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WINTER COLLEGE ON LASERS, ATOMIC AND MOLECULAR PHYSICS

(21 January - 22 March 1985)

Topical Meeting on Multiphoton Processes

MULTIPHOTON PROCESSES IN ATOMS

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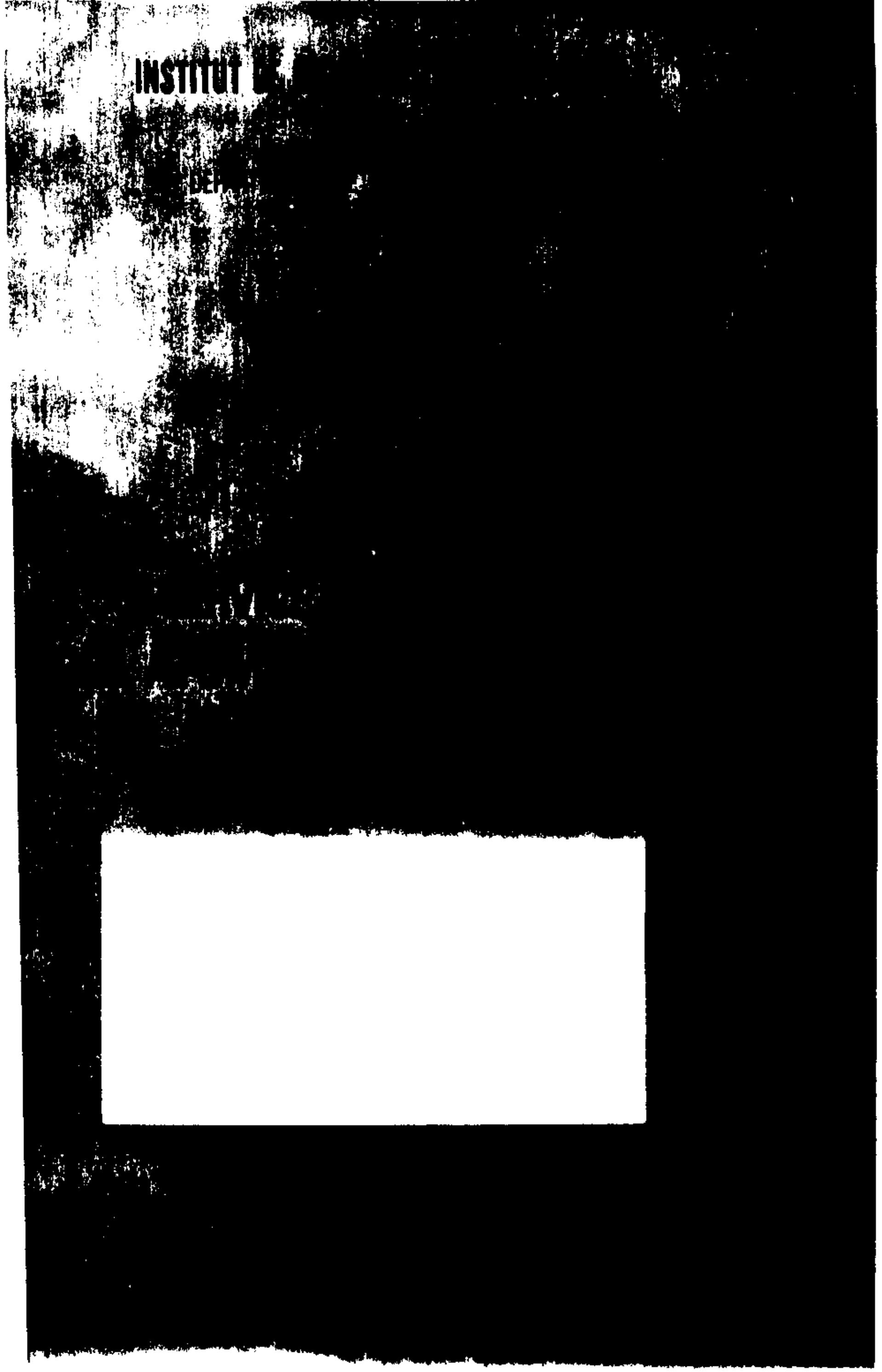
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(TRIESTE)

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MULTIPHOTON PROCESSES IN ATOMS

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MULTIPHOTON PROCESSES IN ATOMS

HISTORICAL BACKGROUND

Long story which began in 1931

1931 M. Göppert-Mayer - theory of 2 photon absorption

1953 B. Cagnac }
1959 J. Winter } Observation of M.A. of R.F. photons (Zeeman sublevels of Na)

------------------------------ Laser ------------------------------

1961 Kaiser and Garrett - 2 photon excitation of CaF_2 in optical range

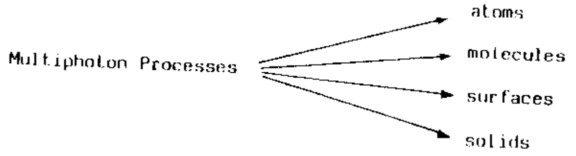
1962 Abella - 2 photon (ruby laser) absorption in C_s vapour

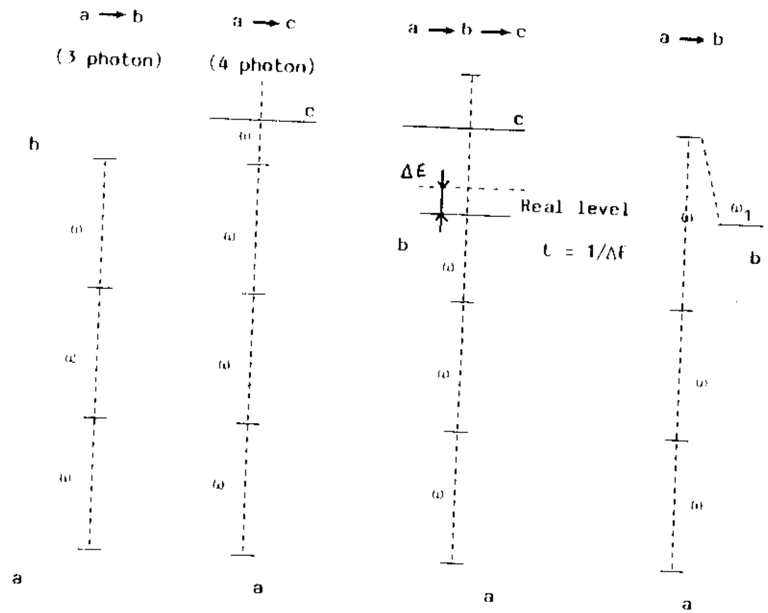
1966 Delone et al. }
1968 Saclay group } M.P. ionization in rare gases

1968 C. Cohen-Tannoudji described the dressed atom formalism which played an essential rôle in theoretical developments

1972-1974 Chebotayev and Cagnac - Spectroscopy without Doppler effect by M.A.

GENERAL CONSIDERATIONS





Bound-Bound

Virtual levels

$t_{life} = \frac{1}{\omega}$

(Incertitude principle)

10^{-15} sec.

Bound-free Non resonant Ionization (Dissociation)

Bound-free Resonant Ionization (Dissociation)

$t_{life} = \frac{1}{\Delta E}$

at resonance

$t_{life} = 10^{-8}$ 10^{-9}

Bound-bound Raman processes

If we call σ the cross-section of an absorbed photon on a virtual state, τ the lifetime of the excited state, F the photon flux, the absorption probability is $(\sigma F).\tau$, with $\tau = 1/\Delta E$.
If 2 photons are absorbed the overall probability of absorption of the 2 photons will be:

(1) $W_2 = (\sigma F)\tau . \sigma F = \sigma^2 . F^2 . \tau$

For an N photon absorption:

(2) $\boxed{W_N = \sigma^N F^N . \tau^{N-1}}$

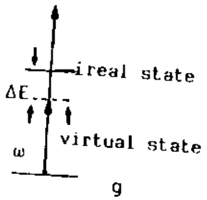


Fig. 1

Multiphoton absorption is an extremely non-linear process, W_N is proportional to F^N.

Let us consider a 2 photon absorption, the energy of the photon is tuned through resonance, τ varies accordingly at resonance W_2 is maximum

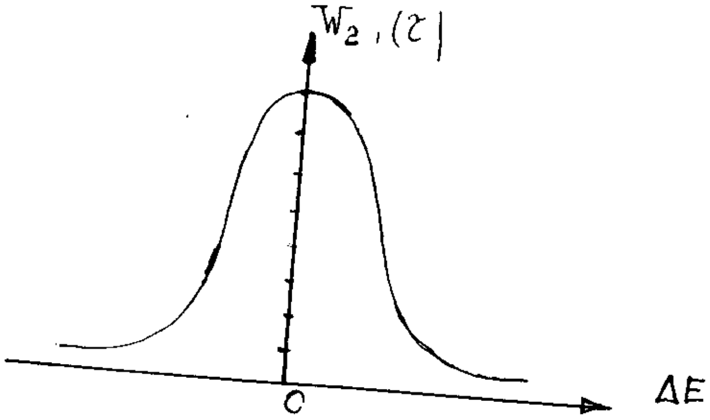


Fig. 2

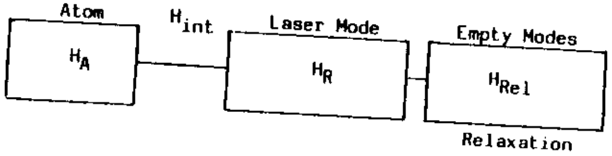
THEORETICAL DESCRIPTION (Dressed atom)

In 1966, Gold and Bebb have developed a time dependent perturbation theory using the concept of evolution operators (in interaction representation). This method is very powerful but fails in resonant situation.

We will use the resolvent operator method, operator technique (Messiah, Golberger, Watson, Mower).

The Atom is coupled to an E.M. field (laser) -
- a field of frequency ω, polarization $\vec{\epsilon}$, n being the occupation mode.

The Dressed Atom is the quantum system formed by the atom and the photon beam in mutual interaction.



Hamiltonian

(3) $$\boxed{H = \overbrace{H_A + H_R}^{H_o} + H_{int}}$$

We do not consider fluorescence here

$H_{rel} = 0$

(4) $$H_R = \sum_{k\epsilon} a^+_{k\epsilon} a_{k\epsilon} \omega_k$$

$\vec{k}$ wave vector

$\vec{\epsilon}$ polarization index

$\omega_k = ck$

Eigenstates of H_R are $| \, .. n_1(k_1 \varepsilon_1), \, n_2(k_2, \varepsilon_2) \ldots >$
or $| \, n(k\varepsilon)>$ with $n(k\varepsilon)$ the number of photons occupying the $(\vec{k\varepsilon})$ mode, (Fock states).

Non coupled states (for t = 0)

$\psi(t=0) = |i> \otimes |n_{k\varepsilon}>$ sometimes written $|i,n>$ in abbreviated way

$\psi(t=0) = |I>$

Energy of non coupled states: $E_i + \sum_{k\varepsilon} n_{k\varepsilon}\omega_k$ (discrete state)
$E + \sum_{k\varepsilon} n_{k\varepsilon}\omega_k$ (continuum state)

Interaction Hamiltonian

In dipole approximation, valid if $\vec{k} \cdot \vec{p} << 1$, $\vec{p}$ impulsion

$H_{int} = \vec{E} \cdot \vec{D}$ with $\vec{D} = e\vec{r}$ (position operator) (5)

$\vec{E}$ the electrical field of the wave in Schrödinger representation ($\vec{E}$ being time independent).

From here on, let us consider the case of a monochromatic wave of frequency ω.

If we express $\vec{E}$ in terms of a and a^+; according to the well-known relation:

In atomic units $H_{int} = V = i \left(\frac{2\pi}{L^3} \omega \right)^{1/2} (a\vec{\varepsilon} - a^+\vec{\varepsilon}^*) . \vec{D}$ (6)

We have considered only one mode, the occupation number being $\underline{n}$

with $F = \frac{n.C}{L^3}$ (photon flux) (7)

If δ^+ and δ^- are the 2 operators acting on the Fock states of the field:

(8) $< m \, \delta^{\pm} \, | \, n > = \delta_{m, n \pm 1}$

(9) $a = \sqrt{n} \delta^- = \left(\frac{FL^3}{C} \right)^{1/2} \delta^-$

(10) $a^+ = \sqrt{n+1} \, \delta^+ = \left(\frac{FL^3}{C} \right)^{1/2} \delta^+$ ($n \cong n+1$) usually $n > 10^{19}$

(11) $H_{int} = -i \left(\frac{2\pi}{C} \omega F \right)^{1/2} \left[\vec{\varepsilon} \vec{D} \delta^- - \vec{\varepsilon}^* \vec{D} \delta^+ \right]$

(12) I intensity = $F.\omega$,

(Absorption of a photon) (Emission of photon)

3) $H_{int} = \boxed{V = V^- + V^+ = \left(\frac{I}{2I_o} \right)^{1/2} \left[\vec{\eta} \vec{D} \delta^- + \vec{\eta} \vec{D} \delta^+ \right]}$ with $\vec{\eta} = -i\varepsilon$

$I_o = 7{,}019.10^{16}$ W/cm^2 - $n_o \cong 10^{24}$ - $F_o = 3.22 \; 10^{34}$/sec cm^2

RESOLVENT FORMALISM

Let us consider the interaction of an atom with an electromagnetic wave. At $t = 0$, the initial state $\psi(t=0)$ is:

(14) $\psi(t=0) = |I> = |g> \otimes | \ldots . n_{k\lambda} \ldots >$

at any time:

(15) $\psi(t) \equiv U(t) \, |I>$

$U(t)$ is the time evolution operator, it satisfies obviously Schrödinger equation:

(16) $H \, U(t) = i \, \dot{U}(t)$.

If H is independent of time $U(t) = e^{-iHt}$. (17)

Therefore $\psi(t) \equiv e^{-iHt}.|I>$ (18)

Let us introduce, by definition, the resolvent operator G(Z), Z being a complex variable:

$$(19)\quad G(Z) = \frac{1}{Z-H}$$

$$(20)\quad H\,G(Z) = Z.G(Z) - 1$$

U(t) is simply related to G(Z) through an inversion integral:

$$(21)\quad U(t) = \frac{1}{2\pi i}\oint e^{-iZt}\,G(Z)dZ$$

where C is the following contour of the complex plane

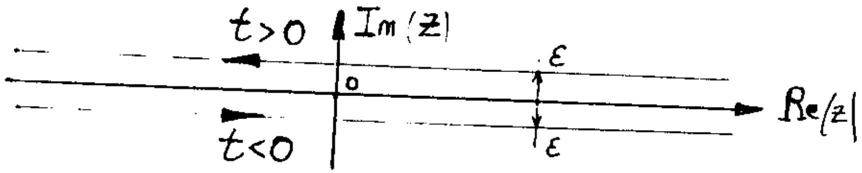


Fig. 3

It can be shown quite generally that for positive values of t, one can replace Z by x+iε and carry out the integration over the real variable x from -∞ to +∞ :

$$(22)\quad \boxed{U(t) = \frac{1}{2\pi i}\int_{-\infty}^{+\infty} e^{-ixt}\,G(x)dx}$$

Proof is given by the following calculation:

$$(23)\quad H\,U(t) = \frac{1}{2\pi i}\int_{-\infty}^{+\infty} H\,G(Z)\,e^{-iZt}dZ$$

with $H\,G(Z) = Z\,G(Z) - 1$

$$(24)\quad H\,U(t) = \frac{1}{2\pi i}\int_{-\infty}^{+\infty} Z\,G(Z)\,e^{-iZt}dZ - \underbrace{\frac{1}{2\pi i}\int_{-\infty}^{+\infty} e^{-iZt}dZ}_{=0}$$

From (22)

$$(25)\quad i\dot{U}(t) = \frac{1}{2\pi}\int_{-\infty}^{+\infty} -iZ\,e^{-iZt}\,.\,G(Z)dZ$$

$$(26)\quad i\dot{U}(t) = \frac{1}{2\pi i}\int_{-\infty}^{+\infty} Z.G(Z)\,.\,e^{-iZt}dZ$$

From (24) and (26) $H.U(t) = i\dot{U}(t)$

We have proved that U(t) expressed through the inversion integral satisfies Schrödinger equation.

From G_{ba}, it is possible to deduce $U_{ba}(t)$.

With $|<b|U|a>|^2 = |U_{ba}|^2$ = Probability of transition from a to b at time t. (G_{ba} is generally easier to calculate than $U_{ba}(t)$).

<u>Closing relation</u> concerning atom with discrete states E_i and continuum E

$$(27)\quad \sum_i |E_i><E_i| + \iint d\gamma dE'\,|\gamma E'><\gamma E'| = \mathbb{1}$$

ensemble of quantum numbers.

Example: $G_{bb}(Z) = \langle b|G(Z)|b\rangle = \langle b| \frac{1}{Z-H} |b\rangle$

using the closure relation :

$$G_{bb}(Z) = \sum_i \frac{|\langle b|E_i\rangle|^2}{Z-E_i} + \int_{E_0}^{\infty} dE' \frac{\int d_\gamma |\langle b|\gamma E'\rangle|^2}{Z-E'} \tag{28}$$

E_0, ionization threshold

$$\boxed{G(Z) = G_0(Z) + G_{0(Z)}\, V\, G(Z)} \tag{29}$$

Very analogous expression to the one obtained by Feynman in quantum electrodynamics.

This famous equation can be obtained by different means. The simplest demonstration uses operational identities.

Let us use the following expression:

$$\frac{1}{A} - \frac{1}{B} = \frac{1}{A}(B-A)\frac{1}{B} \tag{30}$$

If we choose for $A = Z - H_0$

and $B = Z - H_0 - V$

Then $G_0(Z) - G(Z) = -G_0(Z)VG(Z)$

(31) Therefore $\boxed{G(Z) = G_0(Z) + G_0(Z)VG(Z)}$

$$G = \sum_i \frac{|i\rangle\langle i|}{Z - E_i}$$

By iteration $G = G_0 + G_0VG_0 + G_0VG_0VG_0 + G_0VG_0VG_0VG_0$

$$\boxed{G = G_0 \left[1 + \sum_{n=1}^{\infty} (VG_0)^n\right]} \tag{32}$$

with obviously $G_{0\,AB} = (Z-\omega_A)^{-1}\,\delta_{AB}$ G_0 has only diagonal elements.

NON RESONANT TRANSITION

An N order process (N photon) is represented, to lowest order, by the N_{th} term of the series.

A 2 photon transition is represented by the matrix element:

$G_{FI}^{(\varepsilon)} = \langle F|G_0VG_0VG_0|I\rangle$

$$G_{FI}^{(2)} = \frac{1}{Z-\omega_F} \sum^{m} \left[\frac{V_{Fm}V_{mI}}{(Z-\omega_m)}\right] \frac{1}{Z-\omega_I} \tag{33}$$

Let us use the expression of G to derive the N^{th} order probability of transition.

The N^{th} order term $G^{(N)}$ is:

$$G^{(N)} = G_0\underbrace{(V^-G_0)(V^-G_0)\ldots(V^-G_0)}_{\text{N terms}} \tag{34}$$

(35) We recall that $V^- = \sqrt{\frac{I}{2I_0}} \cdot \vec{n}.\vec{D}.\delta^-$

Let us consider the transition between two states $|g\rangle$ and $|f\rangle$, $|f\rangle$ may belong to a continuum

$$\langle f|G^N|g = \left(\frac{I}{2I_0}\right)^{N/2} \underbrace{\langle f|G_0}_{\frac{1}{Z-E_f}\langle f|} \overbrace{\vec{n}\vec{D}\delta^- G_0 \ldots G_0\vec{n}\vec{D}\delta^-}^{M_{fg}(Z)} \underbrace{G_0|g\rangle}_{|g\rangle\frac{1}{Z-E_g}} \tag{36}$$

Let us consider the action of the δ^-.

$$M_{fg}(Z) = \langle f|\,\vec{n}.\vec{D}\; G_0^{(N-1)}\; \vec{n}.\vec{D} \ldots G_0^{(1)}\; \vec{n}.\vec{D}\; |g\rangle \tag{37}$$

with $G_0^{(1)} = \frac{1}{Z-H_{at}-h\omega_p}$

$$(38)\quad G_0^{(N-1)} = \frac{1}{Z-H_{At}-(N-1)h\omega_p}$$

According to the relatibn (21)

$$(39)\quad U_{fg}^N(t) = \frac{1}{2\pi i}\left(\frac{I}{2I_0}\right)^{N/2}\oint dZ\left(\frac{1}{Z-E_f}\right) M_{fg}(Z)\left(\frac{1}{Z-E_g}\right) e^{-iZt}$$

$$(40)\quad U_{fg}^N(t) = \frac{1}{2\pi i}\left(\frac{I}{2I_0}\right)^{N/2}\left[\frac{e^{-iE_gt}}{(E_g-E_f)} M_{fg}^N(E_g) + \frac{e^{-iE_ft}}{E_f-E_g} M_{fg}^N(E_f)\right]$$

with $M^N_{fg}(E_g) = M^N_{fg}(E_g)$

$$(41)\quad |U_{fg}^N(t)|^2 = \frac{1}{4\pi^2}\left(\frac{I}{2I_0}\right)^N\left[M_{fg}^N\right]^2 2\left(\frac{1-\cos\Omega t}{\Omega^2}\right) \quad \text{with } \Omega = E_f-E_g$$

Probability per unit time is $W^N \sec^{-1}$

$$(42)\quad W^N \sec^{-1} = \lim_{T\to\infty} |U_{fg}^N(t)|^2\frac{1}{T} = \frac{1}{4\pi^2}\left(\frac{I}{2I_0}\right)^N (M_{fg}^N)^2\,\delta(E_g-E_f)$$

with:

$$(43)\quad \boxed{|M_{fg}^N|^2 = \sum_{\alpha_{N-1},\ldots\alpha_1}\frac{\langle f|r^{(\lambda)}|\alpha_{N-1}\rangle\ldots\langle\alpha_1|r^{(\lambda)}|g\rangle}{(E_{\alpha_{N-1}} - E_g - (N-1)\omega_p)\ldots(E_{\alpha_1} - E_g - h\omega_p)}}$$

with $\underline{r^\lambda = \vec{\eta}_\lambda . \vec{D}}$

The calculation requires the knowledge of the energy levels and the matrix elements, which presupposes knowledge of the wavefunctions.

Summation over on infinite sets of atomic intermediate states including continuum must be performed. It is only in the case of atomic hydrogen that these calculations can be done exactly in analytical form.

We will not describe the various methods which were derived to perform the calculation of M_{fg}^N.

The 2 most successive approaches have been the use of the Schwartz and Tieman technique, and the use of the Greens function method.

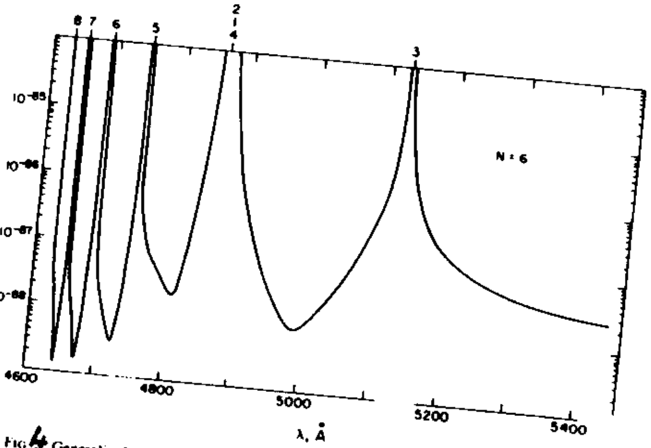
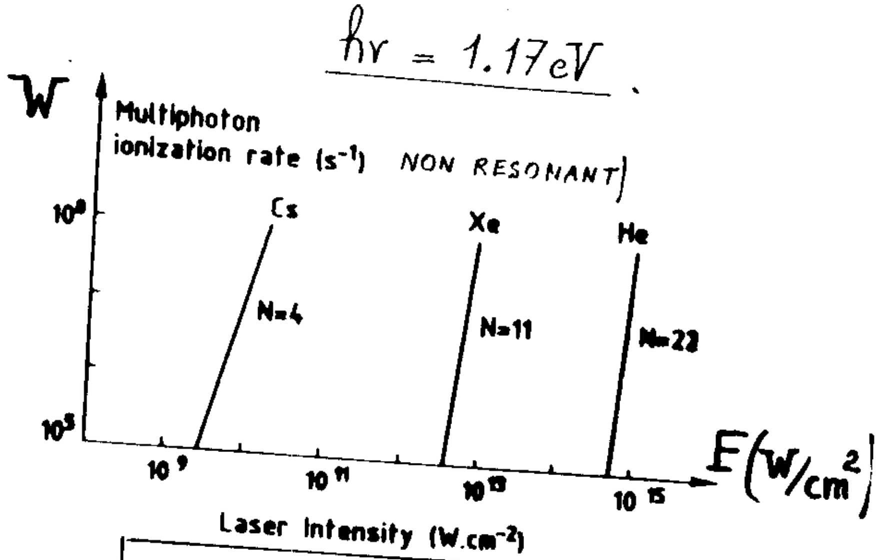


FIG. 4 Generalized cross section for 6-photon ionization of H in the ground state calculated by Karule (1974) using the Green's function method.

Fig. 4

Before considering the theoretical approach to resonant process, let us examine the orders of magnitude of the intensity required to produce 4,11, 21 photon absorption for a duration of light pulse τ = 50 p.sec.



$$\boxed{W = \sigma_N \cdot F^N}$$

Fig. 5

In order to illustrate the degree of accuracy reached by the theoretical predictions based on the utilisation of the formula above, we present the following table:

		Comparison with experimental results	
		Exp.	Theory
			Rachman
Cs	2 hν	$6.7 \pm 1.9\ 10^{-50}$	$6.6\ 10^{-50}\ cm^4.s$
	4 hν	$7.5 \pm 2.8\ 10^{-109}$	$5.3\ 10^{-109}\ cm^8.s^3$
			Aymar and Crance

			Crance
He	2 hν	$2.7 \pm 2\ 10^{-49}$	$2.1\ 10^{-49}\ cm^4.s$
	3 hν	$3 \pm 2\ 10^{-81}$	$3.5\ 10^{-81}\ cm^4.s$
			Crance

RESONANT PROCESSES

General formalism using partitioning technique

By the unitary property of the evolution operator, one can show that:

$$\boxed{P(t) = 1 - |U_{gg}(t)|^2 - |U_{gr}(t)|^2} \quad (44)$$

$|U_{gg}(0)|^2 = 1$
$|U_{gr}(0)|^2 = 0$

ionization probability — Population of fundamental state — Population of resonant state

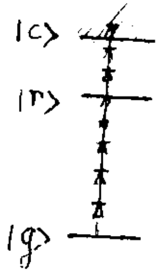


Fig. 6

This is nothing but a conservation relation.

(45) From $Tr(\rho(t)) = 1$ one has

(46) $$\rho_{gg}(t) + \rho_{rr}(t) + \rho_{cc}(t) = 1$$

Owing to the time evolution law for $\rho(t)$

(47) $$\rho(t) = U(t)\,\rho(0)\,U^+(t)$$

At time $t = 0$, only the ground state is populated.

(48) $$\rho(0) = |g><g|$$

one has:

(49) $$\begin{cases} \rho_{gg}(t) = <g|U(t)|g><g|U^+(t)|g> = |U_{gg}(t)|^2 \\ \rho_{rr}(t) = <r|U(t)|g><g|U^+(t)|r> = |U_{\;}(t)|^2 \end{cases}$$

while

(50) $$P(t) = \rho_{cc}(t)$$

is the ionization probability.

(51) $$P(t) = 1 - |U_{gg}(t)|^2 - |U_{gr}(t)|^2$$

The Resolvent method requires the sum of an infinite number of atomic states. One way to avoid such a difficulty is to use the projector method, developed first by Feshbach (Partitioning technique).

This is underlined by the expression of P(t) which requires only the calculation of G_{gg} and G_{gr}.

We will study the evolution of the system described by H, for a restricte number of eigenstates of H_o (which will include $|g\rangle$ and $|r\rangle$). One defines the operators P and Q which project onto and outside the subspace Hp spanned by then eigenstates.

(52) $$P = \sum_{i=1} |e_i\rangle\langle e_i|$$

P is the operator which operates on the subspace Hp

(53) $$\left[\begin{array}{lll} Q = \mathbb{1} - P & & \\ P = P^+ & & Q = Q^+ \\ P^2 = P & & Q^2 = Q \\ P+Q = \mathbb{1} & & PQ = QP = 0 \end{array}\right]$$

$$[H_o, P] = [H_o, Q] = 0$$

We want to calculate G(Z) inside the subspace Hp defined by

(54) $$\tilde{G}(Z) = P\,G(Z)P$$

From here we will only be concerned by the properties and evolution of $\tilde{G}(Z)$.

Starting from $G(Z) = G_0(Z) + G_0(Z)\,V\,G(Z)$

(55) $$\tilde{G}(Z) = \frac{P}{Z-H_o} + \frac{P}{Z-H_o} V\,G(Z)P$$

(56) $$\boxed{\begin{array}{l} \tilde{G}(Z) = \dfrac{P}{Z-H_o-\tilde{R}(Z)} = \dfrac{1}{Z-\tilde{H}_o-\tilde{R}(Z)} \\[2ex] \tilde{R}(Z) = PVP + PV\dfrac{Q}{Z-H_o-QVQ}VP \end{array}}$$

Let us establish the equations (56)

(57) $$(Z-H)G = 1$$

(58) $$(Z-H)(P+Q)G = 1$$

(59) $$P(Z-H)(P+Q)GP = P$$

(60) $$Q(Z-H)(P+Q)GP = 0$$

We deduce:

(61) $$\left\{\begin{array}{l} P(Z-H)P\,\underset{\sim}{PGP} + P(Z-H)Q\,\underset{\sim}{QGP} = P \\ Q(Z-H)P\,\underset{\sim}{PGP} + Q(Z-H)Q\,\underset{\sim}{QGP} = 0 \end{array}\right.$$

2 equations containing the 2 unknowns, PGP and QGP .

From the second, one derives:

(62) $$QGP = -\frac{1}{Q(Z-H)Q}\,Q(Z-H)P\,\underset{\sim}{PGP}$$

By introduction in the first equation:

(63) $$P(Z-H)P\,\underset{\sim}{PGP} - P(Z-H)Q\frac{1}{Q(Z-H)Q}\,Q(Z-H)P.\underset{\sim}{PGP} = P$$

(64) $$PGP = \frac{P}{P(Z-H)P - P(Z-H)Q\dfrac{1}{Q(Z-H)Q}Q(Z-H)P}$$

(65) $$\begin{array}{l} P(Z-H)P = P(Z-H_o)P - PVP \\ P(Z-H)Q = \underbrace{P(Z-H_o)Q}_{0} - PVQ \end{array}$$

We deduce: by PGP $= \tilde{G} = \dfrac{P}{Z-H_o-\tilde{R}(Z)}$ (66)

(67) That $\tilde{R}_{(Z)} = PVP + PV \dfrac{Q}{Z-H_o-QVQ} V P$ C.Q.F.D.

$$\boxed{R_{(Z)} = V + V \frac{Q}{Z-H_o-QVQ} V} \tag{68}$$

By iteration:

$$R_{(Z)} = V + V \frac{Q}{Z-H_o} V + V \frac{Q}{Z-H_o} V \frac{Q}{Z-H_o} V + \dots \tag{69}$$

$$R_{(Z)} = V + \sum_{n=1}^{\infty} V \left(\frac{Q\,V}{Z-H_o} \right)^n \tag{70}$$

$\tilde{R}(Z)$ is called displacement operator (restricted to subspace Hp). This denomination comes from the fact that diagonal values of $\tilde{R}(Z)$ express the level shifts due to their coupling with the E.M non resonant fields (the so-called light shifts).

These relations are exact, there is no approximation so far.

By the structure of R(Z) it is apparent that $\tilde{R}(Z)$ connects two states belonging to the subspace Hp, in-between the system is moving in the orthogonal subspace Q.

APPLICATION TO A 2 PHOTON RESONANT, 3 PHOTON IONIZATION

In order to have a better understanding of the physics we will now describe a 2 photon resonance, 3 photon ionization. Generalisation to higher order process is rather straightforward.

According to expression (51), we have to derive G_{gg} and G_{gr}, then U_{gg} and U_{gr}, it is therefore obvious that the good choice for subspace Hp is

$$\begin{aligned} P &= |r\rangle\langle r + |g\rangle\langle g| \\ Q &= 1 - P \end{aligned} \tag{71}$$

which means that the ground state and resonant state belongs to subspace Hp.

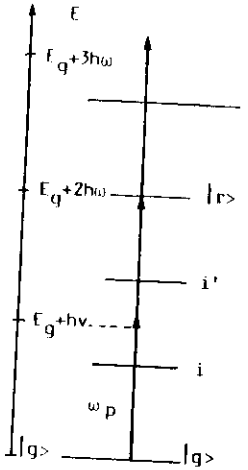


Fig. 7

$$(Z-H_o-\tilde{R})\tilde{G} = P \tag{72}$$

Let us calculate

$$Z-H_o-\tilde{R}_{(Z)}$$

$$\tilde{G}^{-1} = \begin{vmatrix} Z-E_g-n\omega_p-R_{gg}(Z) & -R_{gr}(Z) \\ -R_{rg}(Z) & Z-Er-(n-2)\omega_p-R_{rr}(Z) \end{vmatrix} \tag{73}$$

$$\boxed{\begin{aligned} \tilde{G} &= \begin{bmatrix} Z-E_r-(n-2)\omega_p - R_{rr}(Z) & -R_{gr}(Z) \\ -R_{rg}(Z) & Z-E_g-n\omega_p-R_{gg}(Z) \end{bmatrix} \times \frac{1}{D} \\ \text{with } D &= \left[Z-E_r-(n-2)\omega p-R_{rr}(Z)\right]\left[Z-E_g-n\omega p-R_{gg}(Z)\right] - R_{gr}(Z)\,R_{rg}(Z) \end{aligned}} \tag{74}$$

We deduce:

$$\tilde{G}_{gg}(Z) = \frac{Z-E_r-(n-2)\omega_p - R_{rr}(Z)}{D} \tag{75}$$

$$\tilde{G}_{gr}(Z) = \frac{R_{gr}(Z)}{D} \tag{76}$$

We have three matrix elements of the displacement operator to derive.

$\tilde{R}_{gg}(Z)$, $\tilde{R}_{rr}(Z)$ and $\tilde{R}_{gr}(Z)$.

Let us begin by considering only the two first terms of the R development which contribute significantly to the matrix elements:

$$R(Z) = PVP + PV \frac{Q}{Z-H_o} VP \tag{77}$$

$$\left\{ \begin{aligned} R^{(2)}_{gg}(Z) &= \sum_i \frac{|\langle g|V|i\rangle|^2}{Z-E_i-(n\pm1)\omega_p} \\ R^{(2)}_{gr}(Z) &= \sum_i \frac{\langle g|V|i\rangle\langle i|V|r\rangle}{Z-E_i-(n-1)\omega_p} \\ R^{(2)}_{rr}(Z) &= \underbrace{\sum_i \frac{|\langle r|V|i\rangle|^2}{Z-E_i-(n\pm1)\omega_p}}_{\text{coupling to the discrete states}} + \int_{E_o}^{+\infty} \underbrace{\frac{|\langle r|V|E'\rangle|^2}{Z-E'-(n-1)\omega_p}}_{\text{coupling to the continuum}} dE' \end{aligned} \right. \tag{78}$$

(the contribution of the first order term is nul) V cannot couple |g to |g !

(It is a 2 photon transition from g to r).

It may be proved that the fourth order terms in V, $\tilde{R}^{(4)}_{gg}(Z)$ and $\tilde{R}^{(4)}_{rr}(Z)$, are much smaller than $\tilde{R}^{2}_{gg}(Z)$ and $\tilde{R}^{2}_{rr}(Z)$.

The second term $R_{gr}(Z)$ can be developed according to

$$\mathcal{S}\int_{E_r}^{+\infty} \frac{|\langle r|V|E'\rangle|^2}{Z-E'-(n-1)\omega_p} dE' - i\,\Gamma_R(Z)$$

$$\text{with } \Gamma_R(Z) = \pi|\langle r|V|E'\rangle|^2\,\delta(Z-E') \tag{79}$$

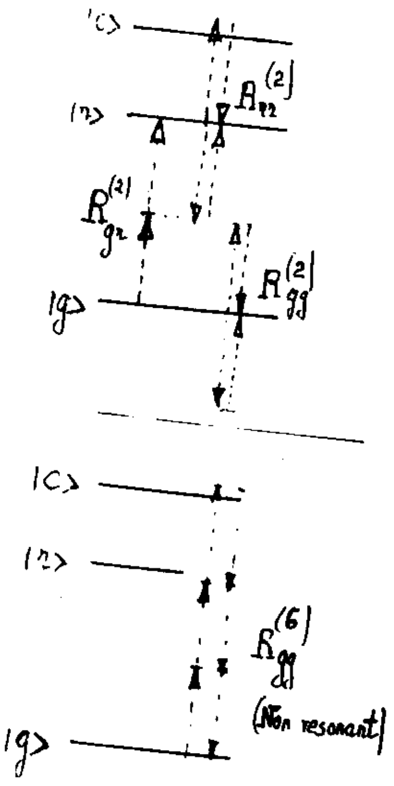


Fig. 8

$\tilde{R}_{rr}$ involves two real parts, one corresponding to a shift induced by coupling with the discrete states, the other by coupling with the continuum. One imaginary part expressing the leakage from |r> to the continuum.

I believe that it is extremely interesting to notice that some authors, mainly those using a different method, the effective Hamiltonian, have deduced different terms, the main difference is that the ground state exhibits an imaginary part, as well as the non diagonal term $\tilde{R}_{gr}$. This can be obtained by considering the development of R to higher order terms.

For R_{gg} we consider the non resonant term coupling the ground state to the continuum it is a V^6 term or I^3. It is nothing but the non resonant ionization channel. We will not fully derive this term which will be expressed as $R^6_{gg} = -i\gamma_g$

$$R_{gg} = R^{(2)}_{gg} + R^{(6)}_{gg} \tag{80}$$

$$\gamma_g = 2\pi|\langle g|R^{(6)}_{gg}|E,(n-3)\,\omega\rangle|^2$$

[redacted]

(81) $R_{gr} = R_{gr}^{(2)} + R_{gr}^{(4)}$ The non diagonal non resonant term, $R_{gr}^{(4)}$, is also quite easy to derive, we will give the result:

$$- i(\gamma_g . \Gamma_r)^{1/2}$$

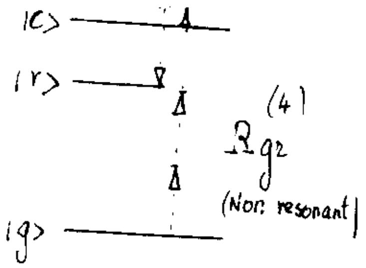


Fig. 9

Some prefer to introduce $q = \frac{|R_{gr}^{(2)}|}{\sqrt{\gamma_g . \Gamma_r}}$ (82)

with $\tilde{R}_{gr} = \sqrt{\gamma_g . \Gamma_r}[q-i]$, we will see later on the influence of this non resonant contribution, which induces interference effects between the 2 channels, resulting curves having Fano structures.

We will consider the one pole approximation to the matrix elements of R. The maximum contribution corresponds to $Z = E_g + n\omega_p$, we will consider only this value. This approximation introduces a considerable simplification.

Let us define $E'_r = E_r + (n-2)\omega_p + \Delta r$ – $\Delta r = R_e(R_{rr}^{(2)})$

$E'_g = E_g + n\omega_p + \Delta g$ – $\Delta g = R_e(R_{gg}^{(2)})$

$G_{gg}(Z)$ and $G_{gr}(Z)$ can be rewritten (with $\gamma_g = 0$, for simplification):

$$(84)\begin{cases} G_{gg}(Z) = \dfrac{Z - E'_r + i\Gamma_r}{(Z-E'_g)(Z-E'_r+i\Gamma_r) - |R_{gr}|^2} \\ \\ G_{gr}(Z) = \dfrac{R_{gr}}{(Z-E'_g)(Z-E'_r + i\Gamma_r) - |R_{gr}|^2} \end{cases}$$

The poles of the denominator are:

$$(85)\ \boxed{Z^{\pm} = \frac{1}{2}\left\{ E'_r + E'_g - i\Gamma_r \pm [(\Delta + i\Gamma_r)^2 + 4|R_{gr}|^2]^{1/2} \right\}}$$

where we have introduced Δ = dynamic detuning = $E'_g - E'_r$. In order to have a better understanding of these poles, let us consider the following situation:

① $\boxed{\Delta >> \Gamma_r, |R_{gr}|}$ non resonant condition. Let us write $Z^+ = E^+ - i\Gamma^+$ and $Z^- = E^- - i\Gamma^-$

$$(86)\ \boxed{\begin{array}{l} Z^+ = E'_g = E_g + n\omega_p + \Delta g \\ Z^- = E'_r - i\Gamma_r = E_r + (n-2)\omega_p + \Delta r - i\Gamma_r \end{array}}$$

with $\Delta g = \alpha_g I$

$\Delta r = \alpha_r I$

It is quite clear that Z^+ is the dressed ground state,

Z^- is the dressed resonant state.

The real part gives the energy of the states, the imaginary part, the width (the inverse being the lifetime).

Z^+ has no imaginary part, due to the fact that we have considered $\gamma g = 0$, for sake of simplicity. It should reflect the width due to coupling with the continuum, i.e. the non resonant ionization probability (80).

Z^+ and Z^- are, generally speaking, bound unstable states of the dressed atom.

E'_g and E'_r are linearly dependent on the intensity, the two lines may cut. If the intensity tends to 0, $E'_r = E_r$ and $E'_g = E_g$.

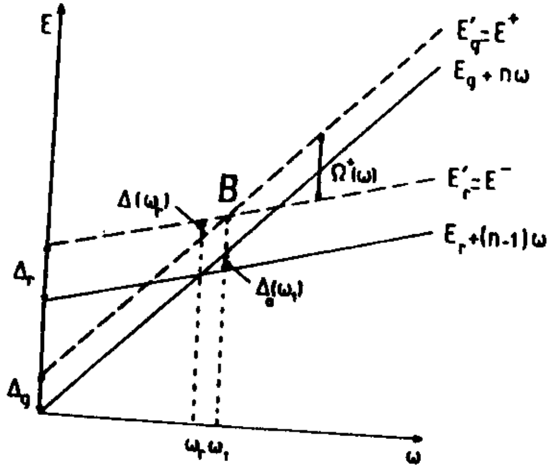


Fig. 10

We may expect a shift in resonances due to intensity effects, the so-called dynamic detuning.

At point B, $E'_g = E'_r$ (Δ=0). Let us consider this situation:

CROSSING AND ANTICROSSING RESONANCE

② $\boxed{\Delta = 0 \text{ (dynamic resonance) with } |R^{gr}| > \Gamma_{r/2}}$, ($E'_g = E'_r$) for Δ=0

One finds with these conditions:

$$\boxed{\begin{aligned} Z^+ &\cong E'_g + |R_{gr}| - i\frac{\Gamma_r}{2} \\ Z^- &\cong E'_r - |R_{gr}| - i\frac{\Gamma_r}{2} \end{aligned}} \qquad \begin{aligned} Z^+ &= E'_g + [(R_{gr})^2 - \frac{\Gamma_r^2}{4}]^{1/2} - i\frac{\Gamma_r}{2} \\ Z^- &= E'_g - [(R_{gr})^2 - \frac{\Gamma_r^2}{4}] - i\frac{\Gamma_r}{2} \end{aligned} \tag{87}$$

It is quite remarkable to underline that, at dynamic resonance, E+ and E⁻ are never equal. The real part of the dressed atomic states present an anticrossing behaviour. (fig 11)

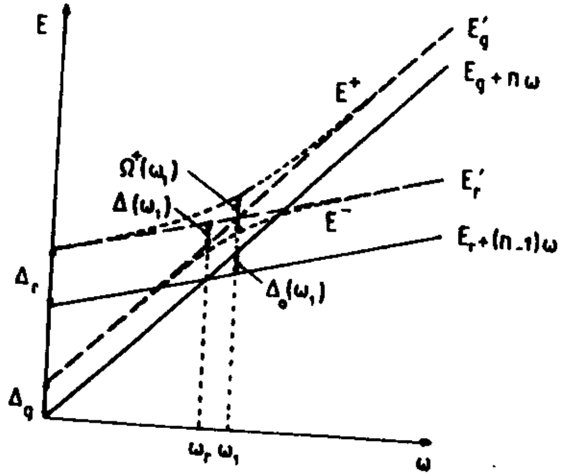


Fig. 11

③ $\boxed{\Delta = 0 \text{ (dynamic resonance) with } |R_{gr}| < \frac{\Gamma_r}{2}}$ $E'_r = E'_g$ (Δ = 0)

We deduced:

$$\boxed{\begin{aligned} Z^+ &= E'_g - i\,[\,\frac{\Gamma_r}{2} - (\frac{\Gamma_r^2}{4} - |R_{gr}|^2)^{1/2}\,] \\ Z^- &= E'_r - i\,[\,\frac{\Gamma_r}{2} + (\frac{\Gamma_r^2}{4} - |R_{gr}|^2)^{1/2}\,] \end{aligned}} \tag{88}$$

$E^+ = E^-$. The real part of the dressed atomic states present a crossing behaviour. (fig. 10)

(R_{gr}) and Γ_r are proportional to I. The anti-crossing situation prevails for a 3 photon ionization, for any value of I (see page 26). A crossing behaviour is presented on page 27, it is a 4 photon ionization, three photon resonant.

On both figures, the energies are represented for three different values of the photon frequency.

The curves have been determined by Trahin and Gontier.

We have everything required to derive the ionization probability according to

$$(89)\quad U_{\substack{gg\\gr}} = \frac{1}{2\pi i}\int_{C'} G_{\substack{gg\\gr}}(Z)\, e^{-iZt} dz$$

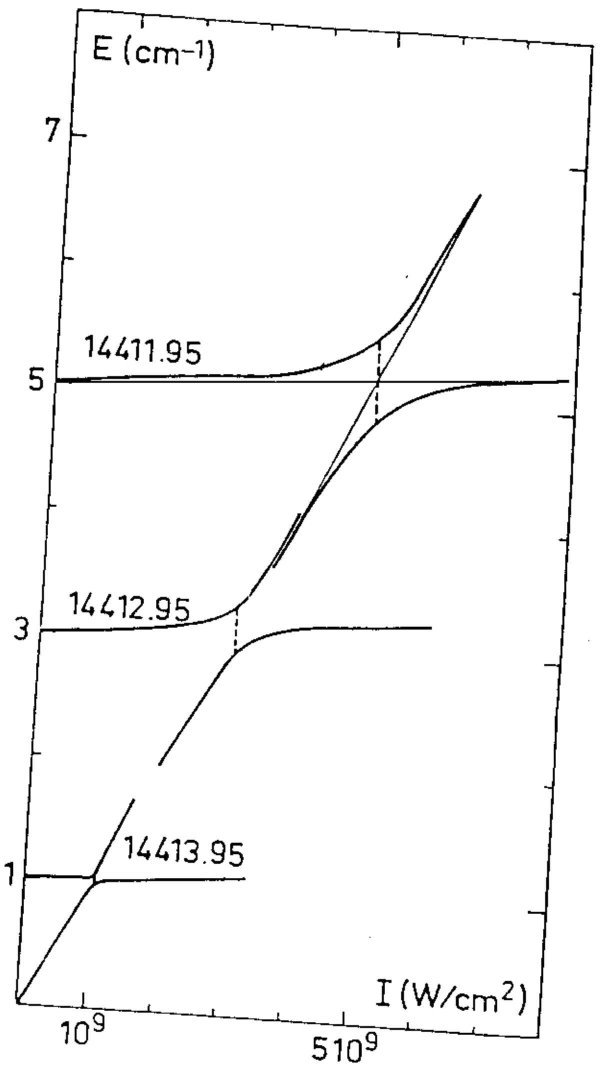
Values of $G_{gg}(Z)$ and $G_{gr}(Z)$ are given on page 21

$$(90)\quad \begin{cases} U_{gg}(t) = \sum\limits_{Z^+,Z^-} \text{Res}\, \dfrac{Z-E'_r + i\Gamma_r}{(Z-Z^+)(Z-Z^-)}\ e^{-iZt} \\[2ex] U_{gr}(t) = \sum\limits_{Z^+,Z^-} \text{Res}\, \dfrac{R_{gr}\ e^{-iZt}}{(Z-Z^+)(Z-Z^-)} \end{cases}$$

$$(91)\quad \begin{cases} U_{gg}(t) = \dfrac{(Z^+-E'_r+i\Gamma_r)\ \exp^{-iZ^+t}}{Z^+ - Z^-} + \dfrac{(Z^--E'_r+i\Gamma_r)\ \exp^{(-iZ^-t)}}{Z^- - Z^+} \\[2ex] U_{gr}(t) = \dfrac{R_{gr}\ \exp^{(-iZ^+t)}}{Z^+ - Z^-} + \dfrac{R_{gr}\ \exp^{(-iZ^-t)}}{Z^- - Z^+} \end{cases}$$

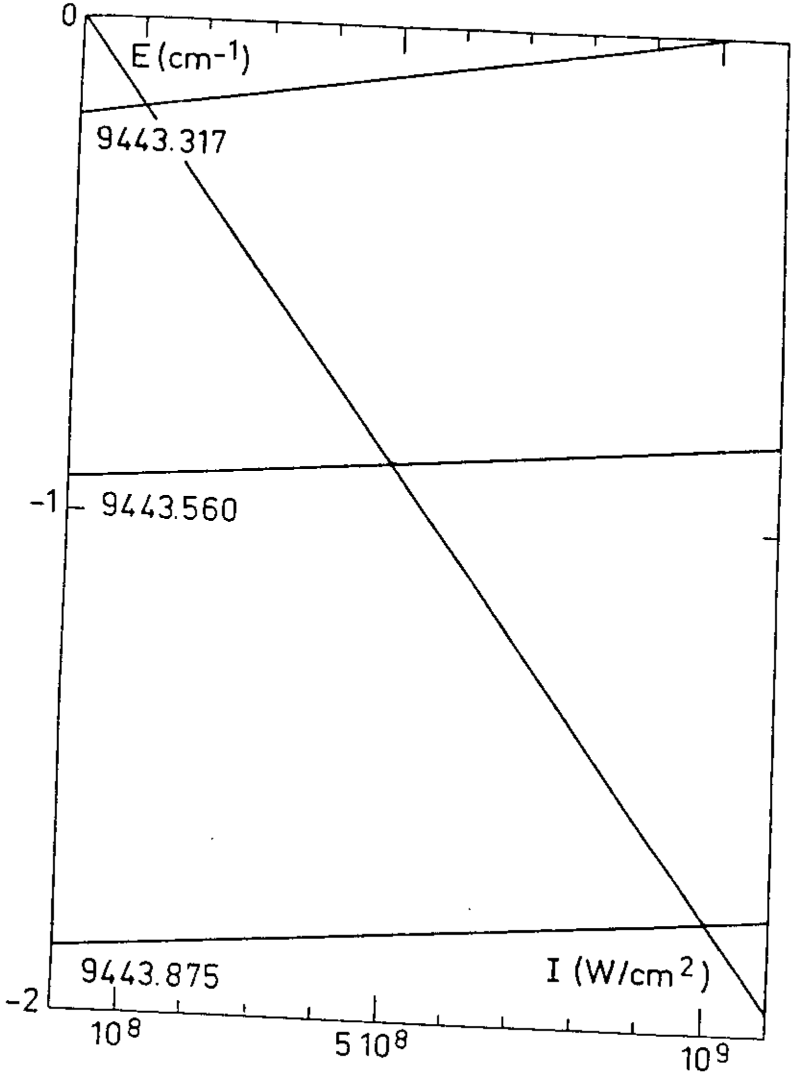
From which according to page 15, after some calculations:

$$(92)\quad \boxed{P(t) = 1 - \frac{1}{|Z^+-Z^-|^2}\Big[|(Z^+-E'_r+i\Gamma_r)e^{-iZ^+t} - (Z^--E'_r+i\Gamma_r)\exp^{-iZ^-t}|^2 + ||R_{gr}|^2[e^{-iZ^+t} - e^{-iZ^-t}]|^2\Big]}$$



Gentier, Trahin

Fig. 12



Gontier, Trahin

Fig. 13

The classical expression that has been widely used is (derived from 92):

$$\boxed{P(t) = 1 - 2\,\frac{e^{-(\Gamma^+ + \Gamma^-)t}}{(E^+ - E^-)^2 + (\Gamma^+ - \Gamma^-)^2}\left\{ a \sin h(\Gamma^+ - \Gamma^-)t + b \sin h(\Gamma^+ - \Gamma^-)t - c \cos(E^+ - E^-)t - d \sin(E^+ - E^-)t \right\}} \tag{93}$$

This expression is valid at any order N of multiphoton process. The difference is taken into account by the constants appearing in P(t). Let us examine how P(t) changes at resonance from a crossing to an anti-crossing situation:

① $\boxed{\Delta = 0,\ |R_{gr}| > \frac{\Gamma_r}{2},\ \text{anticrossing resonance.}}$

After some calculation it may be shown that:

$$(94)\ P(t) = 1 - \frac{e^{-\Gamma_r t}}{\Omega^2}\left[4(R_{gr})^2 - \Gamma_r^2 \cos \Omega t - \Gamma_r\, \Omega \sin \Omega t\right]$$

(95) with $\Omega = [4(R_{gr})^2 - \Gamma_r^2]^{1/2}$ represents the difference $E^+ - E^-$ between two energies of the dressed states, at the anticrossing point, it is very analogous to a general Rabi Frequency of the 2 levels coupled by the E.M. field. One notices that P(t) is oscillating at this very frequency.

For small time intervals at $t \ll 1/\Omega$, $P(t) = 1 - e^{-\Gamma_r t}$, The exponential constant being directly related to the life time of the dressed atomic states ($\Gamma^+ = \Gamma^-$). We are considering a system having two levels with small losses.

② $\boxed{\Delta = 0;\ |R_{gr}| < \frac{\Gamma_r}{2},\ \text{crossing resonance}}$ $(E^+ = E^-,\ \Gamma^+ \neq \Gamma^-)$.

It may be shown that: if $\Omega' = (\Gamma_r^2 - 4(R_{gr})^2)^{1/2}$ (96)

$$(97)\quad P(t) = 1 - \frac{e^{-\Gamma_r t}}{\Omega'^2}\left[-4(R_{gr})^2 + \Gamma_r^2 \,\mathrm{ch}\, \Omega' t - \Gamma_r\, \Omega' \sin h\, \Omega' t\right]$$

<u>There is no oscillating behaviour when the resonant level is more strongly coupled to the continuum than to the ground state.</u>

To sum up, let us consider: $\Omega = [4(R_{gr})^2 - \Gamma_r^2]^{1/2}$

For $\Omega^2 > 0$,

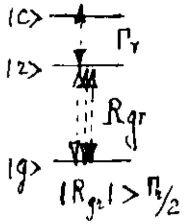


Fig. 14

|g] and |r] strongly coupled, the system oscillates, and leaks slowly in the continuum.

For $\Omega_2 < 0$,

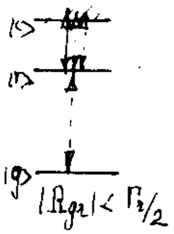


Fig. 15

|r] and |c] are strongly coupled, the oscillating period is longer than the damping term. No oscillation can be sustained. Each particule "arriving" in |r> is immediately ionized !

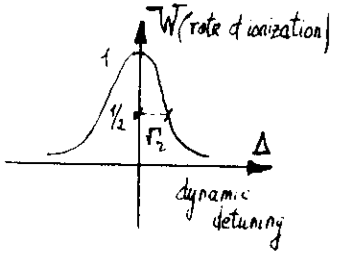
---

③ $\boxed{\Delta \text{ big with } |R_{gr}| << \Gamma_r}$

After a short time, $P(t) = 1 - e^{-Wt}$ for $\Gamma_r t >> 1$

(98) with $W = \dfrac{2\,\Gamma_r |R_{gr}|^2}{\Delta^2 + \Gamma_r{}^2}$ (rate of ionization)

Lorentzian curve



For Wt small

$P(t) = 1 - (1 - Wt) = Wt$

Fig. 16

For 3 photon ionization: $|R_{gr}|^2 \,\vdots\, I^2$

Out of resonance $\Delta >> \Gamma r$, $W \,\vdots\, I^3$ (3 photon process)

At resonance $\Delta = 0$, $W \,\vdots\, I$

It is quite interesting to notice that if we introduce as we did earlier the non resonant contribution of R_{gr} , i.e.

$R_{gr} = \sqrt{\gamma_g \cdot \Gamma_r}\,(q - i)$ with $q = \dfrac{|R_{gr}^{(2)}|}{\sqrt{\gamma_g \cdot \Gamma_r}}$

(99) $\boxed{W = \dfrac{2\,\gamma_g(\varepsilon + q)^2}{\varepsilon^2 + 1}}$ with $\boxed{\varepsilon = \dfrac{\Delta}{\Gamma_r}}$

If q is not too big, W appears as an asymmetric curve Fano profile due to the interference between resonant and non resonant paths, this does not show up for large values of q.

[redacted]

In such a case (q large) $W = \frac{2\gamma_g\, q^2}{\epsilon^2 + 1} = \frac{(R_{gr})^2}{\left(\frac{\Delta^2}{\Gamma_r^2} + 1\right) \cdot \Gamma_r}$

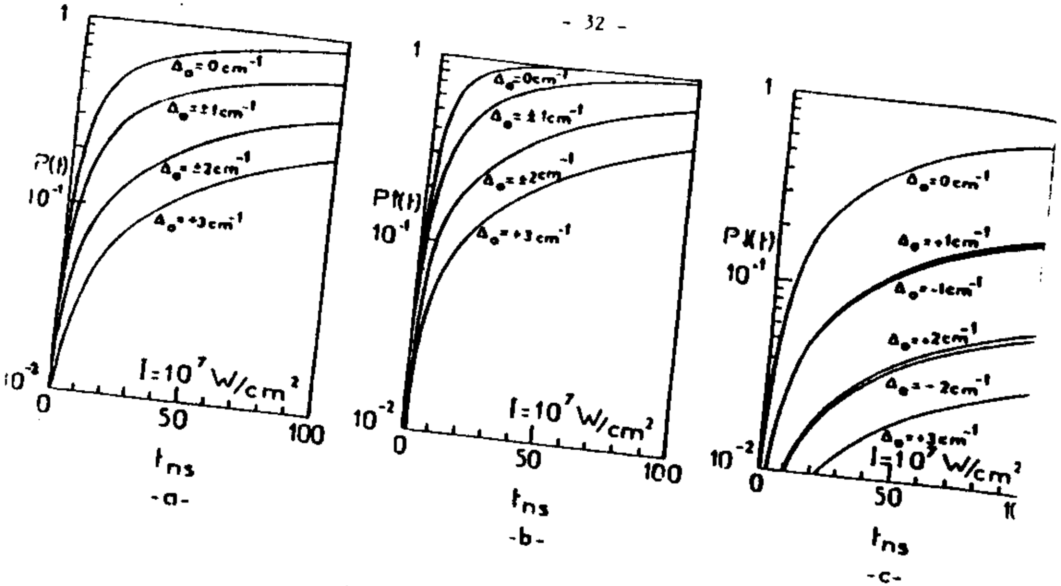
$$W = \frac{2\,\Gamma_r \cdot (R_{gr})^2}{\Delta^2 + \Gamma_r{}^2} \qquad \text{(valid for } q \gg 1\text{)} \tag{100}$$

and we find the previous expression with no asymmetric effects (98).

It is worth noting that, if we refer to the general expression P(t), page 28, the oscillating terms have an argument $\tilde{E}^{+}-E^{-}$ which is equal to $\Omega \simeq [\Delta^2 + 4|R_{gr}|^2 - \Gamma_r^2]^{1/2}$.

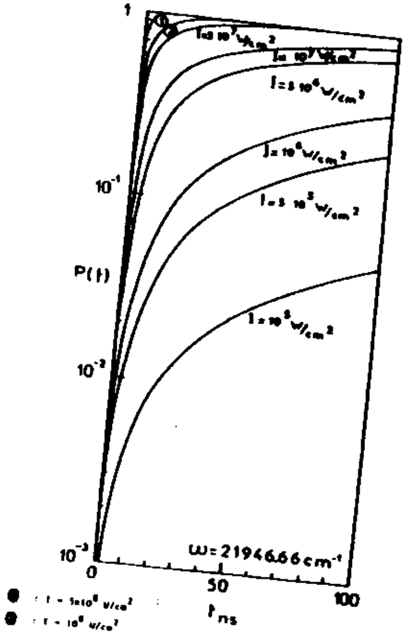
For $\Delta = 0$, we recover the value of Ω page 28, (95)

This generalized Rabi oscillation term increases when the dynamic shift increases. This will be apparent on further theoretical curves. Jaourn and colleagues have carefully studied the temporal dependence of P(t) for a 2 photon ionization of cesium, with a possible one or two photon resonance on intermediate states (7 $P^{1/2}$ and 7 $P^{3/2}$). Γ_r is smaller than R_{gr} , this corresponds to an anti-crossing resonance.

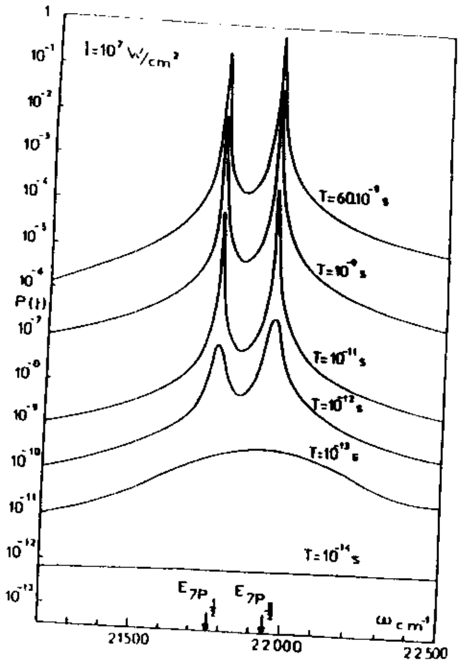


$\Delta_o = 0\ cm^{-1} \leftrightarrow \omega = 21946.66\ cm^{-1}$ Jaouen et al.

Fig.17



These authors have also studied the influence of the interaction time T on resonance from 10^{-14} sec. to 60 nanoseconds.

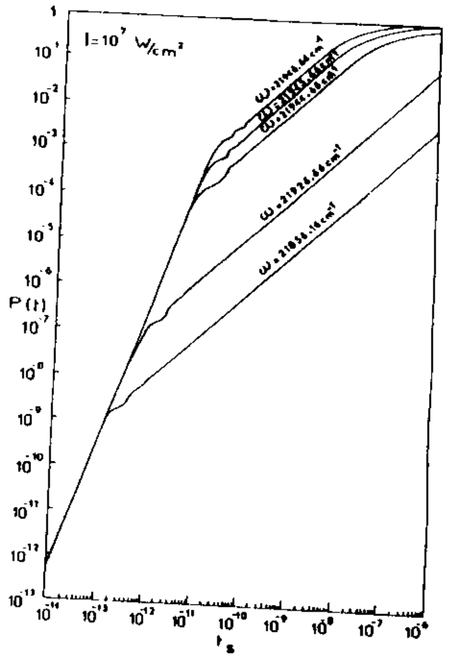


Jaouen et al.

Fig. 19

We notice that for $T = 10^{-14}$ sec., there is no resonance effect. The explanation holds in the fact, that at such small time, the dressed atomic states experience only few Rabi oscillations, the resonant conditions cannot be established !

The authors have also studied P(t) for very small time. We observe two different regimes, at small time a dependence in t^3, later on a t dependence, with the appearance of an oscillatory regime. Still later a saturation appears and P(t)⟶1, ground state is completely depleted. The only part where a rate of ionization can be defined corresponds to the linear regime.



Jaouen et al.

Fig. 20

The critical time corresponding to the transition from t^3 to t, is

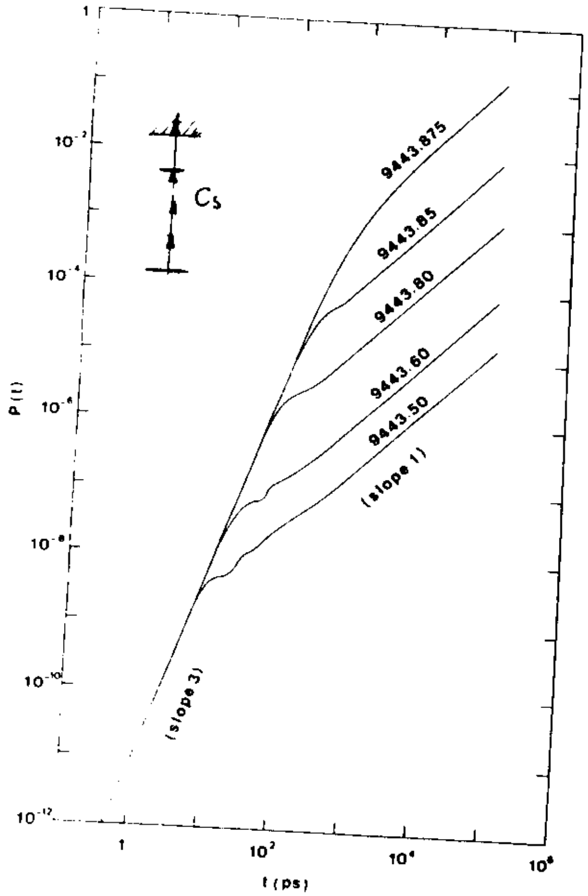
$$(101)\quad t_c = \left(\frac{6}{\Delta^2 + 4|R_{gr}|^2}\right)^{1/2}$$

It may be noticed that t^3 can be forgotten and that the process can be conveniently described by one rate process (which gives the dominant contribution to P(t).

Let us magnify the quantum beat regime.

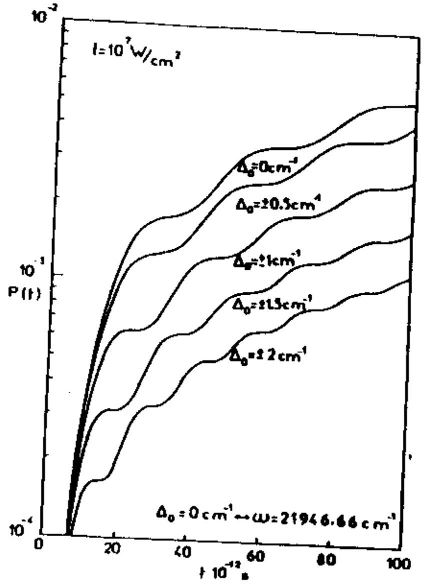
Let us remember that pulsation $\Omega = (\Delta^2 + 4|R_{gr}|^2)^{1/2}$ page 31 It is quite apparent on the figure that ω increases with Δ. (fig. 21)

The whole picture being consistent with our previous remarks: <u>the system presents Rabi oscillation between the dressed non crossing atomic states</u>.



GONTIER, TRAHIN

Fig. 20bis



Jaouen et al.

Fig. 21

They progressively leak in the continuum, the oscillations disappear, saturation is reached, ground state is completely depopulated.

POPULATION OF RESONANT STATES AND CONTINUUM

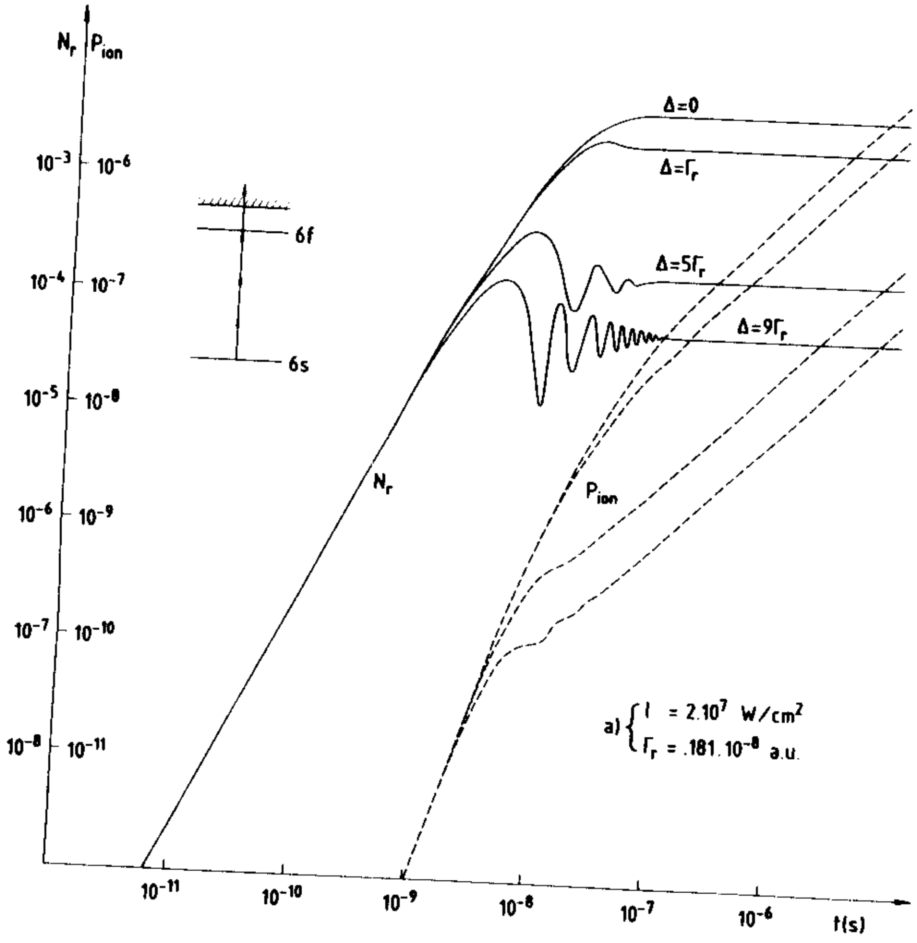
In order to have a complete view of the situation, we need an illuminating insight on the population of the resonant state.

The calculation concerns 3 photon resonant (6f state), 4 photon ionization of cesium. This corresponds to crossing resonance (no oscillation for $\Delta = 0$).

What is remarkable in these results schematized in fig 22 of page 36 is the following observation:

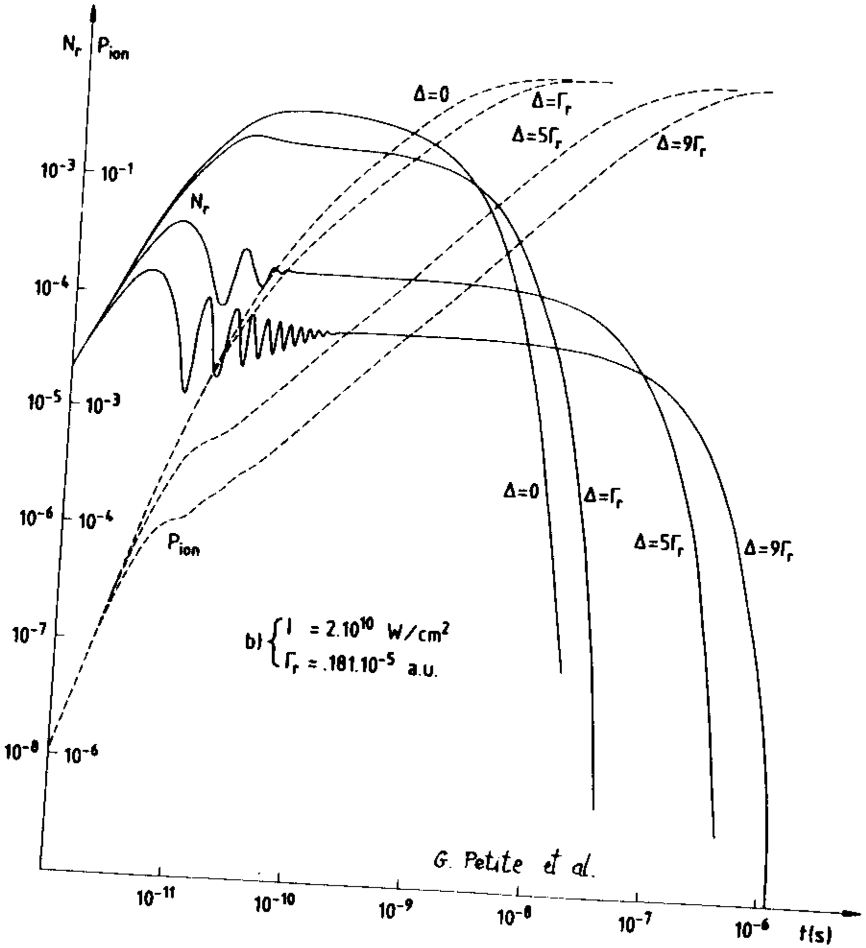
When the linear regime for $N^{+}(t)$ is reached, this corresponds to a plateau of the density of population of the resonant state n_r which

[redacted]



G. Petite et al.

Fig. 22



G. Petite et al.

Fig. 23

remains constant until the ionization saturation sets in, then n_r falls back to zero when $n^+(t) \longrightarrow 1$. In the resonant regime (plateau), transitions are such that the incoming flux to the resonant state equals exactly the outflux to the continuum.

This has been numerically illustrated on figure 22 for $I = 2.10^7 W/cm^2$. It is interesting to notice that oscillations are forced in for $\Delta > \Gamma_r$, for n_r as well as for $N^+(t)$. There is no oscillation, as expected, for $\Delta = 0$.

Figure 23 gives the same evolution for $I = 2.10^{10}$ W/cm^2. Γ_r is 10^3 times larger, plateau is reached for times 10^3 smaller and expected total ionization sets in much sooner.

General behaviour of such processes enlightened by these numerical results is conceptually simple.

EFFECTIVE HAMILTONIAN METHOD

Let us consider the total Hamiltonian

$H = H_o + V$

with $V = Da + D^* a^+$ (interaction Hamiltonian).

Let us consider the dressed atomic states:

initial state: $|g, \mathcal{E}_g + n>$

final state: $|f, E + (n-N)>$

resonant state: $|r, \mathcal{E}_r + (n-p)>$

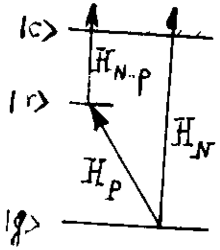


Fig. 24

Close to resonance, the absorption is described by:

1) a 2 stage process (a) p photon absorption described by an effective Hamiltonian H_p, (b) (N-p) photon absorption described by H_{N-p}.

2) a direct process excluding the resonant state $|r>$ described by H_N.

The states are non-degenerate, and one single continuum is introduced.

$$H_o = E_g|g><g| + E_r|r><r| + \int_{E_e}^{\infty} E|f_E><f_E|dE \tag{102}$$

Let us introduce P as we already did, and keep only the energy-conserving terms in the Hamiltonian (rotating-wave approximation)

$$\begin{aligned} H_{INT} = {} & |r><r|H_p|g><g| + \int_{E_o}^{\infty} |f_E><f_E|H_{N-p}||r><r|dE \\ & + \int_{E_o}^{\infty} |f_E><f_E|H_N|g><g|dE + C.C. \\ & + |r><r|H_D|r><r| + |g><g|H_D|g><g| \end{aligned} \tag{103}$$

H_{INT} is the Hamiltonian describing the effective interaction.

If $R = V + VQG_o\,QV + VQG_oQVQG_oQV + \ldots$ (displacement operator) (69)

$$H^D = VQG_oQV + \ldots \text{ (even terms).} \tag{104}$$

In fact $H_{INT} = P\,R^{ND}\,P + P\,R^D\,P$

R^{ND} = non diagonal elements of the displacement operator,

R^D = diagonal " " " .

At the lowest order if we introduce the E.M. field of intensity $\mathcal{E}$, frequency ω and phase $\varphi(t)$, we get for the matrix elements of the Hamiltonian interaction H_{INT}.

$$\left\{ \begin{aligned} & <r|H_p|g> = K \exp[-ip(\omega t+\varphi(t))] \\ & <f_E|H_{N-p}|r> = L_E \exp[-i(N-p)(\omega t+\varphi(t))] \\ & <f_E|H_N|g> = J_E \exp[-iN(\omega t+\varphi(t))] \end{aligned} \right. \tag{105}$$

$\Delta\mathcal{E}_g = <g|H^D|g>$

$\Delta\mathcal{E}_r = <r|H^D|r>$

Let us consider $\tilde{E}_g = E_g + \Delta E_g$, $\tilde{E}_r = E_r + \Delta E_r$.
If $|\Psi(t)>$ is the vector diagonalizing the total Hamiltonian, it is a linear combination of the three wave functions characterizing the dressed atom.

$$i|\dot{\psi}(t)> = (H_o + H_{INT}) \, |\Psi>$$

$$\Psi(t) = a_g(t) \exp^{-i\tilde{E}_g(t)}|g> + a_r(t) \exp^{-i(\tilde{E}_r+p\omega)t}|r> \tag{106}$$

$$+ \quad a_E(t) \exp^{-i(E_g+N\omega)t} dE \; |f_E>$$

The motion equations are obtained by the usual procedure or projection:

$$<g|\dot{\psi}> = \dot{a}_g = \frac{1}{i} [K \, a_r + \int_{E_i}^{\infty} d_E J_E a_E]$$

$$<r|\dot{\psi}> = (\dot{a}_r - i\Delta_r a_r) = \frac{1}{i} [K \, a_g + \int_{E_i}^{\infty} d_E L_E a_E] \tag{107}$$

$$<f_E|\dot{\psi}> = (\dot{a}_E - i\Delta_E a_E) = \frac{1}{i} [J_E \, a_g + L_E a_r]$$

$$\text{With } \Delta_r = \tilde{E}_g - \tilde{E}_r + p\omega \tag{108}$$

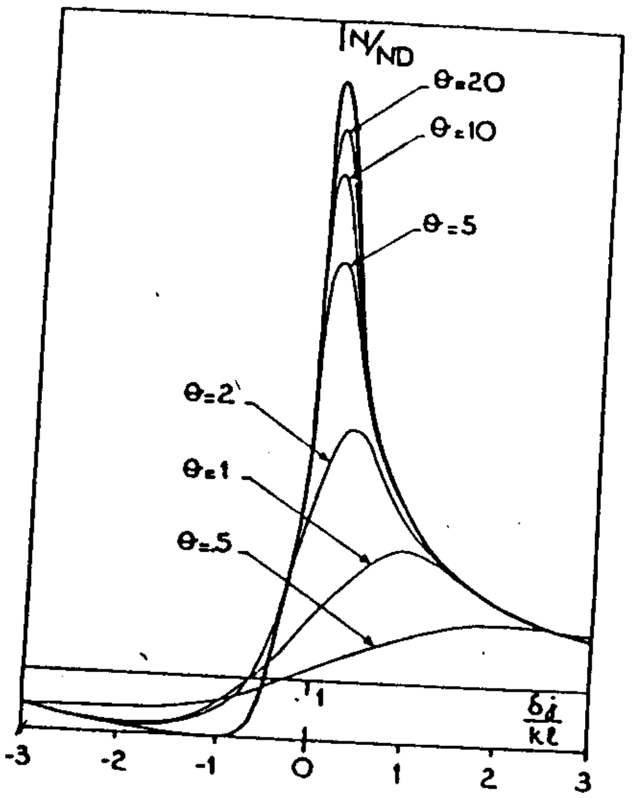
$$\Delta_E = \tilde{E}_g - E_i + N\omega$$

the system can be solved for $a_g(t)$ and $a_r(t)$, as we have already seen, the number of ions produced is:

$$\boxed{N_{(t)} = N_o \int_{E_o}^{\infty} dE \, |a_E \; (t)|^2 = N_o[1 - |a_r \; (t)|^2 - |a_g \; (t)|^2]} \tag{109}$$

The second expression is easier to derive than the first one. By developing the calculations, one finds the result appearing on next figure.

One must remark that for small T, the Rabi oscillations between the coupled states do not appear, the non resonant channel is competing with the resonant one, setting asymmetric Fano profile.



Crance, Feneville

Fig. 25

With $\Theta = L^2 T$

1) When $\Theta < 1$, transient regime.
2) For $\Theta > 1$, continuous regime.
3) Curves correspond to a rectangular pulse.
4) Nd is the number of ions produced by pure non resonant effect.
5) When Θ is not too big, the interference between resonant and non resonant process appears in the characteristic Fano profile.

COMPARISON OF THEORY WITH EXPERIMENT

We will limit ourselves to few examples. Let us consider figure 27 3 photon resonant, 4 photon ionization. The resonance curves are shown on figure 27 for four different intensities. The laser pulse duration is 15 picosecond (laser bandwidth 1.4 cm^{-1}). It is remarkable to notice the shift in resonance due to dynamic Stark shifts.

Let us compare the intensity dependent shift with theoretical predictions.

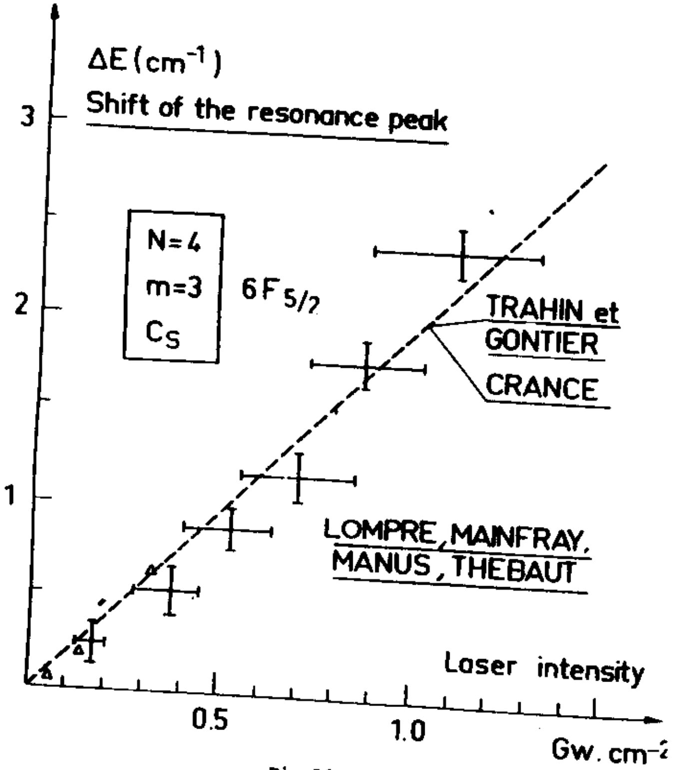
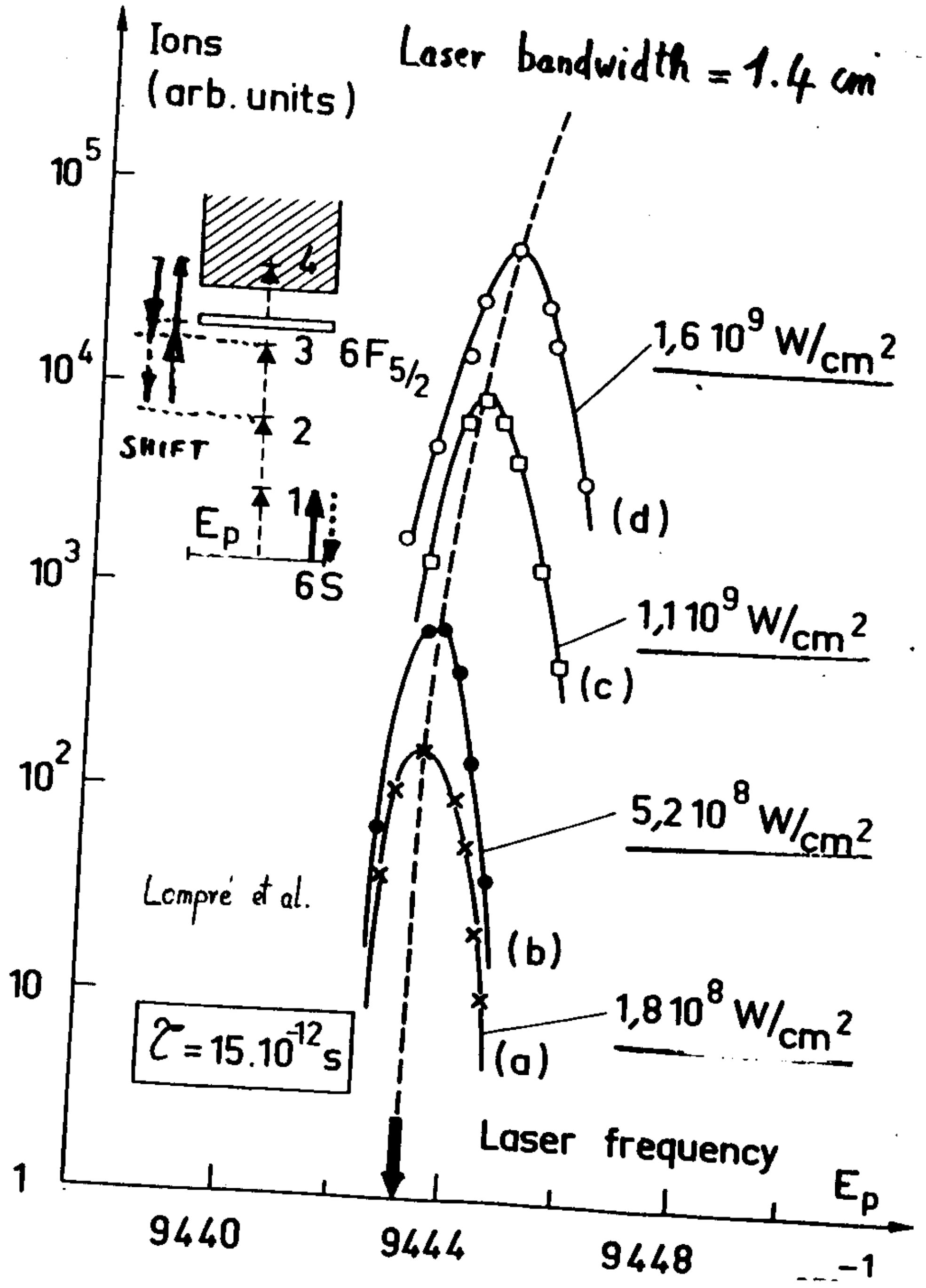


Fig.26

There is another way to describe resonance, by plotting the change in intensity at constant ion rate production (N_i).



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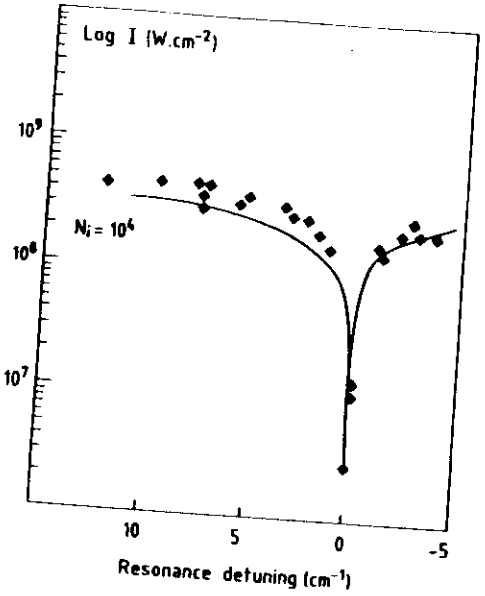
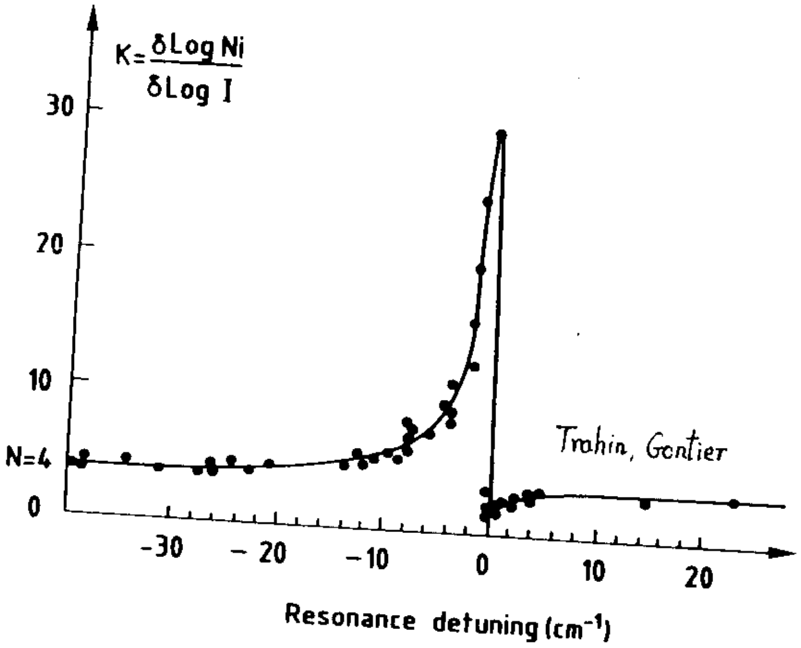


Crance

Fig. 28

The theoretical curve has been derived by the Effective Hamiltonian method (ABF).

The dependence of $K = \frac{d \text{ Log } N_i}{d \text{ Log } I}$ has been studied against resonance detuning.



Trahin, Gontier

Fig. 29

Calculation has been done by Trahin and Gontier, they have taken into account the Gaussian spatial distribution of the laser intensity.

In all these comparisons an excellent agreement between theory and experiment is reached.

We may conclude that the theoretical approach described, as well as the ABF method (Effective Hamiltonian), is perfectly adapted to the understanding of resonant multiphoton process. We have presently an excellent control of the basic physics related to these highly non-linear process.

But so far, we have only been concerned with interaction with one photon of one frequency (pure monomode coherent behaviour). We have to question what would happen in the case of a whole set of frequency, of an "incoherent" beam interacting with atoms ?

INFLUENCE OF THE COHERENCE OF LIGHT

Let us begin with the simplest approach and consider a "chaotic" light beam.

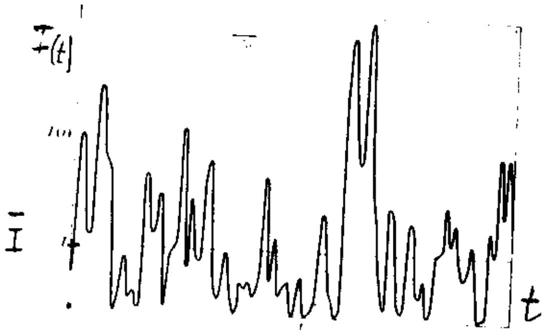


Fig. 30

I(t) is fluctuating erratically around a mean value of the intensity $\bar{I}$. It can be represented as a sum of an infinite number of waves with fluctuating independent phases.

Let us consider a random walk situation corresponding to a Gaussian distribution of the intensity I(t) with $p(I) = \frac{1}{\bar{I}}\, e^{-I/\bar{I}}$

(110)

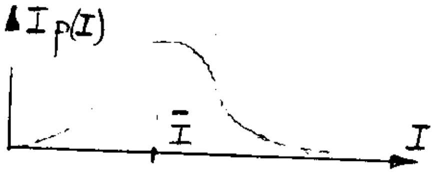


Fig. 31

Multiphoton ionization is proportional to $\langle I^N_{(t)} \rangle$

$$\langle I^N_{(t)} \rangle = \int_0^\infty \frac{1}{\bar{I}}\, I_{(t)}\, e^{-I(t)/\bar{I}}\, dI = N\,!\;\bar{I}^N \qquad (111)$$

$\langle I^N_{(t)} \rangle$ is proportional to the N^{th} order-coherence of the light, $f_N = N!$

In the case of a pure coherent monochromatic beam, where I is non fluctuating, in the case of $\bar{I}_{(t)} = I$, the probability of Multiphoton ionization is directly related to $\bar{I}^N$.

$$\text{Probability of Multiphoton absorption}\begin{cases}\text{coherent beam} \longrightarrow \div\ \bar{I}^N \\ \text{Gaussian incoherent beam} \longrightarrow \div\ N\,!\;\bar{I}^N \\ \text{(of same mean value of the intensity)}\end{cases}$$

$$\boxed{\frac{\text{M.A. (chaotic light)}}{\text{M.A. (coherent light)}} = N\,!} \qquad (112)$$

This very simple although dramatic increase may be derived by a pure quantum approach rigorously, result is the same.

Let us deduce the influence of the coherence on multiphoton ionization.

. The temporal fluctuation of a laser beam can be described by:

$I(t) = \bar{I}_M \cdot G(t) \cdot i(t)$

where $\bar{I}_M$ is the maximum time averaged intensity,

G(t) is the normalized temporal distribution function envelope of the laser intensity, i(t) is a stochastic function related to the incoherence of the beam intensity.

On next figure, a coherent monomode beam is shown, as well as an incoherent beam

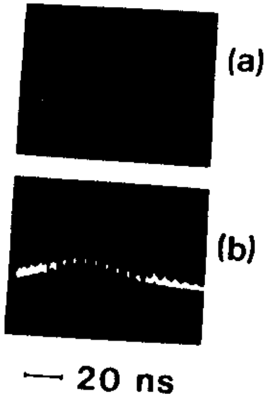


Fig. 32

Let us consider a pure monochromatic coherent light: I_c

$I_c = I_M . G(t)$.

$\mathcal{N}^+_c$ the number of ions formed by N order multiphoton absorption (coherent light).

$$\mathcal{N}^+_c = \beta \int I_M^N \, G^N_{(t)} dt \qquad (113)$$

In the case of an incoherent light with same temporal envelope:

$$\mathcal{N}^+_{inc} = \beta \int I_M^N \, G^N_{(t)} \cdot i^N dt = \mathcal{N}^+_{coh} \cdot \langle i^N_{(t)} \rangle = \mathcal{N}^+_{coh} \cdot f_N \qquad (114)$$

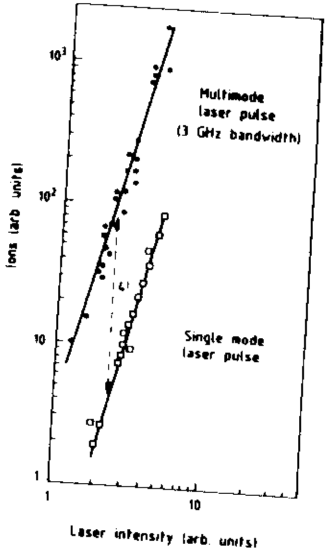
In the case of a Gaussian chaotic light, $f_N = N\,!$

$$\mathcal{N}^+_{(\text{incoherent light})} = \mathcal{N}^+_{(\text{coherent light})} \cdot f_N \qquad (115) .$$

The increase of ionization due to the change in temporal coherence will give a direct measurement of the N the order-coherence of the light.

COMPARISON WITH EXPERIMENT

The demonstration of such an effect is given in figure 33. We have plotted, in the case of a four photon non resonant ionization, the number of ions against the intensity, by using a single mode laser pulse. The laser is then operated with a large number of modes (phase independent). The upper curves show an increase corresponding to 4 ! for each value of the intensity.



Lompré et al.

Fig. 33

The most dramatic experimental demonstration of these effects has been performed in the 11 photon ionization of X_e atoms by varying the mode structure of a Nd glass laser probe from a single-mode to 100 modes !

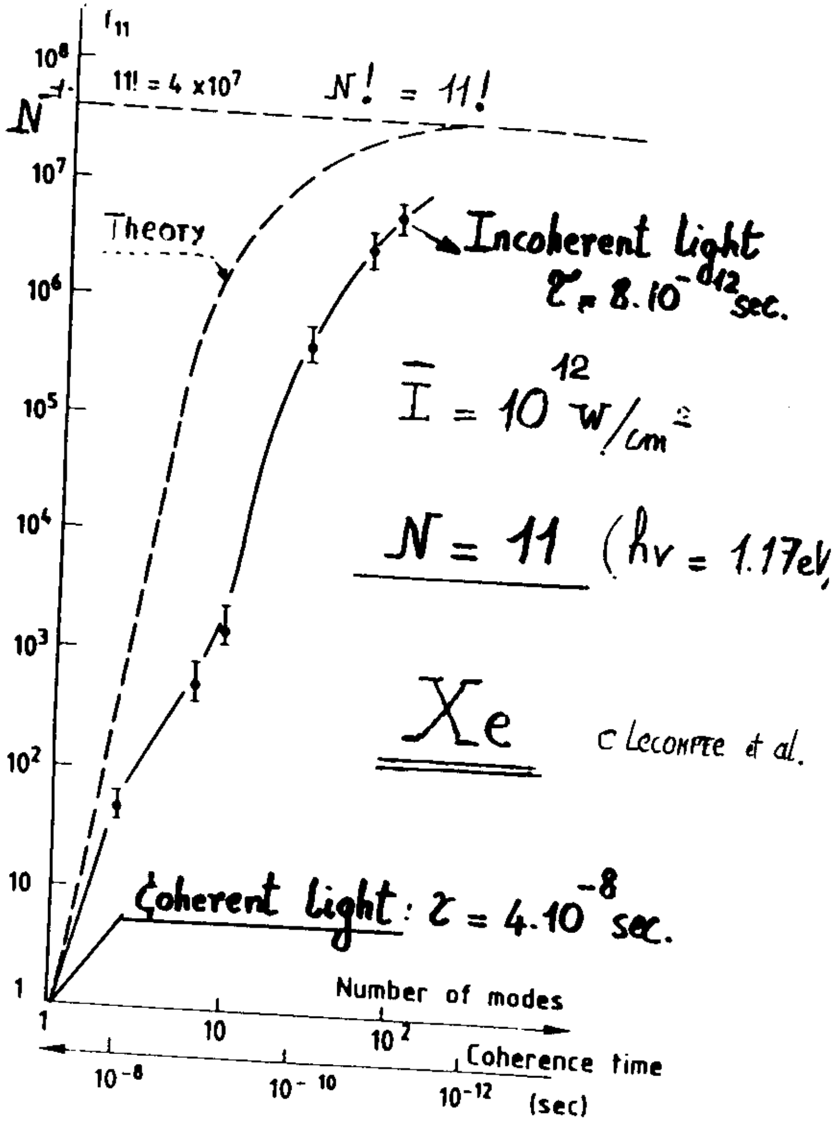


Fig. 34

The enhancement of the number of ions is plotted against the number of modes, or the inverse of the coherence time in sec (Fig. 34).

The number of ion is increased by nearly 10^7 !

A comparison is made with a calculation assuming that modes are phase-independent. Experimental values are systematically smaller than theoretical prediction !

The difference could be due to phase-locking effects between adjacent modes due to the high-intensity field appearing in the laser cavity. We have just examined the rôle of coherence on non resonant M.P. Let us examine the same process in the case of resonance.

COHERENCE EFFECTS IN R.M.I.

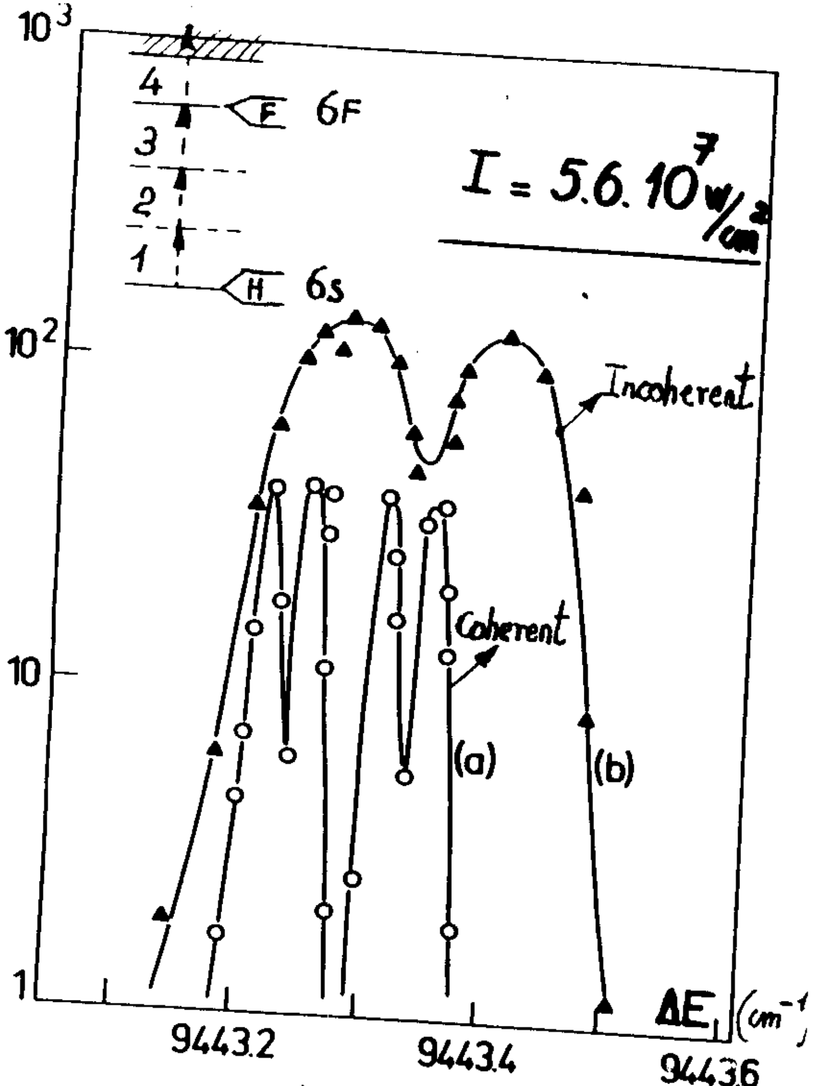
On figure 35 the resonance curves obtained with incoherent laser pulses are observed to be shifted and broadened (b) with regard to those (a) induced by coherent pulses with the same average intensity.

The statistical enhancement of the resonance shift is clearly demonstrated in figure 36 which shows the 2 laws of variation of the resonance shift as a function of the laser intensity, with a coherent pulse (dashed line) and an incoherent laser pulse (full line).

The difference between the 2 lines gives the additional shift due to the laser intensity fluctuations, i.e. $3{,}6 \pm 0.3$ $cm^{-1}/GW/cm^2$.

The ratio between the 2 shifts is ∿ 3.

[redacted]



Lompré et al.

Fig. 35

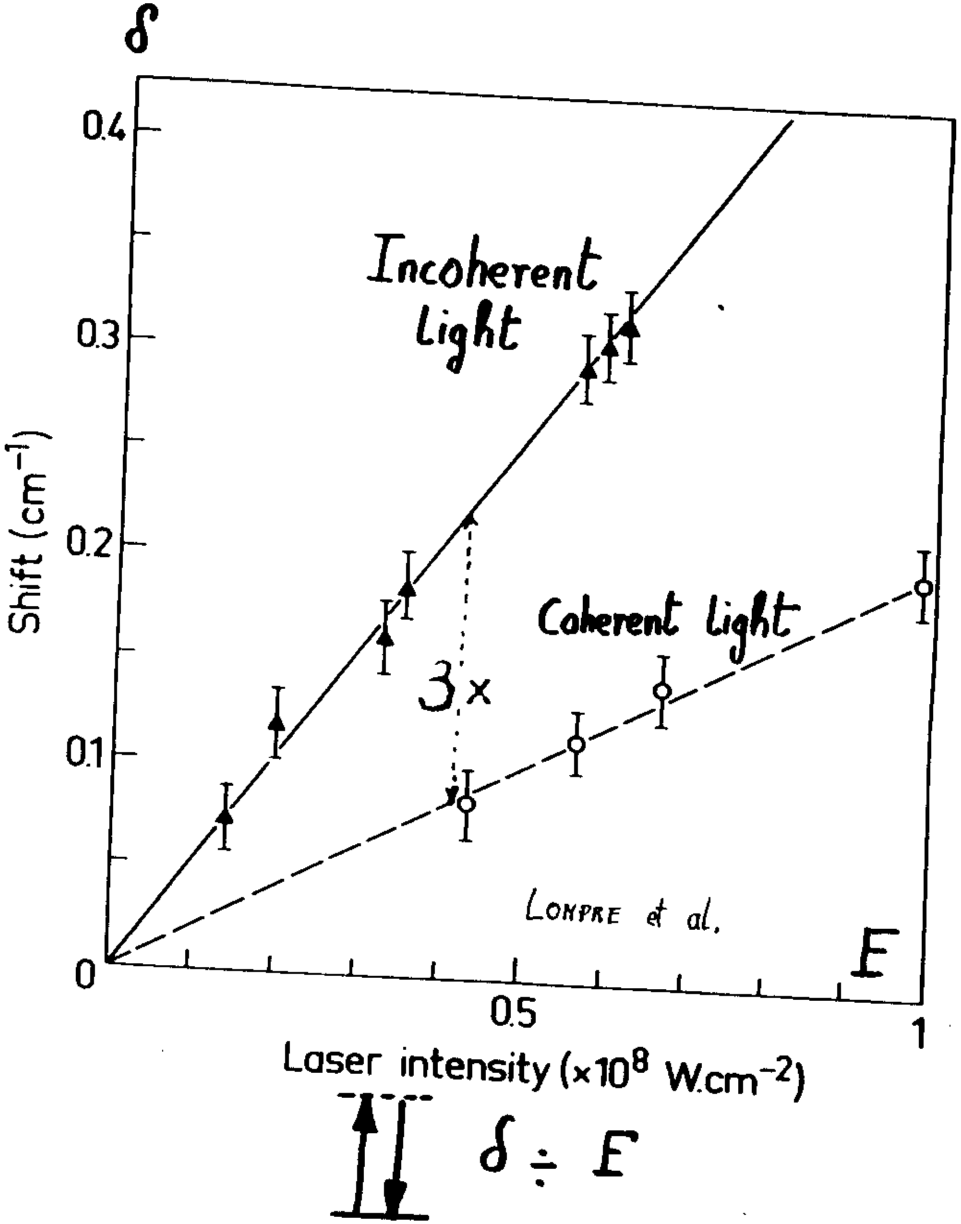


Fig. 36

BOUND-BOUND MULTIPHOTON TRANSITION AND SPECTROSCOPY

Doppler-free transition:

We will consider mainly 2 photon transition, according to figure 37

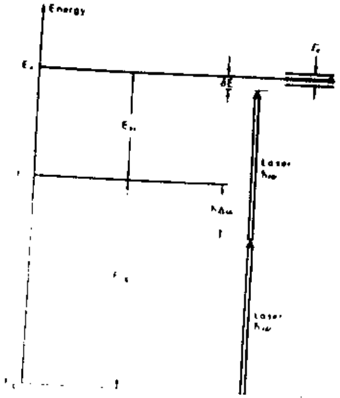


Fig. 37

The transition from E_g to E_e depends strongly on $\Delta\omega_j$.

If we consider the first photon absorbed, the atom, because of the uncertainty principle, stays in the j level a time $\simeq \frac{1}{\Delta\omega_j}$. The two photon transition takes place if a second photon is absorbed during the time spent by the atom in the j state. If $\Delta\omega_j$ is reduced the time increases and therefore the probability also increases.

Many different spectroscopical and collisional properties of the excited states formed have been studied. Lines are broadened by Doppler effect, all the experiments have been performed by using a Doppler-free method.

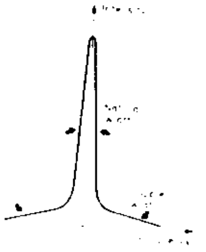
The first order Doppler shift is proportional to $\vec{k}.\vec{V}$, if the direction of the light is inversed, the shift is $-\vec{k}.\vec{V}$.

Let us consider a 2 photon-transition between E_g and E_e. In its rest frame, the atom interacts with 2 opposite travelling waves: $\omega+\vec{k}.\vec{V}$ and $\omega-\vec{k}\vec{V}$.

If the atom absorbs one-photon from each travelling-wave:

$$E_e - E_g = E_{eg} = \hbar(\omega + \vec{k}\vec{V}) + \hbar(\omega-\vec{k}\vec{V}) = 2\ \hbar\omega \qquad (116)$$

- All the atoms, irrespective of their velocities, can absorb 2 photons -



Theoretical two-photon line shape in a standing wave

Fig.38

The superposed Gaussian curves is due to the absorption of photons from the same travelling wave.

Some important results have been obtained:

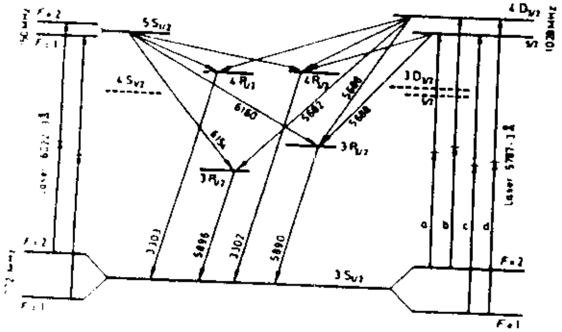
(1) in 5S → nD transitions in Rydberg atoms from n = 25 to n= 85 (by thermoionic detection) . (Collins)

(2) in 1S - 2S transitions in H (1S Lamb shift and 1S-2S isotope shift has been measured

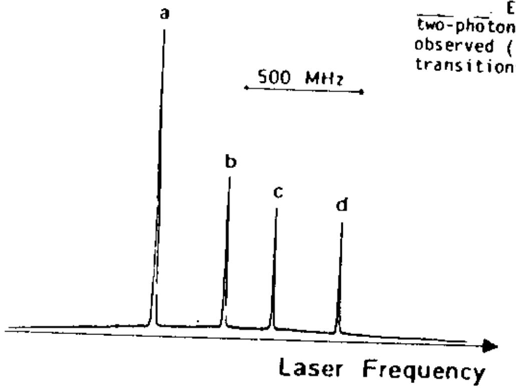
(3) collisional broadening and shift.

Figure 39 gives the Doppler free two-photon transition from 3S → 4D in Sodium.

[redacted]



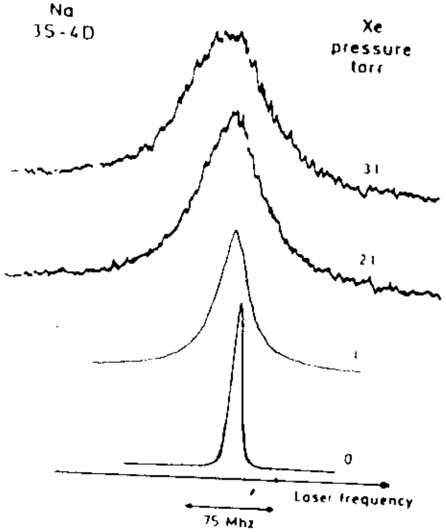
Energy diagram of sodium showing two-photon transitions which can be easily observed (the wavelengths of the optical transitions are indicated in Angström units)



Cagnac et al.

Experimental recording of the Doppler-free two-photon transition 3S → 4D
in sodium (fluorescence intensity vs the laser frequency). It shows the four com-
ponents predicted in Fig.4.6. The small Doppler background produced by absorption
of two photons of the same travelling wave can also be observed [4.28]

Fig. 39



Na(3S-4D) colliding with X_e at different pressure.

Cagnac et al.

Fig. 40

Theoretical approach

Let us derive the population of the excited state and appreciate if the light shift plays or not an appreciable rôle in the type of experiments we have described.

The problem is well suited to the use of the density matrix equation, it is written:

$$\frac{d}{dp} \rho = \frac{1}{i\hbar} [(H_o + R), \rho] - \left(\frac{d}{dt} \rho \right)_{rel.} \qquad (117)$$

R is the displacement operator. It can be seen as an effective Hamiltonian corresponding to a 2 photon-transition.

[redacted]

The laser electrical field is $E(t)$

$$E(t) = R_e \left\{ \underline{\varepsilon}\, \mathcal{E}\, e^{-i\omega t} \right\} = \frac{1}{2} \left(\underline{\varepsilon}\, \mathcal{E}\, e^{-i\omega t} + \underline{\varepsilon}^{*}\, \mathcal{E}^{*} e^{i\omega t} \right) \qquad (118)$$

$\underline{\varepsilon}$ is the polarization of the electromagnetic wave.

The following assumptions are made: $\Delta\omega_j$ is large compared to:

- the natural width of the g-j and j-e transition,
- the saturation width of the g-j and j-e transition,
- the 2-photon detuning $|2\hbar\omega - E_{eg}|$.

The interaction Hamiltonian is $-\vec{D}.\vec{E}$

$\vec{D}$ non relativistic electric dipole moment of the atom.

The last term represents the relaxation:

$$(119) \begin{cases} \left(\dfrac{d}{dt} \rho_{ee} \right)_{rel} = - \Gamma_e \rho_{ee} \\[2ex] \left(\dfrac{d}{dt} \rho_{gg} \right)_{rel} = \Gamma_e \rho_{ee} \\[2ex] \left(\dfrac{d}{dt} \rho_{eg} \right)_{rel} = - \Gamma_{eg} \rho_{eg} \end{cases}$$

Γ_e is the inverse of the radiative lifetime of e level

$\Gamma_{eg} = \Gamma_{e/2}$

All experiments being performed in the steady state regime, let us calculate the steady state solution. If N_e is the number of atoms in the $|e>$ state.

N_o number of atoms in the focal volume:

$$\frac{N_e}{N_o} = \rho_{ee(\omega)} = \frac{\hbar^2 \Gamma_{eg}\, a^2/2}{(E_{eg} + S - 2\hbar\omega)^2 + \hbar\, \Gamma_{eg}^2 (1+a^2)} \qquad (120)$$

S is the light shift:

$$S = \frac{1}{4} \left[<e|R^D|e> - <g|R^D|g> \right] \mathcal{E}^2 \qquad (121)$$

$$\text{and } a^2 = |<e|R^{ND}|g> \mathcal{E}^2|^2 \frac{1}{4h^2\, \Gamma_e\, \Gamma_{eg}} \text{ (saturation parameter)} \qquad (122)$$

The width of the 2 photon absorption line-shape is $\Gamma_{eg} \sqrt{1+a^2}$.

We deduce that the experiments must be performed at low $\mathcal{E}$, in order to have a Lorentzian line which is not enlarged by the saturation parameter a. $\frac{N_e}{N_o} \approx 10^{-6}$ to 10^{-8}.

When a < 1, the shift is normally much smaller than the natural line-width.

Successful attemps have been made at 3 photons, where the light shifts do not exceed the natural line-width in the usual range of operation $\frac{N_e}{N_o} = 10^{-6}$.

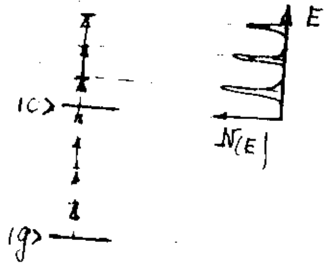
For 4 photon it may be calculated that the light shift is 100 times larger, of the order or larger of the Doppler width. This is the fundamental limitation of multiphoton spectroscopy.

TRANSITIONS IN THE CONTINUUM

Some experimentalists had observed, emission of electrons having energy higher than required by energy conservation, Evans and Thoneman, Martin and Mandel, Hollis...

The question arises: can photon be absorbed above the threshold ionization ?

Before describing the theoretical approach, let us consider the two main experimental results which have contributed to give an answer to such a question.



Let us first imagine what should happen if something like multiphoton in the continuum should exist. The absorption of N+S photons should create a discrete spectrum of electrons, each peak being separate from the other by an energy equal to the energy of a photon.

Fig. 41

This is what may be seen on the next two figures.

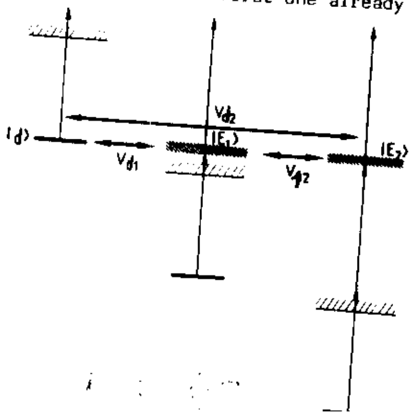
Theoretical description

Let us simplify the problem and consider the following situation, a three-state problem. The atom is ionized by absorption of one photon, and by two-photon absorption. The discrete state is coupled to two different continua. It is worth noting that such a problem had been already encountered in molecular physics and the theory given by Jortner and Beswick...

These states are described by:

$$|\varphi\rangle = C_d|d\rangle + \int dE_1\rho_1(E_1)\, C_{E_1}|E_1\rangle + \int dE_2\rho_2(E_2)C_{E_2}|E_2\rangle \quad (123)$$

Two methods may be successfully applied, the resolvant or the effective Hamiltonian. In order to preserve the unity of these lectures we will restrict ourselves to the first one already used in chapter one (Resolven



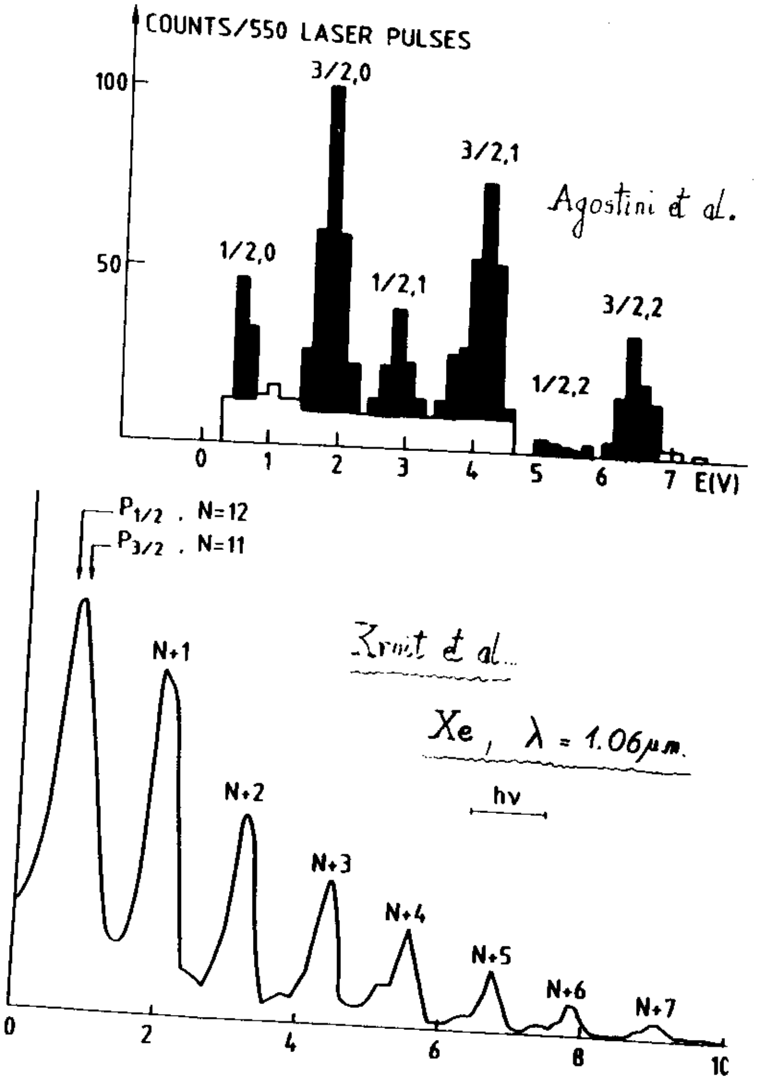


Fig. 43

With
$$V_{d1} = \langle d|V|E_1\rangle$$
$$V_{d2} = \langle d|V|E_2\rangle$$
$$V_{12} = \langle E_1|V|E_2\rangle \qquad (124)$$

The different dressed states of the continuum have the same energy.

We have to derive $C_d(t)$, $C_{E_j}(t)$ by:

$$\boxed{\begin{aligned} C_d(t) &= \frac{1}{2\pi i}\int_{C^+} dE\ e^{-iEt}\ G_{dd}(E) \\ C_{E_j}(t) &= \frac{1}{2\pi i}\int_{C^+} dE\ e^{-iEt}\ G_{jd}(E) \end{aligned}} \qquad (125) \quad \text{with } j = E_1, E_2$$

we use $G = G_0VG + G_0$ (126)

with $G_0(E) = \dfrac{1}{E-H_0+i\eta}$ $\quad \eta \to 0^+$ (127)

By (126), we derive G_{dd} and G_{1d}, G_{2d} - (with ρ density of states in the continuum:

$$(128)\quad \boxed{\begin{aligned} G_{dd}(E) &= \frac{1}{E-E_d+i\eta} + \frac{V_{d1}}{E-E_d+i\eta}\int dE_1\ \rho_1\ G_{1d}(E) + \frac{V_{d2}}{E-E_d+i\eta}\int dE_2\ \rho_2\ G_{20}(E) \\ G_{1d}(E) &= \frac{V_{1d}}{E-E_1+i\eta}G_{dd}(E) + \frac{V_{12}}{E-E_1+i\eta}\int dE_2\ \rho_2\ G_{2d}(E) \\ G_{2d}(E) &= \frac{V_{2d}}{E-E_2+i\eta}G_{dd}(E) + \frac{V_{21}}{E-E_2+i\eta}\int dE_1\ \rho_1\ G_{1d}(E) \end{aligned}}$$

Let us define:

$$\Gamma_{d1} = 2\pi|V_{d1}|^2\rho_1$$
$$\Gamma_{d2} = 2\pi|V_{d2}|^2\rho_2 \qquad (129)$$

$\Gamma_{d\substack{1\\2}}$ is the probability of transition from the discrete state to the continuum, it is the width of the ground state due to the coupling with each continuum.

$$\Gamma_{12} = \pi^2|V_{21}|^2\ \rho_1\rho_2 \qquad (130)$$

It can be considered as the probability of transition from 1 continuum to the other.

We introduce $\Gamma_d = \dfrac{\Gamma_{d1} + \Gamma_{d2}}{1 + \Gamma_{12}}$ (131)

After integrating (128) on all the continua and using (125), after some algebra, one finds:

$$G_{dd}(E) = \frac{1}{E - E_d - D_d + i/2\ \Gamma_d} \qquad (132)$$

D_d being the shift of the ground state due to coupling with the continua.

$$D_d = 2\pi^2\ \rho_1\rho_2\ \frac{V_{12}V_{2d}V_{d1}}{1 + \Gamma_{12}} \qquad (133)$$

$$\boxed{\begin{aligned} P_d(t) &= \exp{-\Gamma_d t} \\ P_1(t) &= \frac{\Gamma_{d1} + \Gamma_{d2}\ \Gamma_{12}}{(1+\Gamma_{12})^2\ \Gamma_d}\left(1 - \exp^{-\Gamma_d t}\right) \\ P_2(t) &= \frac{\Gamma_{d2} + \Gamma_{d1}\ \Gamma_{12}}{\Gamma_d\ (1+\Gamma_{12})^2}\left(1 - \exp^{-\Gamma_d t}\right) \end{aligned}} \qquad P_{(t)} = |C^2_{(t)}| \quad (134)$$

<u>These equations can be generalized to S continua, it is also possible to consider transition from $|d\rangle$ to $|E_1\rangle$ corresponding to a N order process.</u>

[redacted]

Physical meaning of (10)

(1) All the populations evolute according to the same temporal dependence $\exp^{-\Gamma_d t}$.

(2) The branching ratio of the 2 continua is given by R:

$$R = \frac{P_1(t)}{P_2(t)} = \frac{\Gamma_{d1} + \Gamma_{d2}\ \Gamma_{12}}{\Gamma_{d2} + \Gamma_{d1}\ \Gamma_{12}} \tag{135}$$

where $\Gamma_{12} = 0$, $R = \frac{\Gamma_{d1}}{\Gamma_{d2}}$ (136)

If Γ_{12} is large , $R = \frac{\Gamma_{d2}}{\Gamma_{d1}}$ (137)

A strong coupling between the 2 continua induce an inversion between the population of the continua.

(3) It is worth noting that as:

$$\Gamma_d = \frac{\Gamma_{d1} + \Gamma_{d2}}{1 + \Gamma_{12}} \tag{138}$$

Γ_d is reduced when Γ_{12} ceases to be negligible. Coupling between two states of the continua reduces the depopulation of the ground state. This result could not be obtained classically, it underlines an interfering effect between the population of the continua.

When more elaborate and complete calculations are performed (Trahin and Gontier, France) and values of Γ_{N+S} are derived, it is shown that $\Gamma_{N+S} = \Gamma_{N'}$ (for $\Gamma_{N'}$ only one continuum is populated) for N' = N+S, whatever S may be.

This can be understood in the following way: characteristic life time of a state of the continuum is much smaller than a discrete state, i.e. $\sim 10^{-14}$ to 10^{-15} sec, this is the same order of magnitude than the virtual states by which the atom proceeds for non resonant process.

Consequently the pseudo resonant states of the continuum behave almost like virtual states which leads to the idea that transitions in the continuum, if the intensity is not too large, and if only one electron is involved behaves like "classical transitions" below the continuum.

This was experimentally verified by a study on cesium (by F. Fabre et al...

There is some reason to believe that at large intensities, Γ_{12} could play a dominant rôle and change drastically the population in the continuum. Future will tell. Already some calculations have predicted such effects (Eberly, Bianicka-Birula) whereas some experiments could be interpreted as supporting such an interpretation (Van der Wiel...

APPENDIX

/ QUANTUM THEORY OF 2 PHOTON PROCESS / (with ionization)

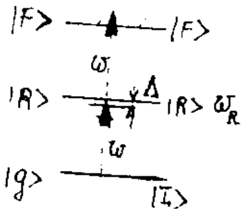


Fig.44

$I = |g,n>$

$F = |f,n-2>$

according to P. Lambopoulos, $R \equiv V$ (139)

The basic equation is:

$$\boxed{G = G_0 + G_0 V G}$$

Let us consider G_{FI} and G_{II}

$$G_{FI} = G^{\circ}_{FF} [\delta_{FI} + \sum_{M} V_{FM} G_{MI}] \qquad (140)$$

$$G_{II} = G^{\circ}_{II} [\ 1 + \sum_{M} V_{IM} G_{MI}] \qquad (141)$$

absorption of one photon excites state |r

State Detuning is $\Delta \equiv \omega - (\omega_a - \omega_g) \equiv \omega - \omega_{ag}$ (142)

The near resonant state $|R> = |r|n-1>$ must be considered. Other non resonant states of same parity have to be considered $|B> = |b,n-1>$

$|g> \rightarrow |R>$, $|b> \rightarrow |f>$

Selection rules allow single-photon transition:

$V_{FI} = 0$, V_{RI}, V_{BI}, V_{FR}, V_{RB} are non-vanishing.

The M states correspond to R, and the ensemble $|B\rangle$.

Let us introduce G_{RI}

$$\begin{cases} (Z-\omega_I)G_{II} = 1 + V_{IR}\, G_{RI} + \sum_{B} V_{IB} G_{BI} & (143) \\ (Z-\omega_A)G_{RI} = V_{RI}\, G_{II} + \sum V_{RF}\, G_{FI} & (144) \\ (Z-\omega_F)G_{FI} = V_{FR}\, G_{RI} + \sum_{B} V_{FB} G_{BI} & (145) \end{cases}$$

Let us neglect the contribution of the B non resonant states. Solving (3) for G_{FI} and substituting into (2), gives:

$$(Z-\omega_R)G_{RI} = V_{RI}\, G_{II} + \sum_{F} \frac{|V_{FR}|^2}{(Z - \omega_F)} G_{RI} \qquad (146)$$

We can replace Z by ω_R because it is only in the vicinity of ω_R that this value is significant.

$\sum_F$ gives a real and imaginary part representing shift and width of $|R>$ due to the coupling with the continuum.

Let us neglect in first approximation the shift (low intensity)

Width $\Gamma_R = \sigma I$ (single photoionization of state $|R>$)

$$\begin{cases} (Z-\omega_I)G_{II} = 1 + V_{IAR}\, G_{RI} & (147) \\ (Z-\omega_R - i\Gamma_R)\, G_{RI} = V_{RI}\, G_{II} & (148) \end{cases}$$

$$G_{II} = \frac{Z-\omega_R + i\Gamma_R}{(Z-\omega_I)(Z-\omega_R - i\Gamma_R) - (V_{RI})^2} \qquad (149)$$

$$G_{RI} = \frac{V_{RI}}{(Z-\omega_I)(Z-\omega_R - i\Gamma_R) - (V_{RI})^2} \qquad (150)$$

Roots of the denominator are:

$$Z^{\pm} = \left\{ \frac{1}{2} \ (\omega_I + \omega_R + i\Gamma_R) \pm [(\Delta + i\Gamma_R)^2 + 4|V_{RI}|^2] \right\} (151)$$

By introducing (4) in (3)

$$\boxed{G_{FI} = \frac{V_{FR}\, V_{RI}}{(Z-\omega_F)(Z-Z^+)(Z-Z^-)}} \qquad (152)$$

From which we can calculate U_{FI} by:

$$U_{FI}(Z) = \frac{1}{2\pi i} \int_C e^{-iZt} G(Z) dZ \qquad (153)$$

[redacted]

From which

$$U_{FI}(t) = \frac{1}{2\pi i} V_{FR}V_{RI} \left[\frac{\exp^{-i\omega_F t}}{(\omega_F - Z^+)(\omega_F - Z^-)} + \frac{\exp^{-iZ^+ t}}{(Z^+ - \omega_F)(Z^+ - Z^-)} + \frac{\exp^{-iZ^- t}}{(Z^- - \omega_F)(Z^- - Z^+)} \right] \qquad (154)$$

$$U_{FI}(t) = \exp^{-i\omega_F t} \frac{V_{FR}\ V_{RI}}{[(\Delta + i\Gamma_R)^2 + 4|V_{RI}|^2]^{1/2}} \left\{ \frac{1 - \exp^{i(\omega_F - Z^+)t}}{\omega_F - Z^+} - \frac{1 - \exp^{i(\omega_F - Z^-)t}}{\omega_F - Z^-} \right\} \quad (15$$

$$Z^+ - Z_- = [(\Delta + i\Gamma_R)^2 + 4(V_{RI})^2]^{1/2}$$

It is easy to check that where the detuning is dominant in $Z^+ - Z^-$, out of resonance conditions, we find:

$$W = \lim_{t\to\infty} \frac{1}{t} |U_{FI}(t)|^2 \cong \frac{2\pi |V_{FR}\ V_{RI}|^2}{\Delta^2} \div F^2 \qquad (156)$$

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