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WINTER COLLEGE ON HIGH RESOLUTION SPECTROSCOPY

(8 January - 2 February 1990)

NON-LINEARITY OF ATOMIC AND MOLECULAR RESONANCES IN OPTICAL TRANSITIONS - I

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OUTLINE OF LECTURES ON "OPTICAL RESONANCE and NONLINEAR PHENOMENA" – G.S. AGARWAL, SCHOOL OF PHYSICS, UNIVERSITY OF HYDERABAD, HYDERABAD, INDIA.

Two Level Approximation – Effective Hamiltonian, time evolution.

Quantized description of Laser Field – Jaynes-Cummings model.

Atomic coherent states – Properties, preparation, Radiative characteristics

Dressed states – Preparation, radiative aspects, use of dressed states for laser action without population inversion

Minimum Uncertainty states – properties, preparation, radiative characteristics.

Optical Bloch EQs – saturated response – power broadening, nonlinear χ's Steady states

Energy absorption from a probe in presence of a coherent pump, parametric gain – models for laser action without population inversion; pressure induced resonances in energy absorption

Spontaneous Emission – Mollow spectra

Four wave and six wave mixing – Effects of saturation and Collisions

Nonlinear Response in Bichromatic Fields of arbitrary intensities – Generation of subharmonic Rabi Resonances of Dressed state interpretation of subharmonic resonances.

Fluorescence in Fully modulated Fields, – New dressed states

Interaction with Resonant EM Fields.

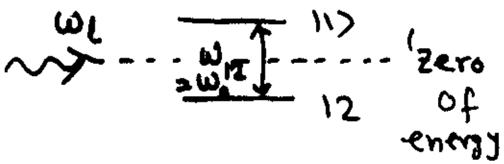
$H_1 = -\vec{d}\cdot\vec{E}(\vec{R},t)$ dipolar H

$\vec{d} = \sum_{ij} d_{ij} |\psi_i\rangle\langle\psi_j|$; unperturbed ψ's

$H_0 = \sum_j E_j |\psi_j\rangle\langle\psi_j|$

$\vec{E} = \vec{\mathcal{E}}\, e^{i\vec{k}\cdot\vec{R} - i\omega_\ell t} + c.c.$ Monochromatic

$\omega_\ell \sim \omega_{12}$



$\vec{d} = d_{12}|1\rangle\langle 2| + H.c.$

$H_1 = -\hbar\,(g|1\rangle\langle 2|\, e^{-i\omega_\ell t} + H.c.)$

$-\hbar\,(g'|1\rangle\langle 2|\, e^{i\omega_\ell t} + H.c.) \leftarrow$ Counter-Rotating

$g = \dfrac{\vec{d}_{12}\cdot\vec{\mathcal{E}}}{\hbar} e^{i\vec{k}\cdot\vec{R}}$, $g' = \dfrac{\vec{d}_{12}\cdot\vec{\mathcal{E}}^*}{\hbar} e^{-i\vec{k}\cdot\vec{R}}$ RWA drop

$\dfrac{\partial\psi}{\partial t} = -\dfrac{i}{\hbar}H\psi$, $\underbrace{\Phi = e^{i\omega_\ell s^z t}\psi}_{\text{Rotating Frame Trans.}}$

$\dot{\Phi} = -\dfrac{i}{\hbar}H_{eff}\,\Phi$

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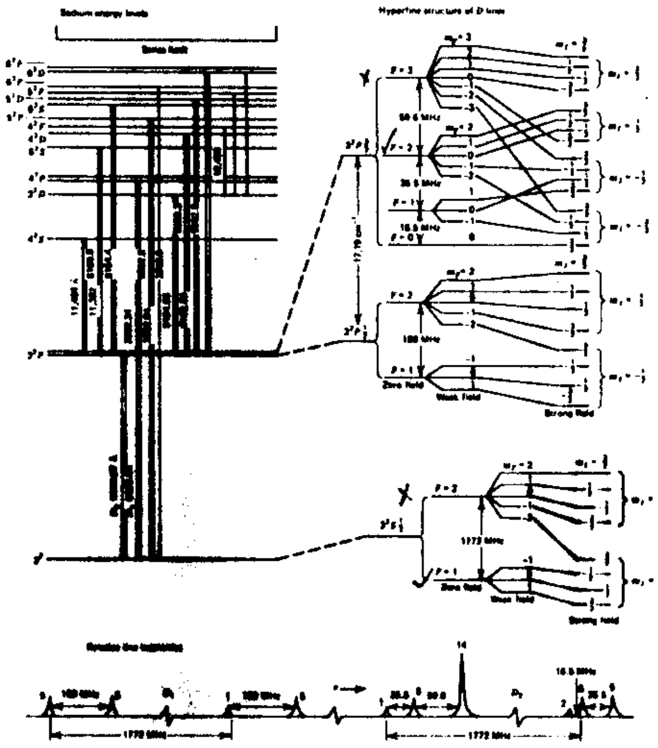


Fig. 2.3 A somewhat simplified diagram of the energy level scheme of sodium, with an expanded representation, not to scale, of the hyperfine structure involved in the D lines. [From F. Schuda and C. R. Stroud, Jr., with permission.]

(Ezekiel et al. observed resonance fluorescence between the levels marked by ([illegible]) ~5890 Å)

F for Hyperfine structure due to the interaction of the electron with the magnetic moment of the proton (I) $\vec{F} = \vec{J} + \vec{I}$, [illegible] $\vec{I}\cdot\vec{J}$

[illegible] subsequently

$$H_{eff} = \hbar \Delta S^z - \hbar (g S^+ + g^* S^-) = \hbar (\vec{\Omega} \cdot \vec{S})$$

$\Delta = \omega_0 - \omega_\ell$, $S^+ = |1\rangle\langle 2|$, $S^- = |2\rangle\langle 1|$

$S^z = \frac{1}{2}(|1\rangle\langle 1| - |2\rangle\langle 2|)$

spin-$\frac{1}{2}$ operators

$\Omega_x = -(g+g^*)$, $\Omega_y = -ig + ig^*$, $\Omega_z = \Delta$

Optical Resonance in 2 level system : view in rotating frame : isomorphic to spin in a magnetic field

Time evolution:

$$U(t) = \exp\left\{-\frac{i}{\hbar} H_{eff}\, t\right\}$$

$$= \cos\frac{\Omega t}{2} - \frac{i H_{eff}}{\hbar} \sin\frac{\Omega t}{2} \Big/ \frac{\Omega}{2},$$

$$|\psi(t)\rangle = \left(\cos\frac{\Omega t}{2} + \frac{i\Delta}{\Omega}\sin\frac{\Omega t}{2}\right) e^{i\omega_\ell t/2} |2\rangle$$

$$+ \frac{2ig}{\Omega} \sin\frac{\Omega t}{2}\; e^{-i\omega_\ell t/2} |1\rangle \quad \text{if } \underline{\psi(0) = |2\rangle}$$

◄ COHERENT SUPERPOSITION OF GROUND + EXCITED STATES. Atomic Coherence created

$$p_1(t) = \frac{4|g|^2}{\Omega^2} \sin^2\left(\frac{\Omega t}{2}\right) \qquad \sim \underline{|g|^2 t^2} \text{ for } \Omega t \ll 1$$

<u>Rabi osc.</u>

t^2 : No density of states (Fermi - Golden Rd t)

$\langle \vec{S}(t) \rangle$ Bloch Vector

$$\langle \vec{S}(t) \rangle \cdot \langle \vec{S}(t) \rangle = \frac{1}{4}$$

$$\langle S^+(t) \rangle = \frac{1}{2} \sin\theta(t)\, e^{i\omega_\ell t + i\varphi(t)}, \quad \text{Bloch } \underline{\text{sphere}}$$

$$\langle S^z(t) \rangle = -\frac{1}{2} \cos\theta(t)$$

$$\cos\theta(t) = 1 - \frac{8|g|^2}{\Omega^2} \sin^2\frac{\Omega t}{2}$$

$$\varphi(t) = \chi + \tan^{-1}\left(-\frac{\Omega}{\Delta} \cot\frac{\Omega t}{2}\right)$$

χ Phase of field $g = |g|\, e^{-i\chi}$

For field on resonance $\Delta = 0$,

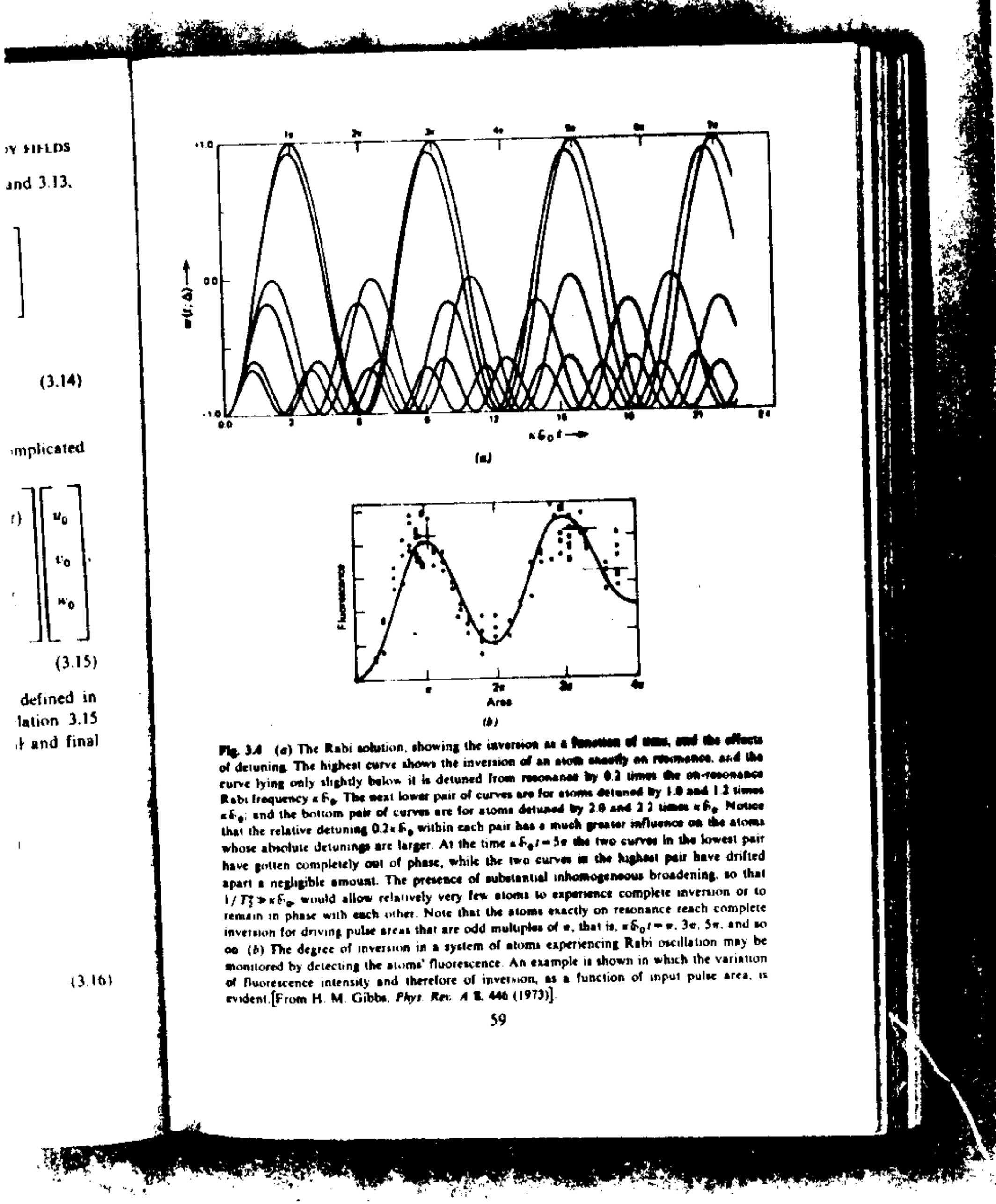
$\underline{\theta(t) = \Omega t}$, $\Omega^2 = 4|g|^2$

Ωt : Area of pulse for $\pi/2$ pulse

$\langle S^z \rangle = 0$, $\langle S^+ \rangle = -\frac{1}{2} i\, e^{i\omega_\ell t + i\chi}$

$$|\psi(t)\rangle = \frac{1}{\sqrt{2}} e^{i\omega_\ell t/2} |2\rangle + \frac{ig}{|g|\sqrt{2}} e^{-i\omega_\ell t/2} |1\rangle$$

Both states equally population; Maximum Coherence.



Y FIELDS

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Fig. 3.4 (*a*) The Rabi solution, showing the inversion as a function of time, and the effects of detuning. The highest curve shows the inversion of an atom exactly on resonance, and the curve lying only slightly below it is detuned from resonance by 0.2 times the on-resonance Rabi frequency $\kappa\mathcal{E}_0$. The next lower pair of curves are for atoms detuned by 1.0 and 1.2 times $\kappa\mathcal{E}_0$; and the bottom pair of curves are for atoms detuned by 2.0 and 2.2 times $\kappa\mathcal{E}_0$. Notice that the relative detuning $0.2\kappa\mathcal{E}_0$ within each pair has a much greater influence on the atoms whose absolute detunings are larger. At the time $\kappa\mathcal{E}_0 t = 5\pi$ the two curves in the lowest pair have gotten completely out of phase, while the two curves in the highest pair have drifted apart a negligible amount. The presence of substantial inhomogeneous broadening, so that $1/T_2^* \gg \kappa\mathcal{E}_0$, would allow relatively very few atoms to experience complete inversion or to remain in phase with each other. Note that the atoms exactly on resonance reach complete inversion for driving pulse areas that are odd multiples of π, that is, $\kappa\mathcal{E}_0 t = \pi, 3\pi, 5\pi$, and so on. (*b*) The degree of inversion in a system of atoms experiencing Rabi oscillation may be monitored by detecting the atoms' fluorescence. An example is shown in which the variation of fluorescence intensity and therefore of inversion, as a function of input pulse area, is evident. [From H. M. Gibbs, *Phys. Rev. A* **8**, 446 (1973)].

QUANTIZED FIELD DESCRIPTION : JAYNES-CUMMINGS MODEL

$H_1 = -\vec{d}\cdot\vec{E}$; $\vec{E}$ mode expansion

Single mode

$$\mathcal{H} = \hbar\omega_0 s^z + \hbar\omega a^\dagger a + \hbar\left(g s^+ a + g^* s^- a^+\right)$$

$$g = i\,\vec{d}\cdot\vec{\varepsilon}\left(\frac{2\pi\omega}{\hbar V}\right)^{1/2}$$

unperturbed state $|n, e\rangle$, $|n, g\rangle$, $n = 0, 1, \ldots \infty$

$$\mathcal{H}|n, e\rangle = \left(\frac{\hbar\omega_0}{2} + \hbar\omega n\right)|n, e\rangle + \hbar g^* \sqrt{n+1}\,|n+1, g\rangle$$

$$\mathcal{H}|n+1, g\rangle = \left(-\frac{\hbar\omega_0}{2} + \hbar\omega(n+1)\right)|n+1, g\rangle + \hbar g\sqrt{n+1}\,|n, e\rangle$$

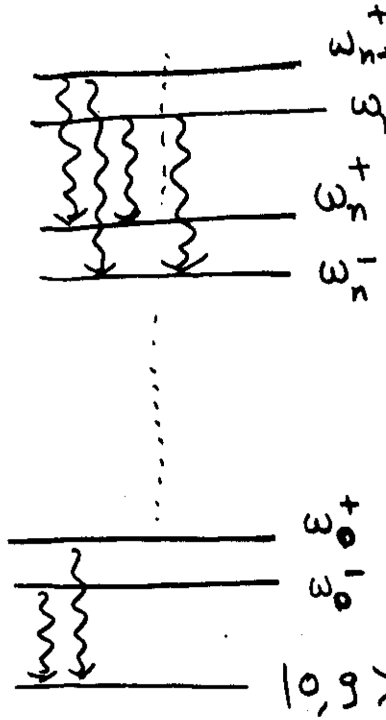
$|n, e\rangle \leftrightarrow |n+1, g\rangle$ Coupling

$$\mathcal{H} = \begin{pmatrix} \boxed{\hbar\omega} & & & & 0 \\ & \boxed{2\times2} & & & \\ & & \boxed{2\times2} & & \\ & & & \boxed{2\times2} & \\ 0 & & & & \ddots \end{pmatrix} ; \left[\begin{array}{c:c} \frac{\hbar\omega_0}{2} + \hbar\omega n & \hbar g^* \sqrt{n+1} \\ \hdashline \hbar g\sqrt{n+1} & -\frac{\hbar\omega_0}{2} + \hbar\omega(n+1) \end{array}\right]$$

diagonalization easy.

$$\hbar\omega_n^{\pm} = \omega(n+\tfrac{1}{2}) \pm \frac{\Omega_{n\Delta}}{2}$$

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



$$\updownarrow = \Omega_{n\Delta} = \sqrt{\Delta^2 + 4g^2(n+1)}$$

$$\xrightarrow{\text{larger}} 2g\sqrt{n}\ , \quad \Delta = 0$$

"For each n doublet"

DRESSEDSTATES

$$|\psi_n^{\pm}\rangle = \begin{pmatrix}\cos\theta_n \\ -\sin\theta_n\end{pmatrix}|n+1, g\rangle + \begin{pmatrix}\sin\theta_n \\ \cos\theta_n\end{pmatrix}|n, e\rangle$$

coherent superposition

$$\tan\theta_n = 2g\sqrt{n+1}\ /(\Omega_{n\Delta} - \Delta)$$

$$\Delta = 0, \quad \theta_n = \pi/4$$

$$U(t) = e^{-iHt}$$

Atomic coherent states

$$|\theta,\varphi\rangle = \exp\{-i\theta\,(S^x \sin\varphi - S^y \cos\varphi)\}\,|S,-S\rangle$$

$$= \sum_{-S}^{+S} \begin{pmatrix}2S \\ S+m\end{pmatrix}^{1/2} \sin^{S+m}\frac{\theta}{2}\,\cos^{S-m}\frac{\theta}{2}\; e^{-i(S+m)\varphi}\,|S,m\rangle$$

$$\langle S^+\rangle = \frac{N}{2}\sin\theta\; e^{i\varphi}$$ Macroscopic dipole moment ⇒

coherent radiation $\propto |\langle S^+\rangle|^2 \propto N^2$: Nonlinear Signals Photon echoes etc.

completeness

$$\frac{2S+1}{4\pi}\int \sin\theta\, d\theta\, d\varphi\; |\theta,\varphi\rangle\langle\theta,\varphi| = 1$$

$$\langle\theta,\varphi|\theta',\varphi'\rangle = e^{iS(\varphi-\varphi')}\left[\cos\frac{\theta-\theta'}{2}\cos\frac{\varphi-\varphi'}{2} - i\cos\frac{\theta+\theta'}{2}\sin\frac{\varphi-\varphi'}{2}\right]^{2S}$$

$\hat{O}\,|\theta,\varphi\rangle = (\ )\,|\theta,\varphi\rangle$ what is $\hat{O}$

$$= S^z\cos\theta - (\sin\varphi\, S^y + \cos\varphi\, S^x)\sin\theta$$

Bosons : Field $|z\rangle = \exp[\alpha a^{+} - \alpha^{*} a]\,|0\rangle$

$$a|z\rangle = z|z\rangle \;\Big|\; = \sum \frac{\alpha^n}{\sqrt{n!}}\, e^{-\frac{1}{2}|\alpha|^2}\,|0\rangle$$

Minimum Uncertainty states:

$$\Delta x\,\Delta p = \frac{\hbar}{2}$$

$b = \mu a + \nu a^{+}$, $\quad b\,|z\rangle = z\,|z\rangle$

$\underline{\mu^2 - \nu^2 = 1}$ $\quad$ if $\underline{\nu = 0}$: usual Coherent States

$\nu \neq 0$; squeezed coherent states

Atomic problem : $\Delta S_x\,\Delta S_y = \frac{1}{2}\,|\langle S_z\rangle|\,\hbar$

$S^{+}|S, S\rangle = 0$, $\quad S^{-}|S, -S\rangle = 0$ only one state

look for linear combination of S^{+}, S^{-}

$$R^{z} = \frac{1}{\sqrt{1-\alpha^2}}\left(S^{x} - i\alpha S^{y}\right) ; \quad \alpha \neq \pm 1$$

$$R^{z}\,|\chi_m, \alpha\rangle = m\,|\chi_m, \alpha\rangle$$

$$\Downarrow$$

$$\underline{e^{\theta S^{z}}\; e^{-\frac{i\pi}{2} S^{y}}\; |s, m\rangle} \;,\quad e^{\theta} = \sqrt{\frac{1-\alpha}{1+\alpha}}$$

α = Real, $\uparrow$ minimum uncertainty state

Non unitary Transformation $\Rightarrow$ $\underline{\text{can not}}$ be generated from Hamiltonian evolution unlike

photon case – Hamiltonian evolution $\Rightarrow$ atomic Coherent states. $\quad \mathcal{H} \sim (\alpha a + \beta a^2 + \text{H.c.})$

Consider a collection of N atoms + spontaneous emission i.e. Vacuum of the radiation field $\Rightarrow$ steady state $|\frac{N}{2}, -\frac{N}{2}\rangle$ all atoms in ground state

However if Vacuum $\rightarrow$ broad band squeezed vacuum, then steady state

$$|\chi_0, \alpha\rangle = e^{\theta S^{z}}\; e^{-i\pi/2\, S^{y}}\; |0\rangle$$

[GSA + RR PURI, Opt. Commun. $\underline{69}$, 267 (1989); Phys. Rev. $\underline{A}$ in press 1990]. Thus eigenstates of $\mu S^{+} + \nu S^{-}$ can be generated.

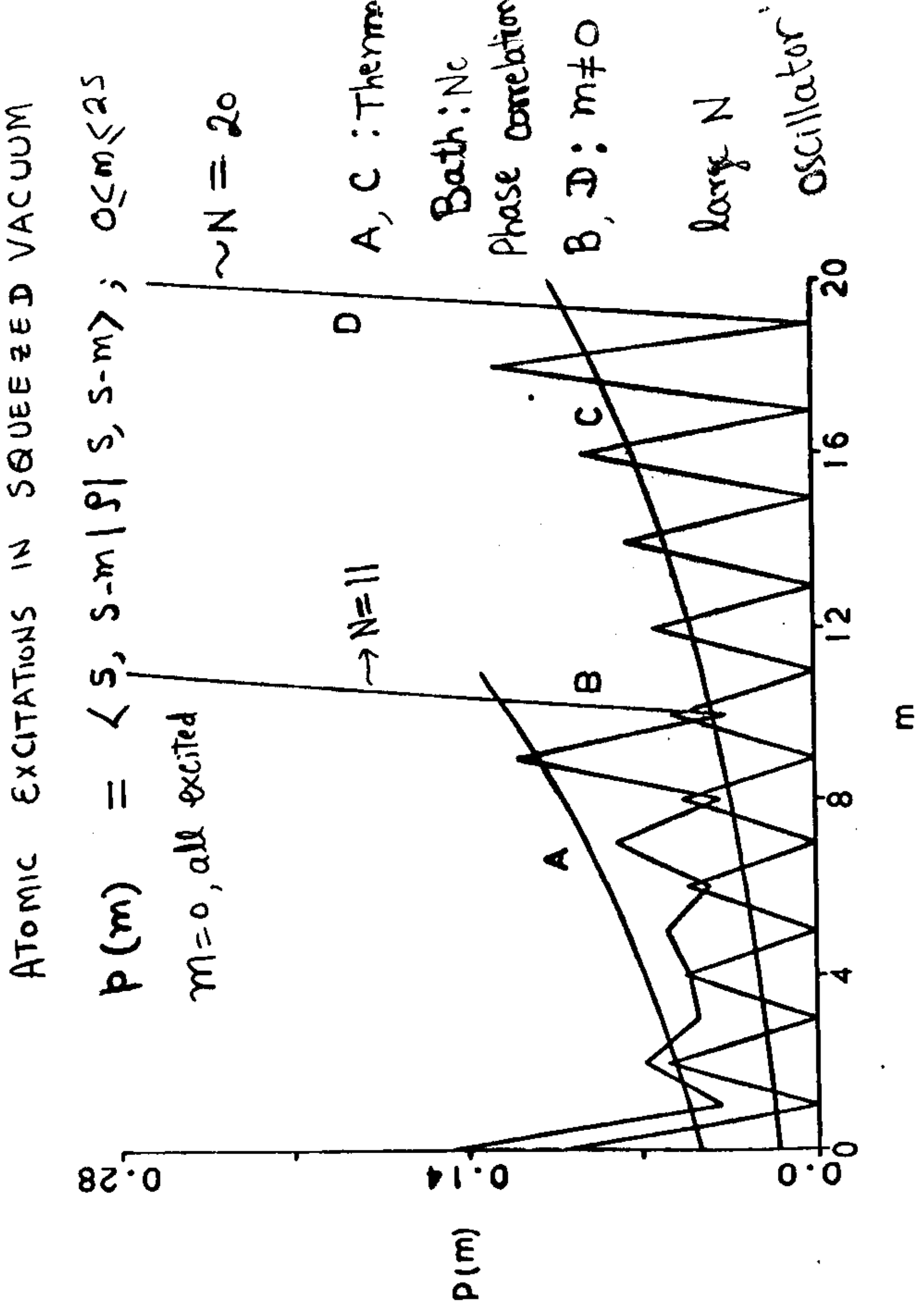
overlap $|\langle m|\chi_0\,\alpha\rangle|^2$: population distribution in the states $|S, m\rangle$

$$\langle m|\psi_0\rangle = A_0\, e^{m\theta}\, d^{S}_{m0}(\pi/2)$$

$$\Downarrow$$

$$\left(\frac{(S+m)!\,(S-m)!\,S!\,S!}{2^{S}}\right)^{1/2} \sum_{p=m}^{S-m} \frac{(-1)^{p}}{(S-p)!\;p!\;(p-m)!\;(S+m-p)!}$$

$= 0$ if N is even and $N/2 + m$ = odd $\Rightarrow$ PAIRWISE EXCITATION !

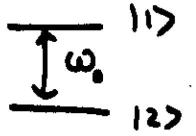


Semi- classical Dressed states

Atom + laser field ; rotating frame

$H_{eff} \sim \hbar \Delta s^z - \hbar (g s^+ + g^* s^-)$

$H_{eff}|\psi_\pm\rangle = E_\pm |\psi_\pm\rangle$



$E_\pm = \pm \frac{\hbar\Omega}{2}$, $|\psi_\pm\rangle = \left(|1\rangle + \frac{\Delta\mp\Omega}{2g}|2\rangle\right)\mathcal{N}$

$|\Omega| = \sqrt{\Delta^2 + 4|g|^2}$

Atom in |1> or |2> + laser field → Time evolution

Transitions possible

Atom in $|\psi_+\rangle$ or $|\psi_-\rangle$ + laser field → No more time evolution - as steady state or stationary state - Time evolution, however, is still possible over times ~ T_1, T_2.

QUESTIONS : (i) HOW TO PREPARE ATOMS IN DRESSED STATES (ii) WHAT ARE THE RADIATIVE PROPERTIES OF SUCH STATES ?

PREPARATION

$\Delta = 0$, $|\Psi_\pm\rangle = \frac{1}{\sqrt{2}}\left(|1\rangle \mp e^{i\chi}|2\rangle\right)$; $g = |g|e^{-i\chi}$

phase is very imp.

atom in $|2\rangle$ + $\frac{\pi}{2}$-pulse phase χ'

$$|\Psi(t)\rangle = \frac{1}{\sqrt{2}}\left(|1\rangle - i\,e^{i\chi'}|2\rangle\right)\left(i\,e^{-i\chi'}\right)$$

choose $\chi = \chi' + \frac{\pi}{2}$ $\quad |\psi\rangle \rightarrow |\Psi_+\rangle$

$\quad = \chi' - \pi/2$ $\quad |\psi\rangle \rightarrow |\Psi_-\rangle$.

change of phase by $\pm\,\pi/2$ leads to $|\Psi_\pm\rangle$.

H_{eff} (before phase change)

$$= -\hbar\,|g|\left(S^+ e^{-i\chi'} + c.c.\right)$$

$$\xrightarrow[\text{of phase}]{\text{after change}} -\hbar\,|g|\left(S^+ e^{-i\chi} + c.c.\right)$$

Monitoring such Dressed states :

(i) Fluorescence from excited state

$I(t)$ or spectrum $I(\omega, t)$

(ii) Probe absorption $\overset{\omega}{\rightsquigarrow}$

$S_A(\omega, t)$ etc. $\omega_\ell \rightsquigarrow$

Choose very long lived atoms

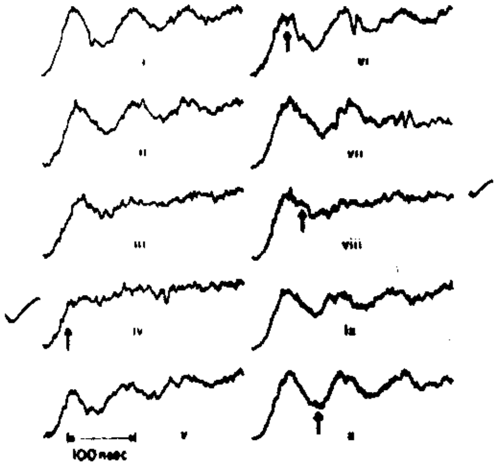


FIG. 4. Effect of a π/2 phase shift applied at successively later times on the fluorescence intensity vs time (see text).

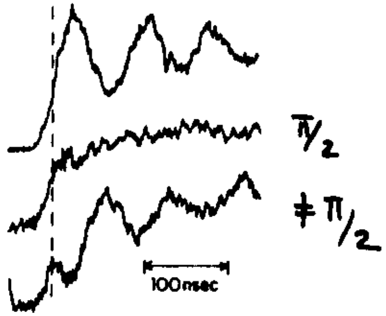


FIG. 6. Effect of phase shifts of various magnitudes on the fluorescence intensity vs time (see text).

optic modulators and phase controlled with an electro-optic modulator. High-voltage power metal-oxide-semiconductor field-effect transistors were employed to introduce variable-amplitude phase shifts with switching times of ~ 8 nsec. To ensure a very uniform laser intensity throughout the laser-Yb interaction region, a 1-mm-square aperture (A1) was positioned to pick out the uniform-intensity central region of the laser beam, and two well-corrected doublet lenses imaged (at unity magnification) the aperture into the atomic beam. Laser power in the interaction region was about 10 mW. The beam contained all the natural isotopes, but they could be easily separated spectrally. Scans of the ^{174}Yb absorption profile revealed a 5-MHz residual beam Doppler width. The effects of stray magnetic fields in the laser-Yb interaction region were minimized by applying a ~ 6-G magnetic field, coaxial with the circularly polarized laser field. Short-focal-length Fresnel lenses provided a fluorescence collection efficiency of about 10%. The excitation sequence was repeated at approximately 10 kHz, and fluorescence intensity was recorded with a gated boxcar integrator. A complete scan of the fluorescence intensity versus time for a particular excitation sequence required about 100 sec. Longer scan times were precluded by laser frequency drift. The atomic beam was run at rather low density; only about 1000 atoms were presented in the interaction region during a particular laser pulse.

In Figs. 4–6, we show recordings of the fluorescence intensity, I_F, versus time during various excitation sequences of the form shown in Fig. 1(b). In Fig. 4, the effect of a fixed $\pi/2$ phase shift $(\varphi_1 - \varphi_0)$ applied at successively larger values of t_{10} (and hence θ_{10}) is examined. In trace (i), there is no phase shift (equivalent to $t_{10} = 0$), and I_F should ideally display the simple oscillatory behavior characteristic of Eq. (9a). The observed damping arises primarily from residual laser-intensity inhomogeneity in the interaction region, but other effects, such as transit time, natural decay, and residual Doppler broadening, also contribute. In the remaining traces, (ii) through (x), the phase shift occurs farther and farther into the pulse. Arrows below several traces indicate the approximate time at which the phase shift was applied. Note the

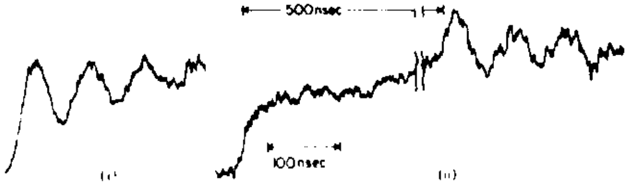
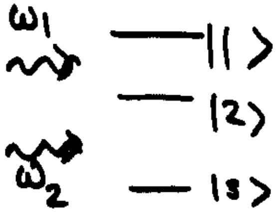


FIG. 5. Reduction of dephasing in stationary atom-field states (see text).

Mossberg + coworkers

Three Level case



$$H_1 = -\hbar g_1 |1><2| e^{-i\omega_1 t} - \hbar g_2 |2><3| e^{-i\omega_2 t} + \text{c.c.} \qquad (\text{RWA})$$

$$A_{\alpha\beta} = |\alpha><\beta| \quad , \quad \dot{\psi} = -\frac{i}{\hbar}(H_0 + H_1)\psi$$

Rotating frame

$$\psi = \exp\{-i((\omega_1+\omega_2)A_{11} + \omega_2 A_{22})t\}\,\phi$$

$$\dot{\phi} = -\frac{i}{\hbar} H \phi$$

$$H = \hbar(\Delta_1 + \Delta_2)A_{11} + \hbar \Delta_2 A_{22} - (\hbar g_1 A_{12} + \hbar g_2 A_{23} + \text{H.c.})$$

$\Delta_1 = \omega_{12} - \omega_1$

$\Delta_2 = \omega_{23} - \omega_2$

Exact eigenvalues of H determine the radiative properties of three level atom.

Probe absorption : ω_1 – weak
ω_2 – strong

Induced Pol at ω_1 : $\rho_{12}(t)$

$$\rho_{12}^{(1)}(t) = i g_1 \int_0^t d\tau\, e^{-i(\Delta_1+\Delta_2)(t-\tau)}\, \psi_2^{(0)}(\tau)\, \psi_2^{(0)*}(t)$$

$\psi_2^{(0)}$ wave function in the absence of probe depends on initial preparation.

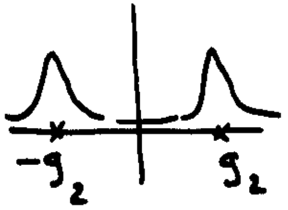
A: Initial state $|3>$; $\Delta_2 = 0$

$$\psi_2^{(0)} = i \sin(g_2 t) \Rightarrow$$

$$\text{Im}\,\rho_{12}(t) = \frac{g_1}{4}\left[\frac{\sin(g_2+\Delta_1)t}{(g_2+\Delta_1)} + g_2 \to -g_2\right] - \frac{g_1}{2}\left[\frac{2}{(\Delta_1+g_2)} \sin\frac{(g_2+\Delta_1)t}{2} \cos\frac{(\Delta_1-3g_2)t}{2} + g_2 \to -g_2\right]$$

Resonances at $\Delta_1 = \pm g_2$

Autler - Townes Doublet



Normally in steady state

$$\lim_{t\to\infty} \text{Im}\,\rho_{12}(t) \sim \delta(g_2 \pm \Delta_1) \Rightarrow \frac{\Gamma_+/\pi}{\Gamma_+^2 + (\Delta_1 \mp g_2)^2} + (\text{other peak})$$

$\Delta_1 = \omega_{12} - \omega_1$

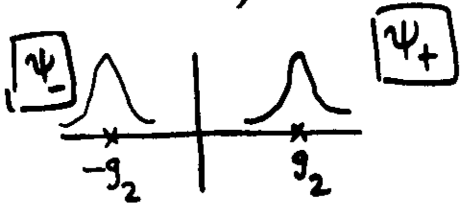
B: Dressed state Preparation

$$h\,\psi_+ = g_2 \psi_+ , \quad h = -\hbar g_2 |2>(3| + \text{H.c.}$$

$$\psi_2^{(0)}(t) = e^{-ig_2 t}\,\psi_+$$

$$\text{Im}\, \rho_{12}(t) = g_1 |\psi_+|^2 \frac{\sin t(\Delta_1 - g_2)}{(\Delta_1 - g_2)}$$

$$\Delta_1 = g_2$$



As t ↑ relaxation effects will mix the states ψ_+ and ψ_- leading to the development of the second peak even if one were there to start with.

Take transparencies from my laser no population inversion paper

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was locked to a magnetically tunable saturation resonance of a Yb reference cell, acousto-optically amplitude gated, electro-optically phase controlled, and resonant with the $(6s^2)^1S_0$-$(6s6p)^3P_1$ transition of ^{174}Yb. A weak cw probe laser (linewidth ≃ 1 MHz) was tuned slowly through resonance with the $(6s6p)^3P_1$-$(6s7s)^1S_0$ transition. Cascade fluorescence [see Fig. 1(a)] constituted the signal observed. In the interaction region, the driving and probe lasers had $1/e$-intensity diameters of ≃ 1.5 and ≃ 0.5 mm, respectively, and were counterpropagating so as to minimize residual Doppler effects. The stainless-steel Yb oven was maintained at 840 K. During each experimental cycle, approximately 10^5 Yb atoms were in the laser excitation volume. A ≃ 5-G magnetic field oriented along the driving-laser propagation axis and the laser polarizations shown in Fig. 1(b) were employed to select a single Zeeman level of the $^3P_1$ state. Ideally, this experiment should be conducted in a sample of equivalent atoms, all of which are excited by the same perfectly controlled laser fields. This situation is approximated fairly well, but not exactly with the experimental system described.

In each experimental cycle, initially ground-state atoms were exposed to a square driving-field pulse several microseconds in duration. The probe field was always on. The time development of the probe-induced fluorescence signal was recorded by a transient digitizer with a 10-nsec sampling interval and a 20-MHz analog bandwidth. About 10 fluorescence photons were detected in each experimental cycle. Typically, 64 000 experimental cycles were averaged for each probe-laser frequency, and a single time-dependent Autler-Townes spectrum (see Fig. 2) combines measurements at 26 probe-laser frequencies. The experiment was operated at 7.5 kilopulses/sec.

In Fig. 2(a), we show a transient Autler-Townes spectra obtained when the driving field (Rabi frequency ≃ 15 MHz at beam center) was switched on and its phase held fixed. Prior to $t = 0$, no probe signal is observed, because the $^3P_1$ state is empty. For $t > 0$, two identical peaks appear. In Fig. 2(b) [2(c)], the phase of the driving field was abruptly shifted by approximately + 90° (− 90°) shortly after turnon. The asymmetry of the peaks is striking. The delay (≃ 20 nsec) of the phase shift relative to the driving-field switch-on was experimentally set to maximize the short-time contrast between the two spectral peaks, but corresponds well with the interval needed for the driving field to have accumulated an area of $\pi/2$.

Measurements of the probe-induced fluorescence signal versus time obtained when the probe laser is tuned to the center of specific spectral peaks are shown in Fig. 3(a). The trace labeled (i) [(iii)] was obtained with the probe tuned to the enhanced (suppressed) peak as in Fig. 2(b) (+ 90° phase change). Trace (ii)

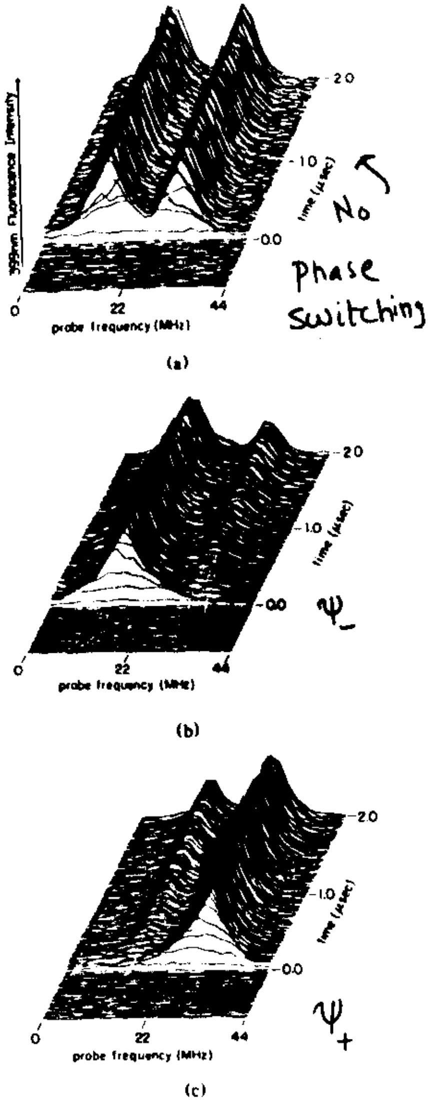


FIG. 2. Transient Autler-Townes spectra. In (a), a constant-phase driving field is employed. In (b) and (c), respectively, the driving field is phase shifted by + 90° and − 90° shortly after switch-on.

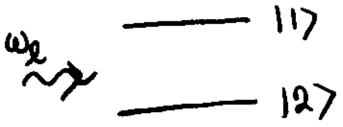
Mossberg and Coworkers

Laser - Maser Action with Dressed States:

(GSA : to be published)

Atom initially prepared with resonant pump

$$\Psi_\pm = \frac{|1\rangle \pm |2\rangle}{\sqrt{2}}$$



$H_{ext} = G\,(s^+ + s^-)$, $H_{ext}\Psi_+ = G\,\Psi_+$

Initial coherence ($\rho_{12} \neq 0$) changes radiative properties

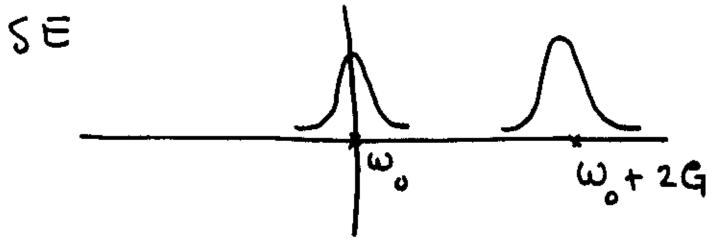
Short Times : spontaneous radiation at ω_0, $\omega_0 \underbrace{+\,2G}_{\text{Side band}}$

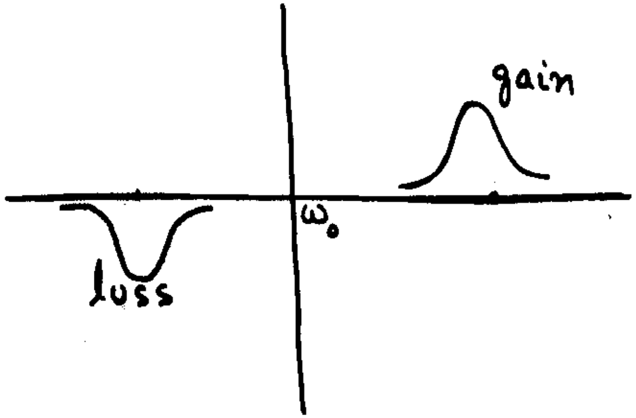
Dipole correlations $\langle s^+(t+\tau)\, s^-(t)\rangle$

SE : $\frac{1}{4}\left[1 + e^{2iG\tau}\right] e^{i\omega_0\tau}$

gain/loss : $\frac{1}{4}\left[e^{-2iG\tau} - e^{2iG\tau}\right] e^{i\omega_0\tau}$

$\langle [s^{(-)}(t), s^+(t+\tau)] \rangle$





Laser Action Choose $\omega_c \sim \omega_0 + 2G$

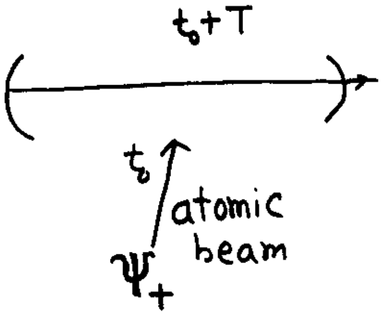
Note $\rho_{11} - \rho_{22} = 0$ Zero Inversion

but $\rho_{++} = 1$

Detailed Theory :

T : Transit Time

Λ : Rate of Injection Of Atoms



$(g s^+ a + g^* s^- a^+)$ (life time $> T$)

FOR RESONANT CASE

$$\dot{a} = -\kappa a + \frac{\Lambda g^2 T^2}{8} \cdot a \Big\{ \overset{\uparrow}{\underset{\text{linear gain}}{1}} - \underbrace{\frac{g^2 T^2}{12} a^+ a}_{\text{Nonlinear Loss}} \Big\} :$$

$\uparrow$ USUAL LASER-MASER EQ.

+ (spont. Em. term)

gain $\frac{\Lambda g^2 T^2}{8}$

mirror loss κ

$$\frac{\Lambda g^2 T^2}{8} > \kappa$$

THRESHOLD CONDITION

distribution function for field amplitudes

$$\frac{\partial P}{\partial t} = \frac{\partial}{\partial \alpha} \left\{ \left(\kappa - \frac{\Lambda g^2 T^2}{8} + \frac{\Lambda g^4 T^4}{96} |\alpha|^2 \right) (\alpha P) \right\} + c.c.$$

$$+ \underbrace{2 \left(\frac{\Lambda g^2 T^2}{8} \right) \frac{\partial^2 P}{\partial \alpha \partial \alpha^*}}_{\text{SE term}}$$

Line width
PHASE DIFFUSION

OPTICAL BLOCH EQS

Relaxations : due to spontaneous emission A coeff

$$= \frac{4}{3} \frac{|d|^2 \omega^3}{c^3 \hbar} = 2\gamma$$

Collisions : Phase interrupting most imp.

Inhomogeneous broadening ; field fluctuations etc.
Thermal Field Induced Transitions

T_2 : transverse relax. time : optical coherence

T_1 : longitudinal " " : population inv.

$\frac{1}{T_1} = 2\gamma$, $\frac{1}{T_2} = \gamma + \gamma_c$, $\eta = \langle s^z \rangle_{eq}$.

$\gamma_c = 0$, $\underline{\underline{\frac{1}{T_1} = \frac{2}{T_2}}}$; many new phenomena arise from $\gamma_c \neq 0$

$\dot{\psi} = M \psi + I$, $\psi^T = (\langle s^+ \rangle, \langle s^- \rangle, \langle s^z \rangle)$

$$I^T = \left(0, 0, \frac{\eta}{T_1}\right), \quad M = \begin{bmatrix} i\Delta - \frac{1}{T_2} & 0 & 2ig^* \\ 0 & -i\Delta - \frac{1}{T_2} & -2ig \\ ig & -ig^* & -\frac{1}{T_1} \end{bmatrix}$$

Steady state

$$\rho_{11} = \frac{1}{2} + \langle S^z \rangle$$

$$\langle S^z \rangle = \frac{(1+(\Delta T_2)^2)\eta}{(1+(\Delta T_2)^2 + 4|g|^2 T_1 T_2)}$$

→ 0 for high fields

Saturation

Power broadening etc.

Fluor. Vs Δ :  , width ~ $2|g|\sqrt{T_1T_2}$

$$\langle \bar{s} \rangle = \frac{-2ig T_2 \eta\,(1-i\Delta T_2)}{(1+(\Delta T_2)^2 + 4|g|^2 T_1 T_2)}$$

← defines susceptibility which depends on Intensity

$$\chi_{\alpha\beta}(\omega_l, \mathcal{E}) = \frac{(-2\eta T_2)(d_{12}^*)_\alpha (d_{12})_\beta\,(i+\Delta T_2)}{\hbar\,[1+(\Delta T_2)^2 + 4|g|^2 T_1 T_2]}$$

Can be used to generate χ's for a number of nonlinear processes

→ linear χ limit $g \to 0$

$$\xrightarrow[\text{correction}]{\text{1st nonvanishing}} \frac{(-2\eta T_2)(d_{12}^*)_\alpha (d_{12})_\beta}{\hbar\,\Delta T_2}\left[1 - \frac{4|g|^2 T_1 T_2}{(\Delta T_2)^2}\right]$$

dispersive limit large ΔT_2

Kerr media $\quad n = n_0 + n_2 I$

$$4|g|^2 T_1 T_2 = (I/I_s)$$ defines saturation intensity line centre

---

Spatial effects

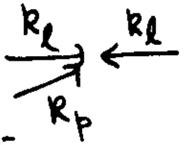
field $\mathcal{E}(\vec{r})\, e^{-i\omega_l t}$

↑ sum of several wave vectors.

$$g \to \frac{d\cdot\mathcal{E}(\bar{r})}{\hbar}$$

$$P = \underline{\chi\,(\omega_l, \mathcal{E}(\vec{r}))}\,\mathcal{E}(\vec{r})$$

⇒ do Fourier decomposition to get χ's for various nonlinear processes.

e.g. degenerate FWM 

$$\mathcal{E}(\bar{r}) = 2\mathcal{E}_l \cos \bar{k}_l\cdot\bar{r} + \mathcal{E}_p\, e^{i\bar{k}_p\cdot\bar{r}}$$

The component of P at $e^{-i\bar{k}_p\cdot\bar{r}}$ will yield the χ for the phase conjugate signal.

Transients : eigen values of M

$$P(z) = \left(z+\frac{1}{T_1}\right)\left[\Delta^2 + \left(z+\frac{1}{T_2}\right)^2\right] + 4|g|^2\left(z+\frac{1}{T_2}\right)$$

$$z = 0, \ \pm i\sqrt{4|g|^2+\Delta^2} \qquad \text{if } T_1, T_2 \to \infty$$

on Res. $\Delta = 0$, $z = -\frac{1}{T_2}$

$$z = -\frac{1}{2}\left(\frac{1}{T_1}+\frac{1}{T_2}\right) \pm \frac{1}{2}\sqrt{\left(\frac{1}{T_1}-\frac{1}{T_2}\right)^2 - 16|g|^2}$$

↑ complex if $16|g|^2 > \left(\frac{1}{T_1}-\frac{1}{T_2}\right)^2$

Responsible for producing new spectral lines in various absorption and emission spectra.

$P(t,\vec{r})$: Various temporal and spatial components ⇒ absorption spectra, gain profiles, nonlinearly generated Coherent signals

Spontaneously emitted radiation

Total fluores $I(t) = (\ )(\text{excited state pop.})$

or time resolved freq resolved spectra

dipole- dipole correlation functions

$$\underline{\langle S^+(t+\tau)\ S^-(t)\rangle}$$

$$\mathcal{E}^+(t) \sim (\ )\,S^-(t) + \text{Free fields}$$

$$\mathcal{E}^-(t) \sim (\ )\,S^+(t) + \text{"}$$

Power spectrum of $\mathcal{E}$ (autocorrelation of $\mathcal{E}$)

$$\sim \langle S^+(t+\tau)\,S^-(t)\rangle$$

$$\Downarrow$$

calculated from Bloch Eqs + Quantum Regression theorem

Mollow
SPECTRUM
Ezekiel
PRL
Nov
77

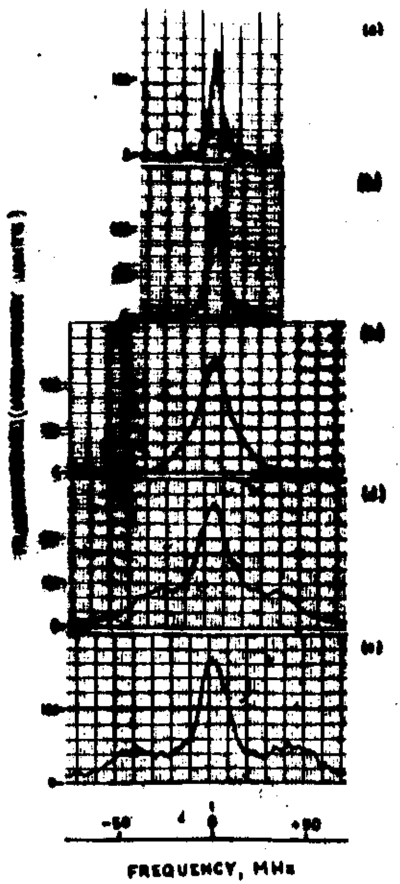
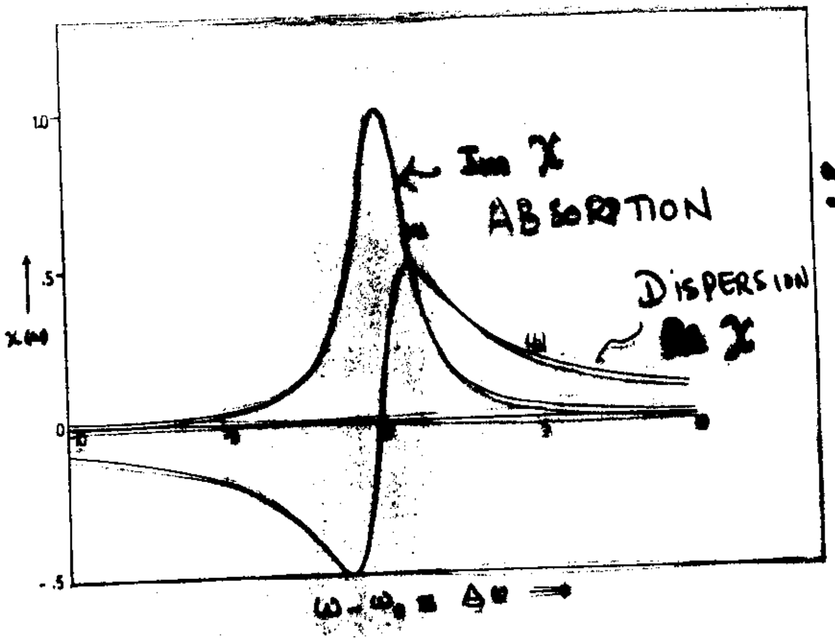


FIG. 2. On-resonance fluorescence spectra at various peak laser intensities. Scan rate, 50 MHz/min; frequency scale, 10 MHz/large division. (a) 0.6 mW/cm², τ = 0.4 sec. (b) 6 mW/cm², τ = 0.4 sec. (c) 66 mW/cm², τ = 3 sec. (d) 400 mW/cm², τ = 3 sec. (e) 900 mW/cm², τ = 3 sec.

Ezekiel et al



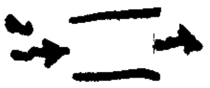


FIG. 3. (a) Imaginary and (b) real parts of the susceptibility [illegible] as a function of the frequency.

$$\chi(\omega) = \sum_{k \neq l} \frac{d_{lk}\, d_{kl}\, (\rho_{kk} - \rho_{ll})}{(\omega + i\Gamma_{kl} - \omega_{kl})} \; ;$$

syst. + Weak Drive

$\omega = \omega_{kl}$, width Γ_{kl}

Increasing Fields ⇒ Power broadened absorption line shapes $\sim \left((\omega-\omega_0)^2 + \Gamma^2 + \beta I\right)^{-1}$

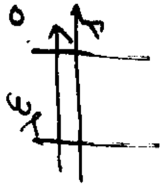
Probe absorption in presence of a pump:

Im $\chi^{(1)}(\omega_s)$: absorption by the medium negligible if ω_s is far detuned from Resonance $\sim \frac{\Gamma}{\Delta^2}$

$$\vec{P}(t) = \vec{P}(\omega_s)\, e^{-i\omega_s t} + c.c.$$

$$P_\alpha(\omega_s) = \chi^{(3)}_{\alpha\beta\gamma\delta}(\omega_s, \omega_\ell, -\omega_\ell)\, E_\beta(\omega_s)\, E_\gamma(\omega_\ell)\, E_\delta(-\omega_\ell)$$

Im $\chi^{(3)}$: absorption by medium in presence of pump if Im $\chi^{(3)} > 0$



Probe is amplified if Im $\chi^{(3)} < 0$

Two Level model simple Result

$$\mathrm{Im}\, \rho_{13}(\omega_s) \approx \frac{\alpha_\ell \Gamma_{13}}{(\Delta+\delta)^2} + \frac{2|\alpha_\ell|^2 \alpha_s}{\Delta(\Delta+\delta)^2} \frac{\delta(2\Gamma_{13}-2\gamma)}{(4\gamma^2+\delta^2)}$$

$\delta = 0 = \omega_\ell - \omega_s$ Resonance   Collision Induced

since $2\Gamma_{13} - 2\gamma \;\alpha\; p$; Dispersive

if $\underline{\Delta > 0}$, then absorbed (amplified) for $\delta > 0\ (<0)$

$\Delta < 0$, probe amplified for $\delta > 0$

Grynberg + Coworkers

Pump – Probe exp

monitor absorption in presence of a pump field

$$\chi^{(1)}(\omega)\ ,\quad \chi^{(3)}(\omega_\ell, -\omega_\ell, \omega)\,|E(\omega_\ell)|^2,$$

$$\chi^{(5)}(\omega_\ell, -\omega_\ell, \omega_\ell, -\omega_\ell, \omega)\,|E(\omega_\ell)|^4 \quad \text{etc.}$$

multiphoton absorption, Renormalize to get all order $\chi^{(1)}(\omega; I_\ell)$

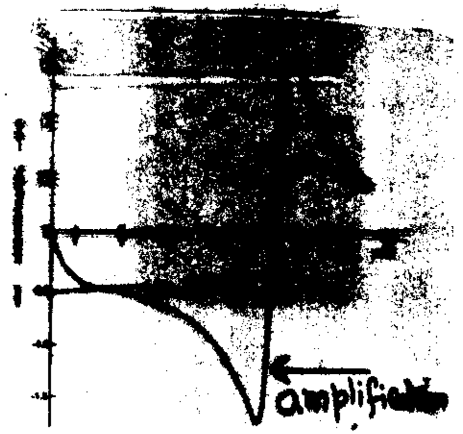


FIG. 3. Absorption spectrum as a function of $\omega-\omega_\ell$ for the radiative relaxation of the atom.

New absorption profile - dispersive

Resonance at Rabi Freq

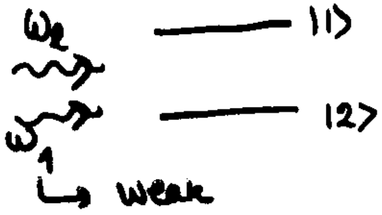
$$\omega - \omega_\ell = \pm\sqrt{\Delta^2 + 4\left(\frac{d\cdot E_\ell}{\hbar}\right)^2}$$

(ω_ℓ pump + Atom)

new system

NOT EXPLAINED BY TRADITIONAL NONLINEAR OPTICS

ω_ℓ : to all orders



$$H_p = -\left(g_p S^+ e^{-i(\omega-\omega_\ell)t} + \text{H.c.}\right)$$

Optical Bloch Eqs + terms from H_p

$$\psi = \psi^{(0)} + \psi^{(+)} e^{-i(\omega-\omega_\ell)t} + \psi^{-} e^{i(\omega-\omega_\ell)t}$$

to 1st order in g_p

Rotating frame

Induced dipole moment $d_{12}^* \langle S^-(t) \rangle$

$$= d_{12}^* e^{-i\omega_\ell t} \psi_2(t)$$

$$= d_{12}^* \psi_2^{(0)} e^{-i\omega_\ell t} + \underset{\substack{\uparrow \\ \text{gain or loss term}}}{d_{12}^* \psi_2^+ e^{-i\omega t}}$$

$$+ \underset{\substack{\uparrow \\ \text{FWM Term}}}{d_{12}^* \psi_2^- e^{-i(2\omega_\ell-\omega)t}}$$

<u>Energy abs.</u> from probe $\hbar\omega S_A(\omega)$

$$S_A(\omega) = 2 \,\text{Im}\, g_p^* \psi_2^+ = 4|g_p|^2 \,\text{Re}\, f[-i(\omega-\omega_\ell)]$$

$$f(z) = (-\psi_3^{(0)}) \Big\{ 2|g|^2 (i\Delta - \tfrac{1}{T_2})^{-1} (z - i\Delta + \tfrac{1}{T_2})^{-1}$$

$$+ 2|g|^2 + (z + \tfrac{1}{T_1})(z - i\Delta + \tfrac{1}{T_2}) \Big\} P^{-1}(z)$$

$\Delta = \omega_0 - \omega_\ell$, $2g$: pump Rabi Freq

$$P(z) = (z + \tfrac{1}{T_1})\left[\Delta^2 + (z + \tfrac{1}{T_2})^2\right] + 4|g|^2 (z + \tfrac{1}{T_2}).$$

All order result

$f(z)$: expand to order g^2 ; $\chi^{(3)}$

$$\left.\begin{array}{l}\text{result} \Rightarrow \text{PIER's} \\ \text{gain regims etc}\end{array}\right\}$$

FWM : Pump $\vec{k}_\ell$, probe $\vec{k}_p$; $2\vec{k}_\ell - \vec{k}_p$
Forward

$$|\text{Polarization}|^2 = \left|4 g^2 g_p^* d_{12}^* \, i \phi_s^{(\omega)} \left(i\Delta + \frac{1}{T_2}\right) P^{-1}\left(i(\omega-\omega_\ell)\right)\right|^2 ; \; 2\omega_\ell - \omega_p$$

depends on all orders of pump; probe weak

Pol $\sim O(g^2 g_p^*)$: $\chi^{(3)}$ Result

$$\frac{\left(\frac{2}{T_2} + i\Omega\right)}{\left(\frac{1}{T_1} + i\Omega\right)} \Big/ \left(\frac{1}{T_2} + i\Delta + i\Omega\right)\left(\frac{1}{T_2} + i\Delta\right)\left(\frac{1}{T_2} - i\Delta + i\Omega\right)$$

$\Omega = \omega_p - \omega_\ell$

↑

PIER : $\frac{2}{T_2} \neq \frac{1}{T_1}$, Collisions needed.

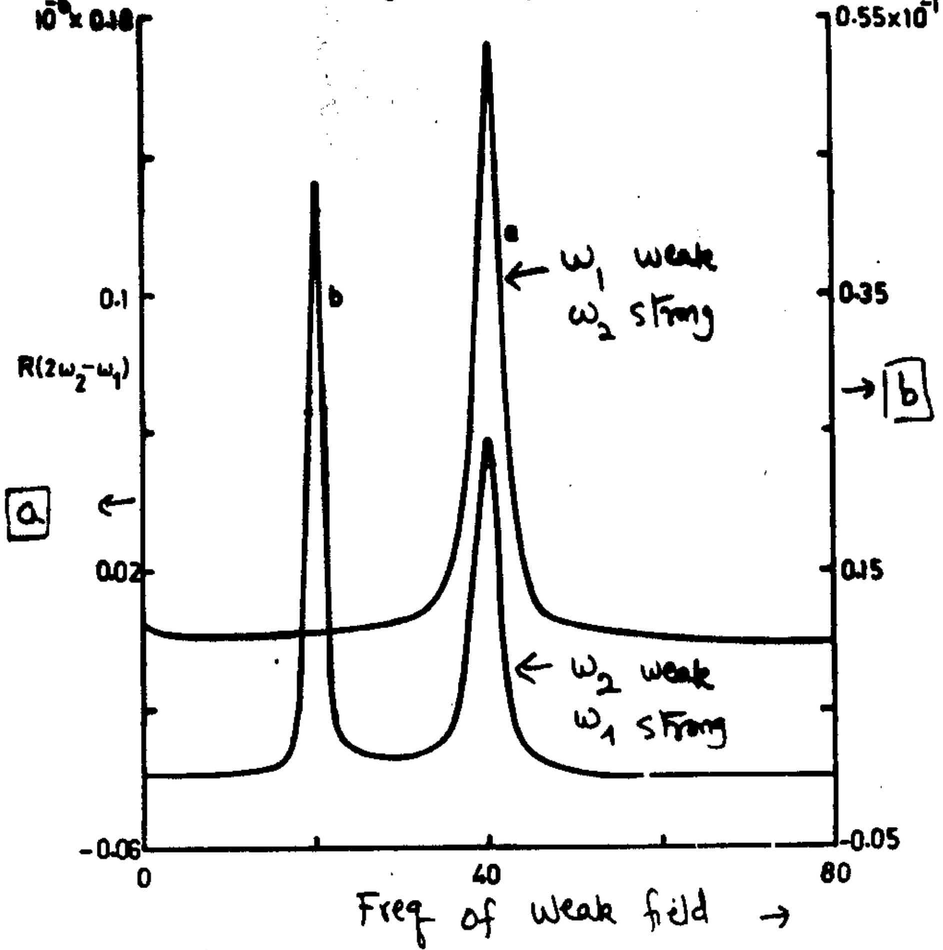
Resonances at Rabi Freq from 'P'

Non degenerate 4 wave mixing $(2\omega_2 - \omega_1)$

Radiative Rel. $\vec{k}_2 + \vec{k}_2 - \vec{k}_1$

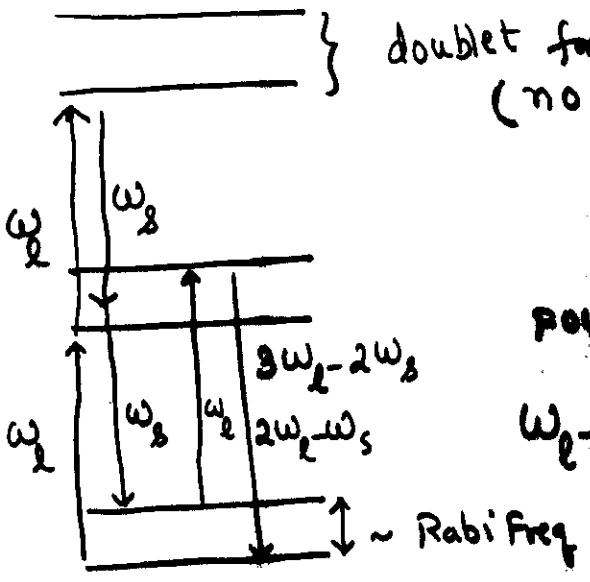
FIG.6

$$P = \underbrace{\chi^{(3)}(\omega_2, \omega_2, -\omega_1)}_{\text{Renormalize it } \chi^{(3)}(\dots ; \mathcal{E}(\omega_2))} \mathcal{E}^2(\omega_2)\, \mathcal{E}^*(\omega_1) \text{ PERTURBATIVE}$$



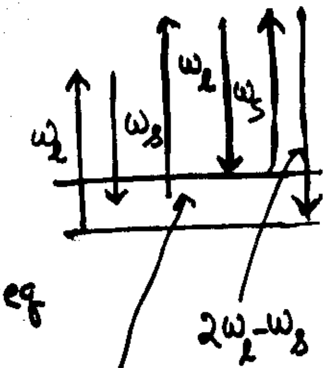
b : Subharmonic bifurcation which is dominant, narrower
strong field on resonance
in each case

DRESSED LEVELS



FOUR WAVE MIXING

$\omega_\ell - \omega_s$ = Rabi Freq



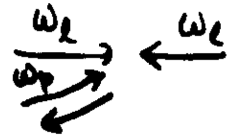
SIX WAVE MIXING

$2\omega_\ell - 2\omega_s$ = Rabi Freq

$\omega_\ell - \omega_s$ = subharmonic

ANALYTICAL RESULTS FOR NONLINEAR RESPONSE ? MOST PHYSICAL PHENOMENA IN TERMS OF APPROPRIATE LINEAR+NONLINEAR

Phase conjugation Geometry



pump : spatial factor $\cos \vec{k}_\ell \cdot \vec{r}$

$g^2 \to \cos^2(\vec{k}_\ell \cdot \vec{r})$; $\psi_3^{(0)}(\xi)$: $P(g^2)$

calculate the Fourier decomposition of Pol

pick up the component independent of $\vec{k}_\ell$

$$\frac{1}{2\pi}\int \frac{\cos^2\theta \, d\theta}{(1+A\cos^2\theta)(1+B\cos^2\theta)} = \frac{((B+1)(A+1))^{-1/2}}{(A+1)^{1/2} + (B+1)^{1/2}}$$

Rabi Resonances

+ Resonances at ± Δ

↑

Extra due to spatial hole burning.

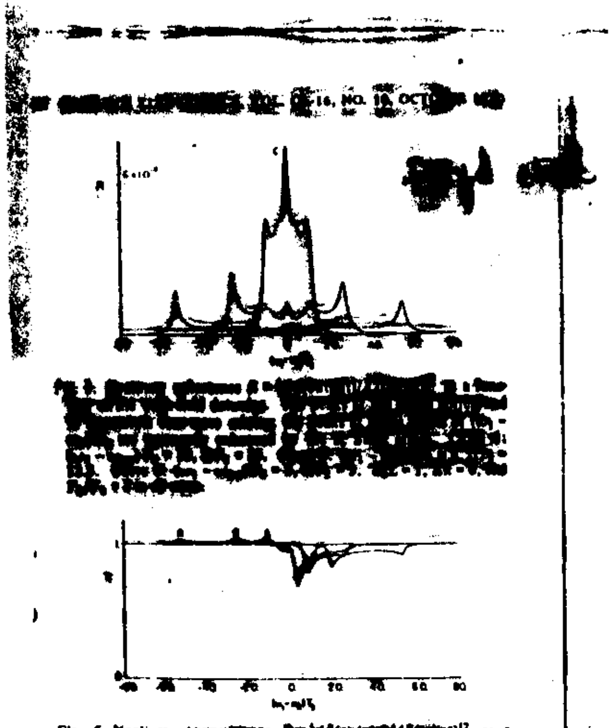


Fig. 6. Nonlinear transmittance $T = |A(\text{output})/A(\text{input})|^2$ as a function of the [illegible] the field ω_4 is amplified; [illegible] $\omega_{ba})T_2 = 19$, [illegible] Curve C: $(\omega_1 -$ [illegible] $= 3$, $\beta T_2 = 5$. $\alpha_0 L = 1$, Δk [illegible] 2 in all cases.

STEEL + LIND

D_2 line 5890 Å

$$3s\ {}^2S_{1/2}\,(F=2) \rightarrow 3p\ {}^2P_{3/2}\,(F=3)$$

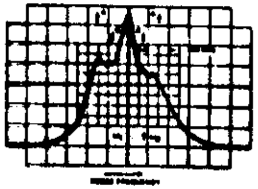


Fig. 6. The pump–probe detuning response as measured at high pump intensity. Five resonances are observed. The central resonance occurs at δ = 0. The two structures designated B occur at δ = ±Δ. The two structures designated A are due to the ac Stark splitting of the atomic levels and occur at δ = ±Ω.

$I \sim 23\ \mathrm{W/cm^2}$

$\Rightarrow \Omega \quad 365\ \mathrm{MHz}$

$$\Delta S^z + \alpha S^x$$

$$\Rightarrow \pm\sqrt{\Delta^2 + 4\alpha^2} = \lambda_i$$

± Δ due to standing waves

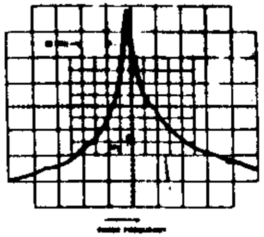
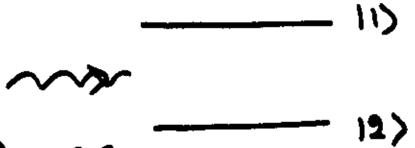


Fig. 7. The bandwidth of the pump-probe detuning signal under high-reflectivity conditions ($R \simeq 150\%$, $\alpha l \simeq 30$).

Modulation exps:



$$\mathcal{E}\, e^{-i\omega_\ell t}\,(1 + \underset{\substack{\downarrow\\ 1}}{M} \cos \nu t) + c.c.$$

↓ 0 for fully modulated field

Total Fluorescence $\propto \rho_{11}$ vs ν or Rabi Freq

d.c. component + $\cos n\nu t$, $\sin n\nu t$:

$\Downarrow$

$$\langle s^{-}\rangle = i\,\frac{\langle s^{z}\rangle_{eq}}{T_1} \sum_n J_n^2(\beta)\,(2g - n\nu)\left[\kappa^2 + (2g - n\nu)^2\right]^{-1}$$

$$\langle s^{z}\rangle = \frac{\langle s^{z}\rangle_{eq}}{T_1} \sum_n J_n^2(\beta)\,\kappa\left[\kappa^2 + (2g - n\nu)^2\right]^{-1}$$

$$\kappa = \frac{1}{2}\left(\frac{1}{T_1} + \frac{1}{T_2}\right) \ ; \quad \beta = \frac{2gM}{\nu}$$

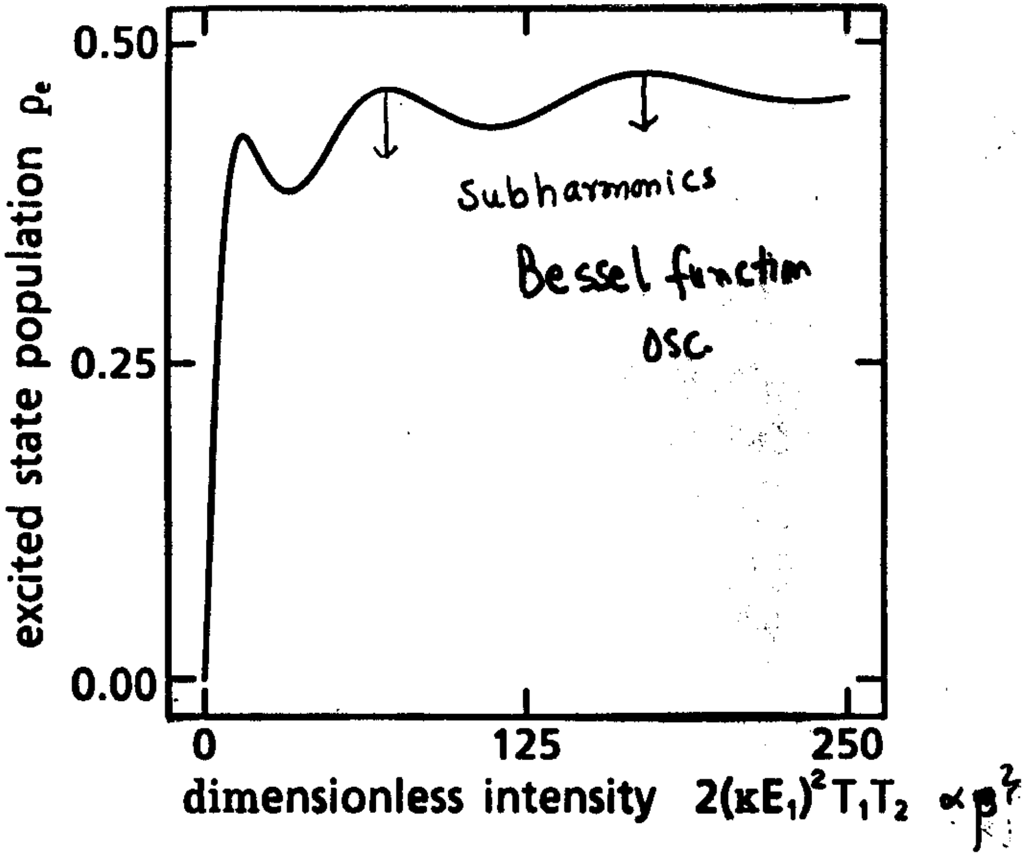
Resonances at $\nu = \dfrac{2g}{n}$ ← Subharmonics

Fully modulated $g = 0$; max - min determined from the oscillations of

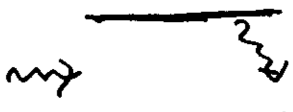
$$J_n^2(\beta) \ : \ \beta = \frac{\text{Rabi Freq}}{\text{Mod. Freq}}$$

Applied field fully modulated

$e^{-i\omega_L t}\,\mathcal{E}\cos(\delta\omega)t\ ;\ \Delta=0$



Amplitude modulation



$\beta=$ (Rabi Freq)/(mod. freq) , $\boxed{\frac{d\cdot\mathcal{E}}{\hbar}\gg\gamma}$

Fig 2

$$\rho_e\sim\frac{1}{2}-\gamma\sum\frac{J_n^2(\beta)\,\kappa}{(\kappa^2+n^2\Omega^2)}$$

$\kappa=3/_2\,\gamma$

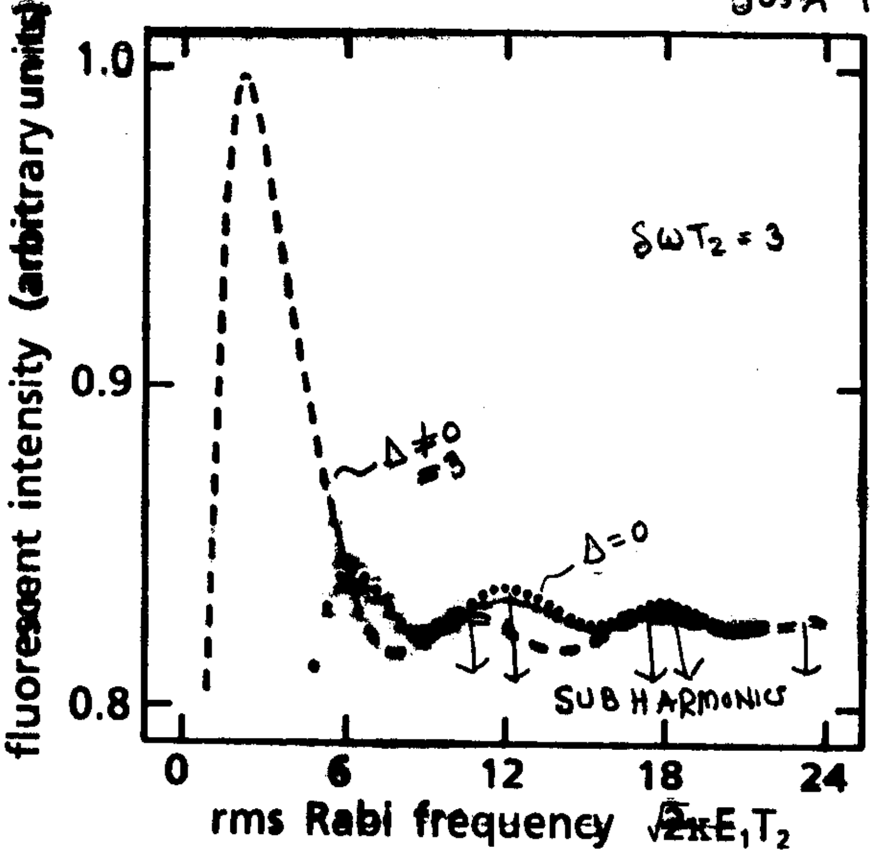
Stroud + Coworkers (1986)

GSA + Nayak : J Phys B 19, 3397

Stroud + Coworkers

JOSA 1988

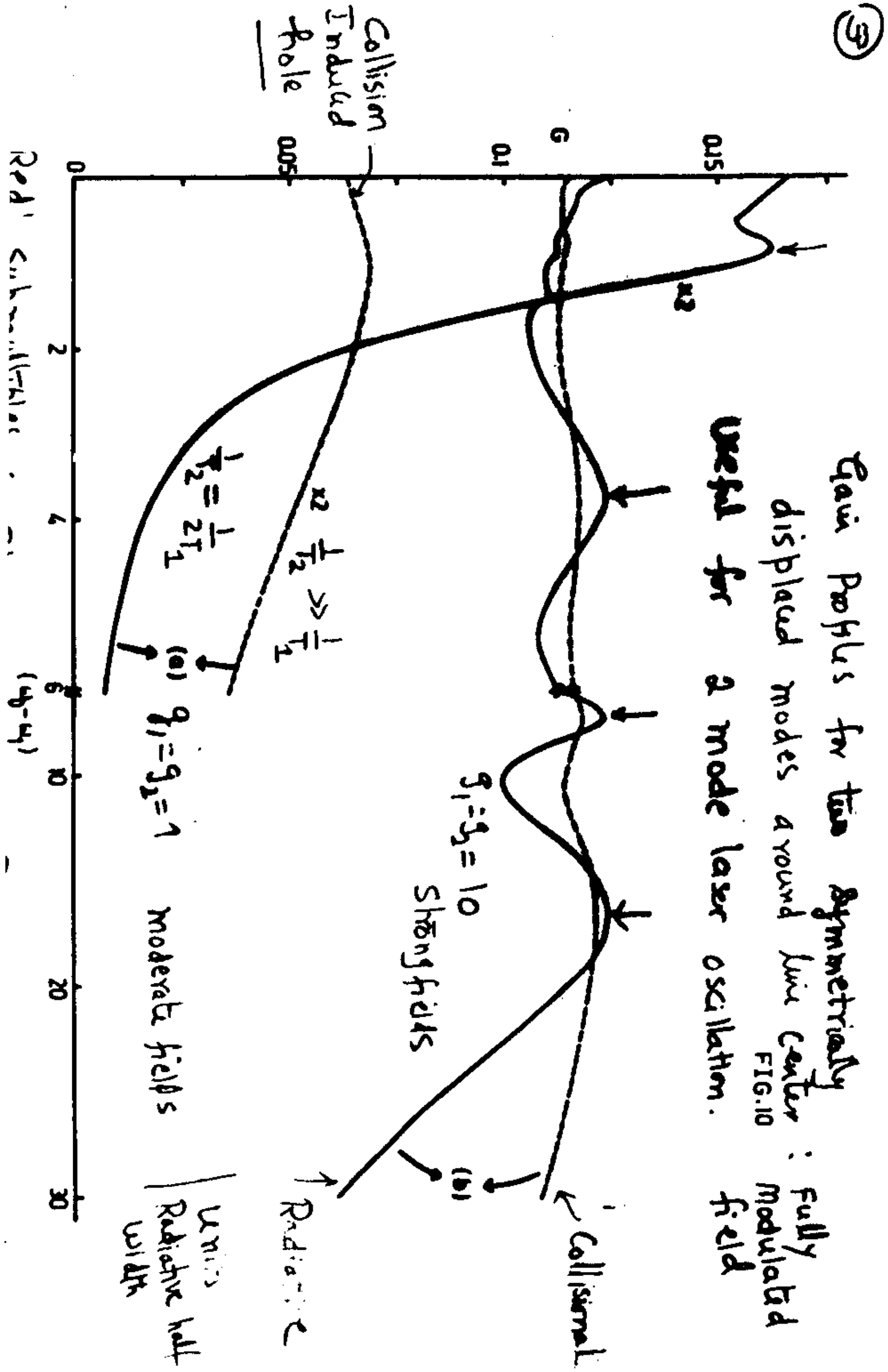
B5, 2015



(Na D_2 line)

Fig 7

data normalized to Single freq. absorption detuned Curve



Gain Profiles for two symmetrically displaced modes around line center : Fully Modulated field

FIG.10

Useful for 2 mode laser oscillation.

Experimental Measurements

0.017

$1/T_1 = 20$ MHz

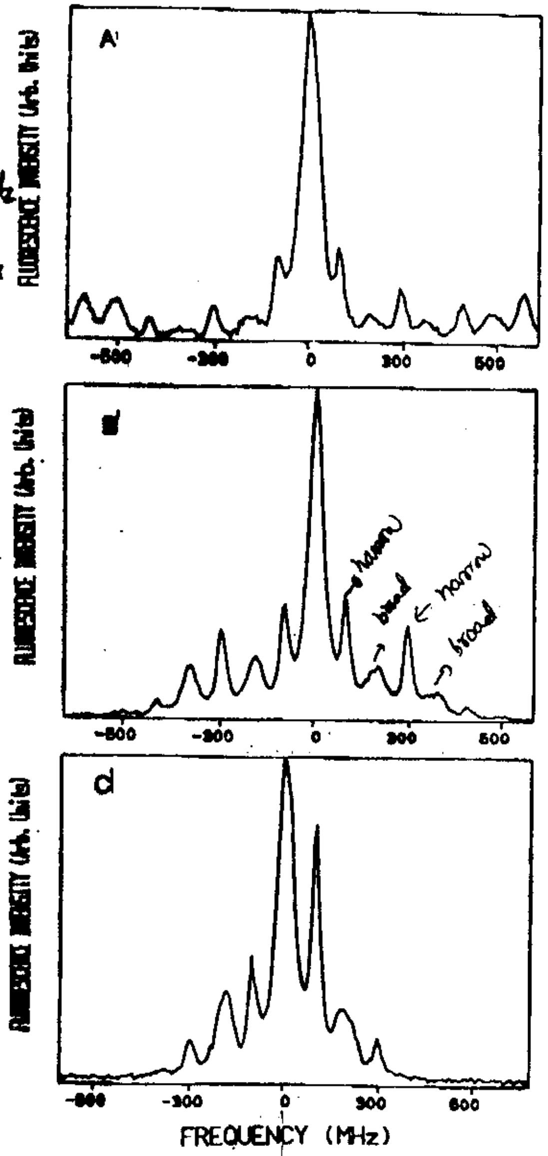
$\Omega_1 = \Omega_2 = 400$ MHz

$\Omega = \nu_1 - \nu_2 = 200$ MHz

$\frac{\nu_1+\nu_2}{2} - \nu_a = 0$

$\Omega_1 = \Omega_2 = 230$ MHz

$\Omega_1 = \Omega_2 = 140$ MHz



Mossberg and coworkers

(Rochester Conf. June 89)

peaks at $\frac{m\Omega}{2}$

m = integer

~~definition of~~

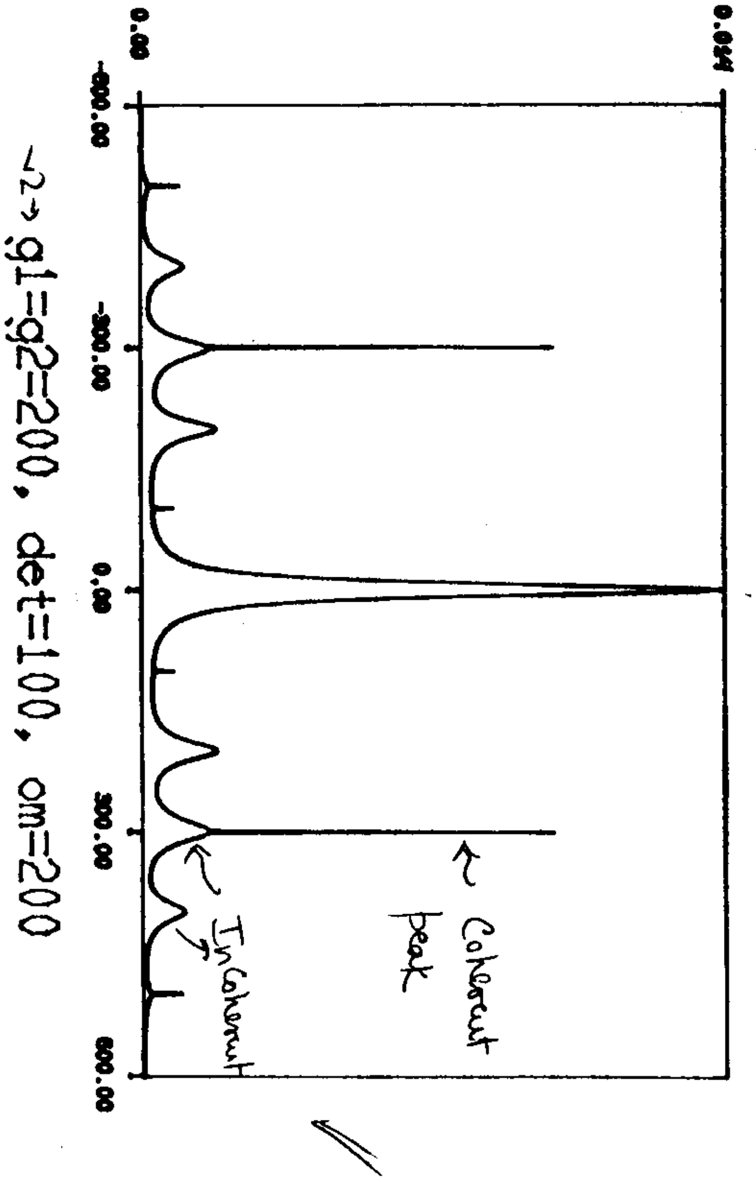
" ~~Grand Rabi Freq~~ "

~~check that~~

~~true correlated~~

zero defined by $\left(\frac{\omega_1+\omega_2}{2}\right) = \omega_0$

(45)



ESA, Y. Zhu, D. Gauthier and T.W. Mossberg

$\nu_1 - \nu_2 = 200$ MHz

$\Omega_1 = \Omega_2 = 200$ MHz. $\gamma_1 = 20$ MHz

method based on specially developed 'CF'

(46)

New Dressed states in modulated field

$$H = -g\left(e^{i\frac{\Omega}{2}t} + e^{-i\frac{\Omega}{2}t}\right)(s^+ + s^-)$$

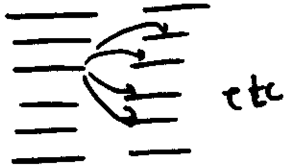
$$\rightarrow \frac{\Omega}{2} b^+ b - 2g_0 (b + b^+) s^x , \quad s^x|\Psi_\pm\rangle = \pm\frac{1}{2}|\Psi_\pm\rangle$$

$$\Phi_\pm^{(n)} = D\left(\pm\frac{2g_0}{\Omega}\right)|n\rangle|\Psi_\pm\rangle \qquad |\Psi_\pm\rangle = \frac{|1\rangle \pm |2\rangle}{\sqrt{2}}$$

$\downarrow$ displacement operator $\exp(\alpha b^+ - \alpha^* b)$

$$E_\pm^{(n)} = \frac{\Omega}{2} n - \frac{2g_0^2}{\Omega} \quad \text{degenerate (doubly)}$$

Transitions



$$\langle \Phi_\pm^{(n)} | s^+ | \Phi_\pm^{(m)} \rangle = \pm \frac{\delta_{mn}}{2} \leftarrow$$ central Peak in the Spectrum

$$\langle \Phi_\mp^{(m)} | s^+ | \Phi_\pm^{(n)} \rangle = \pm\frac{1}{2}\langle m| D\left(\pm\frac{4g_0}{\Omega}\right)|n\rangle \Rightarrow$$

peaks at $\frac{(m-n)\Omega}{2}$

all side peaks

connection with the semi-classical analysis of modulation

$$\langle m | D(\alpha) | n \rangle = J_{m-n}(2\alpha\sqrt{n}), \quad \begin{matrix}\alpha \to 0 \\ n \to \infty \\ \alpha\sqrt{n} \to \text{constant}\end{matrix}$$

SUBHARMONIC (RABI) RESONANCES

$$\omega - \omega_\ell = \pm \frac{1}{n} \underbrace{\sqrt{\Delta^2 + 4\left(\frac{d\cdot\mathcal{E}_\ell}{\hbar}\right)^2}}_{\text{Intensity dependent}}$$

These arise as probe starts becoming strong – probe still perturbative

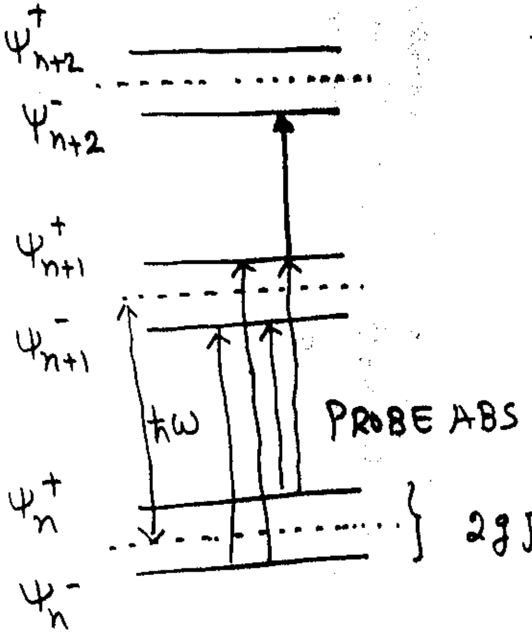
SUBHARMONIC RESONANCES

$\underbrace{\text{Atom + Pump int}}_{\text{diagonalize}}$

ON RESONANCE DRESSED STATES

$\frac{1}{\sqrt{2}}\left(|e,n\rangle \pm |g,n+1\rangle\right) \equiv \psi_n^\pm$: New basis

new selection rules



} $2g\sqrt{n+1} \approx$ RABI FREQ.

TWO PHOTON ABSORPTION FROM PROBE

$2\omega_p = 2\omega \pm 2g\sqrt{n+1}$

$\omega_p = \omega \pm g\sqrt{n+1}$

SINGLE PROBE PHOTON ABS

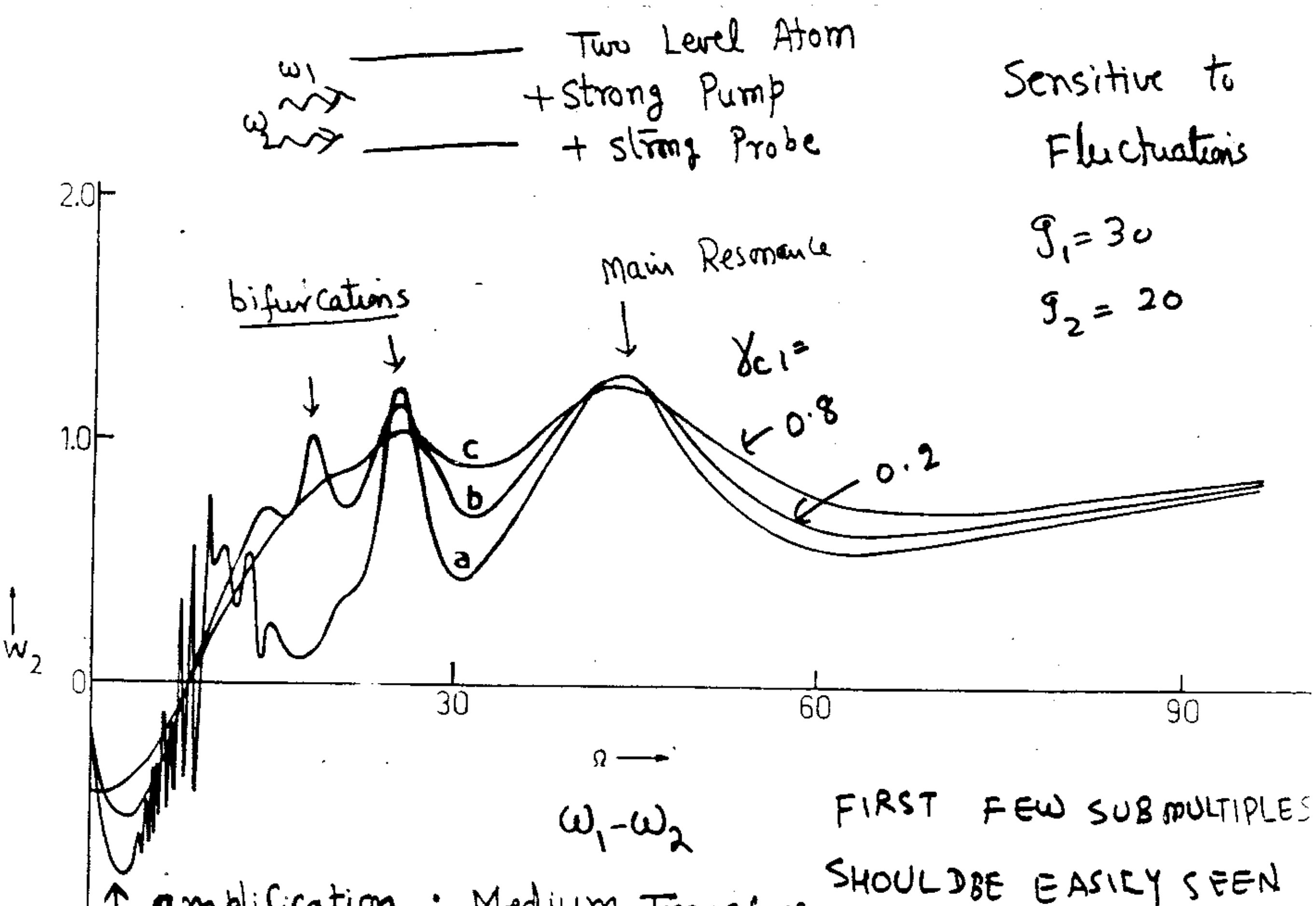
$\omega_p = \omega \pm 2g\sqrt{n+1}$

MULTIPHOTON ABS.

$$M\omega_p = M\omega \pm 2g\sqrt{n+1}$$

IF PROBE IS ALSO STRONG ENOUGH TO SATURATE OPTICAL TRANSITION, THEN DIFFERENT DRESSED STATES ARE TO BE USED - INFINITE ORDER MULTIPHOTON PROCE

Effects of
collisions and saturation in
Nonlinear spectroscopy.



Sensitive to Fluctuations

$g_1 = 30$
$g_2 = 20$

$\omega_1 - \omega_2$

FIRST FEW SUBMULTIPLES SHOULD BE EASILY SEEN

↑ amplification : Medium Transfers energy from one beam to other "OPTICAL BEAM COUPLER"

Agarwal and Nayak
JOSA B (1984)

COLLISION INDUCED COHERENCES IN HIGHER ORDER NONLINEARITIES

(GSA + Nayak 86)

$\chi^{(5)}$, $\chi^{(7)}$ etc can be calculated fm

$$\frac{(-1)^n}{n!} \text{Sym Tr} \Big[Q \Big(\sum_{i=1}^{n} \omega_i - iL_0\Big)^{-1} \tilde{d}^{\alpha_1} \Big(\sum_{i=2}^{n} \omega_i - iL_0\Big)^{-1}$$

$$\tilde{d}^{\alpha_2} \ldots (\omega_n - iL_0)^{-1} \tilde{d}^{\alpha_n} \rho^{(0)} \Big]$$

Too many Terms e.g. $\chi^{(5)}$ will involve $2^5 \times 5!$ $= 3840$. From the structure $(\sum \omega_i - iL_0)^{-1}$ it is clear That $\chi^{(5)}$ etc should also exhibit collision induced coherences. Trebino proved a general theorem on the existence of such Coherences in all orders of perturbation theory.

Consider six wave mixing in 2 level transition $3\omega_l - 2\omega_s$, $3\vec{k}_l - 2\vec{k}_s$

Bloch EQS : Induced pol at $3\omega_l - 2\omega_s$ (RWA)

$$\propto \left(\tfrac{1}{T_1} - i\delta\right)^{-1} \left(\tfrac{1}{T_1} - 2i\delta\right)^{-1} \left(\tfrac{1}{T_2} + i\Delta_l\right)^{-1} \left(\tfrac{1}{T_2} + i\Delta_l - i\delta\right)^{-1}$$

$$\left(\tfrac{1}{T_2} - i\Delta_l - i\delta\right)^{-1} \left(\tfrac{1}{T_2} + i\Delta_l - 2i\delta\right)^{-1} \left(\tfrac{1}{T_2} - i\Delta_l - 2i\delta\right)^{-1}$$

$$\left(\tfrac{2}{T_2} - i\delta\right)\left(\tfrac{2}{T_2} - 3i\delta\right), \quad \delta = \omega_l - \omega_s, \quad \Delta_l = \omega_0 - \omega_l$$

Resonant structures $\delta = 0, \pm \Delta_l, \pm \frac{\Delta_l}{2}$

Large detuning $\Delta_l \gg \frac{1}{T_1}, \frac{1}{T_2}$ etc.

$$|p(3\omega_l - 2\omega_s)|^2 \quad \alpha \quad \frac{(9x^2 + (1+\Gamma)^2)\,(x^2 + (1+\Gamma)^2)}{(x^2+1)\,(4x^2+1)}$$

$\frac{1}{T_2} = \Gamma + \gamma$

$\frac{1}{T_1} = 2\gamma$

$x = \delta T_1$, $\Gamma \propto p$

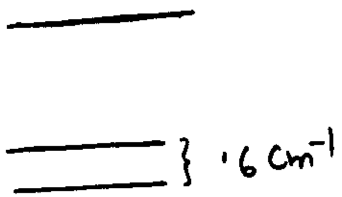
No Collisions signal $\sim \dfrac{9x^2+1}{4x^2+1}$

Inverted Resonance at $x = 0$

Collisions change the character of $\delta = 0$ and make it very prominent

Rahn + Trebino

seeded Na flame

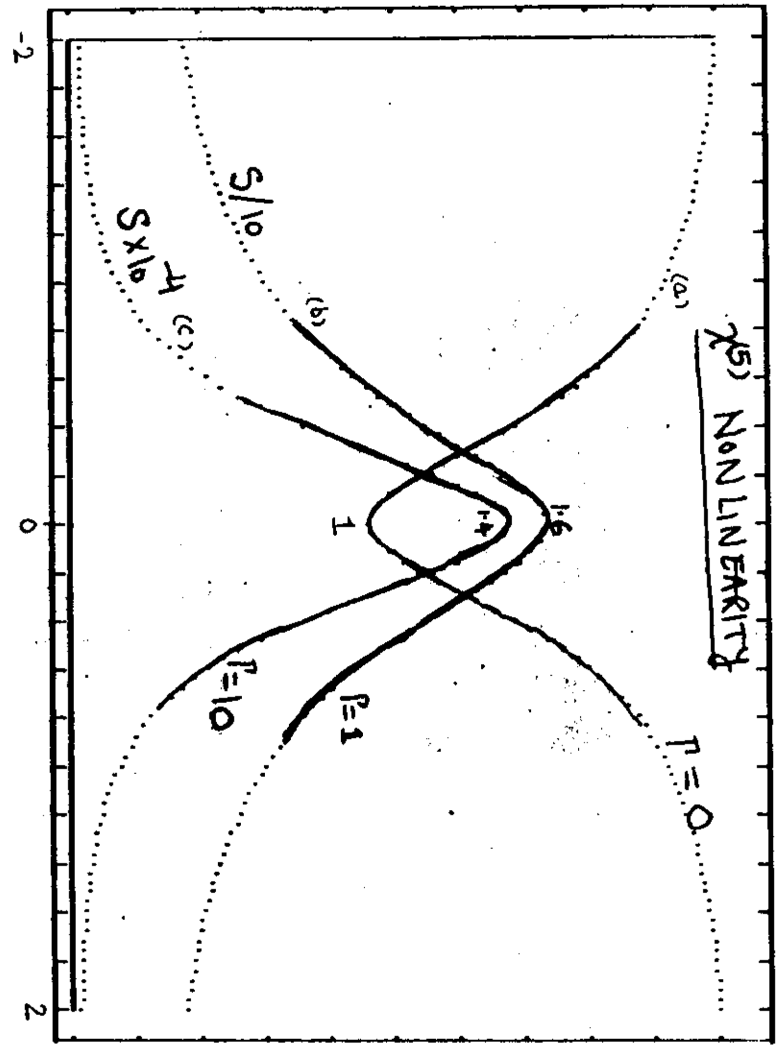


4 wave mixing

CARS

6 wave

8 wave etc.



(b) scaled by a factor of 10.
(c) scaled by a factor of 10^4.

Subharmonic Rabi Resonances in FWM or SWM e.g.

A: signal $2\omega_\ell - \omega_p$

Pol: calculate to all orders in ω_p but 2nd order in ω_ℓ

B: signal $3\omega_\ell - 2\omega_p$

Pol: all orders ω_ℓ :
2nd order ω_p

$$P^{-1}(i\Omega)\underbrace{P^{-1}(2i\Omega)}_{\text{Responsible for 1st Subharmonic}}$$

Show the previous fig &
the interpretation
transparency

II. INTERACTION WITH SEMI-CLASSICAL FIELDS-OPTICAL RESONANCE PHENOMENA

The interaction Hamiltonian between a system with states $|\psi_j\rangle$ with energies E_j and the electromagnetic field $\vec{E}(\vec{R},t)$ can be written in dipole approximation as

$$H_1 \simeq -\vec{d}.\vec{E}(\vec{R},t) \quad , \qquad (2.1)$$

where $\vec{R}$ denotes the position of the atom and $\vec{d}$ is the dipole matrix element which can be expanded as

$$\vec{d} = \sum_{ij} \vec{d}_{ij} \, |\psi_i\rangle\langle\psi_j| \quad . \qquad (2.2)$$

The unperturbed Hamiltonian can be clearly written as

$$H_o = \sum_{j} E_j \, |\psi_j\rangle\langle\psi_j| \quad . \qquad (2.3)$$

Two Level Approximation: Atomic Dynamics in a Monochromatic Field

In resonant physics very often two-level approximation[III] for the atom is adequate. This is so if the frequency ω_1 of the external field is tuned close to the transition frequency ω_o between two levels designated as $|1\rangle$ and $|2\rangle$. We will refer to $|1\rangle$ [$|2\rangle$] as excited (ground) level. The two level approximation works well as long as the width of the external field is small compared to the energy separation between $|1'\rangle$ and $|1\rangle$ where $|1'\rangle$ could be a neighbouring state to which the atom can also get

excited. If the electromagnetic field is a plane wave of frequency ω_1

$$\vec{E}(\vec{R},t) = \vec{\epsilon} \, e^{i\vec{k}.\vec{R}-i\omega_1 t} + \text{c. c.} \quad , \qquad (2.4)$$

and if the two level aproximation is made, then the interaction (2.1) can be expressed as

$$H_1 = -\hbar(g|1\rangle\langle 2| \, e^{-i\omega_1 t} + \text{H.C.})$$

$$- \hbar(g'|1\rangle\langle 2| \, e^{i\omega_1 t} + \text{H.C.}) \quad , \qquad (2.5)$$

where

$$g = \frac{\vec{d}_{12}.\vec{\epsilon} \, e^{i\vec{k}.\vec{R}}}{\hbar} \quad , \quad g' = \frac{\vec{d}_{12}.\vec{\epsilon}* \, e^{-i\vec{k}.\vec{R}}}{\hbar} \qquad (2.6)$$

On choosing the zero of the energy half way between two levels, the Schrodinger equation for the two-level system will be

$$\frac{\partial\psi}{\partial t} = -i \frac{\omega_o}{2} (|1\rangle\langle 1|-2\rangle\langle 2|)\psi - \frac{i}{\hbar} H_1 \psi \quad . \qquad (2.7)$$

On making transformation to a frame rotating with frequency ω_1 of the external field (2.7) becomes

$$\frac{\partial\Phi}{\partial t} = -i \frac{H_{eff}}{\hbar} \phi \quad , \quad \Phi = \exp\left\{ +i\omega_1 S^z t \right\} \psi \qquad (2.8)$$

where

$$H_{eff} = \hbar(\omega_0 - \omega_1)S^z - \hbar(gS^+ + H.C.)$$

$$- \hbar(g'S^+ e^{2i\omega_1 t} + H.C. \quad) \quad . \qquad (2.9)$$

Here we have introduced the operators defind by

$$S^+ = |1\rangle\langle 2| \quad , \quad S^- = |2\rangle\langle 1| \quad , \quad S^z = \frac{1}{2}(|1\rangle\langle 1| - |2\rangle\langle 2|) \qquad (2.10)$$

It can be shown that these operators satisfy spin $\frac{1}{2}$ angular momentum algebra

$$\left[S^+, S^-\right] = 2S^z \quad , \quad \left[S^z, S^+\right] = S^+ \quad , \quad \left[S^z, S^-\right] = -S^- \quad ,$$

$$S^+ S^- = \frac{1}{2} + S^z \quad , \quad S^{+^2} = 0 \quad , \quad S^z S^z = \frac{1}{4} \quad . \qquad (2.11)$$

Note that the Hamiltonian (2.9) contains terms oscillating at twice the optical frequencies. Such terms lead to negligible contribution as long as $|g| << \omega_0$. This is indeed the case for typical optical fields used in resonant experiments. Hence in what follows we ignore these counter rotating terms. Thus the interaction (2.9) is approximated by

$$H_{eff} \sim \hbar \Delta S^z - \hbar (gS^+ + g^*S^-) \quad , \quad \Delta = \omega_0 - \omega_1 \qquad (2.12)$$

This approximation is known as the rotating wave approximation.

We also notice that the effective Hamiltonian (2.12) can be written as

$$H_{eff} = \hbar(\vec{S}.\vec{\Omega}) \quad , \Omega_x = -(g+g^*) \quad , \Omega_y = -ig+ig^* \quad ,$$

$$\Omega_z = \Delta \quad , |\Omega| = \sqrt{\Delta^2 + 4|g|^2} \quad . \qquad (2.13)$$

One thus finds that the problem of spin in a magnetic field and the problem of a two level atom interacting with an electromagnetic field are isomorphic. This was first shown by Feynman, Vernon and Hellwarth.[15] Note that the detuning factor Δ is like the static magnetic field which is used to define the quantization axis of the spin.

We next discuss the dynamical behavior. The time evolution operator U(t) is easily computed

$$U(t) = \exp\left\{-\frac{i}{\hbar} H_{eff}\, t\right\} = \exp(-i\vec{S}.\vec{\Omega}\, t)$$

$$= \cos\frac{\Omega}{2}t - \frac{i(\vec{S}.\vec{\Omega})}{(\Omega/2)} \sin\frac{\Omega t}{2} \quad , \qquad (2.14)$$

$$= \cos\frac{\Omega t}{2} - i\frac{H_{eff}}{\hbar} \sin\frac{\Omega t}{2} \Big/ \frac{\Omega}{2} \quad . \qquad (2.15)$$

The wave function at time t can be obtained from (2.15) and (2.8) assuming that $|\psi(o)\rangle = |2\rangle$:

$$|\psi(t)\rangle = \left(\cos\frac{\Omega t}{2} + \frac{i\Delta}{\Omega}\sin\frac{\Omega t}{2}\right) e^{i\omega t/2}\, |2\rangle$$

$$+ \frac{2ig}{\Omega} \sin\frac{\Omega t}{2}\, e^{-i\omega t/2}\, |1\rangle \quad . \qquad (2.16)$$

The probability $p_1(t)$ of finding the atom in the excited state is

$$p_1(t) = \frac{4|g|^2}{\Omega^2} \sin^2\left(\frac{\Omega t}{2}\right) = \frac{2|g|^2}{\Omega^2}(1 - \cos \Omega t) \qquad (2.17)$$

$$\sim |g|^2 t^2 \qquad \text{if} \quad \Omega t << 1 \quad . \qquad (2.18)$$

Note that for short times p_1 is proportional to the square of time rather than proportional to t. This is because we are dealing with discrete levels and the external field is assumed to have no width. For arbitrary values of the detuning Δ and the field strength g the excitation probability exhibits oscillatory behavior. The oscillation frequency is Ω which is called the (generalized) Rabi frequency.

We next examine the behavior of the Bloch vector $\langle\hat{S}(t)\rangle$. Using (2.16) one can easily show that

$$\langle\hat{S}(t)\rangle \cdot \langle\hat{S}(t)\rangle = \frac{1}{4} \quad ,$$

$$\langle S^+(t)\rangle = \frac{1}{2} \sin \theta(t)\, e^{i\omega_1 t + i\varphi(t)} \quad ,$$

$$\langle S^z(t)\rangle = -\frac{1}{2} \cos \theta(t) \quad , \qquad (2.19)$$

where $\theta(t)$ and $\varphi(t)$ are found to be

$$\cos \theta(t) = 1 - \frac{8|g|^2}{\Omega^2} \sin^2 \frac{\Omega t}{2} \quad , \qquad (2.20)$$

$$\varphi(t) = \chi + \tan^{-1}\left\{-\frac{\Omega}{\Delta} \cot \frac{\Omega t}{2}\right\} \quad , \qquad (2.21)$$

where χ is the phase of the coupling constant

$$g = |g|\, e^{-i\chi} \quad . \qquad (2.22)$$

Thus the Bloch vector $\langle\hat{S}\rangle$ can be represented as a point (θ,φ) on a sphere called Bloch sphere. The north (south) pole gives the ground (excited) state. It should be borne in mind that (2.20) and (2.21) are derived under the initial condition $|\psi(o)\rangle = |2\rangle$. The representation (2.19) holds for arbitrary initial conditions. For a field on resonance $\omega_1 = \omega_0$, $\Delta = 0$, $\Omega^2 = 4|g|^2$,

$$\theta(t) = \Omega t \quad , \qquad (2.23)$$

where Ωt is the area of the pulse. For a $\frac{\pi}{2}$ - pulse $\theta = \frac{\pi}{2}$, we get

$$\langle S^z\rangle = 0 \;\; ; \;\; \langle S^+\rangle = -\frac{1}{2}\, i e^{i\omega_1 t + i\chi} \quad , \qquad (2.24)$$

$$|\psi(t)\rangle = \frac{1}{\sqrt{2}}\, e^{i\omega t/2}\, |2\rangle + \frac{ig}{|g|\sqrt{2}}\, e^{-i\omega t/2}\, |1\rangle \quad . \qquad (2.25)$$

Thus a $\pi/2$ pulse creates equal population between the two states $|1\rangle$ and $|2\rangle$ and leads to maximum coherence. Note further that a state which has finite dipole moment will give rise to coherent radiation. This immediately leads us to the study of the properties of atomic coherent states.

Atomic Coherent States:

Clearly for a two level system we define the coherent states $|\theta,\varphi\rangle$ by

$$|\theta,\varphi\rangle = \cos\frac{\theta}{2}|2\rangle + \sin\frac{\theta}{2}e^{-i\phi}|1\rangle \quad , \quad 0 \le \theta \le \pi ,$$

$$0 \le \theta \le \pi \quad , \quad \langle S^{+}\rangle = \frac{1}{2}\sin\theta\, e^{i\varphi} \quad . \tag{2.26}$$

which are appropriate superpositions of ground and excited states.

These concepts can be generalized to a system of many two-level atoms. A collection of N two-level atoms can be characterized by the angular momentum algebra corresponding to spin value N/2 provided we choose to work with the completely symmetric representation. Thus the atomic coherent states[16,17] for a system of N two-level atoms are defined by

$$|\theta,\varphi\rangle = \exp\left\{-i\theta(S^{x}\sin\varphi - S^{y}\cos\varphi)\right\}|S,-S\rangle \quad , \quad S = \frac{N}{2}$$

$$= \exp\left\{\frac{\theta}{2}(S^{+}e^{-i\varphi} - S^{-}e^{i\varphi})\right\}|S,-S\rangle \quad . \tag{2.27}$$

$$= e^{\tau S^{+}} e^{\ln(1+|\tau|^{2})S^{z}} e^{-\tau^{*}S^{-}}|S,-S\rangle \quad ; \quad \tau = e^{-i\varphi}\tan\frac{\theta}{2} \quad , \tag{2.28}$$

where Baker-Hausdorff Campbell identity has been used. The right hand side of (2.27) on simplification leads to

$$|\theta,\varphi\rangle = \sum_{-S}^{+S}\begin{pmatrix}2S\\ S+M\end{pmatrix}^{1/2}\sin^{S+M}\frac{\theta}{2}\cos^{S-M}\frac{\theta}{2}\,e^{-i(S+M)\varphi}|S,M\rangle \quad . \tag{2.29}$$

The atomic coherent states form a complete set and are nonorthogonal

$$\frac{2S+1}{4\pi}\int|\theta,\varphi\rangle\langle\theta,\varphi|\sin\theta\, d\theta\, d\varphi = 1 \quad , \tag{2.30}$$

$$\langle\theta,\varphi|\theta',\varphi'\rangle = e^{iS(\varphi-\varphi')}\left[\cos\frac{\theta-\theta'}{2}\cos\frac{\varphi-\varphi'}{2} - i\cos\frac{\theta+\theta'}{2}\sin\frac{\varphi-\varphi'}{2}\right]^{2S} \quad . \tag{2.31}$$

We recall that the field coherent states are the eigenstates of the annihilation operator. The question arises – What is the operator of which $|\theta,\varphi\rangle$ are the eigen states. To answer this we notice that

$$S^{z}|S,-S\rangle = -S|S,-S\rangle \quad , \tag{2.32}$$

and hence

$$\tilde{S}^{z}|\theta,\varphi\rangle = -S|\theta,\varphi\rangle \quad , \tag{2.33}$$

where

$$\tilde{S}^{z} = \exp\left\{-i\theta(\vec{S}.\vec{n})\right\}S^{z}\exp\left\{i\theta(\vec{S}.\vec{n})\right\} \quad , \tag{2.34}$$

$$\vec{n} = (\sin\varphi, -\cos\varphi) \quad .$$

On simplification (2.34) reduces to

$$\tilde{S}^{z} = S^{z}\cos\theta - (\sin\varphi S^{y} + \cos\varphi S^{x})\sin\theta \quad . \tag{2.35}$$

This is the operator of which $|\theta,\varphi\rangle$ is the eigenstate.

Minimum Uncertainty States:

The harmonic oscillator coherent states[18] are known to be minimum uncertainty states. The uncertainty relation for the angular momentum operators can be written in the form

$$\Delta S_x\, \Delta S_y \geq \frac{1}{2}\, |\langle S_z\rangle| \quad , \quad \Delta S_x = \sqrt{\langle S^{x^2}\rangle - \langle S^x\rangle^2} \quad . \qquad (2.36)$$

The question arises – are there states that will satisfy equality sign in (2.36). We recall that the nonhermitean operators $S^{\pm}$ have only one eigen state i.e.,

$$S^+|S,S\rangle = 0 \quad , \quad S^-|S,-S\rangle = 0 \qquad (2.37)$$

and thus we search for the eigenvalues of the linear combination of $S^{\pm}$

$$R^z = \frac{1}{\sqrt{1-\alpha^2}} \left[S^x - i\alpha S^y \right] \quad , \qquad (2.38)$$

where α is a complex number $\neq \pm 1$. It can be proved[19] that the eigen states of (2.38) are given by

$$R^z|\chi_m,\alpha\rangle = m|\chi_m, \alpha\rangle \quad , \qquad (2.39)$$

where, apart from a normalization constant,

$$|\chi_m,\alpha\rangle = \exp(\theta S^z)\, \exp(-\frac{i\pi}{2} S^y)|S,m\rangle \quad , \qquad (2.40)$$

$$e^{\theta} = \sqrt{\frac{1-\alpha}{1+\alpha}} \quad . \qquad (2.41)$$

It can proved that if α is real, then the uncertainty equality in (2.36) is satisfied for states $|\chi_m,\alpha\rangle$. In particular for $\alpha=0$, $\theta=0$, $|\chi_m, 0\rangle$ becomes the eigen state of S^x

$$S^x \exp(-\frac{i\pi}{2} S^y)|S,M\rangle = M\, S^x(-\frac{i\pi}{2} S^y)|S,M\rangle \quad . \qquad (2.42)$$

Equation (2.42) can be given the following interpretation–consider a set of N two-level atoms initially in the state $|S,M\rangle$. Let the atoms interact with a resonant $\pi/2$ pulse such that $g=|g|i$, then the state at time t will be (cf. Eq.(2.16))

$$|\Phi(t)\rangle = \exp(-\frac{i\pi}{2} S^y)\, |S,M\rangle \quad . \qquad (2.43)$$

This state at time t is an eigenstate of S^x. Note further that if the phase of the field is changed by $\pi/2$ at the end of the pulse so that $g=-|g|$, then $\tilde{H}_{eff} = +2|g|S^x$. In otherwords we have the system in a state which is the eigenstate of $\tilde{H}_{eff}$.

Semi-Classical Dressed States:

We have seen that in a rotating frame the effective Hamiltonian describing the interaction of a two-level atom with external electromagnetic fields is given by (2.12). The eigen states $|\psi_\pm\rangle$ of (2.12) are called the dressed states. These states and their energies are given by

$$E_\pm = \pm \frac{\hbar\Omega}{2} \quad , \quad |\psi_\pm\rangle = (|1\rangle + \frac{1}{2g}(\Delta\mp\Omega)\ |2\rangle) \left[1 + \frac{(\Delta\mp\Omega)^2}{4|g|^2}\right]^{-1/2} \quad . \tag{2.44}$$

Clearly if a system is prepared in one of these dressed states, then no further evolution takes place unless decay effects are included. Mossberg and coworkers[20] showed how experimentally the system can be prepared in dressed states. Let us consider for simplicity the case $\Delta=0$:

$$E_\pm = \pm\hbar|g| \quad , |\psi_\pm\rangle = (|1\rangle \mp e^{i\chi}\ |2\rangle) \ / \sqrt{2} \quad . \tag{2.45}$$

Let the $\pi/2$ – pulse interact with an atom in the ground state. let χ' be the <u>phase of the field</u>. Then from (2.8) and (2.16) the state at time t is

$$|\varphi(t)\rangle = \frac{1}{\sqrt{2}}(|1\rangle - ie^{i\chi'}|2\rangle)(ie^{-i\chi'}) \quad . \tag{2.46}$$

On comparison with (2.45) we see that

$$|\psi_+\rangle = -e^{i\chi}|\varphi(t)\rangle \qquad \text{if} \qquad \chi = \chi' + \frac{\pi}{2} \quad ,$$

$$|\psi_-\rangle = e^{i\chi}|\varphi(t)\rangle \qquad \text{if} \qquad \chi = \chi' - \frac{\pi}{2} \quad . \tag{2.47}$$

Thus the dressed states $|\psi_\pm\rangle$ can be generated by changing the phase of the field by $\pm\frac{\pi}{2}$. The absorption[21] and scattering[22] of the electromagnetic radiation by atoms in such dressed states have been studied.

Effects of Relaxation:

So far we have considered only the interaction with external electromagnetic fields. In reality one has to account for various sources of decay of the atomic population and coherences. For example the atom can decay radiatively by emitting a photon. The collisions change the populations and coherences. The effects of such decay processes can be included by modifying the basic equations for mean values

$$\frac{d\psi}{dt} = M\psi + I \quad , \tag{2.48}$$

where the matrices M, ψ and I given by

$$\psi = \begin{pmatrix} \langle S^+\rangle \\ \langle S^-\rangle \\ \langle S^z\rangle \end{pmatrix} \ , \ I = \begin{pmatrix} 0 \\ 0 \\ \eta/T_1 \end{pmatrix} \ , \ M = \begin{pmatrix} i\Delta-\frac{1}{T_2} & 0 & 2ig^* \\ 0 & -i\Delta-\frac{1}{T_2} & -2ig \\ ig & -ig^* & -\frac{1}{T_1} \end{pmatrix} . \tag{2.49}$$

Here η is the equilibrium value of $\langle S^z\rangle$ i.e., the value of $\langle S^z\rangle$ in the absence of the external field. T_1 and T_2 are the longitudinal and transverse relaxation times. For radiative relaxation

$$\frac{1}{T_1} = 2\gamma = \frac{2}{T_2} \quad , \qquad \eta = -\frac{1}{2} \quad , \qquad (2.50)$$

where 2γ is the Einstein A-coefficient. Elastic collisions are included if we take

$$\frac{1}{T_1} = 2\gamma \quad , \quad \frac{1}{T_2} = \gamma + \gamma_c \quad , \quad \eta = -\frac{1}{2} \qquad (2.51)$$

where γ_c is the collisional line width.

We first discuss the characteristics of the steady state solution of (2.48). Calculation shows that

$$\langle S^z \rangle = \frac{(1+(\Delta T_2)^2)\eta}{(1+(\Delta T_2)^2+4|g|^2T_1T_2)} \quad , \qquad (2.52)$$

$$\langle S^- \rangle = \frac{-2igT_2(-i\ \Delta T_2+1)\eta}{(1+(\Delta T_2)^2+4|g|^2T_1T_2)} \quad , \qquad (2.53)$$

For high fields $\langle S^z \rangle \longrightarrow 0$ and hence high fields equalize the population in the ground and excited states. The steady state fluorescence is a direct measure of the excited state population. The induced dipole moment (2.53) enables us to define an intensity dependent susceptibility $\chi(\omega_1, E)$ since the induced polarization is $\vec{d}^{\,*}_{12}\langle S^- \rangle$+c.c.. From (2.53) and from the definition of g we get the result

$$\chi_{\alpha\beta}(\omega_1,\mathcal{E}) = \frac{(-2\eta T_2)(\vec{d}^{\,*}_{12})_\alpha(\vec{d}_{12})_\beta(i+\Delta T_2)}{(1+(\Delta T_2)^2 + 4|g|^2T_1T_2)\hbar} \quad . \qquad (2.54)$$

Note that this χ depends on all powers of $\mathcal{E}$ because of $|g|^2$ terms in the denominator. Usual linear χ is obtained by setting g=0. In the limit of large ΔT_2, (2.54) can be used to define Kerr media for which the susceptibility has a term linearly proportional to the intensity of the field

$$\chi_{\alpha\beta}(\omega_1,\mathcal{E}) \simeq \frac{(-2\eta T_2)(\vec{d}^{\,*}_{12})_\alpha(\vec{d}_{12})_\beta}{\hbar\Delta T_2}\left[1 - \frac{4|g|^2T_1T_2}{(\Delta T_2)^2}\right] \quad . \qquad (2.55)$$

Note further that Im $\chi(\omega_1,\mathcal{E})$ is positive and is Lorentzian with a width that is proportional to field intensity. This is known as power broadening. Very often the parameter $4|g|^2T_1T_2$ is expressed as

$$4|g|^2T_1T_2 = I/I_s = \frac{|\mathcal{E}|^2}{|\mathcal{E}_s|^2} \quad , \qquad (2.56)$$

which defines the saturation intensity.

The rate equation behavior results either if the detuning is large or if T_2 is small so that the polarization follows the population adiabatically. In this limit

$$\langle S^-(t) \rangle = -2ig\langle S^z(t)\rangle T_2/(1+i\Delta T_2) \quad , \qquad (2.57)$$

and hence

$$\frac{d}{dt}\langle S^z(t)\rangle = -\frac{1}{T_1}\langle S^z\rangle\left[1+\frac{4|g|^2T_1T_2}{1+\Delta^2T_2^2}\right]+\frac{\eta}{T_1} \quad . \qquad (2.58)$$

The rate equation for $\langle S^z\rangle$ can be easily solved. Even cases of time dependent field envelopes can be handled. Improvements over rate equation approximation are discussed in Ref. III.

The transient behavior of ψ can be obtained by using Laplace transform techniques. The eigenvalues of the matrix M determine the dynamical behavior. The following polynomial which corresponds to det(z-M) is the key to the dynamical behavior:

$$P(z) = (z+\frac{1}{T_1})\left[\Delta^2+(z+\frac{1}{T_2})^2\right]+4|g|^2(z+\frac{1}{T_2}) \quad . \qquad (2.59)$$

In the limit of $T_1, T_2 \longrightarrow \infty$, the roots are

$$z = 0 \quad , \pm\, i\sqrt{4|g|^2+\Delta^2} \quad . \qquad (2.60)$$

The cubic equation can be solved in the limits

$$\text{(a)} \quad T_1 = T_2 \quad , \quad z = -\frac{1}{T_2} \; , \; -\frac{1}{T_2} \pm i\sqrt{4|g|^2+\Delta^2} \quad , \qquad (2.61)$$

$$\text{(b)} \quad \Delta = 0 \quad , \quad z = -\frac{1}{T_2} \quad ,$$

$$z = -\frac{1}{2}(\frac{1}{T_1}+\frac{1}{T_2}) \pm \frac{1}{2}\sqrt{\left(\frac{1}{T_1}-\frac{1}{T_2}\right)^2-16|g|^2} \quad . \qquad (2.62)$$

In other cases the roots can be obtained numerically.

The macroscopic dipole moment p(t) will have the form for $\Delta=0$

$$\langle S^-(t)\rangle = e^{-\frac{1}{2}(\frac{1}{T_1}+\frac{1}{T_2})t}\left[e^{2i|g|t}A_+ + e^{-2i|g|t}A_-\right]+A_0e^{-t/T_2} \; .$$

$$\text{if } 16|g|^2 \gg \left(\frac{1}{T_1}-\frac{1}{T_2}\right)^2 \quad . \qquad (2.63)$$

Classically such a dipole moment will result in radiation at frequencies[23,24]

$$\omega_1 \pm 2|g| \qquad , \quad \text{width} \quad \frac{1}{2}(\frac{1}{T_1}+\frac{1}{T_2})$$

$$\omega_1 \qquad \text{width} \quad \frac{1}{T_2} \quad . \qquad (2.64)$$

The quantum theory[23-26] confirms this and also produces the heights of various spectral peaks.

Response of a strongly pumped two-level system to a weak field

We next consider an important question- How to probe the dynamical properties of a system dressed by a strong pump field. For this purpose we can imagine a probe field of frequency ω acting on the two-level system. We take the probe field weak and thus consider the linear response of the atomic system while treating the pump field to all orders in the perturbation. In the rotating frame the interaction with the probe field ϵ_p can be written as

$$H_p = -(g_p S^+ e^{-i(\omega-\omega_1)t} + \text{H.C.}) \quad ,$$

$$g_p = \frac{\vec{d}_{12}\cdot\vec{\varepsilon}_p}{\hbar} \quad e^{i\vec{k}_p\cdot\vec{R}} \qquad (2.65)$$

The basic equation (2.48) is modified to

$$\frac{d\psi}{dt} = M\psi + I + (M_+ e^{-i(\omega-\omega_1)t} + M_- \; e^{i(\omega-\omega_1)t}) \;\; \psi \quad ,$$

$$(M_+)_{23} = -2ig_p \;,\; (M_+)_{31} = ig_p \;,\; (M_-)_{32} = -ig^*_p \;,$$

$$(M_-)_{13} = 2ig^*_p \quad . \qquad (2.66)$$

where the only non-vanishing elements of $M_\pm$are given. It should be borne in mind that the matrix M depends on the pump field. The solution of (2.66) in the long time limit is

$$\psi = \psi^{(o)} + \psi^{(+)} e^{-i(\omega-\omega_1)t} + \psi^{(-)} \; e^{i(\omega-\omega_1)t} \quad ,$$

$$\psi^{\pm} = \left[\mp i(\omega-\omega_1) - M\right]^{-1} M_\pm \; \psi^{(o)} \quad , \psi^{(o)} = -M^{-1} I \quad . \qquad (2.67)$$

The induced dipole moment in the original frame will be

$$\vec{d}^*_{12}\langle S^-(t)\rangle = \vec{d}^*_{12} \; e^{-i\omega_1 t} \; \psi_2(t)$$

$$= \vec{d}^*_{12}\psi_2^{(o)} \; e^{-i\omega_1 t} + \vec{d}^*_{12}\psi_2^{(+)} \; e^{-i\omega t} + \vec{d}^*_{12}\psi_2^{(-)} e^{-i(2\omega_1-\omega)t} \qquad (2.68)$$

The induced polarization at the probe frequency is given by $\vec{d}^*_{12} \; \psi^+_2 \; e^{-i\omega t}$. Let us now calculate the rate at which the energy is absorbed from the probe field. This rate can be shown to be given by $\hbar\omega S_A(\omega)$ with

$$S_A(\omega) = 2\text{Im} \; g^*_p \; \psi^+_2 \qquad (2.69)$$

From (2.67) and (2.69) one can see that $\psi^+_2 \propto g_p$. The explicit expression for $S_A(\omega)$ can be obtained from (2.67) and (2.49). We give the result[27] below

$$S_A(\omega) = 4|g_p|^2 \; \text{Real} \left\{ f(z)|_{z=-i(\omega-\omega_1)} \right\} \quad ,$$

$$f(z) = (-\psi_3^{(o)}) \left\{ 2|g|^2 (i\Delta - \frac{1}{T_2})^{-1} (z - i\Delta + \frac{1}{T_2}) + 2|g|^2 \right.$$

$$\left. + \; (\text{[illegible]} + \frac{1}{T_1})(z - i\Delta + \frac{1}{T_2}) \right\} \; P^{-1}(z) \quad . \qquad (2.70)$$

where $\psi_3^{(o)}$ and P(z) are given by (2.52) and (2.59) respectively. The absorption spectrum (2.70) has several interesting features. It shows regions of gain[28] for both $\Delta=0$ and $\Delta\neq0$. Thus an input probe can experience amplification. This can infact be used to achieve laser action without population inversion.[29,30] We show in Figs.3 the typical behavior of the absorption spectrum. The absorption spectrum depends on all powers of the pump field. A perturbative expansion of (2.70) will yield nonlinear susceptibilities like $\chi^{(3)}(\omega_1,-\omega_1,\omega)$ etc.

Nonlinear Susceptibility for Four Wave Mixing in a Collection of Two Level Atoms

Let us consider forward four-wave mixing. The field ω_1 with propagation vector $\vec{k}_1$ and the field at ω with propagation vector $\vec{k}$ mix to produce nonlinear signal at the frquency $(2\omega_1-\omega)$ and in the direction $(2\vec{k}_1-\vec{k})$. The nonlinear susceptibility describing the four wave mixing signal can be obtained from (2.68). Note that $\vec{d}^*_{12}\psi_2^{(-)}$ will yield this four wave mixing susceptibility. On simplifiction one finds that the signal is proportional to

$$|\vec{d}^*_{12}\psi_2^{(-)}|^2 = \left| 4g^2 g_p^* \, \vec{d}^*_{12} i\psi_3^{(o)} P^{-1}(i\omega-\omega_1)\Big[i\Delta + \frac{1}{T_2} \Big] \right|^2 . \quad (2.71)$$

The intensity dependence of the pump enters through P and $\psi_3^{(o)}$. The signal as a function of pump-probe detuning $\omega-\omega_1$ will exhibit Rabi side bands.[31,32] This behavior can be understood in terms of the dressed states (Figs.4,5).

The susceptibility for phase conjugation geometry can be obtained by replacing $\epsilon e^{i\vec{k}_1.\vec{r}}$ by $2\epsilon \cos(\vec{k}_1.\vec{r})$ and by finding the $\vec{k}_1$ independent term in the Fourier decomposition of

$$g^2\psi_3^{(o)}P^{-1}(i(\omega-\omega_1))$$

where g^2 has the form $|g|^2\cos^2(\vec{k}_1.\vec{r})$. This averaging procedure yields the nonlinear susceptibility[33] for four wave mixing in phase conjugation geometry. One can rewrite the above expression as

$$g^2\psi_3^{(o)}P^{-1}\Big(i(\omega-\omega_1)\Big) = \frac{|g|^2 \eta T_1 T_2^2}{(zT_1+1)[(\Delta T_2)^2+(zT_2+1)^2]}$$

$$\times \left[\frac{\cos^2\theta}{(1+A\cos^2\theta)(1+B\cos^2\theta)} \right] ,$$

$$z = i(\omega-\omega_1) \quad , \; A = \frac{4|g|^2 T_1 T_2}{1+(\Delta T_2)^2} \quad , \quad B = \frac{4[zT_2+1]|g|^2 T_2^2}{(zT_1+1)[(\Delta T_2)^2+(zT_2+1)^2]}$$

(2.72)

The $\vec{k}_1$ independent term can now be obtained by integrating over θ. The integral can be done by Contour integration with the result

$$\frac{1}{2\pi}\int \frac{\cos^2\theta \; d\theta}{(1+A\cos^2\theta)(1 + B\cos^2\theta)} = \frac{\Big[(B+1)(A+1)\Big]^{-1/2}}{\Big[(A+1)^{1/2}+(B+1)^{1/2}\Big]} ,$$

(2.73)

The final expression for four wave mixing susceptibility is obtained by combining (2.72) and (2.73). The predictions based on this averaged susceptibility have been verified[34]. The signal as a function of pump-probe detuning not only shows resonances at Rabi side bands but also at $\pm\Delta T_2$.

Generation of Subharmonic Rabi Resonances:

We have so far treated the pump field to all orders where as the probe is treated only to first order. Many new effects start appearing as the probe field starts becoming strong. This is an area of research which is being investigated at great length.[32,35-38] Here we discuss some of the more important results. Consider six wave mixing i.e., the generation of coherent signal at the frequency $3\omega_1-2\omega$ and in the direction $3\vec{k}_1-2\vec{k}$ by a system of two-level atoms. Calculations based on optical Bloch equations (2.48) lead to the signal[32]

$$S(3\omega_1-2\omega) = \Bigg| -8i\eta g_p^2 g^3 \frac{T_1}{T_2} \left(1+ \frac{4|g|^2 T_1 T_2}{(\Delta T_2)^2+1} \right]^{-1} P^{-1}(i(\omega-\omega_1))$$

$$\times P^{-1}(2i(\omega-\omega_1))(1 + i\Delta T_2)^{-1}(2 + 3iT_2(\omega-\omega_1))(2 + iT_2(\omega-\omega_1)) \Bigg|^2 . \tag{2.74}$$

Notice the appearance of the polynomial P with the argument $2(\omega-\omega_1)$. Thus the six wave signal not only exhibits resonances at the usual Rabi side bands but also resonances at the subharmonic of Rabi side bands

$$(\omega - \omega_1) = \pm \frac{1}{2} \sqrt{4|g|^2+\Delta^2} \tag{2.75}$$

still higher order nonlinear processes lead to resonances at

$$(\omega - \omega_1) = \pm \frac{1}{n} \sqrt{4|g|^2+\Delta^2} \tag{2.76}$$

where n is an integer. If both ω and ω_1 are strong enough to saturate the transition, then one observes a number of subharmonic resonances in various nonlinear mixing and absorption

spectra. However the frequency is no longer given by the simple formula (2.76) as both ω and ω_1 are strong.

Such subharmonic resonances are also observable in strongly modulated fields.[37] For example consider a field ω_1 which is modulated at the frequency ν i.e.,

$$\varepsilon_1 \longrightarrow (1 + M\cos\nu t)\varepsilon_1 , \tag{2.77}$$

Let us assume that the field ω_1 is on resonance $\Delta=0$. Then the response of the system to such a field is given approximately by

$$\langle S^- \rangle = \frac{i\eta}{T_1} \sum_n J_n^2(\beta)(2g-n\nu)\left[\kappa^2 + (2g-n\nu)^2\right]^{-1} ,$$

$$\langle S^z \rangle = \frac{\eta}{T_1} \sum_n J_n^2(\beta)\kappa \left(\kappa^2 + (2g-n\nu)^2\right]^{-1} ,$$

$$\kappa = \frac{1}{2} \left(\frac{1}{T_1} + \frac{1}{T_2} \right) , \qquad \beta = 2gM/\nu , \tag{2.78}$$

and J_n is the Bessel function of order n.
These are the DC components in the steady state response. The response shows the presence of all the subharmonics (2.76). The weight factor depends on $J_n^2(\beta)$ which in turn depends on the modulation frequency ν, modulation index M and the strength of the driving field. The occurence of various subharmonics in nonlinear mixing and modulation spectroscopy can be understood in terms of the dressed states as is clear from Figure .

Optical Resonance in Three Level Systems:

We have so far considered a variety of physical phenomena that can occur in two-level systems. More complex situations of optical resonance phenomena in multi-level systems can be handled in a similar way. Considerable literature[39-43] exists on optical resonance in three level systems which can be classified into three classes- (i) ladder system, (ii) Raman or Λ-system (iii) Hanle or V-system. Considerations based on parity lead to only two transitions in a three level system. For example in a ladder system with states |1> (top most) |2> (intermediate) and |3> (ground), the allowed transitions are |1> ⟷ |2>, |2> ⟷ |3>. Let the field ε_1 (ε_2), frequency ω_1 (ω_2) act on the transition |1> ⟷|2>,(|2> ⟷ |3>). In rotating wave approximation the interction Hamiltonian is

$$H_1 = -\hbar(g_1|1\rangle\langle 2|e^{-i\omega_1 t} + \text{H.c.}) -\hbar(g_2|2\rangle\langle 3|e^{-i\omega_2 t} + \text{H.C.}) \quad , \tag{2.79}$$

where

$$g_1 = \frac{\vec{d}_{12}\cdot\vec{\varepsilon}_1}{\hbar} e^{i\vec{k}_1\cdot\vec{R}} \quad , \quad g_2 = \frac{\vec{d}_{23}\cdot\vec{\varepsilon}_2}{\hbar} e^{i\vec{k}_2\cdot\vec{R}} \tag{2.80}$$

The operators $A_{ij}=|i\rangle\langle j|$ for the three level system obey SU(3) algebra

$$\left[A_{ij} \ , A_{kl}\right] = A_{il}\,\delta_{jk} - A_{kj}\delta_{il} \quad , \tag{2.81}$$

The Hamiltonian (2.79) can be reduced to static form by a canonical transformation. This can be seen as follows- The Schrodinger equation

$$\frac{\partial\psi}{\partial t} = -\frac{i}{\hbar}\left[H_o + H_1(t)\right]\psi \quad H_o = \sum_j E_j\, A_{jj} \quad , \tag{2.82}$$

can be reduced by defining

$$\psi = \exp\left\{-i\left[(\omega_1+\omega_2)A_{11} +(\omega_2 A_{22})\right]t\right\}\phi \quad , \tag{2.83}$$

so that

$$\frac{\partial\phi}{\partial t} = -\frac{i}{\hbar}H\phi \quad , \tag{2.84}$$

$$H = (\Delta_1+\Delta_2)\hbar A_{11} + \Delta_2\hbar A_{22} -\left[\hbar g_1|1\rangle\langle 2| + g_2\hbar|2\rangle\langle 3| + \text{H.C}\right] \tag{2.85}$$

Here the detunings are given by

$$\Delta_1 = \frac{E_1-E_2}{\hbar} - \omega_1 \quad , \quad \Delta_2 = \frac{E_2-E_3}{\hbar} - \omega_2 \quad . \tag{2.86}$$

The Hamiltonian H(Eq.(2.85)) is static. The Schrodinger eq. (2.84) is easily solved. The eigenvalues of the 3x3 matrix

$$\begin{bmatrix} \Delta_1+\Delta_2 & -g_1 & 0 \\ -g_1^* & \Delta_2 & -g_2 \\ 0 & -g_2^* & 0 \end{bmatrix} \tag{2.87}$$

determine the dynamics of the system.[44]

Let us consider the energy absorption from a probe field applied on the transition 1⟷2 when the strong pump acts on the transition 2⟷3. We assume the probe field to be weak so that it is sufficient to calculate the energy absorption to second order in the probe field. We thus need to know the density matrix element to first order in g_1. The Schrodinger equation (2.83) leads to

$$\rho_{12}^{(1)}(t) = ig_1 \int_0^t d\tau\; e^{-i(\Delta_1+\Delta_2)(t-\tau)}\; \psi_2^{(o)}(\tau)\psi_2^{(o)^*}(t) \quad , \tag{2.88}$$

where $\psi_2^{(o)}(t)$ is to be calculated to all orders in g_2 but to zero order in g_1. The form of $\psi_2^{(o)}(t)$ depends on the initial state of the atom. If we assume that the atom at $t=0$ is in ground state and the pump is on resonance, then

$$\psi_2^{(o)}(t) = i\sin(g_2t) \tag{2.89}$$

and

$$\mathrm{Im}\rho_{12}(t) = \frac{g_1}{4}\left[\frac{\sin(g_2-\Delta_1)t}{(g_2-\Delta_1)} + \frac{\sin(g_2+\Delta_1)t}{(g_2+\Delta_1)}\right]$$

$$-\left(\frac{g_1}{2(\Delta_1+g_2)}\sin\frac{(\Delta_1+g_2)t}{2}\cos\frac{(\Delta_1-3g_2)t}{2} + g_2 \to -g_2\right] \quad . \tag{2.90}$$

Thus the transient energy absorption will exhibit resonances whenever

$$\Delta_1 = \pm g_2 \quad . \tag{2.91}$$

This is the familiar Autler-Townes doublet which has been the subject of many experimental investigations see for example Ref.-40. The appearance of this doublet can be understood in terms of the dressed states of the two-level system $|2\rangle$, $|3\rangle$ under the influence of the pump field. Normally Autler-Townes doublet is studied in the steady state i.e., one includes relaxation of the system and examines $\lim_{t\to\infty} \mathrm{Im}\rho_{12}(t)$. The analysis shows that in the steady state energy absorption exhibits resonant structures at $\Delta_1=\pm g_2$. One may also understand this qualitatively as in the long time limit only the first two terms in (2.90) will survive leading to $\delta(g_2\mp\Delta_1)$ which will then go over to Lorentzians with a finite width.

Clearly the transient absorption depends on the initial state of the atomic system. We next investigate what happens if the atom at $t=0$ is prepared in a dressed state i.e., an eigenstate of

$$h = -\hbar g_2|2\rangle\langle 3| + \text{H.C.} \tag{2.92}$$

for example in the state ψ_+

$$h\psi_+ = +g_2\,\psi_+ \quad , \tag{2.93}$$

then

$$\psi_2^{(o)}(t) = e^{-ig_2t}\,\psi_+ \quad . \tag{2.94}$$

It then follows from (2.88) that

$$\mathrm{Im}\rho_{12}^{(1)}(t) = g_1\,|\psi_+|^2\,\frac{\sin t(\Delta_1-g_2)}{(\Delta_1-g_2)} \quad . \tag{2.95}$$

The probe absorption now exhibits only a single resonance at

$$\Delta_1 = g_2 \tag{2.96}$$

i.e., one of the components of the Autler-Townes doublet is suppressed. This suppression has been observed by Mossberg and coworkers.[21]

In the foregoing we have considered the single time expectation values. We have presented results both in transient and steady state regime. Certain phenomena require the evaluation of the two-time correlation functions like $\langle S^+(t+\tau)S^-(t)\rangle$. Such correlations determine essentially the structure of the spontaneously emitted radiation. These correlations can be computed[23,39,41,42] from the knowledge of the single time mean values and the quantum regression theorem.

Just to illustrate the structure of the dipole-dipole correlation function we consider a simple situation. We assume that the atom has been prepared in one of the dressed states say ψ_+ (Eq.(2.44)). We assume that the external field is on resonance with the atom. We also ignore the relaxation effects i.e., we examine the behavior of the correlation function for times much smaller than the relaxation times. Then one can write

$$\langle S^+(t+\tau)S^-(t)\rangle = \langle\psi_+|e^{iH(t+\tau)} S^+ e^{-iH(t+\tau)} e^{iHt} S^- e^{-iHt} |\psi_+\rangle$$

$$= e^{iE_+\tau/\hbar} \langle\psi_+|S^+e^{-iH\tau} S^-|\psi_+\rangle \quad , \tag{2.97}$$

where we have used the relation $H|\psi_+\rangle = E_+|\psi_+\rangle$. On using the explicit form of $|\psi_+\rangle = \frac{1}{\sqrt{2}}(|1\rangle - \frac{|g|}{g}|2\rangle)$ (2.97) reduces to

$$\langle S^+(t+\tau)S^-(t)\rangle = e^{i|g|\tau}(1/2)\langle 2|e^{-iH\tau}|2\rangle \tag{2.98}$$

$$= e^{i|g|\tau}\,\frac{1}{4}\,\langle\psi_+-\psi_-|e^{-iH\tau}|\psi_+-\psi_-\rangle$$

$$= e^{i|g|\tau}\,\frac{1}{4}\left(e^{-i|g|\tau} + e^{i|g|\tau}\right)$$

$$= \frac{1}{4}\left(1 + e^{2i|g|\tau}\right) \tag{2.99}$$

This result when substituted in the physical definition of the transient spectrum[45] of the scattered radiation will show that the radiation consists of spectral peaks at

$$\omega = \omega_1 \quad , \quad \omega_1 + 2|g| \quad . \tag{2.100}$$

Thus the left side band of the Mollow spectrum is suppressed[22] in the transient domain when the atom is initially prepared in the dressed state $|\psi_+\rangle$.

III INTERACTION OF ACTIVE ATOMS WITH QUANTIZED FIELDS

In this section we consider the interaction of an atom with a quantized electromagnetic field in the cavity^{IV}. This is a very fundamental model the generalization of which can be used to describe a very wide class of phenomena in quantum optics. The physical phenomena depend on the strength of the cavity-atom interaction, losses from mirrors, pumping of atoms, spontaneous emission rate in the cavity, density of atoms, distribution of modes, transit time of the atoms etc. A single mode electric field in the cavity can be expressed as

$$\vec{E} = - i \vec{\epsilon} \left(\frac{2\pi\omega\hbar}{V} \right)^{1/2} a\, u(\vec{r}) + \text{H.C.} \quad , \qquad (3.1)$$

where V is the quantization volume, $\vec{\epsilon}$ the polarization vector of the mode given by the mode function $u(\vec{r})$. Here a and a^+ are the annihilation and creation operators for the field mode satisfying

$$[a , a^+] = 1 \quad . \qquad (3.2)$$

The eigenstates of the number operator a^+a will be denoted by $|n\rangle$. The interaction of a two-level atom with the field (3.1) in the rotating wave approximation can be written as

$$H = \hbar\omega_0 S^z + \hbar\omega\, a^+a + \hbar(gS^+a + g^*S^-a^+) \quad , \qquad (3.3)$$

where

$$g = i\, \vec{d}.\vec{\epsilon} \left(\frac{2\pi\omega}{\hbar V} \right)^{1/2} \quad . \qquad (3.4)$$

The properties of the Hamiltonian (3.3) were first studied by Jaynes and Cummings[46] and the model (3.3) is now known as the Jaynes-Cummings model.

A. EIGENSTATES OF (3.3) : DRESSED STATES

Exact eigenstates of (3.3) can be obtained by noting that the unperturbed states $|n,e\rangle$, $|n,g\rangle$, $n=0,1,2,\ldots,\infty$ are such that

$$H|n,e\rangle = (\frac{\hbar\omega_0}{2} + \hbar\omega n)|n,e\rangle + \hbar g^*\sqrt{n+1}\, |n+1,g\rangle \quad ,$$

$$H|n+1,g\rangle = (- \frac{\hbar\omega_0}{2} + \hbar\omega(n+1))|n+1,g\rangle + \hbar g \sqrt{n+1}\, |n,e\rangle \quad , \qquad (3.5)$$

we have now denoted the excited and ground states of the atom by $|e\rangle$, $|g\rangle$.

Thus the structure of (3.3) is such that only the states $|n,e\rangle$ and $|n+1,g\rangle$ are coupled with each other and therefore the diagonalization of the Hamiltonian (3.3) is equivalent to the diagonalization of 2x2 matrix

$$\hbar \begin{bmatrix} \frac{\omega_0}{2} + \omega n & g^*\sqrt{n+1} \\ g\sqrt{n+1} & -\frac{\omega_0}{2} \quad \omega(n+1) \end{bmatrix} \quad . \qquad (3.6)$$

The diagonalization yields the states

$$|\psi_n^{\pm}\rangle = \begin{pmatrix} \cos\theta_n \\ -\sin\theta_n \end{pmatrix} |n+1,g\rangle + \begin{pmatrix} \sin\theta_n \\ \cos\theta_n \end{pmatrix} |n,e\rangle \quad , \qquad (3.7)$$

$$\tan\theta_n = 2g\sqrt{n+1}/(\Omega_{n\Delta}-\Delta), \quad \Delta=\omega_o-\omega \quad , \Omega_{n\Delta} = \sqrt{\Delta^2+4g^2(n+1)}$$
$$n=0,1,2.... \qquad (3.8)$$

with energies

$$\hbar\omega_n^{\pm} = \hbar\omega(n+\frac{1}{2}) \pm \frac{\hbar\Omega_{n\Delta}}{2} \quad . \qquad (3.9)$$

The states (3.7) are called the dressed states for the quantized system. In addition the state $|\psi_o\rangle = |o,g\rangle$ is also an eigenstate of H with energy $-\omega_o/2$. In Fig. 4 we schematically show these dressed states. These states are coherent superpositions of $|n,e\rangle$ and $|n+1,g\rangle$. Note that the total atomic and field excitation for this set is $(n+1)$. It is easily checked that $\omega S^z + \omega a^+a$ is a constant of motion. Note that for cavity field on resonance $\Delta=0$, $\theta_n = \pi/4$. Having obtained the eigenstates, the time evolution can be studied. We can express time evolution[47] as

$$U(t)|n,e\rangle = A_{ne}(t)|n,e\rangle + B_{ne}(t)\ |n+1,g\rangle$$

$$A_{ne}(t) = \sin^2\theta_n e^{-i\omega_n^+ t} + \cos^2\theta_n e^{-i\omega_n^- t} \quad ,$$

$$U(t)|n+1,g\rangle = A_{n+1,g}(t)|n+1,g\rangle + B_{n+1,g}(t)\ |n,e\rangle$$

$$B_{ne}(t) = B_{n+1,g} = \cos\theta_n \sin\theta_n \left[e^{-i\omega_n^+ t} - e^{-i\omega_n^- t}\right] \quad .$$

$$A_{n+1,g}(t) = \cos^2\theta_n e^{-i\omega_n^+ t} + \sin^2\theta_n e^{-i\omega_n^- t} \quad ,$$

$$A_{og}(t) = e^{i\omega_o t/2} \quad , \quad B_{og}(t) = 0 \quad . \qquad (3.10)$$

We are now in a position to understand various physical phenomena in cavities.

B. ATOMIC EXCITATION PROBABILITIES

Consider an atom in the excited state $|e\rangle$ passing through a cavity in which the field is in a state ρ_F. Let the atom interact for a time t. The probability p_e that the atom comes out in the excited state is obtained from $|A_{ne}(t)|^2$. On averaging over the initial field distribution and on specializing to the resonant situation, we get

$$p_e(t) = \sum_o^{\infty} p(n) \cos^2 gt\sqrt{n+1} \quad , \quad p(n) = \langle n|\rho_F|n\rangle \quad . \qquad (3.11)$$

Note that the distribution p(n) for some typical states of the field is given by

$$p(n) = \delta_{n,n_o} \qquad \text{Fock state } |n_o\rangle \quad ,$$

$$= \frac{\bar{e}^{\bar{n}}(\bar{n})^n}{n!} \qquad \text{coherent state } |\alpha\rangle \quad , \quad |\alpha|^2 = \bar{n} \quad ,$$

$$= (\bar{n})^n/(1+\bar{n})^{n+1} \qquad \text{thermal state} \quad . \tag{3.12}$$

If the field in the cavity is initially in vacuum state $|o\rangle$, then

$$p_e(t) = \cos^2 gt \quad . \tag{3.13}$$

The atom oscillates back and forth between the states $|e\rangle$ and $|g\rangle$ at a frequency determined by the field-atom coupling constant g. The oscillations occur because the atom to start with is in excited state. The atom comes to ground state by emitting a photon. This photon can be reabsorbed by the atom. The series of emissions and absorptions leads to the oscillatory behavior (3.13). These oscillations are known as the vacuum field Rabi oscillations[48-50] and do not require the presence of the field. The atomic excitation energy is exchanged between the atom and the field mode. Note that for reabsorption of the emitted photon, the photon must remain in the cavity. This requires that the escape rate from the cavity must be much smaller than the rate of reabsorption. Otherwise one will get simple exponential decay.

The excitation probability $p_e(t)$ shows remarkable collapse and revival[51] of Rabi oscillations when the distribution $p(n)$ of the cavity photon is distinct from δ_{n,n_o}. For example for a Pois-

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FIGURE CAPTIONS

Fig.1: General structure of the diagrams contributing to $\chi^{(3)}$. The collision induced coherence can be seen by combining the contributions of the triplet of diagrams like these.

Fig.2: Three-level system relevant to transitions in a system like Ruby. This is also an example of an open two-level system since $\gamma_{31}\neq 0$.

Fig.3: The absorption spectrum (2.70) for $\Delta T_2=5$, $T_2/T_1=2$; Rabi frequency = $20/T_2$. The spectral features (b), (c) and (d) respectively, correspond to the resonances at $\omega\approx\omega_1 - \sqrt{\Delta^2+4|g|^2}$, ω_1 and $\omega_1 + \sqrt{\Delta^2+4|g|^2}$. (after Ref. 28b).

Fig.4: Dressed states (3.7) of a single mode field interacting with a two-level atom. The solid lines give the resonant transitions relevant for ground state absorption. In spontaneous emission one will see all the transitions marked by the wavy lines depending of course on the distribution of the input photons. Mollow spectrum in a strong coherent field arises from transitions $|\psi_n^-\rangle \longrightarrow |\psi_{n-1}^{\pm}\rangle$, $|\psi_n^+\rangle \longrightarrow |\psi_{n-1}^{\pm}\rangle$ for large n. Note that for large n, $\Omega_{n\Delta} \sim \sqrt{4|g|^2 n+\Delta^2}$ and the dispersion in photon number $\sim \frac{1}{\sqrt{\bar{n}}}$ which becomes quite small.

Fig.5: The diagrams showing the existence of Rabi side bands in four wave mixing in two-level atoms in presence of a strong pump. Note that strong pump leads to population in both the dressed levels $|\psi_n^{\pm}\rangle$.

Fig.6: (a) The diagrams showing the existence of the first subharmonic of the Rabi side band in six wave mixing. (b) The diagram showing subharmonic of the Rabi side band in two photon absorption from a probe. In modulation spectroscopy the modulated compnent of the pump can be treated as a probe.

Fig.7: The probability of finding the atom in the excited state as a function of time for an input coherent field with average number 5 curve A is for $Q=\infty$, curves B [C] represents $P(t)-\frac{1}{2}$ [$P(t)-\frac{3}{4}$] for κ/g = 0.001 [0.005]. The finite value of Q affects collapses and revivals (after Ref. 53a).

Fig.8: Some of the low lying dressed states of a system of large number of atoms interacting with the cavity mode. Wavy (solid) lines give the transitions due to cavity leakage and spontaneous emission (external fields). The vacuum field Rabi splitting in absorption will correspond to transitions $|o\rangle \longrightarrow |+\rangle$, $|-\rangle$. In two photon absorption $2\omega_o$ can resonate with any of the levels $|1\rangle$, $|2\rangle$ and $|3\rangle$ (after Ref. 56).

Fig.9: Spectrum of spontaneous emission as a function of $\frac{\omega-\omega_c}{g}$ in a resonant cavity with $Q=\infty$ and detector width γ/g = 0.1. Initially The average number of photons in the cavity is 10. The curve a (b) is for coherent (thermal) field. The spectrum depends on the photon statistics of the input field (after Ref. 57).

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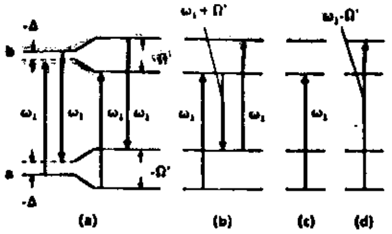

Fig. 1. (a) A laser field at frequency ω_1 that is detuned from resonance by frequency Δ creates a pair of virtual atomic levels, shown as dashed lines. At high laser intensities, the energy-level structure is modified by the ac-Stark effect to become pairs of dressed levels separated by the generalized Rabi frequency Ω', as shown at the right. The possible transitions among these levels are illustrated in (b)–(d). (Both the pump-laser detuning from resonance and the generalized Rabi frequency are negative in this figure.) (b) By simultaneously absorbing two pump-laser photons and emitting a photon with frequency $\omega_1 + \Omega'$, the atom makes a transition to the excited state. As a consequence of this three-photon effect, an incident wave at frequency $\omega_1 + \Omega'$ experiences gain. (c) When the difference between the pump- and probe-wave frequencies is comparable with the inverse of the atomic response time, the strong interaction of the waves' beat frequency with the populations of the atomic levels results in a nearly degenerate absorption feature with a dispersive line shape. (d) The ac-Stark effect shifts the atomic resonance frequency to $\omega_1 - \Omega'$.

Hamiltonian. Each pair of dressed states is split by the generalized Rabi frequency $\Omega' = (\Delta/|\Delta|)(\Delta^2 + \Omega^2)^{1/2}$, where $\Omega = |\mu_{ba}|E_1/\hbar$ is the Rabi frequency, μ_{ba} is the atomic-dipole moment matrix element, and E_1 is the real amplitude of the strong laser field.

The structure of the modified atomic states can be determined by measuring the absorption spectrum with a probe wave whose intensity is weak enough that it does not itself dress the atomic states. For the moment, we ignore the effects of atomic motion. Starting with the density-matrix equations of motion[5,9,27,28] and including the influence of the strong optical field to all orders, while retaining that of the probe field to only first order, results in the following amplitude absorption coefficient for the weak probe wave at ω_3 (Ref. 12)

$$\alpha(\delta) = \frac{2\pi N\omega_3|\mu_{ba}|^2(\rho_{bb} - \rho_{aa})^{c}}{\hbar n_3 c} \times \mathrm{Im}\left[\frac{(\delta + i/T_1)((\delta - \Delta + i)/T_2)(\Delta - i/T_2) - (\Omega^2\delta/2)}{(\Delta - i/T_2)D(\delta)}\right], \tag{1}$$

where $\delta = \omega_3 - \omega_1$ is the probe–pump detuning, T_1 and T_2 are the population lifetime and the dipole-dephasing time, respectively, N is the number density of atoms, n_3 is the index of refraction of the atomic medium at frequency ω_3, and Im[ ] denotes the imaginary part. In Eq. (1), $(\rho_{bb} - \rho_{aa})^{dc}$ is the steady state population inversion induced by the strong laser field:

$$(\rho_{bb} - \rho_{aa})^{dc} = \frac{(1 + \Delta^2 T_2^2)(\rho_{bb} - \rho_{aa})^0}{1 + \Delta^2 T_2^2 + \Omega^2 T_1 T_2}, \tag{2}$$

where $(\rho_{bb} - \rho_{aa})^0$ is the equilibrium population inversion in the absence of the optical fields, and $D(\delta)$ is a cubic polynomial in δ given by

$$D(\delta) = (\delta + i/T_1)(\delta + \Delta + i/T_2)(\delta - \Delta + i/T_2) - \Omega^2(\delta + i/T_2). \tag{3}$$

The probe-wave absorption coefficient $\alpha(\delta)$ for a typical case, shown in Fig. 2, has three features. The ac-Stark-shifted atomic resonance at $\omega_3 = \omega_1 - \Omega'$ corresponds to the transition between the lowest and highest dressed levels [Fig. 1(d)]. Because the ground state is more highly populated than the excited state, the probe wave is strongly attenuated at this frequency.

The central feature in the absorption spectrum occurs where the probe-pump detuning is less than the inverse of some characteristic atomic response time [Fig. 1(c)]. The line shape of this nearly degenerate feature appears dispersive whenever the pump is detuned from resonance. In the high-intensity, large-detuning limit ($\Omega, \Delta \gg 1/T_2$), which corresponds to our experimental conditions, Eqs. (1)–(3) simplify somewhat in the vicinity of the nearly degenerate resonance to give

$$\alpha_{nd}(\delta) \approx A\,\frac{A_1 + A_2\dfrac{\delta}{\Gamma_{nd}} + A_3\dfrac{\delta^2}{{\Gamma_{nd}}^2}}{1 + \delta^2/{\Gamma_{nd}}^2}, \tag{4}$$

where

$$A = \frac{2\pi N\omega_3|\mu_{ba}|^2}{n_3 c\hbar T_1}\,\frac{1}{\left[\dfrac{1}{T_1} + \dfrac{1}{T_2}(\Omega/\Delta)^2\right]^3}, \tag{5}$$

$$A_1 = \frac{1}{\Delta^2 T_1 T_2}\left[\frac{1}{T_1} + \frac{1}{T_2}(\Omega/\Delta)^2\right], \tag{6}$$

$$A_2 = \frac{\Omega^2\Gamma_{nd}}{2\Delta^3}\left\{\frac{1}{T_2}[2 + (\Omega/\Delta)^2] - \frac{1}{T_1}\right\}, \tag{7}$$

③

$$A_3 = \frac{{\Gamma_{nd}}^2}{\Delta^2 T_2}, \tag{8}$$

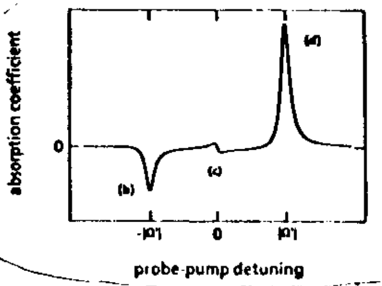

Fig. 2. The probe absorption spectrum as modified by the intense pump laser field for $T_2/T_1 = 2$, $\Omega T_2 = 20$, and $\Delta T_2 = -5$. (b)–(d) Correspondence between the three spectral features and the transitions illustrated in Fig. 1.

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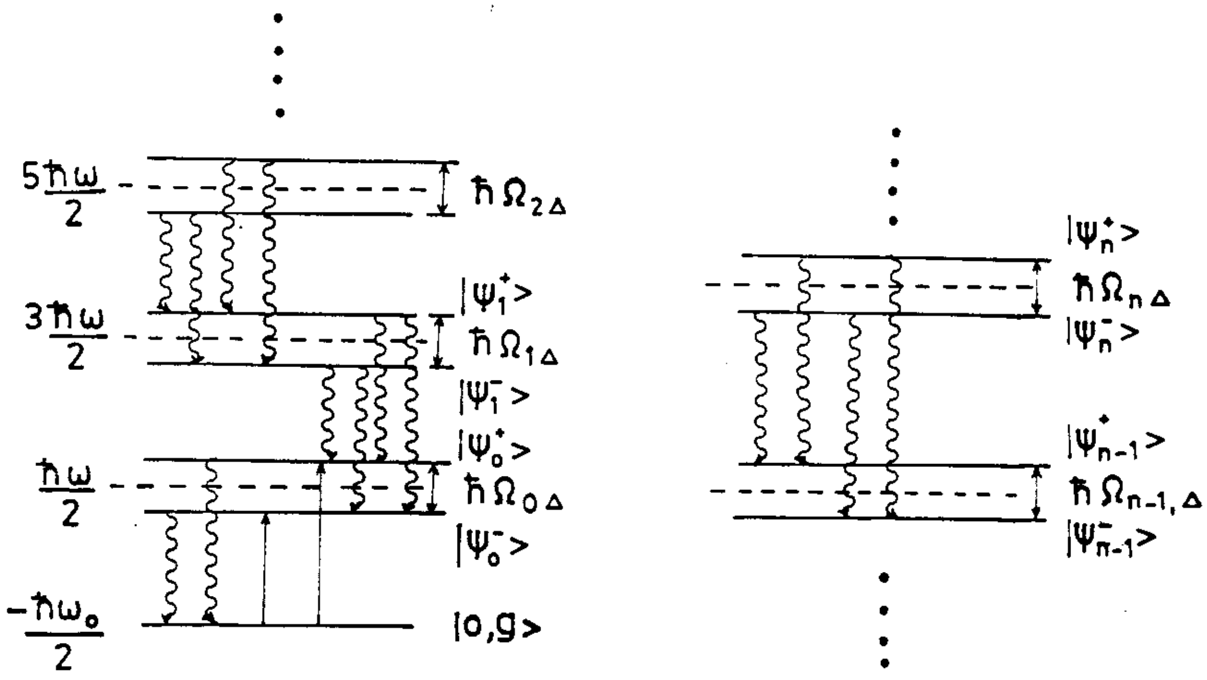
$5\frac{\hbar\omega}{2}$
$\hbar\Omega_{2\Delta}$
$|\Psi_1^+>$
$3\frac{\hbar\omega}{2}$
$\hbar\Omega_{1\Delta}$
$|\Psi_1^->$
$|\Psi_0^+>$
$\frac{\hbar\omega}{2}$
$\hbar\Omega_{0\Delta}$
$|\Psi_0^->$
$-\frac{\hbar\omega_0}{2}$
$|0,g>$
$|\Psi_n^+>$
$\hbar\Omega_{n\Delta}$
$|\Psi_n^->$
$|\Psi_{n-1}^+>$
$\hbar\Omega_{n-1,\Delta}$
$|\Psi_{n-1}^->$

Fig. 4

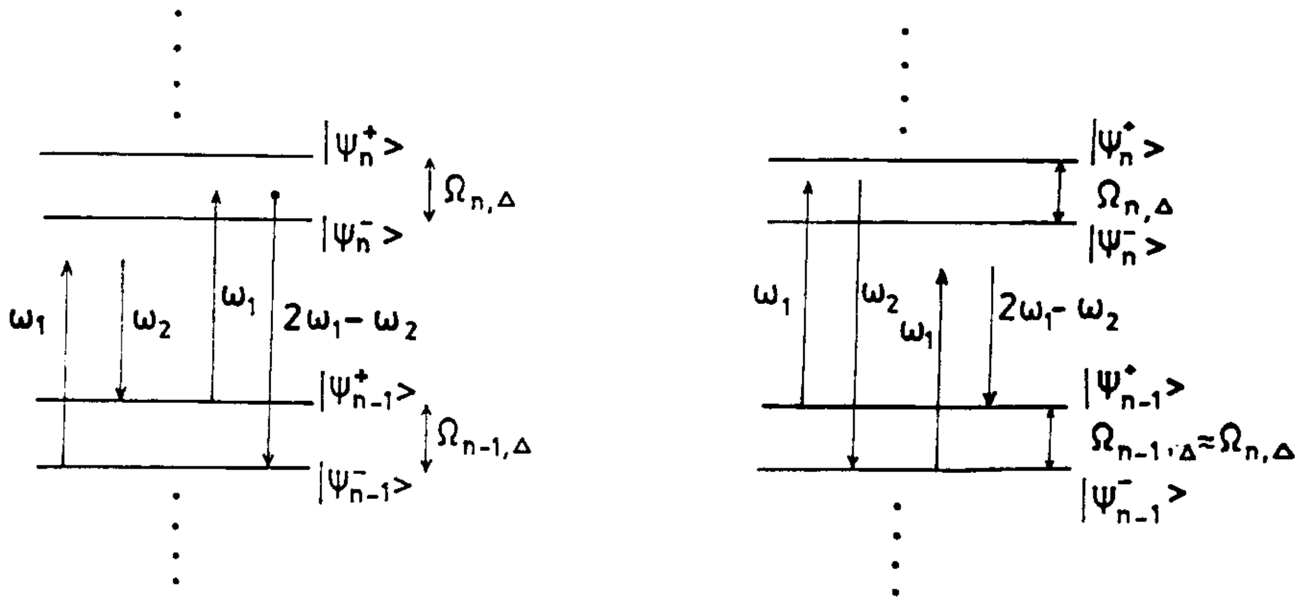
$|\Psi_n^+>$
$\Omega_{n,\Delta}$
$|\Psi_n^->$
ω_1
ω_2
ω_1
$2\omega_1-\omega_2$
$|\Psi_{n-1}^+>$
$\Omega_{n-1,\Delta}$
$|\Psi_{n-1}^->$
$|\Psi_n^+>$
$\Omega_{n,\Delta}$
$|\Psi_n^->$
ω_1
ω_2
ω_1
$2\omega_1-\omega_2$
$|\Psi_{n-1}^+>$
$\Omega_{n-1,\Delta}\approx\Omega_{n,\Delta}$
$|\Psi_{n-1}^->$

Fig. 5

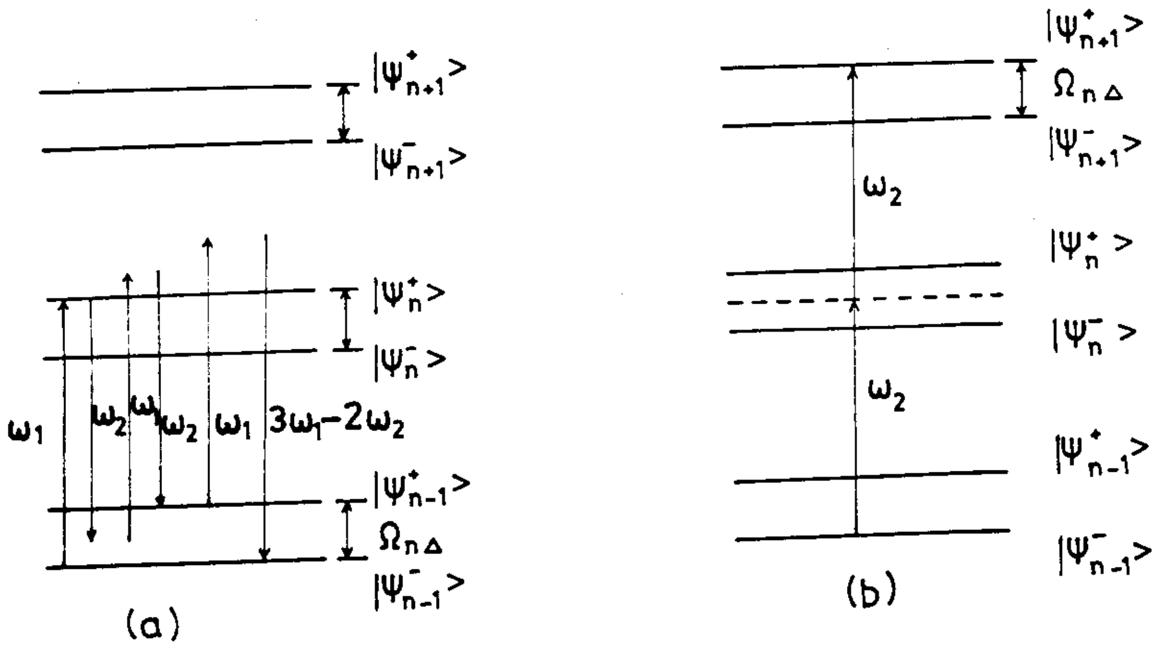
$|\Psi^+_{n+1}>$
$|\Psi^-_{n+1}>$
$|\Psi^+_n>$
$|\Psi^-_n>$
ω_1
ω_2
ω_1
ω_2
ω_1
$3\omega_1-2\omega_2$
$|\Psi^+_{n-1}>$
$\Omega_{n\Delta}$
$|\Psi^-_{n-1}>$
(a)
$|\Psi^+_{n+1}>$
$\Omega_{n\Delta}$
$|\Psi^-_{n+1}>$
ω_2
$|\Psi^+_n>$
$|\Psi^-_n>$
ω_2
$|\Psi^+_{n-1}>$
$|\Psi^-_{n-1}>$
(b)

Fig.6