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Hanle resonances for a $J = 0$ to $J = 1$ transition excited by a fluctuating laser beam

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Abstract. The theoretical method developed in a previous paper is applied to the study of the sensitivity of Hanle resonances to the fluctuations of an intense quasi-monochromatic laser beam. The calculations are performed for the simplest possible transition $J = 0 \leftrightarrow J = 1$. Quantitative expressions are derived for the level-crossing signals which appear to be quite perturbed by the fast phase fluctuations.

1. Introduction

In a previous paper (Avan and Cohen-Tannoudji 1977, to be referred to as I), the behaviour of a two-level atom in an intense fluctuating resonant laser beam has been studied. Two conditions were assumed about the mean Rabi nutation frequency ω_1, the natural width Γ of the excited state and the spectral width $\Delta\nu$ of the laser:

$$\omega_1 \gg \Delta\nu \tag{1.1}$$

$$\Delta\nu \gtrsim \Gamma. \tag{1.2}$$

These two conditions imply that several Rabi nutations occur during the correlation time $1/\Delta\nu$ of the laser beam (very high intensity) and that atoms are sensitive to the laser fluctuations during their lifetime (non-monochromatic laser light). It follows that usual treatments, such as rate equations or Bloch equations, cannot be applied.

The method described in I starts from a description of the laser electric field $E(t)$ as a classical random variable. For a laser well above threshold, $E(t)$ can be written as

$$E(t) = \mathrm{Re}\,(E_0 + e(t)) \exp[-\mathrm{i}(\omega_0 t + \phi(t))] \tag{1.3}$$

where E_0 is the mean amplitude, ω_0 is the mean frequency, $e(t)$ and $\phi(t)$ are the amplitude and phase fluctuations. It can be shown (see I, §3) that the phase exhibits two types of fluctuation: (i) slow fluctuations characterized by a time τ_d

$$\tau_d = 1/\Delta\nu \tag{1.4}$$

and (ii) fast fluctuations characterized by a time $\tau_c \ll \tau_d$. The idea of the calculation presented in I is to apply two different approximations: the adiabatic approximation for slow fluctuations, and a perturbative treatment for fast fluctuations which appear as a relaxation mechanism.

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This method has been applied in I to the computation of the fluorescence spectrum emitted by a two-level atom. In the present paper we study, by the same method, the sensitivity to the laser fluctuations of the level-crossing resonances which appear on the total fluorescence light when the static magnetic field is scanned around the value corresponding to a crossing between two excited sublevels (Hanle and Franken resonances). The shape of these resonances has been studied extensively in the two extreme cases of a broad line excitation ($\Delta\nu \gg \omega_1$) and of a pure monochromatic excitation (Ducloy 1973, Avan and Cohen-Tannoudji 1975) but the situation corresponding to conditions (1.1) and (1.2) does not seem to have been investigated.

Such a problem is discussed in the present paper in the simplest possible case of a $J = 0$ to $J = 1$ transition which leads to a three-level system, as shown in § 2. The problem is reformulated in §3 in terms of a fictitious spin 1. The fast phase fluctuations appear for the fictitious spin as a relaxation process which is evaluated in § 4. The terms describing spontaneous emission are then included. Finally, expressions are derived for the level-crossing signals. The shape of these signals and their sensitivity to the laser fluctuations are discussed in § 5.

2. Notation; expression of the signals

The scheme of the level-crossing experiment is the following: an atomic beam directed along $0z$ is irradiated at right angles by a fluctuating laser beam propagating along $0y$, so that one gets rid of the Doppler effect. A static magnetic field $\boldsymbol{B}_0$ parallel to $0z$ is applied to the atoms (see figure 1).

We consider the particular case of atoms having a ground state g of angular momentum $J_g = 0$, and an excited state e of natural width Γ and angular momentum $J_e = 1$. The energy of the transition $g \leftrightarrow e$ in zero magnetic field is ω_0 (we take $\hbar = 1$) and Ω_e is the Larmor pulsation associated with $\boldsymbol{B}_0$ in the excited state e (see figure 2).

The light wave, of mean pulsation ω_0 in resonance in zero magnetic field with the $e \leftrightarrow g$ atomic transition, has a σ polarization (polarization vector $\boldsymbol{e}_x$ parallel to $0x$); it therefore couples the ground-state Zeeman sublevel $|0\rangle$ (more precisely $|g, m_g = 0\rangle$) to the two excited-state Zeeman sublevels $|\pm 1\rangle$ ($|e, m_e = \pm 1\rangle$). (The third sublevel

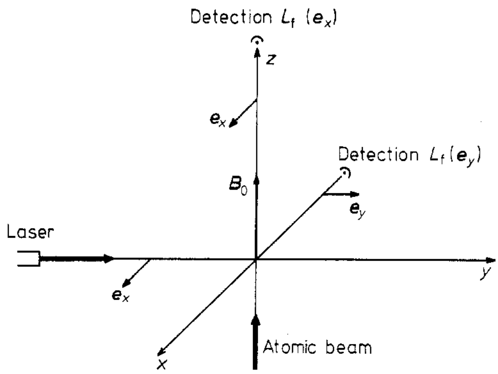

Figure 1. Scheme of the possible experiment: the atomic beam, directed along $0z$, is irradiated at right angles by the fluctuating laser beam propagating along $0y$ and linearly polarized (polarization vector $\boldsymbol{e}_x$ parallel to $0x$). A static magnetic field $\boldsymbol{B}_0$ is applied along $0z$. One monitors the fluorescence signals $L_f(\boldsymbol{e}_x)$ and $L_f(\boldsymbol{e}_y)$ with linear polarizations $\boldsymbol{e}_x$ and $\boldsymbol{e}_y$.

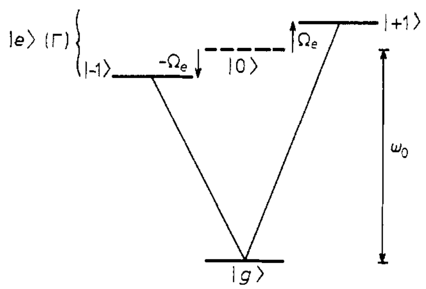

Figure 2. Atomic energy diagram. Γ is the natural width of e. The energy splitting in zero magnetic field between e and g is ω_0 and the Larmor pulsation in the excited state e is Ω_e. The $m = 0$ excited sublevel plays no role in the problem.

$|e, m_e = 0\rangle$ is not excited and can be forgotten in the following, so that we are led to the study of a three-level problem.)

The coupling with the electric field $E(t)$ written in (1.3) is described by the interaction Hamiltonian

$$V = -D_x E(t) \tag{2.1}$$

where $D_x = \boldsymbol{e}_x . \boldsymbol{D}$ is the component of the atomic dipole moment along the polarization vector $\boldsymbol{e}_x$. The relative phases of the two excited sublevels $|+1\rangle$ and $|-1\rangle$ with respect to the ground state $|0\rangle$ will be chosen in such a way that the two matrix elements $\langle 1|D_x|0\rangle$ and $\langle -1|D_x|0\rangle$ (which have the same modulus) are real and positive:

$$\langle +1|D_x|0\rangle = \langle -1|D_x|0\rangle = d \qquad \text{real} > 0. \tag{2.2}$$

We are interested here in the shape of level-crossing signals obtained when the magnetic field $\boldsymbol{B}_0$ is scanned around zero, and particularly in the variation with $\boldsymbol{B}_0$ of the fluorescence signals $L_f(e_x)$ and $L_f(e_y)$ detected in the respective directions 0z and 0x, with the respective polarizations $\boldsymbol{e}_x$ and $\boldsymbol{e}_y$ parallel to 0x and 0y.

These detection signals can easily be related to the elements of the atomic density matrix in the upper state (populations σ_{++} and σ_{--} of $|+\rangle$ and $|-\rangle$, Zeeman coherence σ_{+-} between $|+\rangle$ and $|-\rangle$):

$$L_f(e_{\substack{x\\y}}) = \sum_{\epsilon,\epsilon'} \langle 0|D_{\substack{x\\y}}|\epsilon\rangle\langle\epsilon|\sigma(t)|\epsilon'\rangle\langle\epsilon'|D_{\substack{x\\y}}|0\rangle \qquad \epsilon,\epsilon' = +1, -1. \tag{2.3}$$

With the phase conventions (2.2) (which also fix the matrix elements of D) one immediately gets

$$L_f(e_x) \propto \sigma_{++} + \sigma_{--} + 2\,\mathrm{Re}\,\sigma_{+-} \tag{2.4}$$

$$L_f(e_y) \propto \sigma_{++} + \sigma_{--} - 2\,\mathrm{Re}\,\sigma_{+-} \tag{2.5}$$

$$I_{\parallel} - I_{\perp} = L_f(e_x) - L_f(e_y) \propto 2\,\mathrm{Re}\,\sigma_{+-}. \tag{2.6}$$

As the transit time T through the exciting laser beam is generally long compared to all other characteristic evolution times, it will be possible in expressions (2.4), (2.5) and (2.6) to replace σ_{++}, σ_{--} and σ_{+-} by their steady-state values.

3. Theoretical method

3.1. Formulation of the problem in terms of a fictitious spin 1

The evolution of the atomic density matrix σ is given by (for the moment we neglect spontaneous emission)

$$\mathrm{i}\,\frac{\mathrm{d}\sigma}{\mathrm{d}t} = [H, \sigma] \tag{3.1}$$

where H is represented in the basis $\{|+1\rangle, |0\rangle, |-1\rangle\}$ by the following matrix (in the rotating-wave approximation):

$$H = \begin{pmatrix} \omega_0 + \Omega_e & \alpha \exp(-\mathrm{i}\omega_0 t) & 0 \\ \alpha^* \exp(\mathrm{i}\omega_0 t) & 0 & \alpha^* \exp(\mathrm{i}\omega_0 t) \\ 0 & \alpha \exp(-\mathrm{i}\omega_0 t) & \omega_0 - \Omega_e \end{pmatrix} \tag{3.2}$$

where

$$\alpha = \tfrac{1}{2} d(E_0 + e(t)) \exp[-\mathrm{i}\phi(t)] \tag{3.3}$$

d being defined in equation (2.2).

If to equation (3.1) we apply the unitary transformation

$$U = \begin{pmatrix} \exp[\mathrm{i}(\omega_0 t + \phi(t))] & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & \exp[\mathrm{i}(\omega_0 t + \phi(t))] \end{pmatrix} \tag{3.4}$$

the evolution of the transformed density matrix

$$\sigma'' = U \sigma U^+ \tag{3.5}$$

becomes

$$\mathrm{i}\,\frac{\mathrm{d}\sigma''}{\mathrm{d}t} = [H_c + H_f, \sigma''] \tag{3.6}$$

where

$$H_c = \Omega_e \begin{pmatrix} 1 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & -1 \end{pmatrix} + \frac{dE_0}{2} \begin{pmatrix} 0 & 1 & 0 \\ 1 & 0 & 1 \\ 0 & 1 & 0 \end{pmatrix} \tag{3.7}$$

and

$$H_f = -\dot{\phi}(t) \begin{pmatrix} 1 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 1 \end{pmatrix} + \frac{de(t)}{2} \begin{pmatrix} 0 & 1 & 0 \\ 1 & 0 & 1 \\ 0 & 1 & 0 \end{pmatrix}. \tag{3.8}$$

In analogy with the two-level problem studied in I, we call the new representation 'instantaneous representation' Σ''. In Σ'' the Hamiltonian is split into two parts:

(i) a time-independent term H_c, and

(ii) a fluctuating term H_f depending on the laser fluctuations.

The evolution of the three-level atom in Σ'' can be described in terms of a fictitious spin 1 ($\boldsymbol{S}$) having three levels $|+1\rangle$, $|0\rangle$ and $|-1\rangle$ corresponding to the respective atomic sublevels $|+1\rangle$, $|0\rangle$ and $|-1\rangle$. The Hamiltonians H_c and H_f can be written as

$$H_c = \Omega_e S_z + \omega_1 S_x \tag{3.9}$$

$$H_f = -\dot{\phi}(t) S_z^2 + \sqrt{\tfrac{1}{2}}\, de(t) S_x \tag{3.10}$$

with

$$\omega_1 = dE_0/\sqrt{2}. \tag{3.11}$$

The evolution of the three-level atom in Σ'' is therefore equivalent (see figure 3) to the precession of the fictitious spin 1 subjected to the following fields:

(i) two static fields; a longitudinal magnetic field $\boldsymbol{b}_0$ with an amplitude $-\Omega_e/\gamma$ and a transverse static magnetic field $\boldsymbol{b}_1$ parallel to 0x with an amplitude $-\omega_1/\gamma$, and

(ii) two small fluctuating fields; a magnetic field $\boldsymbol{b}_{\parallel}(t)$ parallel to $\boldsymbol{b}_1$ with an amplitude $-de(t)/\gamma\sqrt{2}$ and an electric field $\boldsymbol{\epsilon}(t)$ parallel to $\boldsymbol{b}_0$ and proportional to $-\dot{\phi}(t)$.

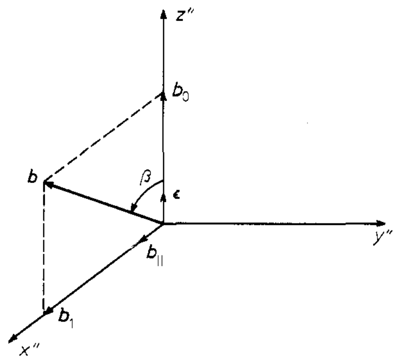

Figure 3. Various fields 'seen' by the fictitious spin 1 in the instantaneous representation Σ''. $\boldsymbol{b}_0$ and $\boldsymbol{b}_1$ are static magnetic fields with respective amplitudes $-\Omega_e/\gamma$ and $-\omega_1/\gamma$. $\boldsymbol{\epsilon}$ is a fluctuating electric field collinear to $\boldsymbol{b}_0$, proportional to $-\dot{\phi}(t)$; $\boldsymbol{b}_{\parallel}$ is a fluctuating magnetic field collinear to $\boldsymbol{b}_1$, proportional to $e(t)$.

3.2. Evolution of the fictitious spin 1

The problem is now to determine the evolution of the fictitious spin 1 under the effect of these various fields. We will follow the method introduced in I. In the representation Σ'', the fields $\boldsymbol{\epsilon}$ and $\boldsymbol{b}_{\parallel}$ associated with fast fluctuations (we neglect the contribution of slow phase diffusion to $\boldsymbol{\epsilon}$) act on $\boldsymbol{S}$ as a relaxation process satisfying the motional-narrowing condition (see I, §3.3). To simplify, from now on we will suppose, as in I, that the laser field is very well stabilized in amplitude so that the effect of $\boldsymbol{b}_{\parallel}$ is negligible. The Hamiltonian in Σ'' therefore becomes

$$H_c + H_f = \Omega_e S_z + \omega_1 S_x - \dot{\phi}(t) S_z^2. \tag{3.12}$$

When coming back to the Schrödinger representation Σ, some phase factors depending on the phase diffusion $\phi(t) - \phi(0)$ can appear but we have shown in I that they are mainly sensitive to slow phase fluctuations and that they can be averaged independently provided that the condition

$$\omega_1 \gg \Delta\nu \tag{3.13}$$

is fulfilled (see I, § 2). Actually it is easy to show, from (3.4) and (3.5) that the three matrix elements of σ, namely σ_{++}, σ_{--} and σ_{+-}, appearing in the fluorescence signals, are insensitive to the phase diffusion since they have the same expression in the two representations Σ'' and Σ:

$$\sigma''_{++} = \sigma_{++} \qquad \sigma''_{--} = \sigma_{--} \qquad \sigma''_{+-} = \sigma_{+-}. \tag{3.14}$$

The signals studied in this paper are therefore insensitive to phase diffusion. Note the difference from the signal studied in I which was related to the correlation function of the atomic dipole moment. Here the total fluorescence light is a quadratic function of the dipole moment components evaluated at the same time (see expression (2.3)).

4. Relaxation of the fictitious spin

4.1. Diagonalization of the non-fluctuating Hamiltonian

It will be convenient first to diagonalize the non-fluctuating Hamiltonian

$$H_c = \Omega_e S_z + \omega_1 S_x \tag{4.1}$$

corresponding to the two magnetic fields $\boldsymbol{b}_0$ and $\boldsymbol{b}_1$ of figure 3. This is easily done by performing a rotation around $0y$ which brings the $0z$ axis along the direction of the total field $\boldsymbol{b} = \boldsymbol{b}_0 + \boldsymbol{b}_1$. Let β be the angle between $0z$ and $\boldsymbol{b}$ (see figure 3). This rotation $R(\beta)$ is given by

$$R(\beta) = \exp(\mathrm{i}\beta S_y) \tag{4.2}$$

and is such that

$$\tilde{H}_c = R(\beta) H_c R(-\beta) = \Omega_1 S_z \tag{4.3}$$

where

$$\Omega_1 = -\gamma b = (\Omega_e^2 + \omega_1^2)^{1/2}. \tag{4.4}$$

The expression $\tilde{H}_f$ of H_f in the new frame $\tilde{\Sigma}''$ is easily calculated by first expanding H_f in a set of irreducible tensor operators $\{T_q^{(k)}\}$ (Messiah 1963, Omont 1976). As S_z^2 is a linear combination of $T_0^{(2)}$ and $T_0^{(0)}$ (see appendix 1 for the expression of $T_q^{(k)}$ in terms of S_z, $S_\pm$) then

$$H_f = -\dot{\phi} S_z^2 = -\dot{\phi}\left(\frac{2}{\sqrt{3}}\, T_0^{(0)} + \sqrt{\frac{2}{3}}\, T_0^{(2)}\right). \tag{4.5}$$

The transformation by rotation of $T_q^{(k)}$ is easily calculated in terms of the rotation matrices $R^{(k)}$ so that

$$\tilde{H}_{\mathrm{f}} = R(\beta)H_{\mathrm{f}}R(-\beta) = -\dot{\phi}\left(\frac{2}{\sqrt{3}}\,T_0^{(0)} + \sqrt{\frac{2}{3}}\sum_q R_{q0}^{(2)}(\beta)T_q^{(2)}\right). \tag{4.6}$$

As S_z is proportional to $T_0^{(1)}$, (4.3) may also be written as

$$\tilde{H}_{\mathrm{c}} = \sqrt{2}\,\Omega_1 T_0^{(1)}. \tag{4.7}$$

Let us finally introduce the expansion of σ'' in the $\{T_q^{(k)}\}$ basis

$$\sigma'' = \sum_{\substack{k=0,1,2\\ q}} c_q^{(k)} T_q^{(k)} \tag{4.8}$$

and the corresponding expansion of

$$\tilde{\sigma}'' = R(\beta)\sigma'' R(-\beta) \tag{4.9}$$

which is given by

$$\tilde{\sigma}'' = \sum_{k,q} \tilde{c}_q^k T_q^k \tag{4.10}$$

with

$$\tilde{c}_q^k = \sum_{q'} R_{qq'}^k(\beta) c_{q'}^k. \tag{4.11}$$

In the absence of fluctuations, the evolution of $\tilde{c}_q^k$ in Σ'' is very simple and is given by

$$\dot{\tilde{c}}_q^k = -\mathrm{i}q\Omega_1 \tilde{c}_q^k. \tag{4.12}$$

The $q = 0$ components are static. The $q \neq 0$ ones oscillate at a very high frequency $q\Omega_1$.

4.2. *Effect of the fluctuating fields*

The relaxation induced by the fluctuating field $\boldsymbol{\epsilon}(t)$ introduces a coupling between the various components of $\tilde{\sigma}$:

$$\dot{\tilde{c}}_q^k = -\mathrm{i}q\Omega_1 \tilde{c}_q^k + \sum_{k'q'} r_{qq'}^{kk'} c_{q'}^{k'}. \tag{4.13}$$

As shown in the following, the coupling coefficients $r_{qq'}^{kk'}$ are of the order of $\Delta\nu$, and consequently (see (3.13)) much smaller than the free evolution coefficients $-\mathrm{i}q\Omega_1$. It is therefore a good approximation to neglect the coupling between coefficients evolving at different frequencies (secular approximation), and (4.13) can be replaced by

$$\dot{\tilde{c}}_q^k = -\mathrm{i}q\Omega_1 \tilde{c}_q^k + \sum_{k'} r_q^{kk'} \tilde{c}_q^{k'}. \tag{4.14}$$

Furthermore, it will be shown later (when the effect of spontaneous emission will be added, introducing some damping and source terms in equation (4.14)) that the steady-state value of $\tilde{c}_0^k$ is larger than the steady value of $\tilde{c}_q^k$ by a factor

$$\frac{q\Omega_1}{r} \sim \frac{q\Omega_1}{\Delta\nu} \gg 1.$$

It is therefore sufficient in the computation of the steady-state value of the detection signals (2.4), (2.5) and (2.6) to keep only the contribution of $\tilde{c}_0^k$.

$\tilde{c}_0^0$ is actually a constant since it is proportional to the total population of the energy sublevels:

$$\tilde{c}_0^0 = 1/\sqrt{3}. \tag{4.15}$$

It follows that is sufficient to study the relaxation of $\tilde{c}_0^1$ and $\tilde{c}_0^2$.

The relaxation equations of $\tilde{c}_0^1$ and $\tilde{c}_0^2$ under the effect of $\tilde{H}_{\mathrm{f}}$ are derived in appendix 2; one finds

$$r_0^{1k'} = -\delta_{k'1}\left(\tfrac{1}{2}\sin^2\beta\cos^2\beta\,\frac{\kappa^2}{\kappa^2+\Omega_1^2} + \tfrac{1}{2}\sin^4\beta\,\frac{\kappa^2}{\kappa^2+4\Omega_1^2}\right)\Delta\nu \tag{4.16}$$

$$r_0^{2k'} = -\delta_{k'2}\tfrac{3}{2}\sin^2\beta\cos^2\beta\,\frac{\kappa^2}{\kappa^2+\Omega_1^2}\Delta\nu \tag{4.17}$$

where κ is the inverse of the correlation time τ_{c} of fast phase fluctuations.

It appears clearly in (4.16) and (4.17) that the order of magnitude of the r coefficients is $\Delta\nu$.

4.3. *Effect of spontaneous emission*

As in I, spontaneous emission is described by adding to the master equation some new terms which have a very simple form when written in Σ'' in the atomic sublevel basis:

$$\begin{aligned}
&\frac{\mathrm{d}}{\mathrm{d}t}\sigma''_{++} = -\Gamma\sigma''_{++} && \frac{\mathrm{d}}{\mathrm{d}t}\sigma''_{--} = -\Gamma\sigma''_{--} \\
&\frac{\mathrm{d}}{\mathrm{d}t}\sigma''_{+-} = -\Gamma\sigma''_{+-} && \frac{\mathrm{d}}{\mathrm{d}t}\sigma''_{0\pm} = -\tfrac{1}{2}\Gamma\sigma''_0 \\
&\frac{\mathrm{d}}{\mathrm{d}t}\sigma''_{00} = \Gamma(\sigma''_{++}+\sigma''_{--}).
\end{aligned} \tag{4.18}$$

The first three expressions describe the damping of the excited state e (Γ is the natural width of this state); the fourth expression describes the damping of optical coherences (with a rate $\Gamma/2$); the last expression describes the transfer of atoms from the upper to the lower state.

It is easy to express each of the matrix elements appearing in (4.18) in terms of the coefficients c_q^k (see appendix 1, (A.6)). Equations (4.18) can easily be transformed into

$$\left(\frac{\mathrm{d}}{\mathrm{d}t}c_q^1\right)_{\text{sp em}} = \sum_{q'}\Lambda_{qq'}^{(1)}c_{q'}^1 \tag{4.19}$$

$$\left(\frac{\mathrm{d}}{\mathrm{d}t}c_q^2\right)_{\text{sp em}} = \sum_{q'}\Lambda_{qq'}^{(2)}c_{q'}^2 + \lambda_0\delta_{q0} \tag{4.20}$$

where $\Lambda^{(1)}$ and $\Lambda^{(2)}$ are the two following diagonal matrices

$$\Lambda^{(1)} = \begin{pmatrix} -\Gamma/2 & 0 & 0 \\ 0 & -\Gamma & 0 \\ 0 & 0 & -\Gamma/2 \end{pmatrix} \qquad \Lambda^{(2)} = \begin{pmatrix} -\Gamma & 0 & 0 & 0 & 0 \\ 0 & -\Gamma/2 & 0 & 0 & 0 \\ 0 & 0 & -\Gamma & 0 & 0 \\ 0 & 0 & 0 & -\Gamma/2 & 0 \\ 0 & 0 & 0 & 0 & -\Gamma \end{pmatrix} \tag{4.21}$$

and where λ_0 is a source term given by

$$\lambda_0 = -\Gamma\sqrt{\tfrac{2}{3}}. \tag{4.22}$$

This source term has a very simple physical meaning: spontaneous emission tends to accumulate atoms in the $|0\rangle$ ground state, creating a longitudinal alignment of the equivalent fictitious spin 1.

It remains now to transform equations (4.19) and (4.20) into the $\tilde{\Sigma}''$ representation. Using (4.11), one immediately gets

$$\left(\frac{\mathrm{d}}{\mathrm{d}t}\tilde{c}_q^k\right)_{\text{sp em}} = \sum_{q'} \tilde{\Lambda}_{qq'}^{(k)}\tilde{c}_{q'}^k + R_{q0}^{(k)}(\beta)\lambda_0\,\delta_{k2} \tag{4.23}$$

with

$$\tilde{\Lambda}^{(k)} = R^{(k)}(\beta)\Lambda^{(k)}R^{(k)}(-\beta). \tag{4.24}$$

Since Γ is, as $\Delta\nu$, small compared to ω_1, and therefore to Ω_1, it is justified to neglect the non-secular terms (coupling by spontaneous emission between coefficients $\tilde{c}_q^k$ evolving at different frequencies).

To summarize, spontaneous emission is included just by adding to the relaxation equations of $\tilde{c}_0^1$ and $\tilde{c}_0^2$ the secular terms $\tilde{\Lambda}_{00}^{(2)}(\beta)$ and $\tilde{\Lambda}_{00}^{(1)}(\beta)$, the source term of equation (4.23) appearing only in the equation giving $\dot{\tilde{c}}_0^{(2)}$:

$$\dot{\tilde{c}}_0^1 = (r_0^{11} + \tilde{\Lambda}_{00}^{(1)})\tilde{c}_0^1 \tag{4.25}$$

$$\dot{\tilde{c}}_0^2 = (r_0^{22} + \tilde{\Lambda}_{00}^{(2)})\tilde{c}_0^2 + R_{00}^{(2)}(\beta)\lambda_0. \tag{4.26}$$

The two terms $\tilde{\Lambda}_{00}^{(1)}$ and $\tilde{\Lambda}_{00}^{(2)}$ are readily evaluated from (4.21), (4.24) and (A.8); one gets

$$\tilde{\Lambda}_{00}^{(1)} = -\tfrac{1}{2}(1 + \cos^2\beta)\Gamma \tag{4.27}$$

$$\tilde{\Lambda}_{00}^{(2)} = -\tfrac{1}{4}[(3\cos^2\beta - 1)^2 + 6\cos^2\beta\sin^2\beta + 3\sin^4\beta]\Gamma. \tag{4.28}$$

By equating to zero the rates of variation of $\tilde{c}_0^1$ and $\tilde{c}_0^2$ in equations (4.25) and (4.26), one immediately gets their steady-state values $\overline{\tilde{c}_0^1}$ and $\overline{\tilde{c}_0^2}$:

$$\overline{\tilde{c}_0^1} = 0 \tag{4.29}$$

$$\overline{\tilde{c}_0^2} = \frac{-R_{00}^{(2)}(\beta)\lambda_0}{r_0^{22} + \tilde{\Lambda}_{00}^{(2)}}. \tag{4.30}$$

The steady-state value of the atomic density matrix in the representation $\tilde{\Sigma}''$ is therefore given by

$$\overline{\tilde{\sigma}''} = \frac{1}{\sqrt{3}}\,T_0^{(0)} - \frac{R_{00}^{(2)}(\beta)\lambda_0}{r_0^{22} + \tilde{\Lambda}_{00}^{(2)}}\,T_0^{(2)} \tag{4.31}$$

i.e. coming back to the Σ'' representation

$$\overline{\sigma''} = \frac{1}{\sqrt{3}} T_0^{(0)} - \frac{R_{00}^{(2)}(\beta)\lambda_0}{r_0^{22} + \tilde{\Lambda}_{00}^{(2)}} \sum_q R_{q0}^{(2)}(-\beta) T_q^{(2)}. \tag{4.32}$$

5. Shape of level-crossing signals

5.1. Calculation of the signals

The total population $\sigma''_{++} + \sigma''_{--}$ of the two sublevels $|\pm 1\rangle$ and the Zeeman coherence σ''_{+-} between them which appear in the detection signals can first be expressed in terms of the c_q^k components of σ'' by using relations (A.6) given in appendix 1:

$$\begin{aligned} \sigma''_{++} + \sigma''_{--} &= \tfrac{2}{3} + \sqrt{\tfrac{2}{3}}\, c_0^2 \\ 2\,\mathrm{Re}\,\sigma''_{+-} &= c_2^2 + c_{-2}^2. \end{aligned} \tag{5.1}$$

From (3.14), the steady-state values of $\sigma_{++} + \sigma_{--}$ and σ_{+-} are the same in Σ and Σ'' and can therefore be evaluated from (4.32). The expressions of $R_{00}^{(2)}(\beta)$ and $R_{20}^{(2)}(\beta)$ are given in appendix 1 (equations (A.8)). Putting into (4.32) the values (4.22), (4.17) and (4.28) of λ_0, r_0^{22} and $\tilde{\Lambda}_{00}^2$, one easily calculates the steady-state values of c_0^2, c_2^2 and c_{-2}^2 which can be inserted in (5.1) to give

$$\begin{aligned} \overline{\sigma_{++}} + \overline{\sigma_{--}} &= \frac{2}{3} + \frac{1}{3}\,\frac{(3\cos^2\beta - 1)^2}{-3\cos^4\beta + 3\cos^2\beta - 2 - 3\sin^2\beta\cos^2\beta\,(1/\Gamma T_1)} \\ 2\,\mathrm{Re}\,\sigma_{+-} &= \frac{(3\cos^2\beta - 1)\sin^2\beta}{-3\cos^4\beta + 3\cos^2\beta - 2 - 3\sin^2\beta\cos^2\beta\,(1/\Gamma T_1)} \end{aligned} \tag{5.2}$$

where

$$\frac{1}{T_1} = \frac{\kappa^2\,\Delta\nu}{\kappa^2 + \Omega_1^2}. \tag{5.3}$$

The level-crossing signals $L_f(e_x)$, $L_f(e_y)$ and $I_{\parallel} - I_{\perp}$ given by equations (2.4), (2.5) and (2.6) follow immediately from (5.2):

$$L_f(e_x) \propto \tfrac{2}{3} + \tfrac{2}{3}(3\cos^2\beta - 1)/D(\beta) \tag{5.4}$$

$$L_f(e_y) \propto \tfrac{2}{3} + \tfrac{2}{3}(3\cos^2\beta - 1)(3\cos^2\beta - 2)/D(\beta) \tag{5.5}$$

$$I_{\parallel} - I_{\perp} \propto (3\cos^2\beta - 1)\sin^2\beta/D(\beta) \tag{5.6}$$

with

$$D(\beta) = -3\cos^4\beta + 3\cos^2\beta - 2 - 3\sin^2\beta\cos^2\beta/\Gamma T_1 \tag{5.7}$$

and, of course (see figure 3)

$$\sin^2\beta = \frac{\omega_1^2}{\Omega_e^2 + \omega_1^2} \qquad \text{and} \qquad \cos^2\beta = \frac{\Omega_e^2}{\Omega_e^2 + \omega_1^2}. \tag{5.8}$$

5.2. Physical discussion

The angle β appearing in (5.4), (5.5) and (5.6) depends on Ω_e (Larmor precession proportional to the static magnetic field) and on ω_1^2 (proportional to the mean laser

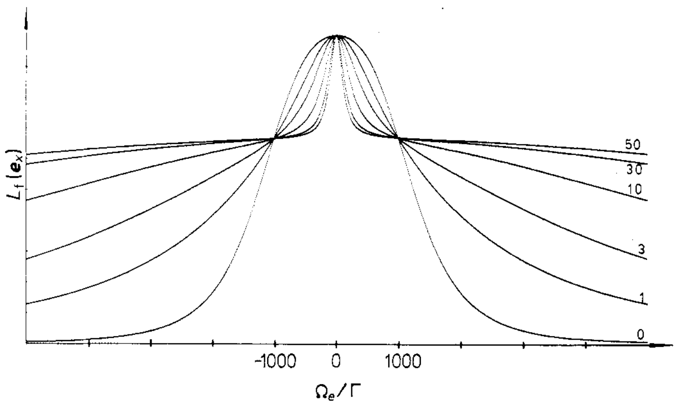

Figure 4. Set of level-crossing resonances detected on $L_f(e_x)$, for a fixed value of the laser intensity ($\omega_1^2/2\Gamma^2 = 10^6$). Each curve corresponds to a different value of the parameter $\Delta\nu/\Gamma$ indicated on the figure. (Ω_e/Γ is a normalized Larmor frequency; $\kappa/\Gamma = 100$.)

intensity). All the effects of the laser fluctuations are contained in the parameter $1/T_1$ which is proportional to the spectral width $\Delta\nu$.

One first checks that for a non-fluctuating laser beam ($1/T_1 = \Delta\nu = 0$), expressions (5.4), (5.5) and (5.6) reduce to the ones obtained in the case of a pure monochromatic laser beam (in the limit $\omega_1 \gg \Gamma$).

We have represented in figures 4, 5 and 6 the shape of level-crossing signals, i.e. the variation with Ω_e of $L_f(e_x)$, $L_f(e_y)$ and $(I_{\parallel} - I_{\perp})$, for a given value of the parameter ω_1^2, i.e. of the mean laser intensity, and for increasing values of the parameter $\Delta\nu/\Gamma$ characterizing the spectral width of the fluctuating laser light (note that $\Delta\nu$ remains always smaller than ω_1 so that the validity condition (3.13) is always fulfilled). The parameter κ (inverse of the correlation time τ_c of fast phase fluctuations) appearing in the expression (5.3) for $1/T_1$ has been chosen as equal to 100Γ.

One first notices that the various curves of each set have several common points:

(i) They all have the same values for $\Omega_e = 0$ and $\Omega_e = \infty$, i.e. according to (5.8), for $\beta = \pi/2$ ($\cos\beta = 0$) and $\beta = 0$ or π ($\sin\beta = 0$). Mathematically, the coefficient of $1/\Gamma T_1$ in $D(\beta)$—$3\cos^2\beta\sin^2\beta$—vanishes when $\beta = 0$, $\pi/2$, π. The physical interpretation of this result is that the fluctuating electric field $\boldsymbol{\epsilon}(t)$ cannot, for these three

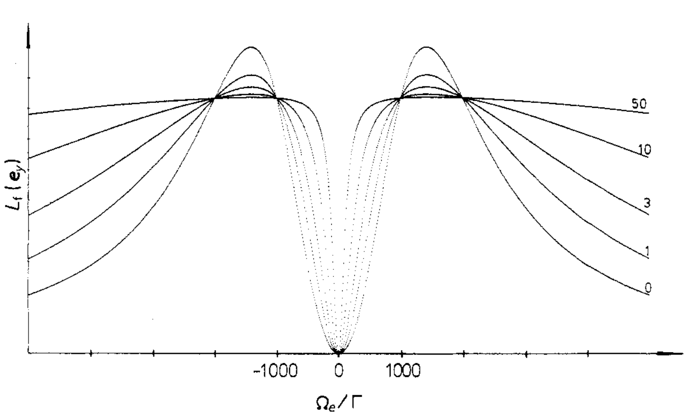

Figure 5. Set of level-crossing resonances detected on $L_f(e_y)$, for a fixed value of the laser intensity ($\omega_1^2/2\Gamma^2 = 10^6$). Each curve corresponds to a different value of the parameter $\Delta\nu/\Gamma$ indicated on the figure.

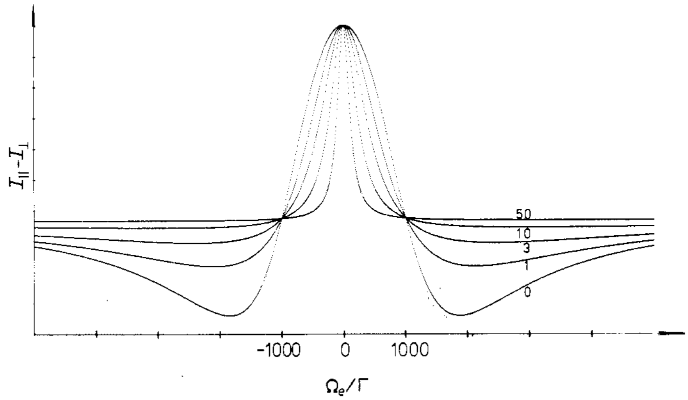

Figure 6. Set of curves giving the difference $I_{\|} - I_{\perp}$ between the two signals $L_f(e_x)$ and $L_f(e_y)$ represented in figures 4 and 5.

values of β, destroy the alignment of the fictitious spin along the total field $\boldsymbol{b} = \boldsymbol{b}_0 + \boldsymbol{b}_1$, so that the level-crossing signals are insensitive to the laser fluctuations. Let us call $|-\tilde{1}\rangle$, $|\tilde{0}\rangle$ and $|+\tilde{1}\rangle$ the energy levels in Σ'', which are the eigenstates of the components of $\boldsymbol{S}$ along $\boldsymbol{b}$. When $\beta = 0$ or π, $\boldsymbol{\epsilon}$ and $\boldsymbol{b}$ are parallel or antiparallel and the perturbation $-\dot{\phi}S_z^2$ associated with $\boldsymbol{\epsilon}$ cannot induce any transition between the $|\tilde{m}\rangle$ states. When $\beta = \pi/2$, $\boldsymbol{\epsilon}$ and $\boldsymbol{b}$ are perpendicular and the perturbation $-\dot{\phi}S_z^2$ connects only the two sublevels $|-\tilde{1}\rangle$ and $|+\tilde{1}\rangle$ without changing their total population $\tilde{\sigma}_{++} + \tilde{\sigma}_{--}$ and consequently the alignment along $\boldsymbol{b}$.

(ii) Two other common points symmetric with respect to 0 correspond to values of β such that

$$3\cos^2\beta - 1 = 0. \tag{5.9}$$

This is obvious from (5.4), (5.5) and (5.6) since in that case $L_f(e_x) = L_f(e_y) = \frac{2}{3}$, $I_{\|} - I_{\perp} = 0$ whatever $\Delta\nu$, the corresponding values of the matrix elements of σ being

$$\sigma_{++} = \sigma_{--} = \tfrac{1}{3} \qquad \sigma_{+-} = \sigma_{-+} = 0. \tag{5.10}$$

Such a result can be interpreted in the following way. In Σ'' the source term for the alignment along $\boldsymbol{b}$ is proportional to $R^{(2)}_{00}(\beta)$ (see equation (4.32)), i.e. to $3\cos^2\beta - 1$. For the values of β corresponding to the zeros of $3\cos^2\beta - 1$, the fictitious spin is not prepared with a longitudinal alignment in Σ''. The steady-state value of its density matrix is isotropic and not affected by the relaxation produced by $\boldsymbol{\epsilon}$. This explains the steady-state values (5.10) corresponding to a completely unpolarized spin.

(iii) Finally, two more common points symmetric with respect to 0 appear in the curves giving $L_f(e_y)$ (see figure 5). From the expression (5.5) of $L_f(e_y)$ these points correspond to

$$3\cos^2\beta - 2 = 1 - 3\sin^2\beta = 0. \tag{5.11}$$

This result is associated with the geometry of the detection. Let us first express the two detection signals $L_f(e_x)$ and $L_f(e_y)$ in terms of the fictitious spin observables. Using expressions (A.4) and (A.6) of appendix 1, one easily derives

$$L_f(e_x) \propto 1 - \langle S_y^2\rangle \tag{5.12}$$

$$L_f(e_y) \propto 1 - \langle S_x^2\rangle. \tag{5.13}$$

$L_f(e_y)$ is therefore proportional to the alignment of $\boldsymbol{S}$ along $0x$. The steady-state value of this alignment is deduced from the steady value of the alignment along $\boldsymbol{b}$ by a rotation around $0y$ with an angle $\frac{1}{2}\pi - \beta$ (see figure 3) which introduces a detection factor

$$R_{00}^{(2)}(\tfrac{1}{2}\pi - \beta) \sim 3\sin^2\beta - 1.$$

Such a detection factor does not appear for $L_f(e_x)$, which is proportional to the alignment along $0y$, since the angle between $\boldsymbol{b}$ and $0y$ remains constant when β varies.

Except for $\Omega_e \simeq 0$ or $\Omega_e \gg \omega_1$ ($\beta = \pi/2, 0, \pi$), the level-crossing signals when $\Delta\nu$ is very large tend to the value corresponding to the common points $3\cos^2\beta - 1 = 0$, i.e. according to (5.10), to the value corresponding to a completely depolarized spin. This means that, when the action of $\boldsymbol{\epsilon}(t)$ is not inhibited by geometrical factors, the corresponding relaxation equalizes the populations of the three sublevels $|+1\rangle$, $|0\rangle$ and $|-1\rangle$, and destroys the coherences between $|+1\rangle$ and $|-1\rangle$.

More precisely, near $\Omega_e = 0$ and with the approximation $\sin^2\beta \simeq 1$, $\cos^2\beta \simeq \Omega_e^2/\omega_1^2$, one finds for example

$$L_f(e_x) \simeq \frac{2}{3} + \frac{1}{3}\left(1 - \frac{3}{\Gamma T_1}\frac{\Omega_e^2}{\omega_1^2}\right)^{-1}. \tag{5.14}$$

This is a Lorentzian shape with a width given by

$$\left(\omega_1^2\frac{\Gamma T_1}{3}\right)^{1/2} \sim \left(\frac{\omega_1^2\Gamma}{\Delta\nu}\right)^{1/2}. \tag{5.15}$$

At first sight, this result seems quite surprising since it implies that the width of the level-crossing resonance decreases when $\Delta\nu$ increases (ω_1^2 being constant). Actually, this behaviour can easily be understood. The fluctuations of $\dot{\phi}$ are always inefficient for $\Omega_e = 0$. The larger the fluctuations, i.e. the larger that $\Delta\nu$ is, the smaller is the value of Ω_e at which the fictitious spin begins to be affected, which explains the narrowing of the level-crossing resonances near $\Omega_e = 0$ when $\Delta\nu$ increases.

To conclude, one can say that the shape of level-crossing resonances are quite sensitive to the fast phase fluctuations of the laser beam. To our knowledge, their effects have not been investigated experimentally. They could provide interesting information on the higher order correlation functions of the light beam. Such a study would also fill the gap between the two extreme situations which are well known and which correspond respectively to a pure monochromatic light beam or to a very broad band excitation ($\Delta\nu \gg \omega_1$).

Appendix 1. Some useful relations on irreducible tensor operators

The irreducible tensor operators $T_q^{(k)}$ can be expressed in terms of the three components $S_\pm = S_x \pm iS_y$, S_z of the spin 1 $\boldsymbol{S}$. With the normalization condition

$$\mathrm{Tr}\, T_q^{(k)} T_{q'}^{+(k')} = \delta_{kk'}\,\delta_{qq'} \tag{A.1}$$

one gets

$$T_0^{(0)} = (1/\sqrt{3})\,\mathbf{1} \tag{A.2}$$

$$T_0^{(1)} = (1/\sqrt{2})S_z \qquad T_{\pm 1}^{(1)} = \mp\tfrac{1}{2}S_\pm \tag{A.3}$$

$$T_0^{(2)} = \sqrt{\tfrac{3}{2}}(S_z^2 - \tfrac{2}{3}) \qquad T_{\pm 1}^{(2)} = \mp\tfrac{1}{2}(S_zS_\pm + S_\pm S_z) \qquad T_{\pm 2}^{(2)} = \tfrac{1}{2}S_\pm^2. \tag{A.4}$$

It follows that, in the expansion of the atomic density matrix σ'' on the basis (see (4.8)),

$$\sigma'' = \sum_{\substack{k=0,1,2\\ q}} c_q^k T_q^{(k)} \tag{A.5}$$

the coefficients c_q^k are related to the matrix elements of σ'' by the following relations:

$$c_0^0 = (1/\sqrt{3})(\sigma''_{++} + \sigma''_{--} + \sigma''_{00}) = 1/\sqrt{3} \qquad c_0^1 = (1/\sqrt{2})(\sigma''_{++} - \sigma''_{--})$$

$$\begin{aligned}
c_1^1 &= -(1/\sqrt{2})(\sigma''_{+0} + \sigma''_{0-}) \quad \text{and} \quad c_{-1}^1 = (1/\sqrt{2})(\sigma''_{0+} + \sigma''_{-0})\\
c_0^2 &= \sqrt{\tfrac{3}{2}}\,(\sigma''_{++} + \sigma''_{--}) - \sqrt{\tfrac{2}{3}}\\
c_1^2 &= (1/\sqrt{2})(\sigma''_{0-} - \sigma''_{+0}) \quad \text{and} \quad c_{-1}^2 = -(1/\sqrt{2})(\sigma''_{-0} - \sigma''_{0+})\\
c_2^2 &= \sigma''_{+-} \quad \text{and} \quad c_{-2}^2 = \sigma''_{-+}.
\end{aligned} \tag{A.6}$$

In the rotation $R(\beta) = \exp(\mathrm{i}\beta S_y)$ introduced in (4.2), the $T_q^{(k)}$ operators transform according to the relation

$$R(\beta)T_q^{(k)}R(-\beta) = \sum_{q'} T_{q'}^{(k)} R_{q'q}^{(k)}(\beta). \tag{A.7}$$

Explicit values of some coefficients $R_{q'q}^{(k)}$ will be useful, particularly in the study of the relaxation of the fictitious spin 1, for example:

$$\begin{aligned}
R_{00}^{(2)}(\beta) &= \tfrac{1}{2}(3\cos^2\beta - 1)\\
R_{10}^{(2)}(\beta) &= \sqrt{\tfrac{3}{2}}\sin\beta\cos\beta\\
R_{20}^{(2)}(\beta) &= (\sqrt{6}/4)\sin^2\beta
\end{aligned} \tag{A.8}$$

with

$$R_{-q0}^{(2)}(\beta) = R_{0q}^{(2)}(\beta) = R_{q0}^{(2)}(-\beta).$$

Appendix 2. Relaxation equations under the effect of the fluctuating fields

We have to determine the equations of evolution of the two coefficients $\tilde{c}_0^1$ and $\tilde{c}_0^2$ of the density matrix $\tilde{\sigma}''$ ($\tilde{\Sigma}''$ representation) under the effect of the two Hamiltonians $\tilde{H}_c$ and $\tilde{H}_f$:

$$\tilde{H}_c = \Omega_1\sqrt{2}\,T_0^{(1)} \tag{A.9}$$

$$\tilde{H}_f = -\dot{\phi}(t)\left((2/\sqrt{3})T_0^{(0)} + \sqrt{\tfrac{2}{3}}\sum_q R_{q0}^{(2)}(\beta)T_q^{(2)}\right). \tag{A.10}$$

According to equations (A.6), it is equivalent to determining the equations of relaxation of the three populations $\tilde{\sigma}_{++}$, $\tilde{\sigma}_{--}$ and $\tilde{\sigma}_{00}$. Since we have seen that the fluctuating Hamiltonian satisfies the motional-narrowing condition, the usual relaxation theory can be applied. One immediately gets for the coupled evolution of these three populations:

$$\begin{aligned}
\dot{\tilde{\sigma}}_{++} &= -(\gamma_1 + \gamma_2)\tilde{\sigma}_{++} + \gamma_1\tilde{\sigma}_{00} + \gamma_2\tilde{\sigma}_{--}\\
\dot{\tilde{\sigma}}_{00} &= -2\gamma_1\tilde{\sigma}_{00} + \gamma_1\tilde{\sigma}_{++} + \gamma_1\tilde{\sigma}_{--}\\
\dot{\tilde{\sigma}}_{--} &= -(\gamma_1 + \gamma_2)\tilde{\sigma}_{--} + \gamma_1\tilde{\sigma}_{00} + \gamma_2\tilde{\sigma}_{++}
\end{aligned} \tag{A.11}$$

where the two coefficients γ_1 and γ_2 are the transition probabilities between levels $|\tilde{0}\rangle\,|+\tilde{1}\rangle$ or $|\tilde{0}\rangle\,|-\tilde{1}\rangle$, and $|-\tilde{1}\rangle\,|+\tilde{1}\rangle$ respectively, induced by the fluctuating Hamiltonian $\tilde{H}_{\mathrm{f}}$ (Abragam 1961), i.e.

$$\gamma_1 = \int_0^\infty \mathrm{d}\tau\, \overline{\langle\tilde{1}|\,\tilde{H}_{\mathrm{f}}(t)\,|\tilde{0}\rangle\langle\tilde{0}|\,\tilde{H}_{\mathrm{f}}(t-\tau)\,|\tilde{1}\rangle}\, \exp(-\mathrm{i}\Omega_1\tau) + \text{Herm conj} \tag{A.12}$$

$$\gamma_2 = \int_0^\infty \mathrm{d}\tau\, \overline{\langle\tilde{1}|\,\tilde{H}_{\mathrm{f}}(t)\,|-\tilde{1}\rangle\langle-\tilde{1}|\,\tilde{H}_{\mathrm{f}}(t-\tau)\,|\tilde{1}\rangle}\, \exp(-2\mathrm{i}\Omega_1\tau) + \text{Herm conj.} \tag{A.13}$$

In these two expressions, one must average over fast fluctuations $\dot{\phi}(t)$. Using equations (A.10) and (A.8), and equation (3.12) of I which gives the correlation function of $\dot{\phi}$:

$$\overline{\dot{\phi}(t)\dot{\phi}(t-\tau)} = \tfrac{1}{2}\kappa\Delta\nu \exp(-\kappa\tau) \tag{A.14}$$

one readily gets the explicit values of γ_1 and γ_2:

$$\gamma_1 = \tfrac{1}{2}\sin^2\beta\cos^2\beta\,\frac{\kappa^2}{\kappa^2+\Omega_1^2}\,\Delta\nu \tag{A.15}$$

$$\gamma_2 = \tfrac{1}{4}\sin^4\beta\,\frac{\kappa^2}{\kappa^2+4\Omega_1^2}. \tag{A.16}$$

The system (A.11) giving the evolution of the populations in $\tilde{\Sigma}''$ can be simply rewritten as

$$\left.\begin{aligned} \dot{\tilde{\sigma}}_{++} - 2\dot{\tilde{\sigma}}_{00} + \dot{\tilde{\sigma}}_{--} &= -3\gamma_1(\tilde{\sigma}_{++} - 2\tilde{\sigma}_{00} + \tilde{\sigma}_{--}) \\ \dot{\tilde{\sigma}}_{++} - \dot{\tilde{\sigma}}_{--} &= -(\gamma_1 + 2\gamma_2)(\tilde{\sigma}_{++} - \tilde{\sigma}_{--}) \end{aligned}\right. \tag{A.17}$$

which, according to (A.6), gives immediately

$$\left.\begin{aligned} \dot{\tilde{c}}_0^2 &= -3\gamma_1\tilde{c}_0^2 \\ \dot{\tilde{c}}_0^1 &= -(\gamma_1 + 2\gamma_2)\tilde{c}_0^1. \end{aligned}\right. \tag{A.18}$$

Finally, coming back to the notation used in §4 (4.14), one finds

$$r_0^{1k} = -\delta_{1k}\left(\tfrac{1}{2}\sin^2\beta\cos^2\beta\,\frac{\kappa^2}{\kappa^2+\Omega_1^2} + \tfrac{1}{2}\sin^4\beta\,\frac{\kappa^2}{\kappa^2+4\Omega_1^2}\right)\Delta\nu \tag{A.19}$$

$$r_0^{2k} = -\delta_{2k}\tfrac{3}{2}\sin^2\beta\cos^2\beta\,\frac{\kappa^2}{\kappa^2+\Omega_1^2}\,\Delta\nu. \tag{A.20}$$

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