NON-LINEAR EFFECTS AND COHERENCE PHENOMENA IN OPTICAL PUMPING

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The shape of resonances observable on atoms and corresponding to the resonant absorption of one optical or RF photon is modified when the intensity of the electromagnetic wave increases : the resonance is shifted and broadened; the variation of its amplitude is no more described by a simple power law. Furthermore, new higher order resonances appear, corresponding to the absorption of several photons by the same atom.

Observation of these non-linear effects becomes easier now, even in the optical range, as an increasing number of high intensity sources is available, and this explains the renewal of interest in the theoretical study of these effects.

I think however that some misleading assertions have recently been published on this subject. I would like in this paper to discuss some of these assertions.

Although the problem of atoms interacting with laser beams seems very different from the one of spins submitted to intense RF fields, the determination of the resonance characteristics is actually very similar in both cases : we have to evaluate the transition probability between 2 energy levels of a system under the influence of a sinusoidal perturbation. The finite number of energy levels in the case of spins simplifies the calculations. This explains why I have chosen to illustrate the discussion by considering some magnetic resonances observed in optical pumping experiments, in particular the "coherence resonances" which result from the preparation of atoms in a coherent superposition of Zeeman sublevels. These resonances turn out to be, in some cases, a very

convenient tool for the study of higher order non linear effects.

In the first part of this paper, I will consider the problem of higher order terms in the radiative shift of a resonance; I will then discuss the possibility of getting non perturbative expressions for the amplitude of multiphoton processes.

I. HIGHER ORDER TERMS IN THE RADIATIVE SHIFT OF A RESONANCE

The radiative shift of a resonance is, at the lowest order, proportional to the intensity of the electromagnetic wave which gives rise to this resonance. For example, the Bloch-Siegert shift of the ordinary magnetic resonance induced by a linear RF field $\vec{B}_1 \cos\omega t$ perpendicular to the static field $\vec{B}_0$ is proportional to B_1^2 ([1]).

Chang and Stehle ([2]) have recently presented a quantum electrodynamics (Q.E.D.) calculation of the higher order terms of the Bloch-Siegert shift (terms in B_1^4, B_1^6...). Their result, which exhibits a kind of oscillatory behaviour, disagrees with those of several other theoretical approaches, using a classical description of the RF field and called, for that reason, "semi-classical" ([3]) ([4])([5]). Chang and Stehle's conclusion is that semi-classical theories are not the appropriate limit of Q.E.D. at high intensities.

It seems very strange that pure quantum effects could appear in the RF domain where the number of quanta is extremely large. The same remark applies to the optical domain : the more intense is the optical wave, the less expected are the pure quantum effects. This does not mean that semi-classical theories are the only ones to be considered in these cases. We have recalculated ([6]) independently the higher order terms of the Bloch-Siegert shift using a quantum description of the RF field and the formalism of the "dressed atom" theory ([7])([8]). Our results are in complete agreement with those of "semi-classical" theories. We have also performed an experimental check ([9]) of the theory by measuring with great precision the position of a coherence resonance observable in transverse optical pumping experiments. The higher order terms in the radiative shift of this resonance, obtained from our calculation, fit very well the experimental results.

The misleading result of Chang and Stehle could give the impression that, in the RF domain, fully quantum mechanical calculations are more difficult to work out than semi-classical ones. This is not our opinion. We think on the contrary that, for high order effects, a correct quantum description of the field leads to calculations which are simpler as they proceed from time independent perturbation treatments.

To illustrate this last point, let's now briefly analyse our method of calculating the higher order radiative shifts.

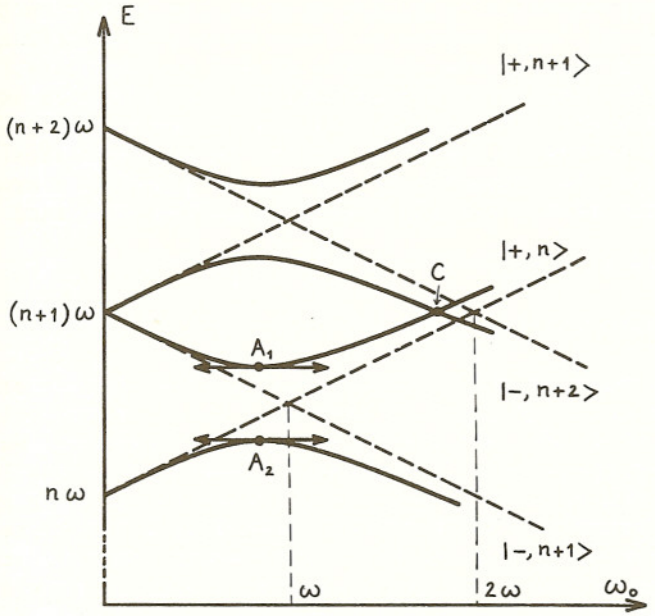

Fig. 1 : Energy levels of the system "spin + RF photons"

The dotted lines of figure 1 represent the unperturbed energy levels of the total system "spin + RF photons", drawn as a function of ω_0 which is the Larmor pulsation in the static field B_0. The $|+, n>$ state for example corresponds to the spin in the $|+>$ state in the presence of n photons; the unperturbed energy of this state is $(\omega_0/2) + n\omega$ where ω is the RF angular frequency. The coupling V between the spin and the field perturbs these energy levels and leads to the energy diagram $E(\omega_0)$ represented by the full lines of fig. 1. The various magnetic resonances observable on the spin correspond to the "anticrossings" and to the crossings on this diagram.

For example, the first anticrossing which arises from the repulsion between the levels originating from $|a> = |+, n>$ and $|b> = |-, n+1>$ is associated to the ordinary magnetic resonance (transition from $|->$ to $|+>$ by absorption of one RF quantum). It can be shown that the center of the resonance is given by the abscissa of the extremum A_1 (or A_2) of the curve $E(\omega_0)$. The difference between this abscissa and ω (abscissa of the crossing formed by $|a>$ and $|b>$) is nothing but the radiative shift and comes from the coupling induced by V between $|a>$ and $|b>$ and the other unperturbed levels.

As shown in reference ([6]), a degenerate perturbation treatment using a Wigner-Brillouin expansion leads to a system of 2 implicit equations giving exactly the abscissa ω_0 and the ordinate E of extremum A_1 . Solving these 2 equations by iteration, one gets ω_0 to any desired order. The matrix elements of the coupling V can be easily related to the parameter ω_1 which, in semi-classical theories, represents the frequency of the Larmor precession around the RF field B_1. Our result for the centre ω_0 of the resonance is

$$\omega_0 = \omega - \frac{1}{\omega}\left(\frac{\omega_1}{4}\right)^2 - \frac{5}{4\omega^3}\left(\frac{\omega_1}{4}\right)^4 - \frac{61}{32\omega^5}\left(\frac{\omega_1}{4}\right)^6 + \ldots \qquad (1)$$

in agreement with the result of semi-classical theories.

It is not easy to check experimentally these higher order effects on the ordinary magnetic resonance which is not only shifted, but also broadened and distorted when ω_1 increases. We preferred studying the coherence resonance associated to the level crossing C of figure 1. This resonance, which can be interpreted as a level crossing resonance of the dressed atom ([7])([8]), is observable in transverse optical pumping experiments and presents the great advantage of being shifted appreciably without considerable broadening when the RF field amplitude is increased over a large range.

Calculations similar to the ones mentioned above give the abscissa of C, i.e. the centre of the coherence resonance at any desired order. The result up to sixth order is :

$$\omega_0 = 2\omega - \frac{1}{6}\frac{\omega_1^2}{\omega} - \frac{7}{54\omega^3}\left(\frac{\omega_1}{2}\right)^4 - \frac{103}{2430\omega^5}\left(\frac{\omega_1}{2}\right)^6 + \ldots \qquad (2)$$

The experimental results obtained on ^{87}Rb are compared to these theoretical predictions on figure 2.

The straight line (noted a) corresponds to the lowest order expression for the shift $2\omega - (\omega_1^2/6\omega)$. Curve b corresponds to expression (2). One sees that the experimental precision is sufficient to test the higher order terms (in ω_1^4 and ω_1^6). When ω_1/ω reaches a value (2.40) corresponding to the first zero of the J_0 Bessel function, it is possible to show that the coherence resonance vanishes at the position $\omega_0 = 0$ ([8]). Expression (2) is no more valid. A perturbative treatment of the Zeeman coupling of the spin with the static field is however possible and gives the theoretical curve c.

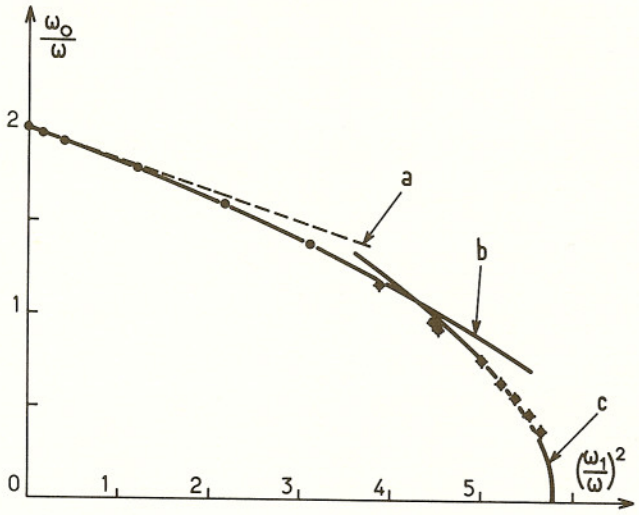

Fig. 2 : Position of the coherence resonance as a function of the RF intensity [measured by $(\omega_1/\omega)^2$]

This good agreement between theory and experiment gives confidence in the theoretical approach described above. Other experimental tests have been performed which check in detail the whole variation with ω_0 of the energy levels represented in fig. 1 ([10]). Let's also mention that the theoretical approach based on continued fractions is very well adapted to a computer calculation of the higher order effects ([5]).

This shows that, for spins, higher order non-linear effects are well understood and can be calculated with a great precision. This is due mainly to the finite number of spin states. In the case of atoms, the greater number of atomic states complicates the perturbation treatment (in particular, the summation over intermediate states). Although some progress has been done in this direction for hydrogen atoms ([11]), it would be very important, for the interpretation of experimental results, to derive new non-perturbative methods. This leads us to the second part of this paper.

II. VALIDITY OF NON-PERTURBATIVE TREATMENTS OF MULTIPHOTON PROCESSES

In 1970, Reiss has proposed a new method for deriving non perturbative expressions for the amplitudes of the multiphoton processes in bound state problems ([12]). The amplitudes appear in a closed form, involving only the initial and final states of the transition; they exhibit a deviation from the simple power law predicted by perturbation theory when the intensity of the electromagnetic wave is sufficiently high. With the powerful laser sources now available, it becomes possible to observe the saturation of the multiphoton processes and this explains the interest for new non perturbative theories : during the last 2 years, a lot of papers have been devoted to the "momentum-translation approximation" (later referred to as m-t-a) developped by Reiss.

Let us briefly outline the idea of the method. An atomic system described by the unperturbed hamiltonian

$$H_0 = \frac{\vec{P}^2}{2m} + V(r) \tag{3}$$

starts at time t_1 from the state $|\phi_i(t_1)\rangle = |\phi_i\rangle e^{-iE_i t_1}$. One switches on adiabatically the potential vector $\vec{A}$ of an incident electromagnetic wave. A time t_2 later, $\vec{A}$ has been switched off. The state vector $|\psi(t_2)\rangle$ of the system is given by the following exact integral equation :

$$|\psi(t_2)\rangle = |\phi_i(t_2)\rangle - i \int_{t_1}^{t_2} e^{-iH_0(t_2-\tau)} H'(\tau) \, |\psi(\tau)\rangle \, d\tau \tag{4}$$

where

$$H' = -\frac{e}{m} \vec{A} . \vec{p} + \frac{e^2}{2m} \vec{A}^2 \tag{5}$$

describes the coupling between the atom and the wave. The transition amplitude from $|\phi_i(t_1)\rangle$ to another field-free state $|\phi_f(t_2)\rangle = |\phi_f\rangle e^{-iE_f t_2}$ is finally given by :

$$U_{fi} = \langle \phi_f(t_2) \, |\psi(t_2)\rangle \tag{6}$$

The main problem is to find a non perturbative expression for the state $|\psi(\tau)\rangle$ appearing in the integral of (4).

The idea of m.t.a is to perform a unitary transformation $T(\tau)$ chosen in such a way that the transformed state

$$|\overline{\psi}(\tau)\rangle = T(\tau) \, |\psi(\tau)\rangle \tag{7}$$

obeys a Schrödinger equation in which the coupling between the atom and the field

$$H_I = T \, H' \, T^+ + i \frac{\partial}{\partial \tau} T \tag{8}$$

is sufficiently small to be neglected. The zeroth order solution of this equation

$$|\bar{\psi}^{(0)}(\tau)> = |\phi_i> e^{-iE_i\tau} \tag{9}$$

is then transformed back to give the approximate expression

$$|\psi(\tau)> \sim T^{+}(\tau)\ |\bar{\psi}^{(0)}(\tau)> = T^{+}(\tau)\ |\phi_i> e^{-iE_i\tau} \tag{10}$$

to be substituded for $|\psi(\tau)>$ in the integral of (4).

The unitary transformation chosen by Reiss is

$$T = \exp\ (-ie\vec{A}.\vec{r}) \tag{11}$$

and leads (in the dipole approximation) to the well known coupling

$$H_I = -e\vec{E}.\vec{r}$$

between the electric field of the incident wave and the atomic electric dipole er. It may be shown that for bound state problems, the ratio H'/H_I is of the order of ω_0/ω , where ω_0 is a typical atomic frequency and ω the frequency of the electromagnetic wave. If we are interested in multiphoton processes involving a large number N of photons, ω_0/ω which is of the order of N is large, and H_I much smaller than H'. Even if H' cannot be treated as a perturbation with respect to H_0, this can occur for H_I and is considered by Reiss as the justification for taking the zeroth order approximation for $|\bar{\psi}(\tau)>$.

Finally, the transition amplitude is approximated by

$$U_{fi} = \delta_{fi} - i\int_{t_1}^{t_2} e^{i(E_f-E_i)\tau} <\phi_f|H'(\tau)\ \exp\ [-ie\vec{A}(\tau).\vec{r}]|\phi_i> d\tau \tag{12}$$

and leads, when $|\phi_f> \neq |\phi_i>$ and when a sinusoïdal time dependence is assumed for $\vec{A}(\tau)$, to the following expression for the transition probability per unit time :

$$W_{fi} = 2\pi \sum_N |T_{fi}^{(N)}|^2\ \delta(E_f - E_i - N\omega) \tag{13}$$

where

$$T_{fi}^{(N)} = i^N(E_i-E_f) < \phi_f|J_N(ea\vec{\varepsilon}.\vec{r})\ |\phi_i> \tag{14}$$

(J_N is the N^{th} order Bessel function, a and $\vec{\varepsilon}$ the amplitude and polarization of the incident wave $\vec{A}$).

We think that the approximations made in m-t-a are not correct. Several objections concerning the validity of this method are explicitly described in reference ([14]). We will just summarize here the conclusions of this discussion.

(i) m-t-a does not predict at all any kind of radiative shift of the type described in the first part of this paper (the δ function of 13 gives $N\omega = E_f - E_i$). In addition, enhancements effects, which can occur when an intermediate state is quasi resonant for a p-quanta process with $p < N$, do not appear in the final closed expression of the transition amplitude.

(ii) When an exact solution is known (for a particular choice of the atomic potential $V(r)$), the results disagree with those given by m-t-a.

(iii) The unitary transformation performed by Reiss is in fact a well known gauge transformation ([13]) which, as quantum mechanics is gauge invariant, does not change the physical content of the theory. Treating in the new gauge H_I to zeroth order amounts to incorporate no interaction processes at all in the wave function (10) which approximates $|\psi(\tau)>$ in the integral of (4) (Note for example that the electronic density associated to this approximate wave function coincides with the unperturbed one) . When (10) is used, the only term which represents the coupling in (4) is $H'(\tau)$ and one does not understand how this procedure can describe correctly processes involving more than one photon.

(iv) One can try to improve the precision by approximating $|\psi(\tau)>$ by

$$T^+(\tau) \left(|\overline{\psi}^{(0)}(\tau)> + |\overline{\psi}^{(1)}(\tau)> \right) \qquad (15)$$

which is better than (10) ($|\overline{\psi}^{(1)}(\tau)>$ is the first order contribution in H_I in the perturbation expansion of $|\overline{\psi}(\tau)>$) . One finds that the $|\overline{\psi}^{(1)}>$ term of (15) leads to expressions which have the same order of magnitude as the ones obtained by keeping only $|\overline{\psi}^{(0)}(\tau)>$. m-t-a is therefore non consistent as one neglects terms comparable to the ones which are kept.

The total expression obtained by using (15) is correct only to first order and, at this order, coincides with the results of ordinary perturbation theory. More generally, we have shown that, for a coherent calculation up to N^{th} order (this is the minimum required for interpreting a N-photon process), one has to use for $|\psi(\tau)>$ an expression at least as precise as

$$T^+(\tau) \sum_{p=0}^{N} |\overline{\psi}^{(N)}(\tau)> \qquad (16)$$

(where $|\overline{\psi}^{(N)}(\tau)>$ is the N^{th} order contribution in H_I to $|\overline{\psi}(\tau)>$).

All these considerations show that, for bound states problems, it seems impossible to bypass perturbation theory by performing a unitary transformation and making some crude approximations in the new representation.

One can now ask about the possibility of applying non perturbative methods to bound state problems. It seems that the coupling between a bound system and the electromagnetic field can be treated to all orders at least in 2 simple cases :

a) situations where the "rotating wave approximation" can be used, and which generalize the simple problem of a spin 1/2 interacting with a rotating RF field perpendicular to the static field. This is interesting for the study of the one photon resonances and, more precisely, for an exact treatment of the saturation effects appearing as a result of successive resonant interactions of the same atom with the electromagnetic field;

b) situations where the sinusoïdal interaction hamiltonian is diagonal in the basis of unperturbed states (generalization of the problem of a spin 1/2 interacting with a linearly polarized RF field parallel to the static field).

We will say a few words about this last situation which is less known than the first one, and which has been investigated in some optical pumping experiments.

The diagonal character of the perturbation, which, on the one hand, permits an exact solution of the equations of motion, excludes, on the other hand, any resonant exchange of energy between the atom and the field : the absorption (or induced emission) of a photon is not accompanied by a jump of the atom from one level to another, and the total energy cannot be conserved. However, virtual processes involving one or several photons can occur, but they require, in order to be detected, that we use a second probing electromagnetic wave. So, we are faced with a new problem where the atom interacts with 2 waves (1) and (2) : we can treat to all orders the coupling with wave (1) which can be as intense as we want, but we must restrict to low intensities for the probing wave (2) and use perturbation theory for describing its effect.

As a first example of such a situation, let us mention the transverse optical pumping of atoms interacting with a RF field $\vec{B}_1 \cos\omega t$ parallel to the static field $\vec{B}_0$ (the wave (1) is the RF field, while the optical pumping beam may be considered as playing the role of wave (2)) . The resonances detected on the light absorbed or reemitted by atoms and which appear when the Larmor frequency ω_0 in $\vec{B}_0$ is equal to $n\omega$ ($n = 0, 1, 2 \ldots$), are associated with the level crossings appearing in the energy diagram of the total system spin + RF photons ([7])([8]) (level crossing resonances of the dressed atom). These coherence resonances can also be interpreted as resulting from the interference between 2 different scattering amplitudes ([15]); each of these amplitudes, which starts from the same initial state and ends at the same final state, involve the absorption (or emission) of one optical photon and of an arbitrary

number of RF photons. The variation of the intensity of the resonances as a function of the RF field amplitude has been calculated exactly and the saturation behaviour has been experimentally checked in great detail ([16]). This shows, as in the first part of this paper, the interest of coherence phenomena associated to transverse optical pumping experiments for the study of higher order non linear effects.

As a second example, I will mention the study by Haroche of the transitions between the 2 hyperfine levels of an alkali atom and corresponding to the absorption of one microwave photon and several radiofrequency photons ([8]). The coupling with the microwave (wave 2) is treated to first order; the coupling with the RF field (wave 1) is treated to all orders (this coupling is diagonal in zero static field, and the effect of an applied weak static field is evaluated by perturbation theory). It has been possible to check with a great accuracy all the theoretical predictions concerning the radiative shifts and the saturation behaviour of these multiphoton resonances for arbitrary values of the RF power.

This leads us to the following remark. The calculations of non linear effects observed in the 2 experiments described above are in fact equivalent to a partial resummation of the perturbation series : one sums the contributions of all processes involving only one photon of the wave 2 but an arbitrary number of photons of the wave 1. One might then ask whether it would be possible, in some other cases, to pick out by some physical arguments a certain class of processes which are the most important for the phenomenon being studied, and to sum up all the corresponding contributions. There is perhaps in this direction a possibility for getting the non perturbative expressions which are urgently needed at the present time.

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