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Optical Pumping with a Multimode Laser Beam: Saturation Resonances by Magnetic Mode Crossing

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The resonances of saturation which occur when the mode spacing is equal to the Zeeman splitting are theoretically studied for a mode spacing $\lesssim$ the natural width broadened by collisions. Two kinds of resonances occur. The first set, due to a population effect (crossing of holes), is not resolved and is unobservable. The second set, due to a Zeeman coherence effect, is well resolved since the widths of the resonances are of the order of the Hanle-effect width. These resonances are very sensitive to the relative phases of the modes.

When a gas (Ne in our experiments) excited by a discharge is placed in a magnetic field and is submitted to a resonant laser beam (He-Ne laser) linearly σ polarized, a resonance appears on the fluorescence lines emitted from one of the laser levels each time the Zeeman splitting is equal to the frequency difference between two modes. These resonances, first observed by Fork, Hargrove, and Pollack,[1] have been used to measure Landé g factors.[2] However, some aspects concerning their widths and the influence of mode locking on their amplitudes were not clearly understood. For that reason we have performed a semiclassical calculation, in the formalism of irreducible tensors, up to the fourth order of perturbation in the laser electric field (as previously done to second order to study the Hanle effect[3]).

In this Letter we summarize the results in the case $J_a=0$, $J_b=1$ (a, lower level; b, upper level). The details of the calculation and the discussion of the positions of resonances when both levels have a Zeeman structure will be published later.[4] The hypotheses are the following:

(a) As our detection does not resolve the spectral shape of the fluorescence lines, and furthermore as the detection is perpendicular to the laser beam, it is not necessary to take into account the frequency correlation between the laser and the fluorescent light.[5,6] The calculation of the atomic density matrix is sufficient to determine the fluorescence.

(b) The perturbation development is performed according to the usual methods with the rotating-wave and the Doppler-limit approximations.[7,8]

(c) We take into account spontaneous emission from the upper to the lower level.

(d) Although we must write the equations separately for each atomic velocity along the laser axis, we assume that the relaxation is sufficiently isotropic to produce no coupling between different tensorial orders[9] and to give rise to relaxation rates independent of Q (we write the irreducible tensor as T_Q^k; k is the tensorial order and Q the component).

(e) We take into account velocity changes due to collisions or trapping of fluorescence lines with the help of the strong-collision model.[8,10]

With the last two assumptions, the relaxation of the components on the ${}_\alpha T_Q^k$ basis of the density matrix for the atomic quantities (α stands for aa,

or bb and refers to the submatrices within each level) can be written[8,4]

$$[(d/dt)\ {}_{\alpha}\rho_{Q}^{k}(v)]_{\mathrm{relax}} = -\Gamma_{\alpha}'(k)\,{}_{\alpha}\rho_{Q}^{k}(v) + \gamma_{\alpha}'(k) W_{M}(v) \int {}_{\alpha}\rho_{Q}^{k}(v')\,dv', \tag{1}$$

where $W_M(v)$ is the Maxwellian velocity distribution; $\Gamma_\alpha'(k)$ is the relaxation rate taking into account radiative decay, destruction by collisions, and velocity changes; and $\gamma_\alpha'(k)$ is the probability of survival of the quantity ${}_{\alpha}T_{Q}^{k}$ after velocity changes by collisions or reabsorption of a photon. The usual relaxation rates, which can be measured by Hanle-effect experiments and which are insensitive to velocity changes, are given by

$$\Gamma_{\alpha}(k) = \Gamma_{\alpha}'(k) - \gamma_{\alpha}'(k). \tag{2}$$

Expressions like (1) can be written for optical quantities ${}_{ab}\rho_{Q}^{k}$ (off-diagonal submatrices). Since $J_a = 0$ and $J_b = 1$, the three components of the optical dipole, ${}_{ab}\rho_{Q}^{1}$, are only to be considered and we write $\Gamma_{ab}'(1) = \Gamma_{ab}'$. The imaginary part of Γ_{ab}' (pressure shift) is assumed to be included in the optical atomic frequency ω. Because of the Doppler-limit approximation, which is expressed by $\Gamma_{ab}' \ll \Delta\nu$ (Doppler width), the effect of velocity changes on optical quantities can be shown to be negligible.

The relaxation terms (1) are inserted in the Schrödinger equation which is solved by iteration.[3,8] As the fluorescence is detected with an analyzer for polarization π (or σ in high magnetic field), the observed saturation signal is a linear superposition of longitudinal quantities at fourth order, ${}_{\alpha}\rho_{0}^{k\,(4)}$. With a linearly σ-polarized laser, the orientation ${}_{b}\rho_{0}^{1}$ (aa and bb will be abreviated as a and b) vanishes so long as the Zeeman splitting ω_b is small compared to the Doppler width $\Delta\nu$. ${}_{a}\rho_{0}^{0}$, ${}_{b}\rho_{0}^{0}$, and ${}_{b}\rho_{0}^{2}$ are proportional to each other (in the special case $J_a = 0$, $J_b = 1$), and therefore the saturation signal does not depend on the fluorescence line studied. It contains two sets of unmodulated resonant terms of the form

$$A(p,s) = \sum_{\nu} \frac{L(\nu,p,s)}{\Gamma_b'(2) + i(p+2\omega_b)} \left\{ \frac{X(\delta_\nu - (p+s)/2)}{2\Gamma_{ab}' + i[p-s+2\omega_b]} + \frac{\gamma_b'(2)}{\Delta\nu} \frac{X(\delta_\nu - p/2)X(\delta_\nu - s - p/2)}{\Gamma_b(2) + i(p+2\omega_b)} \right\} + \text{c.c.}, \tag{3}$$

$$B(p,s) = \sum_{\nu} L(\nu,p,s) f(p) X(\delta_\nu - (p+s)/2)[2\Gamma_{ab}' + i(p-s+2\omega_b)]^{-1} + \text{c.c.} \tag{4}$$

ν, p, and s determine the four modes (denoted ν, μ, λ, and κ) according to Fig. 1; p and s are multiples of the mode spacing $\Delta\omega$; $X(\Omega) = \sqrt{\pi}\exp[(\Omega/\Delta\nu)^2]$ is the Doppler line shape; and $\delta_\nu = \omega_\nu - \omega$ (ω is the optical atomic frequency). $f(p)$ is given by

$$f(p) = (\Gamma_a' + ip)^{-1}\{1 - \gamma_{ba}[\Gamma_b'(0) + ip]^{-1}\} + \tfrac{1}{3}[\Gamma_b'(0) + ip]^{-1} - \tfrac{1}{2}[\Gamma_b'(1) + ip]^{-1} + \tfrac{1}{6}[\Gamma_b'(2) + ip]^{-1}, \tag{5}$$

where γ_{ba} is the probability of spontaneous emission for the laser line $b \to a$. The influence of the phase φ and the amplitude $\mathcal{E}$ of modes is contained in

$$L(\nu,p,s) = \mathcal{E}_\nu \mathcal{E}_\mu \mathcal{E}_\kappa \mathcal{E}_\lambda \cos(\varphi_\nu - \varphi_\mu + \varphi_\lambda - \varphi_\kappa). \tag{6}$$

Let us first discuss the terms $A(p,s)$. The resonant factor $[\Gamma_b'(2) + i(p+2\omega_b)]^{-1}$ expresses the ability of modes ν (σ^- component) and μ (σ^+ component) to produce, to second order of perturbation, transverse alignment ${}_{b}\rho_{2}^{2\,(2)}$ modulated at the frequency $p = \omega_\nu - \omega_\mu$; the resonance occurs when the beat frequency is equal to the Larmor frequency. We call this factor the "Zeeman factor." It is a manifestation of the Hanle effect ($p = 0$), and of the resonances of modulation[1] ($p \neq 0$). The factor $[2\Gamma_{ab}' + i(p-s+2\omega_b)]^{-1}$ is resonant when $p - s = \omega_\kappa - \omega_\mu = -2\omega_b$, i.e., when the modes (μ, σ^+) and (κ, σ^-) interact with atoms of the same velocity. This factor expresses the need of an *optical coincidence* (within a $2\Gamma_{ab}'$ width) between

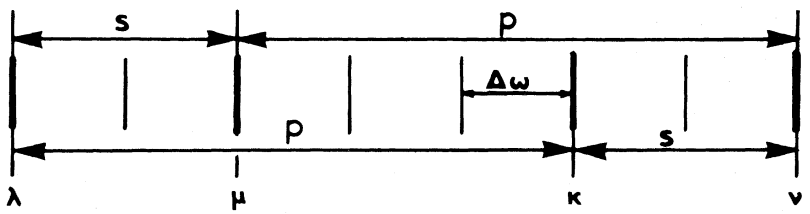

FIG. 1. Modes in the frequency scale, definition of the notation. The condition $\omega_\nu - \omega_\mu + \omega_\lambda - \omega_\kappa = 0$ insures the fourth-order terms to be unmodulated. Modes are assumed to be equidistant.

the first pair of modes (ν, μ) and the second one (κ, λ). The condition $\omega_\nu - \omega_\mu + \omega_\lambda - \omega_\kappa = 0$ (unmodulated saturation signal) imposes the frequency of the fourth mode λ and insures that the second pair has the exact beat frequency to demodulate ${}_b\rho_2{}^{2(2)}$. The X Doppler factor expresses the number of atoms with the correct velocity to interact with the four modes.

The last term of the last factor in $A(p, s)$ arises from atoms whose velocity has changed at second order before the interaction with the second pair of modes. The optical coincidence factor is replaced by a second Doppler X function which is the probability for atoms to get the correct velocity to interact with the second pair of modes (strong-collision model). The factor $[\Gamma_b(2) + i(p + 2\omega_b)]^{-1}$ is a part of the Zeeman factor.

Because of their origin we call resonances of the type $A(p, s)$, "Zeeman coherence effect" (ZCE) resonances (transverse alignment; coherence between Zeeman sublevels $\Delta m = 2$). On the other hand, $B(p, s)$-type resonances are due to a "population effect" (PE); as it is expressed by the factor $f(p)$, which takes the place of the Zeeman factor in $A(p, s)$, these terms arise from second-order longitudinal quantities, ${}_\alpha\rho_0{}^{k(2)}$, or in other words from the populations of the Zeeman sublevels. These terms are produced by beating of the σ^+ components of modes ν and μ (up to second order) followed by an interaction with modes (λ, σ^-) and (κ, σ^-). The optical coincidence between the two pairs is insured by the factor $[2\Gamma_{ab}' + i(p - s + 2\omega_b)]^{-1}$. For the most important term, $p = 0$ $(\nu = \mu; \lambda = \kappa)$, this factor produces a Lorentzian resonance which is interpreted as the crossing, by magnetic scanning, of the holes burned in the Doppler distribution by modes (μ, σ^+) and (κ, σ^-). Note that the contribution to the PE of atoms whose velocity has changed presents nonresonant behavior and has been omitted in (4).

Let us now focus our attention on $L(\nu, p, s)$. When modes are free running their phases are randomly distributed. Moreover, modes are in general not exactly equidistant and their relative phases can be regarded as slowly varying. Therefore, except for $s = 0$ $(\nu = \kappa; \mu = \lambda)$ or for $p = 0$ $(\nu = \mu; \kappa = \lambda)$, the quantities $L(\nu, p, s)$, $A(p, s)$, and $B(p, s)$ disappear when averaged (summed over ν and averaged over time). The remaining terms are those involving no more than two modes so that the phases cancel out in (6). On the other hand if modes are phase locked in such a manner that they are all in phase at regular time intervals (laser modulated in pulses), all relative phases are equal for a correct choice of the origin of time. Therefore the cosine term in (6) is equal to 1 in every case and all values of s and p are possible. Consider now two cases:

(1) *Large mode spacing.*—If $\Gamma_\alpha'(k), \Gamma_{ab}' \ll \Delta\omega$, $f(p)$ is negligible for $p \neq 0$ and the two factors in the first term of $A(p, s)$ must be simultaneously resonant $(s = 0)$. Furthermore the contribution of atoms whose velocity has changed is negligible.[11] Therefore the only important terms are $A(p, 0)$ and $B(0, s)$; they are not phase sensitive and produce well-resolved sets of resonances for $2\omega_b = r\Delta\omega$ (r is an integer). Resonances from $B(0, s)$ are Lorentzian curves of width $2\Gamma_{ab}'$ (twice the width of holes).

(2) *Mode spacing of the order of the hole width.*—The usual conditions in our experiments ($\Delta\omega \sim 80$ MHz, pressure of the order of a few Torr) are defined by

$$\Gamma_\alpha'(k) < \Gamma_{ab}' \sim \Delta\omega. \tag{7}$$

Resonances arising from the PE, $B(0, s)$, are no longer observable since they all overlap because of their broad width $2\Gamma_{ab}'$ [Fig. 2(a)]; terms sensitive to the phase of modes $(p \neq 0)$ remain small as long as $\Gamma_\alpha(k)$ is smaller than $\Delta\omega$.

In the expressions $A(p, s)$, from the ZCE, it is not possible to neglect terms arising from velocity diffusion or those which contain two denominators which are resonant in different magnetic fields. If modes are phase locked, all values of p and s are possible. It is easy to see that the quantity $\sum_s A(p, s)$ represents a narrow symmetric resonance centered at $2\omega_b = -p$. The magnitude of the resonance in zero magnetic field $(p = 0)$ and that of the lateral resonances $(p \neq 0)$ are of the same order; their width is of the order of $\Gamma_b'(2)$ so that they are well resolved [Fig. 2(b)]. When modes are free running, the central resonance $[\sum_s A(0, s)]$ is not affected as it involves only two modes $(\nu = \mu; \kappa = \lambda)$. On the contrary, resonances for $p \neq 0$ become much smaller as the only nonvanishing term is $A(p, 0)$ [Fig. 2(a)].

At the limit of very close and very numerous modes $[\Gamma_\alpha'(k) < \Delta\omega \ll \Delta\nu \lesssim N\Delta\omega$, where N is the number of modes], it is possible to show that resonances tend to be Lorentzian curves of width $\Gamma_b(2)$, that is, they have the same shape as in the Hanle effect (this is true for phase-locked modes and in any case for the central resonance). Of course for large laser intensity these curves are broadened as in the Hanle effect itself, but this cannot be shown with a calculation limited at the fourth order of perturbation.[13]

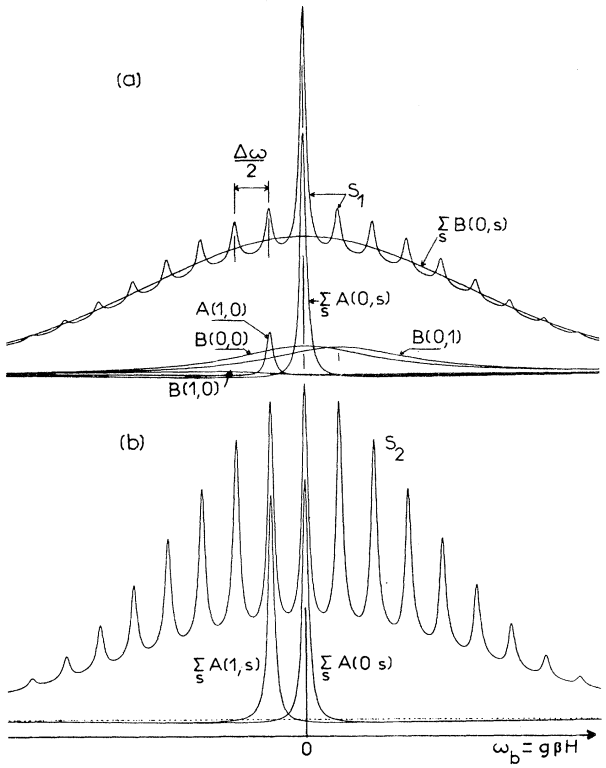

FIG. 2. Computer calculation of saturation resonances produced by eleven modes ($I_\nu = E_\nu{}^2 = 3$ for $-3 \leq \nu \leq 3$, $E_\nu{}^2 = 2$ for $\nu = \pm 4$, $E_\nu{}^2 = 1$ for $\nu = \pm 5$). The Doppler width is $\Delta\omega = 800$ MHz. The mode spacing $\Delta\omega = 80$ MHz and the relaxation rates are (in MHz) $\Gamma_{ab}' = 100$, $\Gamma_b'(0) = 9.6$, $\Gamma_b'(1) = 11$, $\Gamma_b'(2) = 11.5$, $\Gamma_b(2) = 7.5$, $\Gamma_a' = 15$. These conditions correspond approximatively to the laser line at 7305 Å with 1.5 Torr of Ne (see Ref. 12). (a) Modes free running. Resonances from ZCE and PE are compared. ZCE resonances are narrow and resolved; the resonance in zero magnetic field [$\sum_s A(0, s)$] is much higher than the others [$A(p, 0)$]. PE resonances [$B(0,0)$ and $B(1,0)$] are not resolved, as is shown by the curve $\sum_s B(s, 0)$. Off-resonance terms $B(p, 0)$ are negligible.

$$S_1 = \sum_{s \text{ or } p=0} [A(p, s) + B(p, s)]$$

is the resulting signal. (b) Modes phase locked. ZCE resonance for $p = 1$ has the same order of magnitude as that in zero magnetic field. $S_2 = \sum_{sp} [A(p, s) + B(p, s)]$ is the total signal.

When the cell is inside the laser cavity, the standing-wave aspect of the laser beam does not qualitatively change the results. The contrast between free-running– and phase-locked–mode resonances is increased approximately by a factor of 2. Resonances such as the Lamb dip do not appear as they overlap like other PE.

We point out the importance of the relative magnitude of the mode spacing and of the hole width. In the experimental cases [Eq. (7)], the resonances are due only to the ZCE's. The resonances in nonzero magnetic field are very sensitive to the phases of modes, as is illustrated by Fig. 2. This behavior is well verified experimentally (Fig. 3).

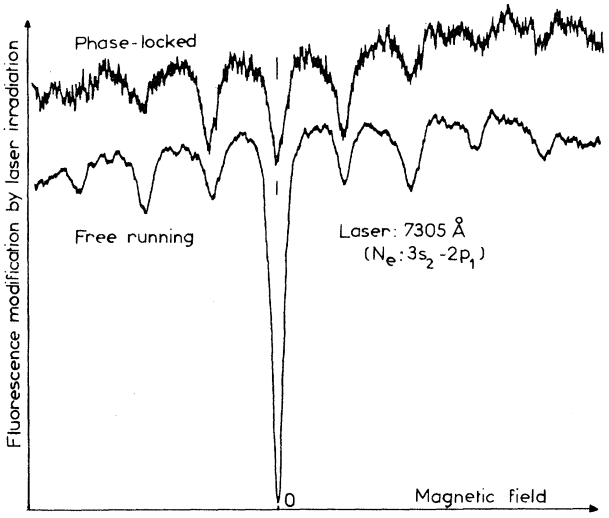

FIG. 3. Experimental records of the saturation signal on fluorescence lines (π polarization). To obtain a spontaneously phase-locked oscillation, we must strongly decrease the laser intensity. Therefore the intensity of the phase-locked signal is much less than that of the free-running signal. Nevertheless these curves clearly demonstrate the difference of behavior between the lateral resonances and the central one. The position of resonances provides a measurement of the Landé g factors (see Refs. 2 and 4). The width of resonance appears to be of the same order of magnitude as the Hanle-effect width. A quantitative study of the shape of resonances has been done by Ducloy (see Ref. 13).

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[11]Each term is of the order of $\Delta\nu^{-1}$ and the summation over s produces n terms with $n \lesssim \Delta\nu/\Delta\omega$. Therefore the sum is of the order of (or less than) $\Delta\omega^{-1}$, which is to be compared with the main term (no velocity changes) of the order of $\Gamma_{ab}'^{-1}$.

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Ultrasonic Impedometric Studies in Cholesteric Liquid Crystals*

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The real part of the shear mechanical impedance has been measured for nematic-cholesteric mixtures as a function of concentration. At low concentrations the results are found to be in good agreement with the theoretical expression derived by Brochard.

In this Letter we report on the first observation of pitch dependence of the real part of the shear mechanical impedance of a cholesteric liquid crystal. In a Cartesian frame $O(xyz)$, this impedance can be represented by a matrix Z as shown recently by Brochard.[1] When the ultrasonic shear-wave vector and the helicoidal axis are parallel to the Oz direction, the diagonal elements of Z are

$$Z_{xx} = iM\frac{k_1+k_2}{1+(i\omega - q_0^2M/\rho)(k_1k_2N/\rho)^{-1}}, \quad (1a)$$

$$Z_{yy} = iN\frac{k_1+k_2}{1+(i\omega - q_0^2N/\rho)(k_1k_2M/\rho)^{-1}}, \quad (1b)$$

where M and N are linear combinations of the Leslie-Ericksen viscosity coefficients α_i,[2,3]

$$N = \tfrac{1}{2}\alpha_4,$$

$$M = \tfrac{1}{2}(\alpha_3+\alpha_4+\alpha_6) - \tfrac{1}{2}\alpha_3(1+\gamma_2/\gamma_1).$$

ρ is the density, $p_0 = \Pi/q_0$ is the helicoidal pitch, and ω is the circular frequency of shear waves; k_1 and k_2 are wave vectors of the two transverse modes and are given by the roots of the dispersion equation:

$$k^4 - k^2\left(\frac{i\omega\rho}{N} + \frac{i\omega\rho}{M} + 2q_0^2\right) + \left(q_0^2 - \frac{i\omega\rho}{M}\right)\left(q_0^2 - \frac{i\omega\rho}{N}\right) = 0.$$

The case $q_0 = 0$ corresponds to the nematic phase for which the shear impedance has been calculated.[4] The real parts R_{xx} and R_{yy} of Z_{xx} and Z_{yy} can be determined from the reflection coefficient of transverse ultrasonic waves at an interface fused silica-liquid crystal sample.

Two geometries can be used: (a) The director at the interface can be parallel to the flow. The amplitude of the reflection coefficient is then

$$r = \frac{Z_s - R_{xx}}{Z_s + R_{xx}}.$$

(b) The director at the interface can be perpendicular to both flow and gradient. Then,

$$r = \frac{Z_s - R_{yy}}{Z_s + R_{yy}},$$

where Z_s is the shear mechanical impedance of the fused silica.

The behavior of Z depends critically on the ratio p_0/e, where e is the penetration depth of shear waves, given by $e = (\bar{\eta}/\omega\rho)^{1/2}$ ($\bar{\eta}$ being a viscosity coefficient). If

$$p_0/e \ll 1, \quad R_{xx} = R_{yy},$$

$$p_0/e \gg 1, \quad R_{xx} \neq R_{yy}.$$

We have measured R_{xx} and R_{yy} by using a reflectance technique described elsewhere,[4] for mixtures of parametoxybenzylidenebutylaniline (MBBA) and cholesteric propionate (CP), in a concentration range from 0 to 0.95. The orientation of the samples has been achieved by rubbing the surface of the silica bar. The accuracy on R_{xx} and R_{yy} is of the order of 5%. At very high concentration, the liquid-crystal mixture becomes waxy and needs to be applied from a solution in a fast evaporating solvent. In that case the error on the determination of R_{xx} and R_{yy} can be as high as 20%.

The pitch is known to vary with weight concen-