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WINTER COLLEGE ON ATOMIC AND MOLECULAR PHYSICS

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INTRODUCTION TO QUANTUM OPTICS: Part III

QUANTUM OPTICS: COHERENT RESONANT SPECTROSCOPY

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3 - Quantum Optics : Coherent resonant spectroscopy

3.1 Introduction

In this Part we survey the relevant spectroscopic effects induced by a coherent field near resonance to an atomic or molecular transition.

The main outcomes are

1) precise measurements of electric dipole strengths
   $\mu = \langle 2 | e \vec{r} | 1 \rangle$
2) lifetime measurements (spontaneous; collision broadening and its behaviour versus gas pressure which sheds light on interatomic potentials)
3) absolute positioning of atomic levels (new wavelength standards, accurate measurements of isotope shifts)
4) fine and hyperfine structure (f s , h f s) : Landè factors, Lamb shift, tensor polarizability
5) metrology : new accurate values for α , Ry , c (finer structure constant, Rydberg constant, light velocity)
6) preparation of particular atomic states (coherent excitation) or field states (modulation)

We shall cover the following subjects:

i) coherent resonant effects { local effects / propagation effects

ii) non linear spectroscopy : saturation and two-photon

iii) perturbed fluorescence spectroscopy.

3.2 The interaction model

Let us consider an e.m. field and a set of atoms confined in a cavity. We discuss how to go from a physical Hamiltonian as

$$H = H_{field} + H_{atoms} + H_{f-a} ,$$

where $$H_{f-a} = -\sum_i \frac{e}{m} \vec{A}(x_i) \cdot \vec{p}_i$$

x_i being the position of atom i , $\vec{A}(\mathbf{x}_i)$ the value of the vector potential at x_i and p_i the momentum of the i-th electron to a suitable Hamiltonian .

We expand the field in plane waves as (take $\vec{A}$ as a scalar)

$$A(x,t) = \sum_k \sqrt{\frac{\hbar}{2 \varepsilon_0 \omega_k V}} \left[a_k e^{-i(\omega_k t - kx)} + c.c \right]$$

(V = cavity volume ; mks system used) and consider a_k and the conjugate a^+_k as Bose operators

$$\left[a_k , a^+_{k'} \right] = \delta_{kk'}$$

For simplicity, we skip vector relations and give a scalar theory. ω_k and k are related by the dispersion relation

$$\omega_k = c\, k$$

We consider the atoms as two-level atoms, so that the Hilbert space of a single atom is fully described by the identity operator I plus the three Pauli operators

$$\sigma^+ , \sigma^- , \sigma_3$$

$$\left[\sigma^+ , \sigma^- \right] = \sigma_3 \quad , \quad \left[\sigma^\pm , \sigma_3 \right] = \mp 2 \sigma^\pm$$

It is then a straightforward matter to obtain the following model Hamiltonian

$$\frac{H}{\hbar} = \sum_k \omega_k a_k^+ a_k + \frac{\omega_0}{2} \sum_{i=1}^{N} \sigma_3(i) +$$

$$+ \sum_{k,i} g_k \left(a_k \sigma_i^+ e^{ikx_i} + a_k^+ \sigma_i^- e^{-ikx_i} \right) \qquad (3.1)$$

We introduce the collective operators.

$$J_k^{\pm} = \sum_i \sigma_i^{\pm} e^{\pm ikx_i}$$

$$J_z = \sum \sigma_3(i) \qquad (3.2)$$

When the space dependence can be neglected, that is, for the three cases

i) point laser (cavity length $\ll \lambda$)

ii) single mode case (one $k = k_0$ value)

iii) traveling plane wave field (again, one k_0 value)

one has closed commutation relations for $J^{\pm} = \sigma_i^{\pm}$ and J_3 , and the Heisenberg equations of motion are obtained.

$$\dot{a} = -i\omega a - ig J^-$$

$$\dot{J}^- = -i\omega_0 J^- + ig a J_2 \qquad (3.3)$$

$$\dot{J}_2 = -ig\left(a J^+ - a^+ J^-\right)$$

They do not form a closed set, because one would need also an eq. for the motion of bilinear operators as a J^+ etc.

We here study the S C F A (self consistent field approximation)

$$\text{S C F A :} \quad \langle aJ \rangle \simeq \langle a \rangle \langle J \rangle \qquad (3.4)$$

Table 3.1

classical current	Maxwell →	field state	$\langle a \rangle \neq 0$ ← coherent states
" field	Bloch →	atomic "	$\langle J^{\pm} \rangle \neq 0$ ←

For discussion of coherent states we refer to Part IV. Here we want to discuss eq. (3.5), analogous to the Bloch eq. in N M R, which is the vector form of the atomic equations in (3.3) once averaged with the SCFA:

$$\frac{d}{dt} \langle \vec{J} \rangle = \vec{\Omega} \times \langle \vec{J} \rangle \qquad (3.5)$$

$$\Omega \equiv \left(g\langle a \rangle, \; 0, \; \omega_0 \right)$$

3.3 The two-level atom

Under what conditions can a real atom be approximated by a two-level atom ? Consider e.g. the allowed electric dipole Dlines of Na atom

$I = 3/2$

$\vec{F} = \vec{I} + \vec{J}$

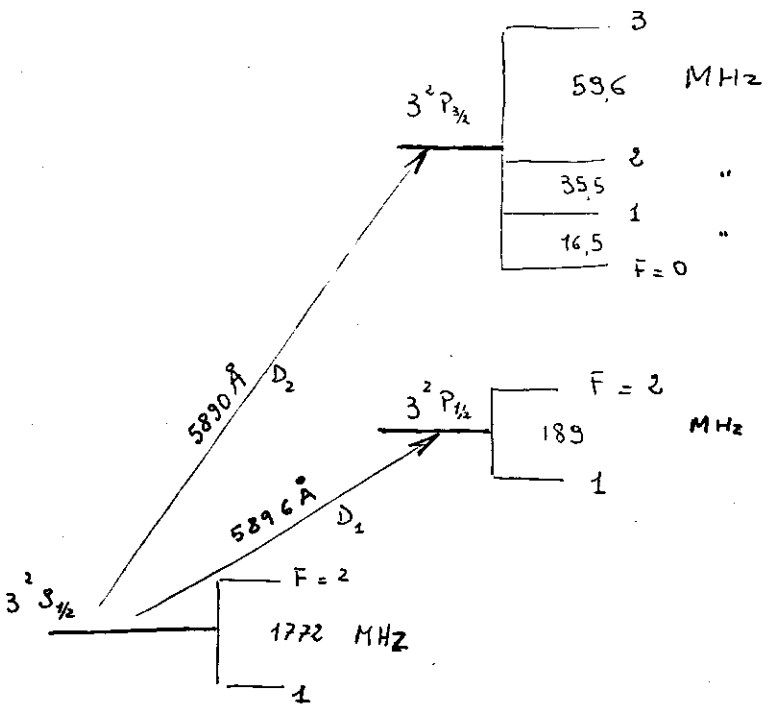

Fig. 3.1a

$$3p - 3s \begin{cases} {}^2P_{3/2} - {}^2S_{1/2} & 5890 \text{ Å} \\ {}^2P_{1/2} - {}^2S_{1/2} & 5896 \text{ Å} \end{cases}$$

In dilute gas (10^{12} atoms / cm^3) the collision time is $\sim 10^{-7}$ sec. The associated homogeneous linewidth is

$$\frac{1}{T_2} = \delta\omega_c \sim 10^8 \ s^{-1}$$

Further, there is an inhomogeneous Doppler broadening which is typically

$$\frac{1}{T_2^*} = \delta\omega_D \sim \frac{\omega}{c}\sqrt{\frac{KT}{M}} \sim 10^{10}\ s^{-1}$$

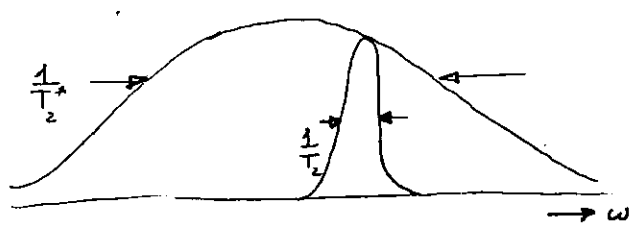

Fig. 3.1b

The nuclear spin I = 3/2 combines with J to produce hfs. Each of the F = I + J hfs levels is then magnetically degenerate (2 F + 1) times.

If we want to study coherent excitation on the $^2S_{1/2}$ (F = 1) to $^2P_{3/2}$ (F = 2) transition avoiding overlap with F = 1 , the pulse must be narrower than 35.5 MHz , that is, the pulse duration must be longer than 0.5×10^{-8} s.

On the other hand, coherence lasts for less than the collision time, so $\bar{t}$ pulse $< 10^{-7}$ sec.

Only under such limits we have coherent 2 level excitation.

3.4 The Bloch equations

We approach the Bloch eq. directly by Schrödinger, rather than Heisenberg, method. This way, we attribute a physical meaning to the 3 components of $\vec{J}$.

The time dependent wave function

$$\psi = a(t)|+\rangle + b(t)|-\rangle$$

$$\mu \equiv \langle + | e\vec{r} | - \rangle \qquad (3.6)$$

of a 2 level atom, under a coherent field excitation,

$$E = \mathcal{E} \cos \omega t \qquad (3.7)$$

obeys the Schrödinger eq. We set up a linear combination

$$\vec{J} \equiv \begin{pmatrix} a^* b + b^* a \\ -i\left(a^* b - b^* a\right) \\ |a|^2 - |b|^2 \end{pmatrix} \qquad (3.8)$$

of the two complex amplitudes a and b which weight the eigenstates + and - of the free atom Hamiltonian. It obeys the vector equation (3.6)' , which is as (3.5)

$$\dot{\vec{J}} = \vec{\Omega} \times \vec{J} \qquad \Omega \equiv \left(-\frac{\mu \mathcal{E}}{\hbar}, 0, \omega_0\right), \qquad (3.6)'$$

by evaluating

$$\langle \psi | e\vec{r} | \psi \rangle$$

with the help of (3.6), it is easy to see that the three components of J have the following meaning

J_1 is proportional to the in-phase polarization (dispersion)

J_2 is proportional to the out-of-phase polarization (absorption or gain)

J_3 is proportional to the population inversion

We then write $\cos \omega t = \frac{e^{i\omega t} + e^{-i\omega t}}{2}$, go to a frame rotating around z at angular velocity ω_0

$$\Omega = \begin{pmatrix} -\frac{\mu}{\hbar}\mathcal{E} \\ 0 \\ \Delta = \omega - \omega_0 \end{pmatrix} \qquad (3.9)$$

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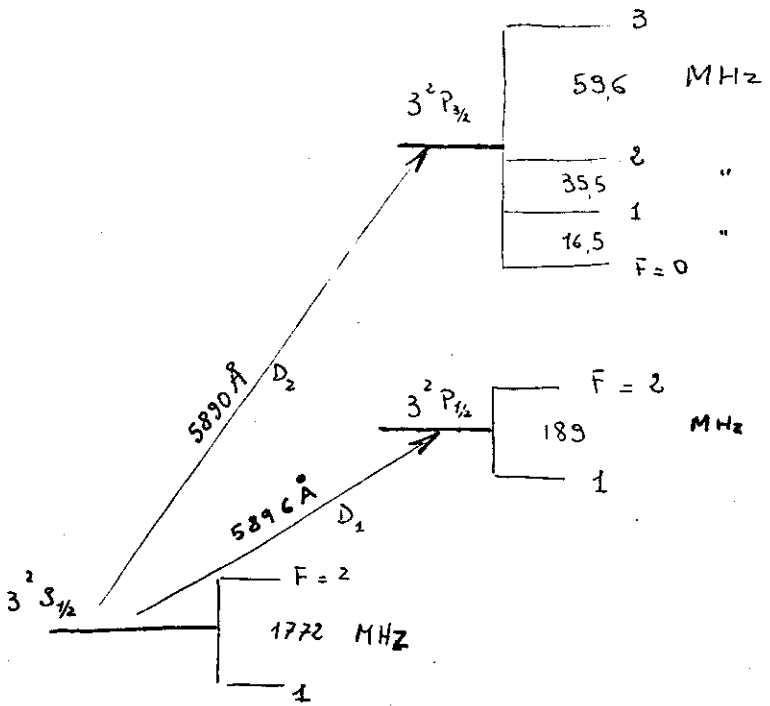

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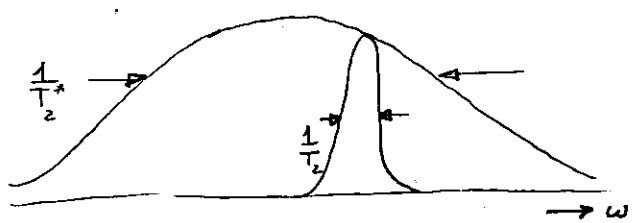

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We then write $\cos \omega t = \frac{e^{i\omega t} + e^{-i\omega t}}{2}$, go to a frame rotating around z at angular velocity ω_0

$$\Omega = \begin{pmatrix} -\frac{\mu}{\hbar} \mathcal{E} \\ 0 \\ \Delta = \omega - \omega_0 \end{pmatrix} \qquad (3.9)$$

which will yield as solutions Jacobi elliptic functions. In particular we may have a hyperbolic secant solution. We emphasize the role of the cooperation time τ_c

$$\boxed{\frac{1}{\tau_c^2} = \frac{\omega N \mu^2}{\hbar \varepsilon_0}} \qquad (3.17)'$$

It is the characteristic time of rad. - atoms interaction.
For a linearized problem ($w = -1 = \text{cont}$), (3.17) reduces to $\ddot{\Theta} = -\Omega^2 \Theta$

$$\boxed{\Omega^2 = N e^2 / m \varepsilon_0} \qquad \text{where now} \qquad (3.17)''$$

is the usual plasma frequency for a gas of Lorentz oscillators. Here, m is the electron mass, and μ has been replaced by the classical cross section for free electrons.

In propagation, we transform to a coordinate frame moving at velocity V_0 and look for undistorted pulses or solitons ($\frac{\partial}{\partial \zeta} = 0$).

$$(z\ ,\ t\ ) \longrightarrow \left(\zeta = z\ ;\ \tau = t - \frac{z}{V_0}\right) \qquad (3.18)$$

From (3.14) it is an easy matter to obtain (3.19) and hence (3.20).

$$\left(1 - c/V_0\right) \frac{\partial \mathcal{E}}{\partial \tau} = \frac{\omega N \mu}{2 \varepsilon} \mathcal{V} \qquad (3.19)$$

$$\ddot{\Theta} = \frac{1}{1 - \frac{c}{V_0}} \frac{1}{\tau_c^2} \sin \Theta \qquad (3.20)$$

The hyperbolic secant solution of the soliton equation (3.20) implies the following constrain between the pulse velocity V_0 and the velocity c in vacuum, the linear attenuation α (→ attenuation) and the pulse duration τ_p (→ pulse duration):

$$\frac{1}{V_0} = \frac{1}{c} + \frac{1}{2} \alpha \tau_p \qquad (3.21)$$

Further, it is easy to prove

1) the area of the radiation pulse obeys the space propagation eq.

$$\Theta \equiv \int_{-\infty}^{+\infty} \frac{\mu}{\hbar} \mathcal{E}\, dt\ ;\ \frac{\partial \Theta}{\partial z} = -\frac{\alpha}{2} \sin \Theta(z) \qquad (3.22)$$

which gives solitons for 2π, 4π etc.

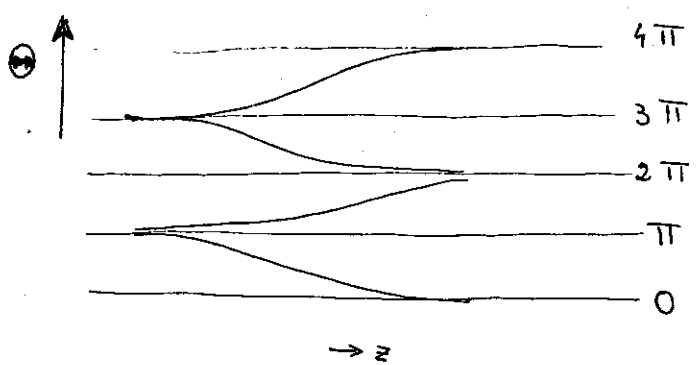


Fig. 3.4

2) The 2π pulse corresponds to an atomic system starting from the South Pole and returning to the South Pole without global exchange of energy with the field (S I T = self induced transparency). The pulse velocity is (3.21) where α is the linear attenuation

$$\underset{cm^{-1}}{\alpha} = \underset{cm^{-2}}{\sigma} \quad \underset{cm^{-3}}{N}$$

3.5 Irreversible processes in presence of dampings

Let us introduce phenomenological lifetimes T_2 for polarization components, T_1 for inversion ($T_2 \leq T_1$), and $1/k$ for the field. In case the material is confined in a region of size L without mirrors then $1/k$ is the transit time L/c.

Further, let us introduce, besides the atoms and field, a third partner, the "pump" which in the absence of field yields a steady state value of w

$$w = w_0$$

The Bloch-Maxwell eqs. become

$$\frac{\partial}{\partial t}\begin{pmatrix} u \\ v \\ w \end{pmatrix} = \begin{pmatrix} \Delta \cdot v & -u/T_2 \\ -\Delta \cdot u + \mu \mathcal{E} w & -v/T_2 \\ -\mu \mathcal{E} v & -\frac{w - w_0}{T_1} \end{pmatrix} \qquad (3.22)$$

$$\left(\frac{\partial}{\partial t} + \varepsilon \frac{\partial}{\partial z}\right)\mathcal{E} = \frac{\omega N \mu}{\hbar \epsilon} v - k \mathcal{E} \qquad (3.23)$$

In the absence of a pump, we must put

$$w_0 = -1$$

because the atoms will spontaneously decay toward the South Pole of the Bloch sphere, with a lifetime T_1.
If $T_2 \ll (T_1, 1/k)$, we can solve at steady state the eqs. for u, v and have two coupled rate - eqs. for w, $\mathcal{E}$.

$$\begin{cases} \dot{w} = \dfrac{-\alpha}{T_1} I w - \dfrac{w - w_0}{T_1} \\ \left(\dfrac{\partial}{\partial z} + \dfrac{1}{c}\dfrac{\partial}{\partial t}\right) I = -\alpha I \end{cases} \qquad (3.24)$$

where

$$\begin{cases} I \equiv \left(\dfrac{\mu \mathcal{E}}{\hbar}\right)^2 T_1 T_2 & \text{(normalized intensity)} \\ \alpha = \dfrac{1}{1 + \Delta^2 T_2^2} & \text{(Lorentzian line shape)} \end{cases} \qquad (3.24)'$$

Many approaches to lasers are based on (3.24). On the other hand, if the escape time of the field is faster than T_1 , T_2) We solve (3.23) at steady state, replace in (3.22), neglect atomic losses and get

$$\dot{\Theta} = \frac{1}{K\tau_c^2} \sin\Theta \qquad (3.25)$$

whose solution is

$$\mathcal{E} \sim \operatorname{sech}\left(\frac{\tau - \tau_M}{\tau_R}\right) \qquad (3.26)$$

with

$$K\tau_c^2 \equiv \tau_R \simeq \frac{\tau_{sp}}{N\lambda^2 L} \quad (\text{pulse width}) \qquad (3.27)$$

(†)

$$\tau_M = \tau_R \ln N \quad (\text{pulse delay}) \qquad (3.28)$$

Here, time $\tau = t - z/c$ is the local time. We have a soliton solution or π pulse.

If no applied field is present but the atoms are prepared in the excited state, we have spontaneous π pulse (superradiance) after a time τ_M.

(†) $\tau_{sp}^{-1} \sim \frac{M^2}{\lambda^3 \hbar \varepsilon_0}$ is the spontaneous life time, τ_c was given in eq. (3.17)', K is replaced by c/L

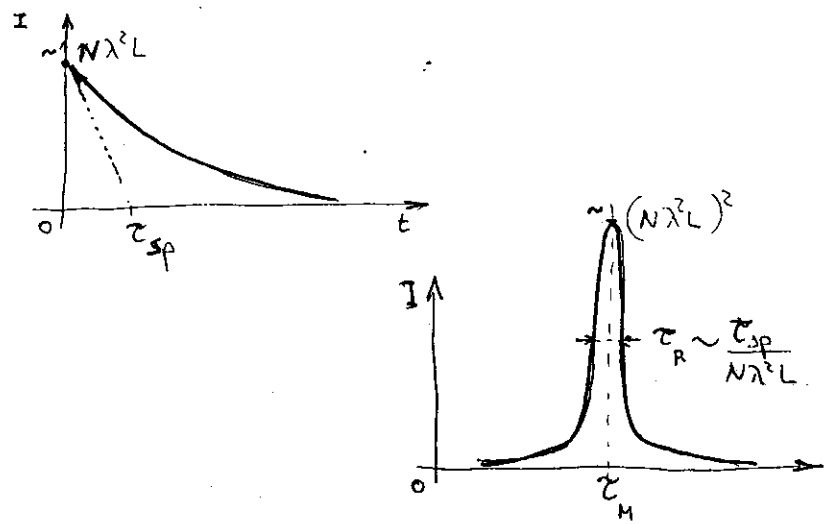
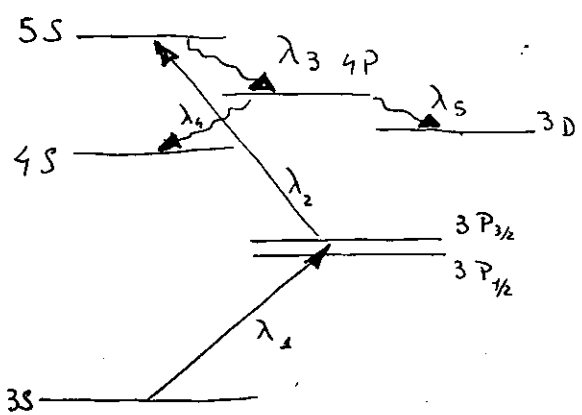


Fig. 3. 5

In this figure we compare spontaneous uncorrelated (Weisskopf - Wigner) and correlated emissions.

Fig. 3.6 shows the experiment by Haroche et al.



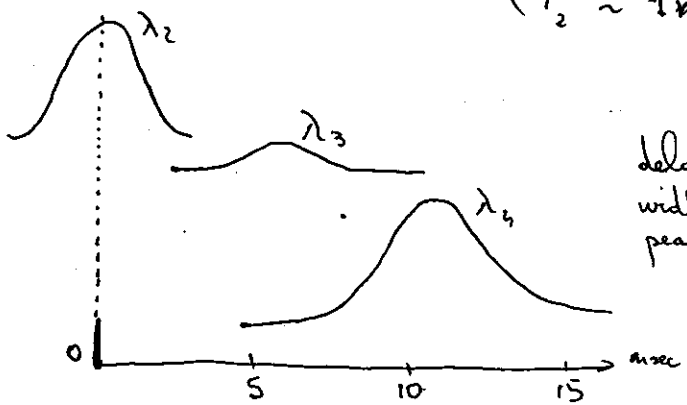
$\lambda_3 = 3.41\,\mu$

$\lambda_4 = 2.21\,\mu$

$$\tau_R = \frac{1}{W_c^2} \sim \frac{\tau_{sp}}{N \mathcal{L} \lambda^2} < T_2^*$$

for $N \sim 3 \cdot 10^9\ cm^{-3}$

($T_2^* \sim 1$ msec)



delay, width, peak } agree with theory

3.6 Saturation and non-linear spectroscopy

The steady state value of w has an intensity dependent denominator (see solution (3.29) of eq.(3.24)

$$W(\Delta) = \frac{W_0}{1 + \mathcal{L} I} \xrightarrow{W_0 = -1} - \frac{1 + (\Delta \cdot T_2)^2}{1 + (\Delta \cdot T_2)^2 + I} \qquad (3.29)$$

where $\mathcal{L}$ and I are given in eq (3.24)' .

As we shine a strong laser field in a Doppler broadened line, we dig a hole in the line, whose width depends on T_2 and I, but in anyway smaller than $1 / T_2^*$. By a probe field, we can detect the hole, localizing transitions better than in regular linear spectroscopy (limited by the Doppler width).

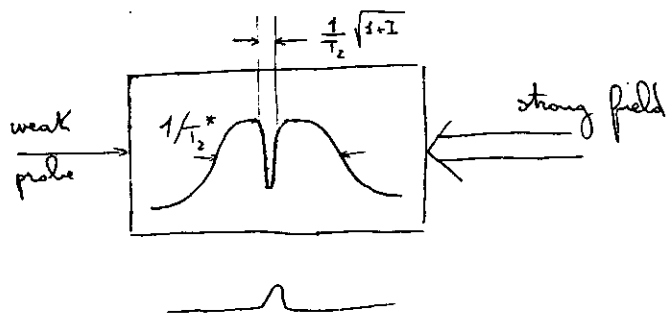


Fig. 3.8

3.7 Two photon spectroscopy. Comparison with saturation

A transition between two levels of equal parity, as 3 s ⟶ 4 s in Na , forbidden for one photon, becomes allowed for the simultaneous absorption of two photons. If two lasers are shined in opposite directions into a cell, the Doppler shifts cancel and we have a resonance exactly at the same frequency:

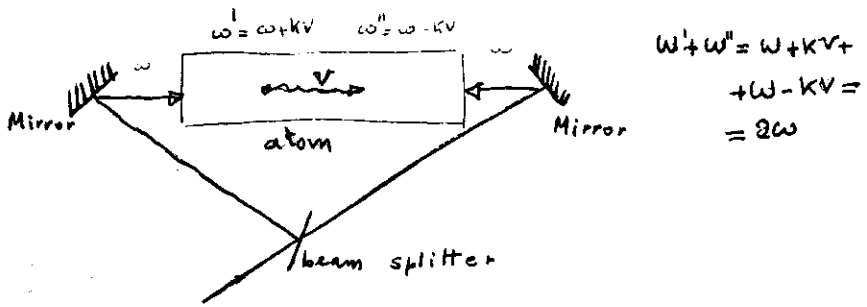


Fig. 3.9

We show a comparison between the two methods, and list the applications.

Table 3. 2

Two photon spectroscopy

vs. saturation

+	contrast 100%
+	no saturation broadening
-	level shifts (a.c. Stark)
+	no recoil
-	2^{nd} order Doppler (red shift $\propto v^2$)

Table 3. 3

Applications

1	fs , hfs
2	splitting in low B , E fields (Zeeman, Stark)
3	collisional broadening & shift at low pressures
4	selective population of single level within Doppler line
5	Isotope shift
6	precise position levels ⟶ metrological standards

3.8 Perturbed fluorescence spectroscopy

Hitherto, we have described systems which decay after preparation by a laser pulse.

Here we consider cases whereby the atoms, after laser preparation, undergo an extra perturbation. This could be an applied B or E field (Zeeman or Stark splitting), internal couplings of the constituent angular momenta of the atom, atomic collissions etc. We restrict our attention to detection of polarized or anisotropic light.

In particular, for simultaneous excitation of two f. s. levels, we can consider three techniques:

- quantum beats
- double resonance (light + r. f.)
- modulated light beam

These are illustrated in figures 3.10 and 3.11, and comparisons are given in the captions.

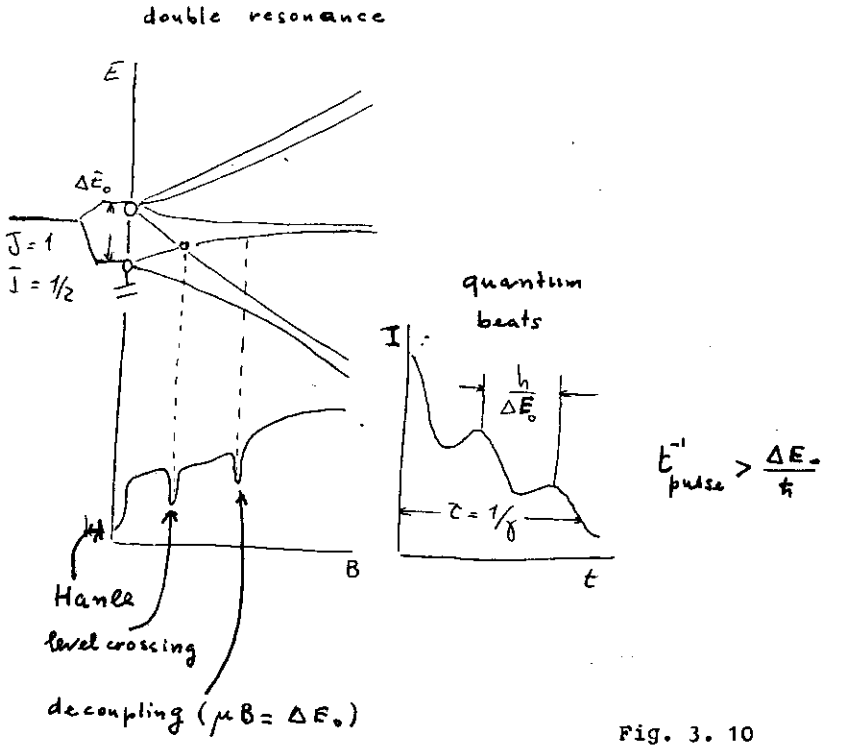


Fig. 3. 10

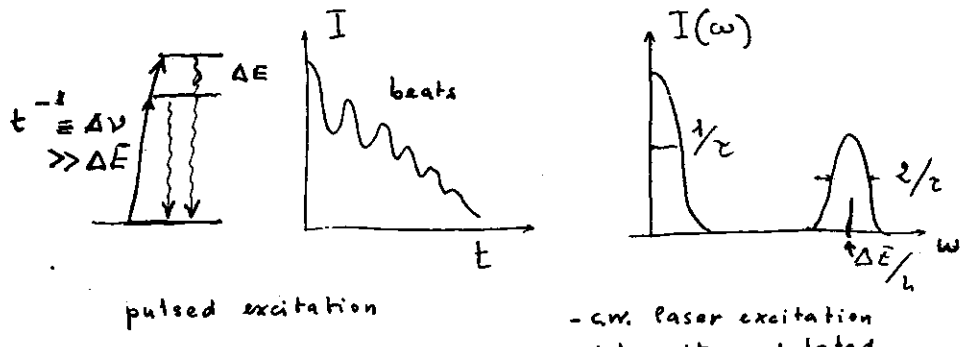


Fig. 3. 11

3.9 Dynamic Stark shift

Spontaneous fluorescence after laser excitation is predicted to have a three peaked spectrum

- Semiclassical $(\langle E^- E^- \rangle \sim \langle E^+ \rangle \langle E^- \rangle)$

$$I(t) = I_0 \left[e^{-i\omega_c t} + \frac{1}{2} e^{-\frac{3}{4}\gamma t} \left(e^{-i\left(\omega_c \pm 2\frac{\mu\mathcal{E}}{\hbar} t\right)} \right) \right] \qquad (3.31)$$

- **Quantum.Above a threshold**

$$\frac{\mu\mathcal{E}}{\hbar} \geq \gamma/4 = 1/T_2 \qquad (3.32)$$

There are stationary correlations given by a power spectrum (see fig. 3.12)

$$I(\omega) = \mathcal{F}\left\{ G^{(2)}(\tau) \propto 3 e^{-(\gamma/2 + i\omega_c)\tau} + \right. \qquad (3.33)$$

$$\left. + 1 e^{-\left[\frac{3}{4}\gamma + i\left(\omega_c \pm \frac{\mu\mathcal{E}}{\hbar}\right)\right]\tau} \right\}$$

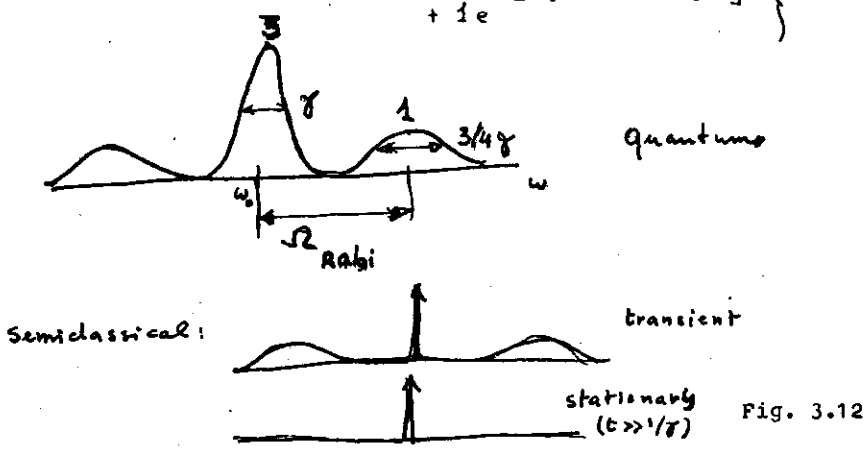


Fig. 3.12

- This last equation suggest correlation spectroscopy as an alternative to ordinary spectroscopy.

The corresponding outcomes are depicted in fig. 3. 13

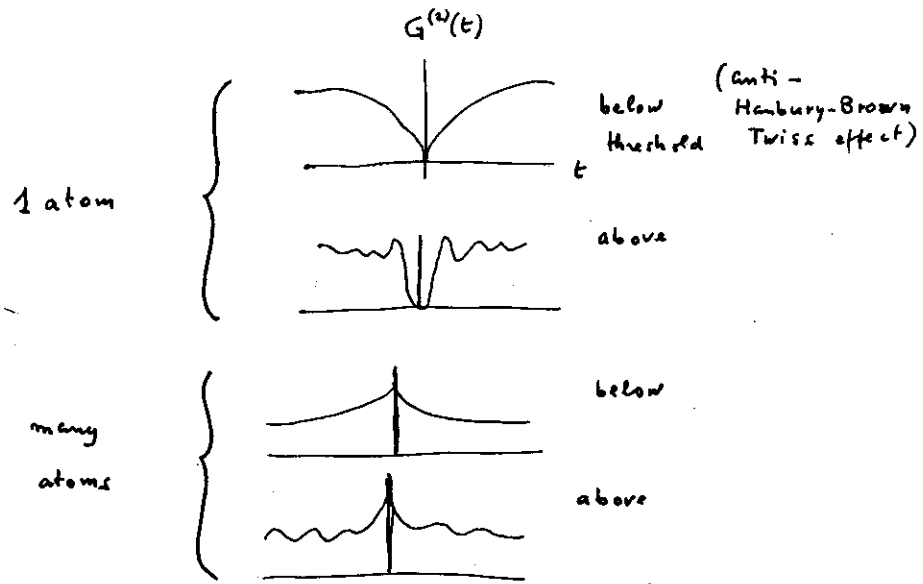


Fig. 3. 13