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Paul Avan, Claude Cohen-Tannoudji

Institutions: Collège de France

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P. Avan, C. Cohen-Tannoudji

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HANLE EFFECT FOR MONOCHROMATIC EXCITATION. NON PERTURBATIVE CALCULATION FOR A J = 0 TO J = 1 TRANSITION

P. AVAN and C. COHEN-TANNOUDJI

Laboratoire de Spectroscopie Hertzienne de l'E.N.S.
et Collège de France, 24, rue Lhomond, 75231 Paris Cedex 05, France

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Résumé. — On établit des expressions théoriques exactes pour les signaux de fluorescence observés dans une expérience de croisement de niveaux effectuée avec une excitation monochromatique sur une transition $J_g = 0 \leftrightarrow J_e = 1$. On discute l'effet de l'élargissement radiatif et du désaccord Zeeman.

Abstract. — Exact theoretical expressions are derived for the fluorescent light observed in a level crossing experiment performed on a $J_g = 0$ to $J_e = 1$ transition using monochromatic excitation. The effect of radiative broadening and Zeeman detuning is discussed.

1. **Introduction.** — The development of narrow band tunable laser sources results in the possibility of achieving an almost monochromatic excitation of free atoms. Recently, Rasmussen, Schieder and Walther observed [1] zero field level crossing resonances on an atomic beam of Ba^{138} irradiated perpendicularly by a single mode laser beam ([1]). The available laser intensity was sufficient to saturate the atomic transition ($6s^2\ ^1S_0 \leftrightarrow 6s\,6p\ ^1P_1$); in such a case, the radiative broadening of the resonances may become important and the lowest order theory (with respect to the electric field of the light wave) is insufficient. It is necessary to go beyond the Born approximation [2, 3, 4] for the scattering amplitude.

The necessity of computing such higher order effects also occurs for a broad-line excitation (spectral width of the incident beam very large compared to the natural and Doppler widths). In this case, the shortness of the correlation time of the excitation simplifies the problem : it is possible to obtain rate equations coupling the density matrices σ_e and σ_g representing the collection of atoms in the excited and ground states. Their steady state solution gives non perturbative expressions for the various detection signals and allows a quantitative interpretation of several higher order effects (radiative broadening, saturation resonances... [5] to [9]).

For monochromatic excitation, the problem is more difficult : the coherence time of the light is very long, so that it is impossible to obtain rate equation coupling only σ_e and σ_g. The optical coherences σ_{eg} (off diagonal elements of the density matrix between e and g, which represent the motion of the atomic dipole moments driven by the incident light wave) must be taken into account, as they are now not negligible [9]. In this case it is better to start from the equations describing the coupled evolution of σ_e, σ_g and σ_{eg} under the influence of the various processes : Larmor precession, spontaneous emission, interaction with the laser light, and to try to solve them for arbitrary values of the light intensity. This is what we do in the present letter, for the simple case of a $J_g = 0 \rightarrow J_e = 1$ transition (corresponding to the experiment of reference [1]). From the steady state solution of the equations of evolution, we derive exact theoretical expressions for various detection signals allowing a quantitative interpretation of the experimental results.

2. **Notations.** — We consider an atomic beam propagating along Oz. A magnetic field $\mathbf{B}_0$ is applied in the same direction. The atoms are irradiated by a single mode laser beam of pulsation ω propagating along Oy and having a linear polarization $\mathbf{e}_x$ parallel to Ox. One monitors the fluorescence $L_F(\mathbf{e}_y)$ emitted by the atoms along Ox with a linear polarization $\mathbf{e}_y$ parallel to Oy. Another possible detection signal could be the fluorescence $L_F(\mathbf{e}_x)$ emitted along Oz with a linear polarization parallel to Ox.

The $J_g = 0$ atomic ground state has only one Zeeman sublevel $|0\rangle$ which is coupled by the σ-polarized exciting light to the $|+1\rangle$ and $|-1\rangle$

([1]) We have heard that similar work has been performed by BRAND H. *et al.*, at Hannover Technical University (*Optics Communications*, to be published).

sublevels of the $J_e = 1$ excited state (we can forget the $m = 0$ excited sublevel). The $|0\rangle \leftrightarrow |+1\rangle$ and $|0\rangle \leftrightarrow |-1\rangle$ transitions correspond to energies respectively equal to $\omega_0 + \Omega_e$ and $\omega_0 - \Omega_e$, where ω_0 is the energy of the optical line in zero magnetic field and Ω_e the Larmor pulsation in the excited state (we take $\hbar = 1$).

Let σ be the density matrix representing the ensemble of atoms in the illuminated zone of the atomic beam. The diagonal elements σ_{++}, σ_{--}, σ_{00} of σ represent the populations of the 3 Zeeman sublevels $|+1\rangle$, $|-1\rangle$, $|0\rangle$. $\sigma_{+-} = \sigma^*_{-+}$ is the Zeeman coherence between the 2 excited sublevels, $\sigma_{0+} = \sigma^*_{+0}$ and $\sigma_{0-} = \sigma^*_{-0}$ are the optical coherences between $|0\rangle$ and $|+1\rangle$, and $|0\rangle$ and $|-1\rangle$.

From the theory of spontaneous emission, one can show that $L_F(\mathbf{e}_y)$ and $L_F(\mathbf{e}_x)$ are proportional to some linear combinations of the density matrix elements of σ_e :

$$L_F(\mathbf{e}_y) \propto \sigma_{++} + \sigma_{--} + 2\,\mathrm{Re}\,\sigma_{-+} \tag{1}$$

$$L_F(\mathbf{e}_x) \propto \sigma_{++} + \sigma_{--} - 2\,\mathrm{Re}\,\sigma_{-+}\,. \tag{2}$$

Therefore a quantitative interpretation of the experimental results requires the calculation of σ_{++}, σ_{--}, σ_{-+}.

3. **Master equation.** — The rate of variation of the various elements of σ is given by :

$$\frac{\mathrm{d}}{\mathrm{d}t}\sigma = -\,i[\mathcal{H}, \sigma] \tag{3}$$

where $\mathcal{H}$ is the total hamiltonian of the system, including the effects of the atomic hamiltonian $\mathcal{H}_0$ (free evolution), and the coupling with the incident beam; some extra terms describing the effect of spontaneous emission must be added to (3). The following equations are easily derived :

$$\begin{array}{l|l|l}
\dot\sigma_{++} = & -\Gamma\sigma_{++} & -iv(\sigma_{0+}\,e^{-i\omega t} - \sigma_{+0}\,e^{i\omega t}) \\
\dot\sigma_{--} = & -\Gamma\sigma_{--} & +iv(\sigma_{0-}\,e^{-i\omega t} - \sigma_{-0}\,e^{i\omega t}) \\
\dot\sigma_{-+} = 2i\Omega_e\,\sigma_{-+} & -\Gamma\sigma_{-+} & +iv\,e^{-i\omega t}\sigma_{0+} + iv\,e^{i\omega t}\sigma_{-0} \\
\dot\sigma_{00} = & \Gamma(\sigma_{++} + \sigma_{--}) & -iv(\sigma_{+0} - \sigma_{-0})\,e^{i\omega t} + iv(\sigma_{0+} - \sigma_{0-})\,e^{-i\omega t} \\
\dot\sigma_{0+} = i(\omega_0 + \Omega_e)\,\sigma_{0+} & -\Gamma\sigma_{0+}/2 & -iv(\sigma_{++} - \sigma_{-+} - \sigma_{00})\,e^{i\omega t} \\
\dot\sigma_{0-} = i(\omega_0 - \Omega_e)\,\sigma_{0-} & -\Gamma\sigma_{0-}/2 & +iv(\sigma_{--} - \sigma_{+-} - \sigma_{00})\,e^{i\omega t}
\end{array} \tag{4}$$

Γ is the natural width of the excited state and v is a coupling parameter proportional to the product of the atomic dipole moment by the amplitude $\mathcal{E}$ of the laser light wave. More precisely,

$$v^2 = 3\,\mathcal{E}^2 e^2 f_{ge}/16\,m\omega_0$$

where f_{ge} is the oscillator strength of the transition g → e. We have supposed that the laser frequency is not too far from the atomic frequency, so that the so-called *rotating wave approximation* (valid if $|\omega - \omega_0| \ll \omega_0$ and $v \ll \omega_0$) can be used in deriving the terms of the last column of eq. (4) describing the effect of the coupling between the atoms and the laser. The first column of eq. (4) describes the free evolution of the atoms, the second one, the effect of spontaneous emission (damping of σ_{++}, σ_{--}, σ_{-+} and $\sigma_{0\pm}$ with a time constant equal to $1/\Gamma$ or $2/\Gamma$, and transfer of atoms from the excited state e to the ground state g).

If we put : $\sigma_{0\pm} = \rho_{0\pm}\,e^{i\omega t}$ and $\sigma_{\pm 0} = \rho_{\pm 0}\,e^{-i\omega t}$, we can easily transform (4) into a set of linear differential equations with time independent coefficients giving $\dot\sigma_{++}\ \dot\sigma_{--}\ \dot\sigma_{-+}\ \dot\rho_{0\pm}$ (equivalent of the transformation to the rotating frame in NMR). By equating these rates of variation to zero, one then finds the steady state solution for the various matrix elements of σ. Reinserting these steady values of $\sigma_{++}\ \sigma_{--}\ \sigma_{-+}\ \sigma_{+-}$ into eq. (1) and (2), one finally obtains the following theoretical expressions for the detection signals $L_F(\mathbf{e}_x)$ and $L_F(\mathbf{e}_y)$

$$L_F(\mathbf{e}_y) \propto \frac{\Omega_e^2(D-1) - \Omega_e\,Y}{A + (3D-1)\,B} \tag{5}$$

$$L_F(\mathbf{e}_x) \propto \frac{(\Omega_e + Y)\,Y + (D-1)(\Omega_e^2 + B)}{A + (3D-1)\,B} \tag{6}$$

where $\Delta\omega = \omega - \omega_0$

$$x^{\pm}(\Omega_e) = [(\Gamma/2)^2 + (\Delta\omega \pm \Omega_e)^2]\,v^{-2}$$

$$D(\Omega_e) = 1 + (1 + x^+)^{-1} + (1 + x^-)^{-1}$$

$$Y(\Omega_e) = (\Delta\omega - \Omega_e)(1 + x^-)^{-1} - (\Delta\omega + \Omega_e)(1 + x^+)^{-1}$$

$$A(\Omega_e) = 4\,\Omega_e(Y + \Omega_e D) + 3\,Y^2$$

$$B(\Omega_e) = \frac{\Gamma^2}{4}\left(2 + \frac{1}{x^+} + \frac{1}{x^-}\right) + \frac{(\Delta\omega - \Omega_e)^2}{x^-(1 + x^-)} + \frac{(\Delta\omega + \Omega_e)^2}{x^+(1 + x^+)}\,.$$

4. **Discussion.** — Expressions (5) and (6) give the dependence of the signal on 3 important parameters, the static magnetic field (Ω_e), the light intensity (v^2), and detuning of the laser from the atomic frequency ($\omega - \omega_0$). If one fixes v^2 and $\omega - \omega_0$ and varies Ω_e, one obtains the shape of the zero field level crossing resonance. This shape is clearly not lorentzian (for

a broad line excitation, this shape would be always lorentzian in the case of a $J_g = 0 \leftrightarrow J_e = 1$ transition [5, 6, 9]). Figure 1 gives the shape of the level crossing resonances for a zero detuning ($\omega = \omega_0$); each curve corresponds to a different value of the light intensity. One clearly sees a radiative broadening of the resonances. Figure 2 gives the shape of the resonances for a fixed value of the intensity and increasing values of the detuning $\omega - \omega_0$.

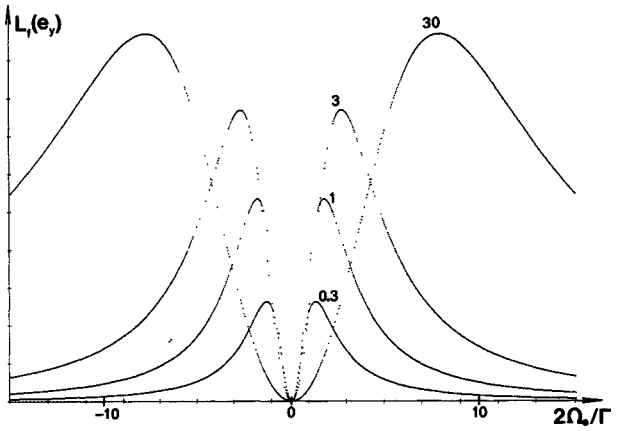

FIG. 1. — Set of zero field level crossing resonances detected on $L_F(\mathbf{e}_y)$ for a zero detuning. $2\,\Omega_e/\Gamma$ is a normalized Larmor pulsation; each curve corresponds to a different value of the parameter $4\,v^2/\Gamma^2$ (proportional to the light intensity) indicated on the figure. One clearly sees a radiative broadening of the resonances. As the polarizations of the incident and fluorescent light are orthogonal, the signal vanishes for $\Omega_e = 0$.

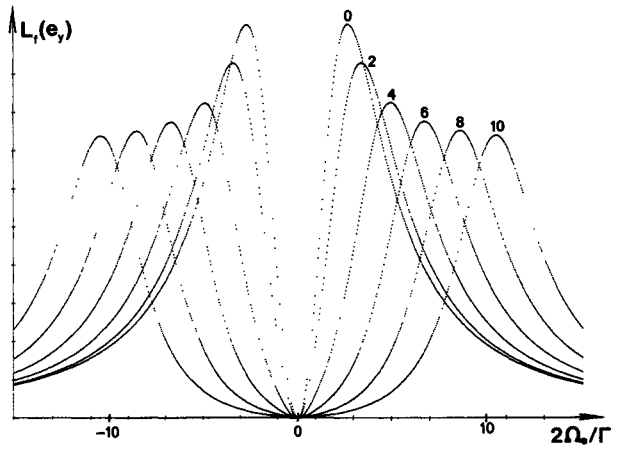

FIG. 2. — Set of Hanle resonances detected on $L_F(\mathbf{e}_y)$ for different detunings $2\,\Delta\omega/\Gamma$, indicated on the figure. This set corresponds to a fixed value of the light intensity ($4\,v^2/\Gamma^2 = 3$). For a very large detuning one gets two Lorentz curves corresponding to the incoherent excitation of the $|+1\rangle$ and $|-1\rangle$ Zeeman sublevels.

It is interesting to study some limiting cases.

— For vanishing light intensities ($v \to 0$), one finds for example for $L_F(\mathbf{e}_y)$ the following expression valid to lowest order in v :

$$L_F(\mathbf{e}_y) \propto \propto \frac{v^2\,\Omega_e^2}{[(\Gamma/2)^2 + (\Delta\omega - \Omega_e)^2]\,[(\Gamma/2)^2 + (\Delta\omega + \Omega_e)^2]} \qquad (7)$$

(7) coincides with the expression obtained from the Born amplitude for the resonant scattering [2, 3, 4]. The width of the resonance is of the order of $\Gamma/2$, i.e. half of the width obtained in the same conditions with a broad line excitation.

— At very high intensities ($v \gg \Gamma, |\omega - \omega_0|$), the signal depends only on the dimensionless parameter Ω_e/v. For example :

$$L_F(\mathbf{e}_y)_{v\ \Gamma,\Delta\omega} \propto \frac{(\Omega_e/v)^2}{(\Omega_e/v)^4 + (\Omega_e/v)^2 + 4}. \qquad (8)$$

In this case the shape of the level crossing resonance does not change when v increases, provided that the scale of the horizontal axis is contracted proportionally to v. In other words, the height of the resonance saturates and its width increases as the square root of the light intensity. We find a behaviour similar to the one observed with a broad line excitation [5, 6, 9].

When the detuning of the laser is very high ($|\omega - \omega_0| \gg \Gamma, v$), the Zeeman splitting required to put an excited sublevel in resonance with the laser light is so large that Zeeman coherence can no longer appear between the two excited sublevels. Thus $L_F(\mathbf{e}_y)$ tends to a sum of two Lorentzians centered on $\Omega_e = \pm\,\Delta\omega$, corresponding to the resonant excitation of $|+1\rangle$ or $|-1\rangle$ sublevels :

$$L_F(\mathbf{e}_y)_{\Delta\omega \gg v,\Gamma} \propto \frac{v^2}{(\Delta\omega - \Omega_e)^2 + 2\,v^2 + \Gamma^2/4} + \frac{v^2}{(\Delta\omega + \Omega_e)^2 + 2\,v^2 + \Gamma^2/4}. \qquad (9)$$

At very high magnetic field

$$(\Omega_e \gg \Gamma, |\omega - \omega_0|, v)\ L_F(\mathbf{e}_y) \quad \text{and} \quad L_F(\mathbf{e}_x)$$

tend to zero. This is due to the fact that the frequencies $\omega_0 \pm \Omega_e$ of the optical lines $0 \leftrightarrow +1$ and $0 \leftrightarrow -1$ are out of resonance with the laser frequency ω. Let us recall that for a broad line excitation, L_F would tend to a non-zero limiting value corresponding to the light reemitted incoherently from $|-1\rangle$ and $|+1\rangle$ sublevels.

The preceding calculation could be easily generalized to atomic transitions corresponding to higher values of J_g and J_e. It seems difficult to get explicit analytical expressions for the steady state solution of the coupled equations of evolution of σ_e, σ_g, σ_{eg}, but, as these equations are linear, it is always possible to compute this solution, at least numerically, by inverting a matrix.

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