the lower level (due to the fact that the removed atom carries a certain orientation) and in the upper level. This explains the presence of a term with $\bar\rho_a$ in (I.54a) and (I.54b).

(2) Likewise, the ${}_b\bar\rho$ term represents the effect of the laser on the atoms in the upper level: this is the effect of stimulated emission. This process and the absorption one are symmetrical. In ordinary optical pumping, such a process was not taken into account. Let us point out that absorption and stimulated emission do not interfere and appear in an uncorrelated way. Contrary to the coupling by spontaneous emission which is isotropic (term with Θ in (I.54a), the coupling by stimulated emission is anisotropic, because of the laser polarization.

(3) The term with n is the source term at the second order in the electric field: it represents the effect of both the absorption and the stimulated emission on the laser-off populations inversion, n.

C. Free-Running Modes

With the help of (I.13), (I.34) can be written

$$ {}^{s}_{p'}S^{k'}_{Q'}(v,\omega_Z) = \sum_{\mu} \frac{P^2_{ab}|\mathcal{E}^{\mu}||\mathcal{E}^{\mu-s}|e^{-i(\phi_\mu-\phi_{\mu-s})}}{\Gamma_{ab}(k')+i(\omega_\mu+p'\Delta\omega-\omega+Q'\omega_Z-\bar k v)}\,. \tag{I.55} $$

When the modes are free running, the phase ϕ_μ is a random function of μ.

1. $s=0$

For zero s, $\phi_\mu-\phi_{\mu-s}=0$ and the free-running-modes case is equivalent to the phase-locked one. What has been said in Sec. IIIA is still valid.

2. $s\neq 0$

${}^{s}_{p'}S^{k'}_{Q'}$ is the sum of terms such as $f(\mu)e^{i\psi}\mu$, where $f(\mu)$ is slowly varying with μ and ψ_μ a random function of μ. S cannot be expressed as an integral anymore. In the BLA conditions ($\Gamma_{ab}>\Delta\omega$), the various terms of (I.55) vanish by interference and $S(s\neq 0)\ll S(s=0)$. We finally get

$$ {}^{s}_{p'}S^{k'}_{Q'}(v,\omega_Z) = \gamma\delta_{s,0}\,, \tag{I.56} $$

where γ is defined in Sec. IIIA.

The exact validity of (I.56) is analyzed in a more detailed manner in Ref. 26. As long as the discharge excitation does not create modulation, the density-matrix modulations at the $p\Delta\omega$ frequencies disappear, due to (I.56): Equations (I.54) are still valid, provided that $p=s=0$. Subsequently, in the free-running-modes case, the condition (I.39b) is verified very well.

D. Physical Meaning of the Broad-Line Approximation

Relation (I.49) means that internal and external variables are separated: The atomic response to the laser irradiation does not depend on the velocity. This can be interpreted in the following way.

Let us consider the velocity distribution of one of the laser levels (Fig. 2). A mode μ interacts with the atoms having such a velocity that the mode frequency shifted by Doppler effect is equal to the atomic frequency, up to the natural width of the atomic transition: $\omega_\mu - k_\mu v = \omega$. Subsequently each laser mode creates a hole in the velocity distribution (the well-known "Bennett hole"[27]). The hole width is proportional to twice the optical coherence relaxation rate Γ_{ab} and their distance is proportional to the mode splitting $\Delta\omega$. If $\Gamma_{ab} > \Delta\omega$, the holes are overlapping and the hole structure of the atomic response disappears. If, in addition, the modes cover all the velocity distribution ($N\Delta\omega > \bar{k}u$), the atomic response does not depend on the velocity: we get the relation (I.49).

In the previous discussion, we have shown that the hole structure disappears if $\Gamma_{ab} > \Delta\omega$. For the atoms, everything happens as if the mode structure of the laser line were vanishing, and the laser line becomes equivalent to an ordinary light source of width $N\Delta\omega$. Subsequently, an analogy appears between the laser pumping and the optical pumping by an ordinary light source described by Barrat and Cohen-Tannoudji.[28] In an ordinary source, as the relative phase of the various field components are random, this analogy will be complete in the case when the laser modes are free running. This point, which is studied in a more detailed manner in Ref. 26 had previously been analyzed by Dumont.[5]

As shown in Ref. 26 this analogy leads to considering γ and δ [Eq. (I.42)], respectively, as the probability of real and virtual laser-induced transitions. It also leads to a physical interpretation of conditions (I.39), (I.41), and (I.45).

As in the case of an ordinary source, we can associate to the free-running multimode laser field a correlation time of about $T_L \simeq 2\pi(N\Delta\omega)^{-1}$. In the same way as in usual pumping, this correlation time determines the decay time of the transient phenomena appearing in the a and b levels after an interaction between an atom and a laser photon.[29] This means that the absorption or the stimulated emission, seen from the laser levels, lasts a time of the order of T_L.

This property is also valid for a phase-locked laser, in the BLA approximation. It is no longer possible to associate the atom-photon-interaction transient time to the correlation time of the laser field, because the latter is infinite. But we can

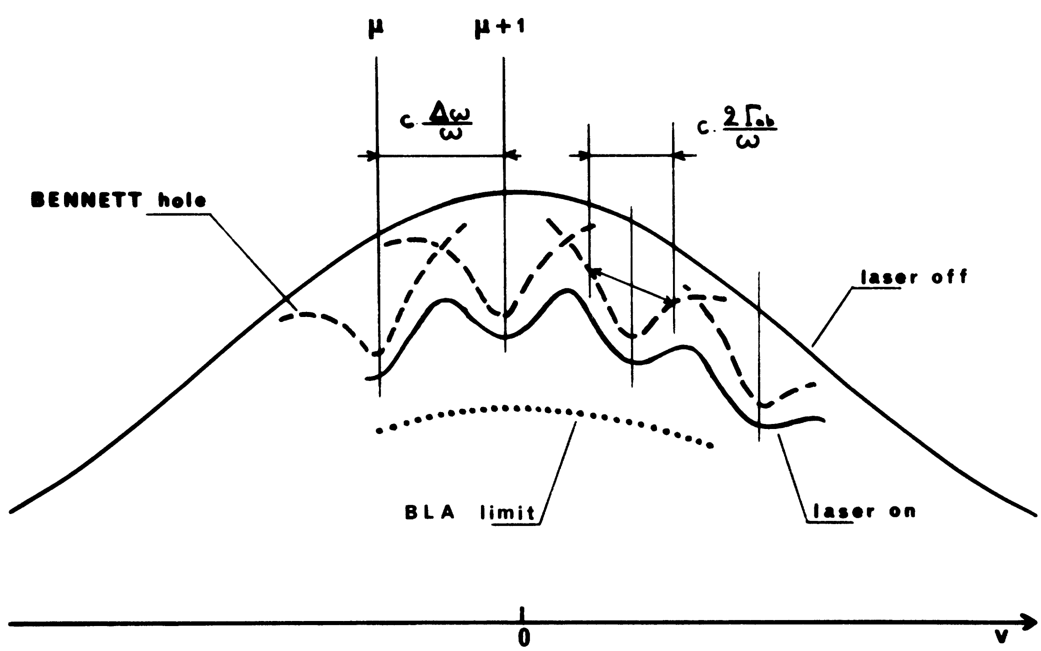

FIG. 2. Velocity distribution of a laser level. The laser-off distribution is proportional to the Maxwell repartition function $W_M(v)$. When the laser is turned on, a set of equidistant "Bennett holes" appears, the position of which is determined by $\bar{k}v = \omega_\mu - \omega$ (dashed curves). When Γ_{ab} is greater than $\Delta\omega$, the hole structure disappears and the atomic response is proportional to $W_M(v)$ (BLA limit, dotted line).

use another simple physical argument.

Let us analyze the time behavior of the laser electric field. At position r_0, the field is given by [see (I.15)]

$$\mathcal{E}(t) = e^{-i\bar{\omega}(t-t_0)} \sum_{\mu} \mathcal{E}^{\mu} e^{-i\mu\Delta\omega(t-t_0)} , \quad \text{(I.57)}$$

where $r_0 = ct_0$, $\bar{\omega}$ is the laser mean frequency, and μ varies between $-N/2$ and $N/2$ by integer values. $\mathcal{E}^{\mu}$ is real and positive for a phase-locked laser. For $t = t_0$ all the modes are in phase and the field amplitude is maximum. For $t - t_0 \gtrsim 2\pi/N\Delta\omega$, every phase has increased differently and there is a destructive interference. The field amplitude vanishes. For $t - t_0 = 2\pi/\Delta\omega$, all the modes get the same phase again and the amplitude is maximum. The laser field is modulated in short periodic pulses of width of the order of T_L and of period $2\pi/\Delta\omega$ (see Fig. 3).

Two cases are possible.

(1) $\Gamma_{ab} \ll \Delta\omega$. The optical dipole created by a pulse has a lifetime long enough to wait for the next pulse. Then a second interaction with the laser field may occur and the atom will undergo a transition between levels a and b: A time-delayed photon absorption may appear.[30]

(2) $\Gamma_{ab} \gtrsim \Delta\omega$. The latter process is no longer possible because the optical dipole vanishes before the transit of the second pulse. (For $\Gamma_{ab} = \Delta\omega$, the dipole is already reduced by a factor $e^{-2\pi} \simeq 2\times 10^{-3}$). The two successive interactions with the electric field must be realized during the transit time of one pulse. As it is in the free-running case, the photon absorption time is of the order of T_L. The difference between the two operations comes from the time modulations of the phase-locked laser intensity, which involve the modulations of the atomic quantities.

Introducing the characteristic interaction time T_L, we can write (I.39a) and (I.39b) as

$$\frac{\omega_z}{2\pi} T_L \ll 1 \quad \text{(I.58a)}$$

and

$$\frac{p\Delta\omega}{2\pi} T_L \ll 1 . \quad \text{(I.58b)}$$

Equation (I.58a) means that the optical coherence does not precess under the effect of the magnetic field during the interaction time. Since the magnetic field has not enough time to operate, the transition probability γ does not depend on H. From this, we can obtain the condition which alows us to neglect the coupling between the various ${}_{ab}\rho^{k'}_{Q'}$ in (I.20c). The $(\omega_a - \omega_b)$ factor means that this coupling is induced by the magnetic field, due to the anomalous Zeeman effect. If

$$\frac{\omega_a - \omega_b}{2\pi} T_L = \frac{\omega_a - \omega_b}{N\Delta\omega} \ll 1 , \quad \text{(I.59)}$$

the magnetic field has not enough time to induce this coupling during the characteristic interaction time and it can be neglected. A rigorous proof is given in Ref. 26.

Similarly, (I.58b) infers that, during the absorption time, the density matrix does not evolve at

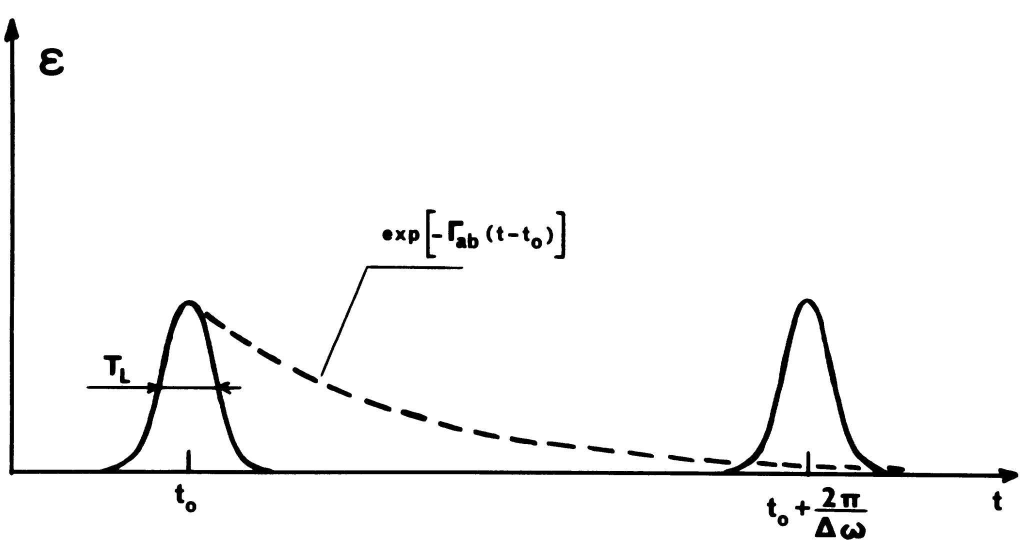

FIG. 3. Electric field amplitude of a phase-locked multimode laser as a function of time. The amplitude is modulated in short periodic pulses of width T_L. The dashed curve represents the decay of the optical coherence created by the t_0 pulse.

the modulations frequencies: γ is independent of these modulations. Condition (I.41) is equivalent to $(\bar{k}u/2\pi)\,T_L \ll 1$ (or else $uT_L \ll \lambda$). During the absorption time, the atom travels across a distance much smaller than the laser wavelength. The Doppler shift of the optical dipole does not appear and γ does not depend on the velocity.

Lastly, (I.45) can be written $\Gamma_{ab}T_L \ll 1$. Thus γ is not sensitive to the optical dipole decay and appears as the product of the interaction strength, $P_{ab}^2 I$, by the interaction time $(N\Delta\omega)^{-1}$. But in the contrary case $(\Gamma_{ab}^{-1} \ll T_L)$ the optical coherence lifetime is much smaller than the pulse duration: now Γ_{ab}^{-1} is the photon-atom interaction time. This explains the shape of (I.48): $P_{ab}^2 I(\Gamma_{ab})^{-1}$.

IV. LINEAR POLARIZATION

In the following, we shall analyze Eqs. (I.54) in the more compact form ($\beta = a$ or b):

$$[\Gamma_\beta(k) + i(Q\omega_\beta + p\Delta\omega)]\,{}_\beta\bar{\rho}_Q^k(p) = \delta_{\beta,a}\Theta(b,a,k)\,{}_b\bar{\rho}_Q^k(p) + \frac{n\gamma}{\sqrt{3}}\,{}_\beta g_Q^k + \gamma\sum_{k''Q''s}\left[{}_{\beta\alpha}h_{QQ''}^{kk''}\,{}_\alpha\bar{\rho}_{Q''}^{k''}(p-s) - {}_\beta h_{QQ''}^{kk''}\,{}_\beta\bar{\rho}_{Q''}^{k''}(p-s)\right], \tag{I.60}$$

with

$${}_b g_Q^k = -2\sum_{qq'} e_{-q}e_{q'}^{*}\,{}_{ba}^{\;q}G_{q'Q}^{1k}\,, \tag{I.61}$$

$${}_{ba}h_{QQ''}^{kk''} = 2\sum_{qq'} e_{-q}\,e_{q'}^{*}\,{}_{ba}^{qq'}H_{QQ''}^{kk''}\,, \tag{I.62}$$

$${}_b h_{QQ''}^{kk''} = \sum_{qq'}\left[e_{-q}e_{q'}^{*} + (-)^{k+k''+Q+Q''}e_{-q'}e_q^{*}\right]{}_b^{qq'}H_{QQ''}^{kk''}\,, \tag{I.63}$$

and similar expressions for the a level.

When the laser is linearly polarized, the polarization vector $\tilde{e}$ is real and

$$e_q = (-)^q e_{-q}^{*}\,. \tag{I.64}$$

Thus

$${}_\beta g_Q^1 = 0 \tag{I.65}$$

and

$${}_{\beta\alpha}h_{QQ''}^{kk''} = 0 \;\;\text{for odd } (k-k'')\,. \tag{I.66}$$

Equation (I.65) means that, up to the second order in the electric field, the laser creates population, $k=0$, and alignment, $k=2$ (no orientation, $k=1$). This is a well-known result in optical pumping. On the other hand, (I.66) means that a coupling only appears between such tensorial quantities as $k-k''=0$ or ± 2. At all orders in the field, the laser only creates even tensorial orders.

A. π Polarization

When the polarization vector is parallel to the magnetic field, $e_0 = 1$ and $e_{\pm 1} = 0$. The selection rules of the $3J$ coefficients prescribe $Q = Q'' = 0$ in (I.54). The equations of motion do not depend on the magnetic field anymore.[31] When the modes are free running, there are no modulations and one obtains

$$\Gamma_a(k)\,{}_a\bar{\sigma}_0^k = \Theta(b,a,k)\,{}_b\bar{\sigma}_0^k + \frac{2n\gamma}{\sqrt{3}}\,{}_{ab}^{\;0}G_{00}^{1k} + 2\gamma\sum_{k''}\left[{}_{ab}^{00}H_{00}^{kk''}\,{}_b\bar{\sigma}_0^{k''} - {}_a^{00}H_{00}^{kk''}\,{}_a\bar{\sigma}_0^{k''}\right], \tag{I.67a}$$

$$\Gamma_b(k)\,{}_b\bar{\sigma}_0^k = -\frac{2n\gamma}{\sqrt{3}}\,{}_{ba}^{\;0}G_{00}^{1k} + 2\gamma\sum_{k''}\left[{}_{ba}^{00}H_{00}^{kk''}\,{}_a\bar{\sigma}_0^{k''} - {}_b^{00}H_{00}^{kk''}\,{}_b\bar{\sigma}_0^{k''}\right], \tag{I.67b}$$

where ${}_\beta\bar{\sigma}$ is the density matrix for the π polarization. In the phase-locked case, Eqs. (I.67) are still valid if

$$\Gamma_\beta(k) \ll \Delta\omega\,. \tag{I.68}$$

The density-matrix modulations contain the nonresonant factor $[\Gamma_\beta(k) + ip\Delta\omega]^{-1}$ and are negligible.

Due to the selection rules of the $3J$ coefficients, the tensorial order of the optical coherences must be odd.[32] Subsequently, the H coefficients are defined by

$${}_{\beta\alpha}^{00}H_{00}^{kk''} = \sum_{k'\,\text{odd}} {}_{\beta\alpha}^{\;0}G_{00}^{k'k}\,{}_{\alpha\beta}^{\;0}G_{00}^{k'k''} \tag{I.69}$$

and

$${}_\beta^{00}H_{00}^{kk''} = \sum_{k'\,\text{odd}} {}_{\beta\alpha}^{\;0}G_{00}^{k'k}\,{}_{\beta\alpha}^{\;0}G_{00}^{k'k''}\,. \tag{I.70}$$

An interesting case corresponds to the transitions verifying

$$J_a + J_b \le 2\,. \tag{I.71}$$

They are the $J=0 \leftrightarrow J=1$, $J=1 \leftrightarrow J=1$, and $J=\frac{1}{2} \leftrightarrow J=\frac{3}{2}$ transitions. Owing to the selection rules $(k' \le J_a + J_b)$, k' must be equal to 1 and the H coefficients factorize. The ${}_{ab}\xi$ coefficients [Eq. (I.51)] can be reintroduced in (I.67):

$$\Gamma_a(k)\,{}_a\bar{\sigma}_0^k = 2\gamma\,{}_{ab}\bar{\xi}_0^1\left[{}_{ab}^{\;0}G_{00}^{1k} - \frac{\Theta(b,a,k)}{\Gamma_b(k)}\,{}_{ba}^{\;0}G_{00}^{1k}\right], \tag{I.72a}$$

$$\Gamma_b(k)\,{}_b\bar{\sigma}_0^k = -2\gamma\,{}_{ab}\bar{\xi}_0^1\,{}_{ba}^{\;0}G_{00}^{1k}\,. \tag{I.72b}$$

It is easy to show that

$${}_{ab}\bar{\xi}_0^1 = \frac{n}{\sqrt{3}}\left[1 + 2\gamma\sum_k\left(\frac{({}_{ba}^{\;0}G_{00}^{1k})^2}{\Gamma_b(k)} + \frac{({}_{ab}^{\;0}G_{00}^{1k})^2}{\Gamma_a(k)} - \Theta(b,a,k)\frac{{}_{ab}^{\;0}G_{00}^{1k}\,{}_{ba}^{\;0}G_{00}^{1k}}{\Gamma_a(k)\Gamma_b(k)}\right)\right]^{-1}. \tag{I.73}$$

All the density-matrix components show the same variations with the laser intensity and their ratios are constant.[33]

This last result can be interpreted by simple physical considerations. In Fig. 4 the different laser transitions have been represented. It is essential to note that, in every case, all the allowed transitions have the same probability. Due to this fact and to the spherical symmetry of the relaxation, the Zeeman sublevels of a laser level, which are connected by these transitions, undergo identical populations modifications. This characteristic is valid at all orders in the laser electric field. The other sublevels may be populated by the depolarizing relaxation processes. Subsequently, at every order in the electric field, the proportion between the laser-induced modifications of the sublevels populations is constant: Their ratio is independent of the laser intensity.

From Eq. (I.67), we can obtain the atomic density matrix for any linear polarization, at zero magnetic field. This can be done by performing on the density-matrix components, the rotation which brings the Oz axis on the selected polarization axis.[33] Particularly, for the polarization along the Oy axis.

$$
{}_\beta\bar{\rho}_Q^k(H=0)=R_{Q0}^{(k)}(\pi/2,\pi/2,0)\,{}_\beta\bar{\sigma}_0^k
=(-)^{k/2}\frac{[(k+Q)!\,(k-Q)!\,]^{1/2}}{2^k[(k+Q)/2]!\,[(k-Q)/2]!}\,{}_\beta\bar{\sigma}_0^k\,; \tag{I.74}
$$

k and Q are even. $R_{QQ'}^{(k)}$ are the rotation matrix components.[34] For the transitions verifying $J_a+J_b \leqslant 2$, the ratios between the density-matrix components are constant. This is the reason why the anisotropy ratio of the laser-induced fluorescence variations does not depend on the laser intensity. (For experimental verifications, see Fig. 4 of Ref. 1a, and Ref. 10, Figs. 7 and 14.)

B. σ Polarization

If the polarization axis is perpendicular to Oz, $e_0=0$. Thus, we easily show that Q must be even. When the field is equal to zero, by carrying out a rotation which brings the polarization direction on Oz, we reduce the problem to the previous one.

In the other cases, we must consider Eqs. (I.60). For a laser polarization along the Oy axis, $e_{\pm1} = -i/\sqrt{2}$, $e_0=0$, and subsequently

$$
{}_{\beta\alpha}h_{QQ''}^{kk''}=\sum_{\substack{q'\neq 0\\ q'\neq 0}}{}^{qq'}_{\beta\alpha}H_{QQ''}^{kk''}\quad(\beta,\ \alpha=a\text{ or }b)\,. \tag{I.75}
$$

For each particular transition, Eqs. (I.60) are entirely solvable. In practice, for high-J values, the calculations become too complicated. The

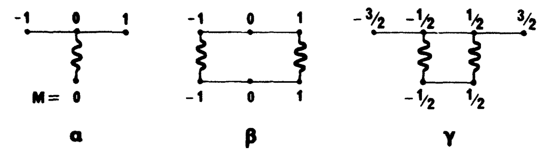

FIG. 4. Diagram of the π transitions when $J_a+J_b \leq 2$. (α) $J=1$-$J=0$, (β) $J=1$-$J=1$, (γ) $J=\frac{3}{2}$-$J=\frac{1}{2}$.

equations of motion have been solved at all orders in the field by an analytic calculation for $J=1$-$J=0$ (Ref. 17) and $J=1$-$J=1$ (Ref. 48) transitions, and by a computer calculation for a $J=1$-$J=2$ (Ref. 35) transition. The exact solutions and the experimental verifications will be presented in forthcoming papers. In the following, we study the behavior of the solution for any J value, by means of a fourth-order perturbation expansion which is only valid at weak laser intensities. The results will be compared with previous calculations.

1. Nonlinear Effects in Weak Magnetic Field

We maintain condition (I.68): $\Gamma_\beta(k) \ll \Delta\omega$. Therefore, in weak field $(\omega_\beta \simeq \Gamma_\beta(k))$, due to the nonresonant factor $[\Gamma_\beta(k)+i(Q\omega_\beta+p\Delta\omega)]^{-1}$ the density-matrix modulations are negligible and we set $p=s=0$ in (I.60). Moreover when the modes are free running, condition (I.68) is not necessary.

To the second order in the electric field (i.e., first order in γ) the laser irradiation induces a population modification (ρ_0^0) and creates a longitudinal (ρ_0^2) and transverse alignment $(\rho_{\pm2}^2)$. At higher orders, first a coupling between these quantities appears and secondly, for high-J levels $(J \geqslant 2)$, the laser creates high-order multipole moments $(k \geqslant 4)$. Generation and detection of such moments will be studied in other publications.[35] These high-order multipole moments are created only from the fourth order in the field. Subsequently, they appear in the equations of motion of the population and alignment at the sixth order: *they will be neglected in the fourth-order calculation.*

a. Hanle-effect broadening. Up to the fourth order, the equation of motion of the b transverse alignment is given by Eq. (I.60), in which we set $k=Q=2$ and we neglect the ρ_Q^k components for $k \geqslant 4$:

$$
[\Gamma_b(2)+2i\omega_b]\,{}_b\bar{\rho}_2^2=\frac{n\gamma}{\sqrt{3}}\,{}_bg_2^2
+\gamma\sum_{k''=0,2}\left[{}_{ba}h_{20}^{2k''}\,{}_a\bar{\rho}_0^{k''}-{}_bh_{20}^{2k''}\,{}_b\bar{\rho}_0^{k''}\right]
+\gamma\left[{}_{ba}h_{22}^{22}\,{}_a\bar{\rho}_2^2-{}_bh_{22}^{22}\,{}_b\bar{\rho}_2^2\right]. \tag{I.76}
$$

We shall see later on that, from the fourth order in the electric field, the longitudinal components depend on the magnetic field. Nevertheless, these variations appear into Eq. (I.76) at the sixth order (i.e., third order in γ) only. Consequently, to study the Hanle-effect shape up to the fourth order, we only take into account the coupling between the transverse alignments.

${}_bh_{22}^{22}$ is always positive:

$$ {}_bh_{22}^{22}=5\sum_{k'}(2k'+1) \times\left[\begin{pmatrix}k' & 1 & 2\\ 1 & 1 & -2\end{pmatrix}^2+\begin{pmatrix}k' & 1 & 2\\ 3 & -1 & -2\end{pmatrix}^2\right]\begin{Bmatrix}k' & 1 & 2\\ J_b & J_b & J_a\end{Bmatrix}^2 . \tag{I.77}$$

The coupling of ${}_b\overline{\rho}_2^2$ with itself corresponds to a linear broadening of the Hanle effect. Its width becomes $\Gamma_b(2)+{}_bh_{22}^{22}\gamma$.[36] The physical meaning is evident: due to the laser-induced transitions between a and b, the alignment mean lifetime is reduced. If ${}_{ba}h_{22}^{22}$ is different from zero, there is a coupling between the a and b transverse alignments, and the shape of Hanle effect of b is no longer Lorentzian.[37]

Using the symmetry relations of H (Ref. 24), we obtain

$$ {}_{ba}h_{22}^{22}=2\,{}_{ba}^{-1 1}H_{22}^{22}=10\begin{Bmatrix}3 & 1 & 2\\ J_b & J_b & J_a\end{Bmatrix}\begin{Bmatrix}3 & 1 & 2\\ J_a & J_a & J_b\end{Bmatrix} . \tag{I.78}$$

This last relation means that, in BLA, the a and b alignments are coupled through the $k'=3$ optical coherence (${}_{ab}\rho_3^3$) only. For the transitions verifying $J_a+J_b\leq 2$ [Eq. (I.71)], the coupling coefficient vanishes: there is no direct coupling of the transverse alignments. (At higher orders in the field, there is an indirect coupling through the longitudinal quantities: ${}_b\rho_2^2\rightarrow{}_\beta\rho_0^k\rightarrow{}_a\rho_2^2$.) In this case, up to the fourth order, the Hanle effect keeps a Lorentzian shape and its width is given by $\Gamma_b(2)+{}_bh_{22}^{22}\gamma$.

The apparent relaxation rate is the sum of the effects due to spontaneous emission, collisions, and laser irradiation. For $J_a+J_b\leq 2$, up to the fourth order, the laser effect is equivalent to a relaxation process which does not depend on the other relaxation processes: the corresponding term has only to be added to the other relaxation terms.[38]

The absence of coupling is evident for the $J=0\leftrightarrow J=1$ and $J=\frac{1}{2}\leftrightarrow J=\frac{3}{2}$ transitions: an alignment cannot exist in one of the laser levels. The $J=1\leftrightarrow J=1$ case can be explained by simple physical considerations. In Fig. 5 we have drawn the transitions induced between the two levels by a σ-polarized laser.

The b transverse alignment, which corresponds to the Zeeman coherence between the $m=-1$ and $m=1$ sublevels, is coupled by the laser to the population of the $m=0$ sublevel of level a only (dashed transitions). Likewise, the a transverse alignment is coupled to the population of the $m=0$ sublevel of b only (solid lines). Thus the laser does not couple the alignments.[39]

b. Saturation resonances in a zero field. If we neglect the tensorial components of order $k\geq 4$, the equations of motion of the longitudinal quantities are given by

$$\Gamma_b(k)\,{}_b\overline{\rho}_0^k=\frac{n\gamma}{\sqrt{3}}{}_bg_0^k+\gamma\sum_{k''=0,2}\left[{}_{ba}h_{00}^{kk''}\,{}_a\overline{\rho}_0^{k''}-{}_bh_{00}^{kk''}\,{}_b\overline{\rho}_0^{k''}\right] +\gamma\left[{}_{ba}h_{02}^{k2}({}_a\overline{\rho}_2^2+{}_a\overline{\rho}_2^{2*})-{}_bh_{02}^{k2}({}_b\overline{\rho}_2^2+{}_b\overline{\rho}_2^{2*})\right]. \tag{I.79}$$

To the fourth order, due to the coupling with ${}_a\overline{\rho}_2^2$ and ${}_b\overline{\rho}_2^2$, the longitudinal quantities present a resonant variation around the zero magnetic field. This resonance is the sum of two Lorentzian shapes proportional to the a Hanle effect (${}_a\overline{\rho}_2^2+{}_a\overline{\rho}_2^{2*}$), and the b Hanle effect (${}_b\overline{\rho}_2^2+{}_b\overline{\rho}_2^{2*}$), respectively. The relative importance of these contributions is given by

$$-\frac{{}_{ba}h_{02}^{k2}\,{}_a\overline{\rho}_2^2(H=0)}{{}_bh_{02}^{k2}\,{}_b\overline{\rho}_2^2(H=0)}=\frac{{}_{ba}^{-1-1}H_{02}^{k2}\;{}_{ab}^{1}G_{12}^{12}}{{}_{b}^{-1-1}H_{02}^{k2}\;{}_{ba}^{1}G_{12}^{12}}\frac{\Gamma_b(2)}{\Gamma_a(2)}. \tag{I.80}$$

For high values of J, (I.80) is equal to $\Gamma_b(2)/\Gamma_a(2)$, and the importance of the contribution of the two Hanle effects is inversely proportional to their width. From (I.79), it is evident that the power broadening of the Hanle effects must also appear in the saturation resonances.

2. *Nonlinear Effects in Strong Field*

If the magnetic field is strong, we cannot neglect the modulations of the density matrix (in the phase-locked operation). Indeed, ${}_\beta\overline{\rho}_Q^k(p)$ is resonant if $Q\omega_\beta+p\Delta\omega=0$. First we shall study the case where either the Landé factors are equal, or the laser transition is a $J=1$-$J=0$ transition.

a. $g_a=g_b=g$. ${}_a\overline{\rho}_Q^k(p)$ and ${}_b\overline{\rho}_Q^k(p)$ are simultaneously resonant for $Q\omega_a=Q\omega_b=-p\Delta\omega$. In particular, the first modulations of the transverse alignments appear for $2\omega_a=2\omega_b=-\Delta\omega$. For such a value of the field, ${}_\beta\overline{\rho}_Q^k$ presents a resonant modulation at the frequency $p\Delta\omega=\frac{1}{2}Q\Delta\omega$ (Q is even).

If $\Gamma_\beta(k)\ll\Delta\omega$, the other modulations are negligible and Eqs. (I.60) are reduced to

$$\left[\Gamma_\beta(k)+iQ(\omega_\beta+\tfrac{1}{2}\Delta\omega)\right]{}_\beta\overline{\rho}_Q^k(Q/2) =(n\gamma/\sqrt{3})\,{}_\beta g_Q^k+\delta_{\beta,a}\Theta(b,a,k)\,{}_b\overline{\rho}_Q^k(Q/2) +\gamma\sum_{k''Q''}\left[{}_{\beta\alpha}h_{QQ''}^{kk''}\,{}_\alpha\overline{\rho}_{Q''}^{k''}\left(\frac{Q''}{2}\right)-{}_\beta h_{QQ''}^{kk''}\,{}_\beta\overline{\rho}_{Q''}^{k''}\left(\frac{Q''}{2}\right)\right]. \tag{I.81}$$

When $\omega_a=\omega_b$, these equations are exactly equivalent to the weak-field equations, if we replace $(\omega_\beta+\frac{1}{2}\Delta\omega)$ by ω_β and ${}_\beta\overline{\rho}_Q^k(Q/2)$ by ${}_\beta\overline{\rho}_Q^k(0)$. The behavior of the solutions are the same. All that has been said about the Hanle effect can be applied to the modulated transverse alignment, ${}_B\bar{\rho}_2^2(1)$. For instance, if $J_a+J_b\leq 2$, up to the fourth order, the behavior of ${}_B\bar{\rho}_2^2(1)$ is Lorentzian and is centered on the following value of the field:

$$H^{(1)}=\Delta\omega/2g\mu_B\,. \tag{I.82}$$

Because of equations similar to (I.79) [where we replace ${}_B\bar{\rho}_2^2$ by ${}_B\bar{\rho}_2^2(1)$], this modulation of the alignment is responsible of a saturation resonance on the nonmodulated longitudinal quantities. For $H=H^{(1)}$, a "lateral saturation" appears on ${}_B\bar{\rho}_0^k$, having exactly the same behavior as the "central saturation" ($H=0$).

In the same way, the modulations of the alignment at the $s\Delta\omega$ frequency appear for $H^{(s)}=s\Delta\omega/2g\mu_B$. Thus ${}_B\bar{\rho}_Q^k$ evolves at the frequency $p\Delta\omega=s(Q/2)\Delta\omega$. Equations (I.81) are still valid, provided that we replace $\Delta\omega$ by $s\Delta\omega$, and ${}_B\bar{\rho}_Q^k(Q/2)$ by ${}_B\bar{\rho}_Q^k(sQ/2)$. In particular, the longitudinal quantities present a "lateral saturation" resonant for $H=H^{(s)}$. [Nevertheless, let us remember that these results are valid only if condition (I.39) is fulfilled: $s\ll N$.]

The previous study shows that when (1) the modes are phase locked, (2) the Landé factors are equal, and (3) $\Gamma_B(k)\ll\Delta\omega$, the general solution can be deduced from the solution of the equations of motion in weak field [(I.60) with $p=s=0$].

If we call ${}_B^0\bar{\rho}(H)$ the solution in weak field, the general solution is, for $Q\neq 0$,

$${}_B\bar{\rho}_Q^k(H)=\sum_s {}_B^0\bar{\rho}_Q^k(H-H^{(s)})\exp[i(sQ/2)(t\Delta\omega-r\Delta k)] \tag{I.83}$$

and, for $Q=0$,

$${}_B\bar{\rho}_0^k(H)={}_B^0\bar{\rho}_0^k(\infty)+\sum_s[{}_B^0\bar{\rho}_0^k(H-H^{(s)})-{}_B^0\bar{\rho}_0^k(\infty)]\,. \tag{I.84}$$

${}_B^0\bar{\rho}_0^k(\infty)$ is the solution of (I.60) when the longitudinal quantities are the only ones kept ($Q=0$); this corresponds to a magnetic field such that $\Gamma_B(k)\ll\omega_B\ll\Delta\omega$ $[{}_B^0\bar{\rho}_0^k(H-H^{(s)})-{}_B^0\bar{\rho}_0^k(\infty)]$ represents the sth lateral saturation resonance. Its shape is given at all orders by (I.60), and at fourth order by (I.79).

When the modes are free running, there is no more modulation and the general solution is given by ${}_B^0\bar{\rho}_Q^k(H)$. On the longitudinal quantities appears only a "central saturation" ($H=0$) but no lateral saturation.[4]

b. $g_a\neq g_b$. If the Landé factors are different, the modulations of ${}_a\bar{\rho}$ and ${}_b\bar{\rho}$ are not resonant for the same value of the field. Indeed, the first modulations of ${}_a\bar{\rho}$ appear when $-2\omega_a=2g_a\mu_BH\simeq\Delta\omega$,

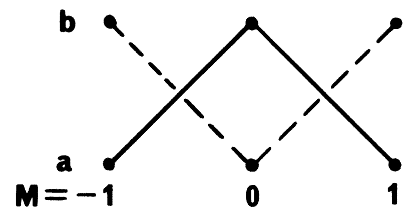

FIG. 5. Diagram of the transitions induced by a σ-polarized laser beam between two $J=1$ levels.

and the first modulations of ${}_b\bar{\rho}$ when $2g_b\mu_BH\simeq\Delta\omega$. If $|\omega_a-\omega_b|\lesssim\Gamma_B(k)$, Eqs. (I.81) are still valid; but, as $\omega_a\neq\omega_b$, they are no more equivalent to the weak-field equations. The saturation resonances from levels a and b are not centered on the same value of the field.

On the other hand, if $|\omega_a-\omega_b|\gg\Gamma_B(k)$, the nonresonant transverse quantities can be eliminated from (I.81). For instance, if the b transverse quantities are the only resonant ones, a coupling between the transverse alignments cannot exist. Even if $J_a+J_b>2$, up to the fourth order, the modulated transverse alignment of b keeps a Lorentzian shape.[40]

V. GENERAL CASE

When the laser polarization is elliptic, the study is much more complicated. In general, there is no more selection rule. All the density-matrix tensorial orders are excited. In particular, at the second order in the electric field, the laser creates both orientation ($k=1$) and alignment ($k=2$), and couples them at higher orders: from the fourth order, the Hanle effects do not keep a Lorentzian shape. On the other hand, the saturation resonances on the longitudinal quantities come from all the second-order Hanle effects: transverse orientation ($\bar{\rho}_1^1$) and alignment ($\bar{\rho}_1^2$, $\bar{\rho}_2^2$).

When (I.68) is fulfilled, for a given magnetic field, there is at most one resonant modulation in ${}_B\rho_Q^k$. Then, in theory, Eqs. (I.54) can be completely solved.[26] But the calculation is complicated. As experiments have been performed using a linear polarization, we shall not analyze the general case in a more detailed way.

VI. VALIDITY OF THE APPROXIMATIONS–LIMITS OF THE THEORY

In this section, we analyze, in a detailed manner, the validity of the approximations introduced in Sec. III.

A. Atomic Transitions

In a first part, we consider the case of visible or near-infrared laser transitions of neutral atoms. More particularly, all the numerical values come from the following neon transitions (in Paschen notation): $3s_2-2p_4$ ($\lambda=6328$ Å), $3s_2-2p_2$ ($\lambda=6401$ Å), $3s_2-2p_1$ ($\lambda=7305$ Å), and $2s_2-2p_1$ ($\lambda=1.52$ μ).

To write S as an integral, the conditions (I.36) and (I.37) are necessary. For the visible lines, (I.37) is well verified in the usual experimental conditions. (For a 2-m-long laser, $N\simeq 10-13$ for $\lambda=7305$ Å, $\simeq 15$ for 6401 Å, and $\gtrsim 20$ for 6328 Å). On the other hand, if the atomic linewidth is greater than the mode spacing [Eq. (I.36)], the hole structure of the atomic response vanishes (see Fig. 2). For a 2-m-long laser cavity, $\Delta\omega$ is equal to 75 MHz. But the atomic linewidth is strongly broadened by the dephasing atomic collisions. By Lamb-dip experiments on the 6328-Å laser line, Smith[41] measures a broadening $d\Gamma_{ab}/dp$ about 70 MHz/Torr in a 7 : 1 mixture of He : Ne; for the same mixture, Shank[42] gives 90 MHz/Torr. Dietel[43] finds 61 MHz/Torr for a 5 : 1–He : Ne mixture and Cordover[44] finds 58 MHz/Torr for a 8 : 1 mixture. On the 1.52-μ line, Carroll[45] gives 80 MHz/Torr for a 10 : 1–He : Ne mixture. Subsequently, the condition (I.36) is valid in the usual laser pressure range (1 to 5 Torrs) and the expression of S by means of an integral [Eq. (I.38)] is well proved.

Now, let us consider the second set of conditions: $N\Delta\omega\gg\omega_Z$, $p'\Delta\omega$, $s\Delta\omega$ (I.39). Most of the experimental results will concern the Hanle effect and the zero-field "saturation resonance". In this case, (I.39) is verified very well, because the magnetic field is weak: $|H|\lesssim 5$ G and subsequently $|\omega_Z|\lesssim 10$ MHz, while $N\Delta\omega\simeq 1000-2000$ MHz. Moreover, as condition (I.68) is valid in the usual experimental situation [$\Gamma_\beta(k)\simeq 10$ MHz and $\Delta\omega \simeq 80$ MHz (Refs. 1 and 10)], the density-matrix modulations are not resonant and $p'=s=0$.

On the other hand, for the analysis of modulations and "lateral saturations", the validity of (I.39) must be carefully studied. For the first modulations ($p=1$), $p\Delta\omega\simeq 80$ MHz and the resonant field corresponds to $\omega_Z\simeq 40$ MHz: (I.39) is still valid, but becomes less and less valid as the modulation order grows up and, subsequently, as the magnetic field at resonance becomes higher. To show the limits of the present theory, let us compare the results of Sec. IV with the interpretation that Dumont proposes for the saturation resonances using a more sophisticated fourth-order perturbation calculation.[4] Dumont has shown these resonances are produced by two phenomena.

(1) A population effect which is resonant when the σ^+ component of a laser mode and the σ^- component of another one interact with the atoms of same velocity (Bennett hole crossing). The width of these resonances is $2\Gamma_{ab}$. In the BLA conditions ($\Gamma_{ab}>\Delta\omega$), these resonances are not resolved, as shown by both the theoretical calculations and the experimental results of Dumont.[4]

(2) A Zeeman coherence effect: to the second order, the laser creates a modulated transverse alignment, and, to the fourth order, this alignment produces unmodulated longitudinal quantities exhibiting resonances when the magnetic field is scanned. The saturation resonances appearing in Sec. IV, have exactly the same origin. However, when the modes are phase locked, in the BLA approximation, all the resonances have the same intensity, whereas the more realistic calculation of Ref. 4 shows that, due to the finite number of modes, the intensity of the resonances slowly decreases when their order (p) grows (see Figs. 5, 7, and 9 of Ref. 4). This feature agrees with the validity conditions of the BLA: *the modulation order must be much smaller than the modes number.*

$N\Delta\omega\gg\Delta\nu$ is the last condition introduced in the BLA (Eq. I.41). It is the most crude one because, for the usual neon laser lines, $N\Delta\omega$ is of the order of $2\Delta\nu$. This condition is necessary for the laser-induced transition probability $S(v)$ to be independent of the velocity. Indeed there are other ways to get the velocity-averaged equations, (I.50) and (I.51), from the motion equations, (I.35). For instance, it is sufficient that $S(v)$ and $\rho(v)$ are not correlated: the velocity average of $S(v)\rho(v)$ is then written as a product of the two velocity averaged quantities $\langle S(v)\rangle_{av}\langle\rho(v)\rangle_{av}$.

This situation is realized in the optical pumping of the ground state.[29] There are many collisions which change the velocity without disturbing the internal state of the atom. The velocity distribution is quickly thermalized and $\rho(v)$ is proportional to the Maxwell repartition function. To a certain extent, this situation may also be realized in the excited states when the fluorescence lines are strongly reabsorbed in the gas: in this case, it is the trapping of the fluorescence lines that ensures the thermalization of the velocity of the excited atoms.

Another possibility is that the intensities of the modes are fairly equal. Indeed, if all the modes have the same amplitude, it is shown in Ref. 26 that, to get Eqs. (I.50), condition (I.41) can be replaced by $N\Delta\omega\gg\Gamma_{ab}$.

In other respects, when the laser intensity grows, the correlations which may exist between $\rho(v)$ and $S(v)$ decrease. Indeed, the ρ dependence on the modes intensity is essentially nonlinear. This nonlinear behavior softens off the depth difference of the Bennett holes coming from the difference between the modes intensities. Moreover, these holes are powerbroadened and more overlapping. $\rho(v)$ comes nearer to the Maxwell distribution. On the other hand, $S(v)$ is always a linear function of the intensity of the modes: this explains the vanishing of the $S(v)$-$\rho(v)$ correlations. Subsequently, the BLA approximation is particularly convenient for high laser intensities.

In conclusion of this part, the use of the BLA equations seems to be correct for the atomic transitions.

B. Molecular Transitions

Recently, a few molecular levels have been successfully pumped with ionic lasers. McClintock *et al.*[6] have observed the zero-field level crossing in the excited $B^1\pi_u$ state of the Na_2 molecule pumped by an argon-ion laser. Broyer *et al.*[7] have studied the same effect in some excited states of I_2 with Ar^+ and Kr^+ lasers.

Is the BLA valid for these experiments? The width of the ion lasers is very large: $N\Delta\omega \simeq 3000$–5000 MHz. The Doppler widths are about 300 MHz for I_2 and 800 MHz for Na_2. Subsequently the condition (I.41) is fulfilled: $N\Delta\omega \gg \Delta\nu$. On the other hand, due to the weak-g values of these molecules, $N\Delta\omega$ is much greater than ω_Z. The principal difficulties come from the relative values of Γ_{ab} and $\Delta\omega$. In the pumping of molecules, the pressures are smaller (10^{-2}–1 Torr) than for the atoms, and the collision broadening of the optical linewidth must be weaker.

For the Na_2 experiments, McClintock *et al.* give $\Delta\omega = 60$ MHz and measure a natural width of the $B^1\pi_u$ state of about 25 MHz. The problem is that the effect of the dephasing collisions is not known (the authors do not give the experimental pressures). It is possible that the BLA conditions are fulfilled: this is necessary for the validity of the zero-field optical pumping theory of Drullinger and Zare using a rate-equation approach (see note[23]).

With regard to the I_2 molecule, the laser-saturated absorption experiments[46] show that the optical linewidth stays in the 1–10 MHz range as long as the pressure is lower than 1 Torr. Due to the 100 MHz mode spacing, the BLA validity is highly unlikely. In particular, the Zeeman-tuned holes crossing effect must be taken into account. The observed saturation effects[47] cannot be explained in the frame of the present theory.

VII. CONCLUSION

In this paper, we have presented a theoretical study of the optical pumping with a multimode laser. The so-called "broad-line approximation" allows us to write a set of equations of motion which are theoretically solvable at all orders in the laser field. The fourth-order solution has been analyzed here and the results have been compared with previous theories. The validity of BLA must be considered in a detailed way for each particular transition.

This theory seems well fitted to the laser pumping of the neon levels. We have entirely solved the motion equations in the case when the laser is linearly polarized and resonant for the following transitions: $J=0$-$J=1$ (Ref. 17), $J=1$-$J=1$ (Ref. 48), and $J=1$-$J=2$ (Ref. 35). The experimental verifications have been worked out on the neon laser lines. They will be described in a detailed manner in following papers.

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[15] The polarization vector may be complex (case of a circular polarization). Let us recall the definition of the standard components e_q:
$e_0 = e_z$, $e_{\pm 1} = \mp(1/\sqrt{2})(e_x \pm i e_y)$.

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[23]The introduction of a laser-induced transition probability which is independent of the atomic velocity leads to equations equivalent to those usually called "rate equations." These equations have been used by several authors [see, for instance, R. E. Drullinger and R. N. Zare, J. Chem. Phys. **51**, 5532 (1969); also, P. W. Smith, IEEE J. Quantum Electron. **QE-2**, 62 (1966)]. The use of these rate equations is only right in the BLA, as described in Sec. III.

[24]Using the symmetry reactions of the $3J$ and $6J$ coefficients (Messiah, Ref. 34, pp. 907–917), we can get ${}^{q'q}_{\beta\alpha}H^{kk''}_{QQ''} = (-)^{k+k''+Q+Q''}\,{}^{qq'}_{\beta\alpha}H^{kk''}_{QQ''}$, ${}^{q'q}_{\alpha\beta}H^{k''k}_{Q''Q} = {}^{qq'}_{\beta\alpha}H^{kk''}_{-Q-Q''}$ $= (-)^{k+k''}\,{}^{-q\,-q'}_{\beta\alpha}H^{kk''}_{QQ''}$, ${}^{q'q}_{\beta}H^{k''k}_{Q''Q} = (-)^{k+k''}\,{}^{qq'}_{\beta}H^{kk''}_{-Q-Q''}$ $= {}^{-q\,-q'}_{\beta}H^{kk''}_{QQ''}$.

[25]The following relation is used: ${}_\beta\rho^{k}_{-Q}(-p) = (-)^{Q}_{\beta}\rho^{k*}_{Q}(p)$ (it is deduced from (I.27) and from ${}_\beta\rho^{k}_{-Q} = (-)^{Q}_{\beta}\rho^{k*}_{Q}$).

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[30]Interaction with the laser field and interaction with laser photons must be carefully distinguished. If the atom is in one of the laser levels, one interaction with the field creates optical coherence. A second interaction implies a photon absorption (or a stimulated emission) and, at the same time, the atom undergoes a transition between the two laser levels.

[31]Relation $Q = 0$ is imposed by the selection rules, whatever the approximations are. The only magnetic-field dependence comes from the coupling between the different optical coherences [anomalous Zeeman effect, (I.20c]. In the BLA conditions ($\Gamma_{ab} \gtrsim \Delta\omega$ and $|\omega_a - \omega_b| \ll N\Delta\omega$), this coupling disappears and the equations do not depend on H. But in the contrary case ($\Gamma_{ab} \ll \Delta\omega$), a coupling appears for $|\omega_a - \omega_b| \simeq \Gamma_{ab}$ and the density matrix may depend on the magnetic field.

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[33]Let us point out that this property is due to the selection rules and is maintained even if BLA is not valid. Nevertheless BLA is necessary to get the homographic variations of ${}_{ab}\zeta^{1}_{0}$ with γ (Eq. I.73).

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[36]For high-J values (case of molecular pumping), ${}_b h^{22}_{22} \simeq 0.571(2J_b + 1)^{-1}$ if $J_a = J_b \pm 1$, or $0.857(2J_b + 1)^{-1}$, if $J_a = J_b$. The power-broadening ${}_b h^{22}_{22}\gamma$ remains finite because γ behaves like $|P_{ab}|^2 \propto (2J_b + 1)\gamma_{ba}$ (γ_{ba} is the b-a transition probability by spontaneous emission). Nevertheless, let us point out that, in molecule pumping, the BLA conditions are not always fulfilled (see Sec. VI).

[37]That is the case for the xenon laser lines. The coupling between the transverse alignments has been experimentally observed by M. Tsukakoshi and K. Shimoda (Ref. 2). However, in addition to the laser-induced coupling, a very strong coupling exists which is induced by the spontaneous emission from b to a (Ref. 12). [(See also M. Ducloy, C. R. Acad. Sci. B **268**, 1575 (1969)].

[38]To a certain extent, the present work arises from the experimental observation of this independence between the power broadening and the collision broadening of the Hanle effect [B. Decomps, thesis (Paris, 1969) (unpublished)].

[39]Let us point out that this property is valid at all orders in the electric field, if the relaxation processes do not induce any transitions between the Zeeman sublevels. On the other hand, the alignment of a may be coupled to that of b by spontaneous emission from b to a (Ref. 10).

[40]If $J_\beta \geqslant 2$, ${}_\beta\bar{\rho}^{4}_{4}$ can be created by the laser at higher orders in the electric field (Ref. 35). From (I.83), ${}_\beta\rho^{4}_{4}$ only presents modulations at the even frequencies, $2s\Delta\omega$. Is it possible to induce odd-frequency modulations, at $\Delta\omega$ for instance? This modulation should be resonant for $\omega_\beta = -\Delta\omega/4$. If $g_a \simeq g_b$ and $\Gamma_\beta(k) \ll \Delta\omega$, the transverse alignment modulations are resonant for $\omega_\beta = -s\,\Delta\omega/2$. When $\omega_\beta = -\Delta\omega/4$ there is no transverse alignment and subsequently no ρ^{4}_{4}. Nevertheless, if $\Gamma_\beta(k) \simeq \Delta\omega$, it will be possible to create modulations of ρ^{4}_{4} at the $\Delta\omega$ frequency. Another possibility is that $g_a = 2g_b$; $\omega_b = -\Delta\omega/4$ is equivalent to $\omega_a = -\Delta\omega/2$. For such a value of magnetic field (to the second order), the laser induces (into the a level), a transverse alignment modulated at the $\Delta\omega$ frequency, and, subsequently (to the fourth order), a ${}_b\rho^{4}_{4}$ component modulated at the same frequency.

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