

Optical parametric resonances

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Abstract. The response of a two-level atom driven by a strong resonant amplitude-modulated field is calculated in the Bloch model. The population difference and dipole moment show a resonant behaviour when the Rabi frequency is a multiple of the modulation frequency. This phenomenon is related to the resonant AC Stark effect and is the optical counterpart of the coherence resonances known in radiofrequency magnetic resonance.

1. Introduction

The problem of a two-level atom interacting with a strong resonant light wave of constant amplitude has been the subject of a number of theoretical papers (Newstein 1968, Mollow 1969, 1975, Stroud 1971, Cohen-Tannoudji 1975, Carmichael and Walls 1975, 1976a,b, Hassan and Bullough 1975, Smithers and Freedhoff 1975, Swain 1975) as well as experimental ones (Schuda *et al* 1974, Walther 1975, Wu *et al* 1975, Hartig *et al* 1976).

The main feature of such a system lies in the power spectrum of the light scattered by the atom, which exhibits two sidebands, symmetrically placed with respect to the usual fluorescence line, and separated from the latter by the Rabi frequency of the system (dynamic Stark effect). At the same time, however, the long-term expectation values for the inversion and dipole moment are time-independent.

When replacing the constant-amplitude field by an amplitude-modulated one, further information can be obtained about the evolution of the atomic observables. In the steady state these undergo oscillations at each harmonic of the modulation frequency. The amplitude of these oscillations goes through a maximum when the Rabi frequency is a multiple of the modulation frequency. Moreover, the static component of the inversion also undergoes such a resonant variation, which can be measured directly through the average scattered intensity.

This problem has been treated in two limiting cases (Armstrong and Feneuille 1975, Feneuille *et al* 1976). In the first case, the modulation depth is small and serves as a perturbation parameter in the Bloch equations; in the second case, an exact solution is obtained for an infinite modulation depth (suppressed carrier field).

The solution presented here extends the amplitude range of the fields to all combinations of carrier and modulation that result in a permanently saturating field.

2. Statement of the problem

The two atomic states $|0\rangle$ and $|1\rangle$ are eigenstates of the atomic Hamiltonian H_0 , with the energy difference $E_1 - E_0 = \hbar\omega_0$. The coupling of the atom with the coherent resonant light wave is described by the electric dipole Hamiltonian in the semiclassical approximation:

$$H_1(t) = -\boldsymbol{\mu} \cdot \mathbf{E}(t). \quad (1)$$

The following definitions are used: the dipole moment operator is given by

$$\boldsymbol{\mu} = \begin{pmatrix} 0 & \boldsymbol{\mu}_{10} \\ \boldsymbol{\mu}_{10}^* & 0 \end{pmatrix} \quad (2)$$

and

$$\mathbf{E}(t) = E(t) \operatorname{Re}(\boldsymbol{\epsilon} e^{-i\omega_L t}) \quad (3)$$

is the (real) electric field of the light wave; its time dependence is split into two parts: a constant-amplitude light wave of angular frequency ω_L and polarization vector $\boldsymbol{\epsilon}$ ($\boldsymbol{\epsilon} \cdot \boldsymbol{\epsilon}^* = 1$), and a slowly varying envelope $E(t)$. Here $E(t)$ will take the following form:

$$E(t) = E_0[1 + a \cos(\omega t + \theta)] \quad (4)$$

where a is the modulation depth ($a > 0$) and θ an arbitrary phase.

The evolution of the coupled system will be determined by solving the density-matrix equation

$$i\hbar \frac{\partial \rho}{\partial t} = [H(t), \rho] \quad (5)$$

in the 'rotating frame' evolving at the laser frequency ω_L . The rotating-wave approximation is fully justified here since $\boldsymbol{\mu} \cdot \mathbf{E} \ll \hbar\omega_0$ for current CW lasers in the optical range. Thus the only explicit time dependence in $H(t)$ arises from the slowly varying envelope of $E(t)$. The equations of motion then read:

$$i\hbar \frac{\partial \rho_{11}}{\partial t} = -\frac{1}{2}E(t)(\boldsymbol{\mu}_{10} \cdot \boldsymbol{\epsilon} \rho_{01} - \boldsymbol{\mu}_{10}^* \cdot \boldsymbol{\epsilon}^* \rho_{10}) - i\hbar\gamma\rho_{11} \quad (6a)$$

$$i\hbar \frac{\partial \rho_{00}}{\partial t} = \frac{1}{2}E(t)(\boldsymbol{\mu}_{10} \cdot \boldsymbol{\epsilon} \rho_{01} - \boldsymbol{\mu}_{10}^* \cdot \boldsymbol{\epsilon}^* \rho_{10}) + i\hbar\gamma\rho_{11} \quad (6b)$$

$$i\hbar \frac{\partial \rho_{10}}{\partial t} = -\hbar(\omega_L - \omega_0)\rho_{10} + \frac{1}{2}E(t)\boldsymbol{\mu}_{10} \cdot \boldsymbol{\epsilon}(\rho_{11} - \rho_{00}) - i\hbar\frac{1}{2}\gamma\rho_{10} \quad (6c)$$

$$i\hbar \frac{\partial \rho_{01}}{\partial t} = \hbar(\omega_L - \omega_0)\rho_{01} - \frac{1}{2}E(t)\boldsymbol{\mu}_{10}^* \cdot \boldsymbol{\epsilon}^*(\rho_{11} - \rho_{00}) - i\hbar\frac{1}{2}\gamma\rho_{01}. \quad (6d)$$

The relaxation terms are introduced phenomenologically; they arise exclusively from spontaneous emission from $|1\rangle$ to $|0\rangle$ (Mollow and Miller 1969). $\hbar\gamma$ is the natural width of the excited level.

These equations can be written in terms more relevant from a physical point of view, i.e. the population inversion

$$z = \rho_{11} - \rho_{00} \quad (7a)$$

and the components of the atomic dipole in phase and $\pi/2$ out of phase with respect

to the applied field

$$x = \rho_{10} + \rho_{01} \quad (7b)$$

$$y = i(\rho_{10} - \rho_{01}). \quad (7c)$$

For convenience we assume $\mu_{10} \cdot \epsilon$ is a real number and define $\Omega(t)$ and Ω_0 by

$$\frac{\mu_{10} \cdot \epsilon}{\hbar} E(t) = \Omega(t) = \Omega_0 [1 + a \cos(\omega t + \theta)]. \quad (8)$$

Equations (6) then take the well known form of the optical Bloch equations

$$\dot{x} = -\frac{1}{2}\gamma x + \alpha y \quad (9a)$$

$$\dot{y} = -\alpha x - \frac{1}{2}\gamma y + \Omega(t)z \quad (9b)$$

$$\dot{z} = -\Omega(t)y - \gamma(z + 1) \quad (9c)$$

with

$$\alpha = \omega_L - \omega_0. \quad (10)$$

Such a description of the atomic behaviour, limited to its averaged parameters, is not intended to give information on the spectral properties of the scattered light. But, as we shall see in the following sections, the introduction of a modulated driving field leads to a possible measurement of the strength of the atom–field coupling by observing the mere total intensity scattered by the atom, which in turn is proportional to the inversion $z(t)$.

3. Solution of the Bloch equations

Let us first recall the exact steady-state solution when $a = 0$:

$$x = -\frac{4\alpha\Omega_0}{\gamma^2 + 4\alpha^2 + 2\Omega_0^2} \quad (11a)$$

$$y = -\frac{2\gamma\Omega_0}{\gamma^2 + 4\alpha^2 + 2\Omega_0^2} \quad (11b)$$

$$z = -\frac{\gamma^2}{\gamma^2 + 4\alpha^2 + 2\Omega_0^2}. \quad (11c)$$

If $a \neq 0$, the system (9) is not solvable analytically. However, at resonance ($\alpha = 0$) the first equation uncouples, leading to the trivial solution $x = 0$. The method used here for solving the two remaining equations consists in introducing a trial solution whose form is suggested by the linearity of the system and the periodicity of the coefficients:

$$\begin{aligned} y &= Y e^{if(t)} \\ z &= Z e^{if(t)} \end{aligned} \quad (12)$$

where Y , Z and $f(t)$ are to be determined. By introducing (12) into the homogeneous part of the system (9), the necessary condition for Y and Z to differ from zero is

$$if(t) = -\frac{3}{4}\gamma \pm \frac{1}{4}(\gamma^2 - 16\Omega^2(t))^{1/2}. \quad (13)$$

The analytical integration of (13) is not possible in the general case; however, an approximate solution is readily obtainable in two cases.

(i) *Low-field case*

$$\Omega^2(t) \ll \frac{1}{16}\gamma^2. \quad (14)$$

The atom stays in the vicinity of its ground state. A perturbative calculation is more suitable, but will not be developed here.

(ii) *Strong-field case*

$$\Omega^2(t) \gg \frac{1}{16}\gamma^2 \quad (15)$$

The condition must be verified at all times, which implies

$$\Omega_0^2(1 - a)^2 \gg \frac{1}{16}\gamma^2.$$

This is no severe restriction on the modulation depth provided that the carrier field has a saturating amplitude.

Under the assumption (15), equations (9b, c) are integrated by the standard method, and only terms of the first order in γ/Ω_0 are kept. We also neglect the transient regime, which decays with a characteristic time of the order of γ^{-1} .

In order to give the solution for $z(t)$ and $y(t)$ in a compact form, let us define the following list of symbols:

$$\beta_n = \frac{\Omega_0 - n\omega}{\frac{3}{4}\gamma} \quad (16)$$

$$\tan \phi_n = \frac{4\Omega_0}{\gamma} \left(\frac{\Omega_0 - n\omega + \frac{3}{16}\gamma^2/\Omega_0}{2\Omega_0 + n\omega} \right) \quad (17)$$

$$\tan \psi_n = -\beta_n^{-1} \quad (18)$$

$$r_n \cos \phi_n = \frac{4}{3} \left(\frac{\frac{2}{3}(1 + \frac{1}{2}n\omega/\Omega_0)}{1 + \beta_n^2} \right) \quad (\cos \phi_n > 0) \quad (19)$$

$$r_n \sin \phi_n = \frac{4}{3} \left(\frac{\beta_n + \frac{1}{4}\gamma/\Omega_0}{1 + \beta_n^2} \right) \quad (20)$$

$$s_n \cos \psi_n = \frac{4}{3} \left(\frac{\beta_n}{1 + \beta_n^2} \right) \quad (\cos \psi_n > 0) \quad (21)$$

$$s_n \sin \psi_n = -\frac{4}{3} \left(\frac{1}{1 + \beta_n^2} \right). \quad (22)$$

The approximate steady-state solutions of equations (9b, c) then read:

$$z(t) = - \sum_{\substack{p,n \\ -\infty \\ +\infty}} (-1)^p J_n \left(\frac{a\Omega_0}{\omega} \right) J_{n-p} \left(\frac{a\Omega_0}{\omega} \right) r_n \cos [p(\omega t + \theta) + \phi_n] \quad (23)$$

$$y(t) = - \sum_{\substack{p,n \\ -\infty \\ +\infty}} (-1)^p J_n \left(\frac{a\Omega_0}{\omega} \right) J_{n-p} \left(\frac{a\Omega_0}{\omega} \right) s_n \cos [p(\omega t + \theta) + \psi_n] \quad (24)$$

where J_n denotes the Bessel function of order n .

As can be seen from the dependence of r_n and s_n on Ω_0 (see equations (16), (19)–(22)) the inversion z and the dipole moment y show a resonant behaviour when $\Omega_0 = n\omega$, i.e. when the average Rabi frequency Ω_0 is a multiple of the modulation frequency.

Since equations (23) and (24) are long-term solutions they describe correctly the behaviour of atoms entering the interaction region in arbitrary initial states.

An interpretation of these resonances is easily given in the Bloch picture. Note first that the solutions (23) and (24) are very similar to the solution of the Bloch equations for magnetic dipoles pumped transversely in a magnetic field having a constant and an oscillating component parallel to each other. There an exact solution is obtained when the longitudinal and transverse relaxation rates are equal (Favre and Geneux 1964, Aleksandrov *et al* 1964, Polonsky and Cohen-Tannoudji 1965). In the optical case the spontaneous emission introduces two unequal relaxation constants and pumps, at the same time, the system in its ground state.

Let us then consider a collection of atoms described by their average coordinates (y, z) and driven by the field $\Omega(t)$. Before the modulation is switched on, $y(\infty)$ and $z(\infty)$ are given by equations (11b, c) and result from an equilibrium between the applied field and the relaxation. This equilibrium is destroyed by the modulation. The evolution is then alternatively dominated by relaxation (during the low-field half-period) and by the driving field (high-field half-period). If the average field E_0 is such that the nutation angle, integrated over one modulation period, amounts to an integral number of cycles, the periodic effect of the relaxation is cumulative and contributes to sustain large oscillations of $y(t)$ and $z(t)$. Thus the modulation can lead to a permanent optical nutation. The origin of parametric resonances is seen to be purely kinematic. Figure 1 illustrates the behaviour of $z(t)$ when the condition for the first resonance is met ($\Omega_0 = \omega$).

The parametric resonances give a means of measuring the Rabi frequency in a given field. In this respect, they are related to the dynamic Stark effect. But, whereas in the latter the Rabi frequency Ω_0 appears in the *power spectrum* of the scattered light, for which the fluctuations only are responsible, in the parametric resonances

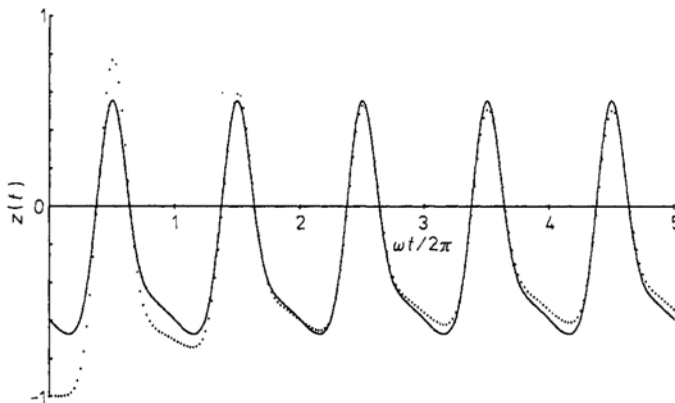


Figure 1. Time dependence of the inversion ($x = 0$, $a = 1$, $\Omega_0 = \omega = 5\gamma$, $\theta = \pi$). Dotted curve: numerical integration of equations (9b, c) with initial conditions $z(0) = -1$, $y(0) = 0$. Full curve: equation (23) (steady state, no transient regime), with $|n \pm p| \leq 4$. The effect of relaxation during the lower half-period (low field) is clearly visible (note that $a = 1$ does not fulfill condition (15) at all times).

Ω_0 shows up through a variation of the *total intensity* scattered by the atom. This variation affects the time-independent part as well as the modulated part of the intensity. Of course the modulation of the scattered light would also lead to sidebands in the spectrum, but their origin would be coherent oscillations of the atomic dipole moments instead of fluctuations around a constant value.

Another characteristic feature of parametric resonances is that they are not subject to power broadening. Their width is $\frac{3}{4}\gamma$ (HWHM) and is independent of a , the modulation depth. This is due to the kinematic origin of the resonances; they result from a coincidence between the modulation frequency of the field and the frequency of the evolution it forces. Such a coincidence can occur only when $\Omega_0 - n\omega \lesssim \gamma$, whatever the strength of the modulation.

4. Special cases

In order to examine the approximate solution for $z(t)$ (equation (24)) in a few characteristic situations, let us rewrite equation (24) so as to express z in terms of the amplitude $z_{p\omega}$ and phase $\Phi_{p\omega}$ of its component at frequency $p\omega$:

$$z(t) = z_0 + \sum_{p=1}^{\infty} z_{p\omega} \cos [p(\omega t + \theta) + \Phi_{p\omega}]. \quad (25)$$

4.1. Static component ($p = 0$)

$$z_0 = -\frac{4}{3} \sum_{n=-\infty}^{+\infty} \frac{J_n^2(a\Omega_0/\omega) \frac{1}{3} (1 + \frac{1}{2}n\omega/\Omega_0)}{1 + [(\Omega_0 - n\omega)/\frac{3}{4}\gamma]^2}. \quad (26)$$

Figure 2 gives an estimate of the error introduced when condition (15) ceases to be verified ($a = 0$); z_0 is plotted as a function of Ω_0/ω and can be compared with the exact solution (11c). The relative error increases when Ω_0 goes to zero but remains less than 11%.

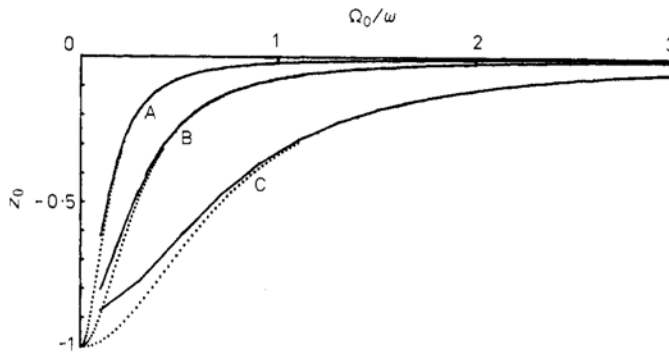


Figure 2. Inversion z in the unmodulated case ($a = 0$, $\alpha = 0$, $z(t) = z_0$). The full curves are given by equation (26), the dotted curves by equation (11c). A, $\omega/\gamma = 5$; B, $\omega/\gamma = 2.5$; C, $\omega/\gamma = 1$.

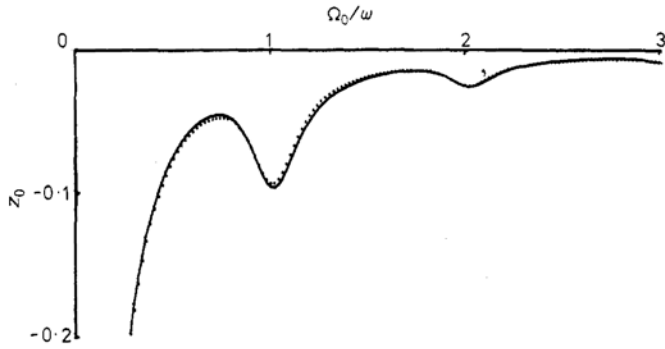


Figure 3. Static part of the inversion in the modulated case ($x = 0$, $\omega/\gamma = 5$). Equation (25) (full curve) is compared with the exact solution obtained by numerical integration (dotted curve) ($a = 0.5$).

Figures 3 and 4 show $z_0(\Omega_0/\omega)$ for $a = 0.5$ and 1.5 respectively (the sum over n in equation (26) is limited to $|n| \leq 3$). The dotted curves were obtained from the numerical integration of the Bloch equations (equations (9b, c)). The approximation remains close to the exact solution even for modulation depths that violate condition (15) (figure 4).

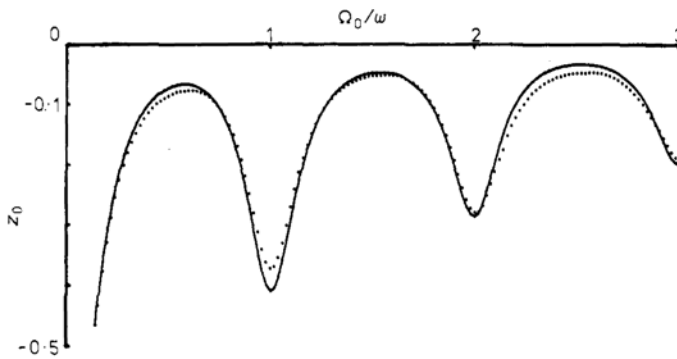


Figure 4. As figure 3 with $a = 1.5$.

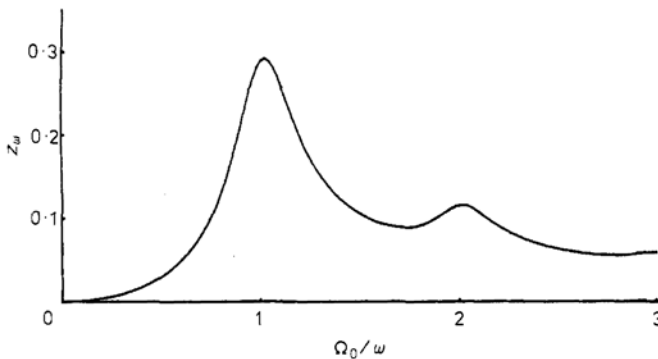


Figure 5. Amplitude of the first harmonic component of the inversion $z_\omega(x = 0, a = 0.5, \omega/\gamma = 5)$.

4.2. First harmonic ($|p| = 1$)

The detailed expressions for z_ω and Φ_ω can be deduced from the comparison of equations (25) and (23). They are plotted on figure 5 ($z_\omega(\Omega_0/\omega)$; $a = 0.5$) and figure 6 ($\Phi_\omega(\Omega_0/\omega)$; $a = 0.01, 0.7, 1.0, 1.5$). Note that the phase Φ_ω goes through zero in principle at all resonances, provided that the dominant contribution to z_ω is the resonant term. This requires well separated resonances and a sizable value of the n th Bessel function at the n th resonance (curve D figure 6).

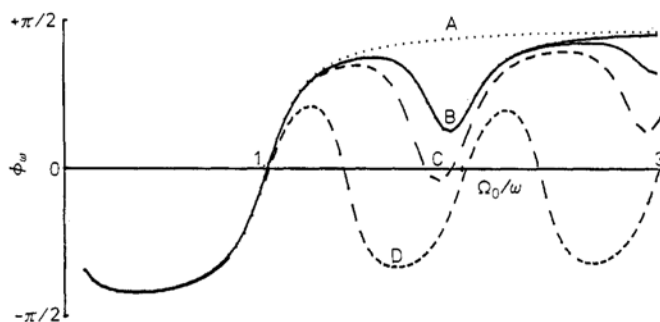


Figure 6. Phase of the first harmonic oscillations of z_ω , A, $a = 0.01$; B, $a = 0.7$; C, $a = 1$; D, $a = 1.5$.

5. Experimental aspects

Considerable work has been done on parametric resonances with magnetic fields (Aleksandrov *et al* 1964, Favre and Geneux 1964, Polonsky and Cohen-Tannoudji 1965, Chapman and Series 1970) and electric fields (Faist 1972) in the radiofrequency range, where they proved to have useful applications in spectroscopy and metrology (Dupont-Roc *et al* 1969).

In the optical range they provide a means of relating the strength of a coupling between atom and field (Ω_0) to a measurable frequency (ω). As calculated here, they could be best observed on the two-level transition in atomic sodium where $|0\rangle = |3S_{1/2}, F = 2, m_F = 2\rangle$ and $|1\rangle = |3P_{3/2}, F' = 3, m'_F = 3\rangle$ ($\lambda = 589$ nm). This transition can be isolated by optical pumping with circularly polarized light falling at right angles on an atomic beam in zero magnetic field. With a saturating laser field, complete depletion of all $|F = 2, m_F \neq 2\rangle$ states is achieved in a few tens of natural lifetimes (Thomann 1976). A power of about 1 W cm^{-2} is needed to obtain a saturation parameter Ω_0/γ of 10. Other atomic transitions would be suitable, as the transition at 553.5 nm in Ba I: $|6^1S_0, m_L = 0\rangle \rightarrow |6^1P_1, m_L = 0\rangle$.

Although the calculation has been presented here for a non-decaying lower level, this is no fundamental restriction and it can be extended to the case of two decaying levels.

Finally, with the present development of tunable cw lasers in the infrared, one might expect to apply parametric resonances to the measurement of electric dipole matrix elements in molecules, where the radiative lifetime is too long to be measured directly.

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