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Winter College on Quantum Optics: Novel Radiation Sources

3-21 March 1997

Subnatural linewidth spectroscopy and quantum noise quenching

G. S. Agarwal

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Physics and Applications of Man-Made Quantum Interferences

G.S. Agarwal
Physical Research Laboratory, Ahmedabad- 380 009, India

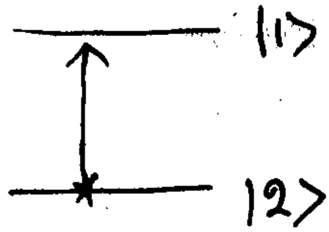
[illegible] been known from time immemorial. Interference is very [illegible] physics as we deal with super position of states all the [illegible] techniques such as quantum beat and Hanle [illegible]. The interferences leads to, for example, vanishing of spectroscopic lines in nonlinear spectroscopy of atoms/molecules. In the last few years, one has recognised the possibility of creating and controlling interferences by using electromagnetic fields. I will describe the basis of this new idea and present some applications which not only include laboratory applications but also practical ones in the context of the correction of pulse distortion, soliton amplification, isotope discrimination.

Quantum Interference

Transition amplitudes

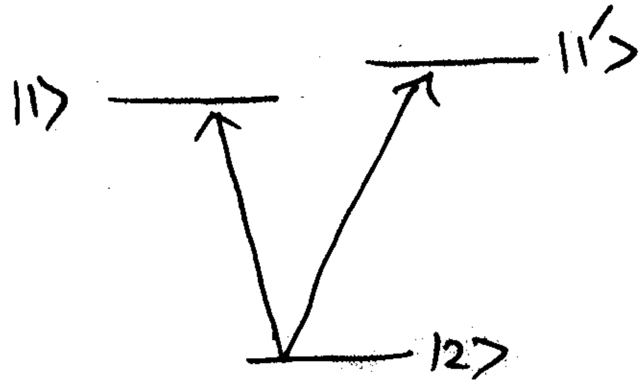
Optical Absorption

$$\propto \mathrm{Im}\,\frac{(\text{dipole matrix})^2(\text{density})}{(\omega_{12}-\omega_l-i\Gamma_{12})}$$



$\sum$

Sum over all final states



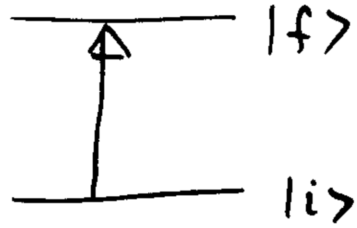
No Interference "yet"

unless "final states" interact or prepared as coherent superposition

(FANO INTERFERENCE — INTERACTING FINAL STATES)

TWO PHOTON FERMI GOLDEN RULE

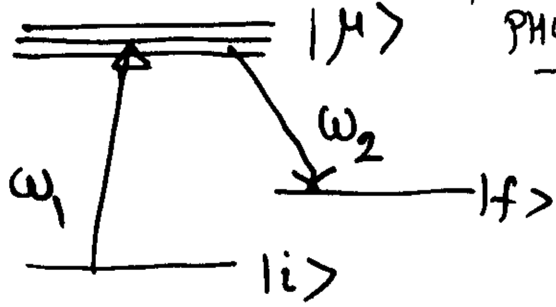
$$\frac{2\pi}{\hbar} |\langle f | H_1 | i \rangle|^2 \, \delta(E_f - E_i)$$



(i,f include field Quantum no.)

MUCH MORE NATURAL IN MULTI PHOTON --

"Tunable" ⟶



$$\frac{2\pi}{\hbar} \left| \sum_{\mu} \frac{\langle f | H_1 | \mu \rangle \langle \mu | H_1 | i \rangle}{(E_{\mu i})} \right|^2 \delta(E_f - E_i)$$

ω_2 : Could be stimulated or spont.

If 2 photon matrix element

$$\sum_{l} \frac{\langle f | \vec{d} | l \rangle \langle l | \vec{d} | i \rangle}{(E_{li} - \hbar\omega_1)} = 0 \quad \Bigg| \quad \delta(E_f - E_i - \hbar\omega_1 + \hbar\omega_2)$$

<u>No</u> transition

special case 2 intermediate states

Tuning & strength of matrix elements

Bjorkholm + Liao

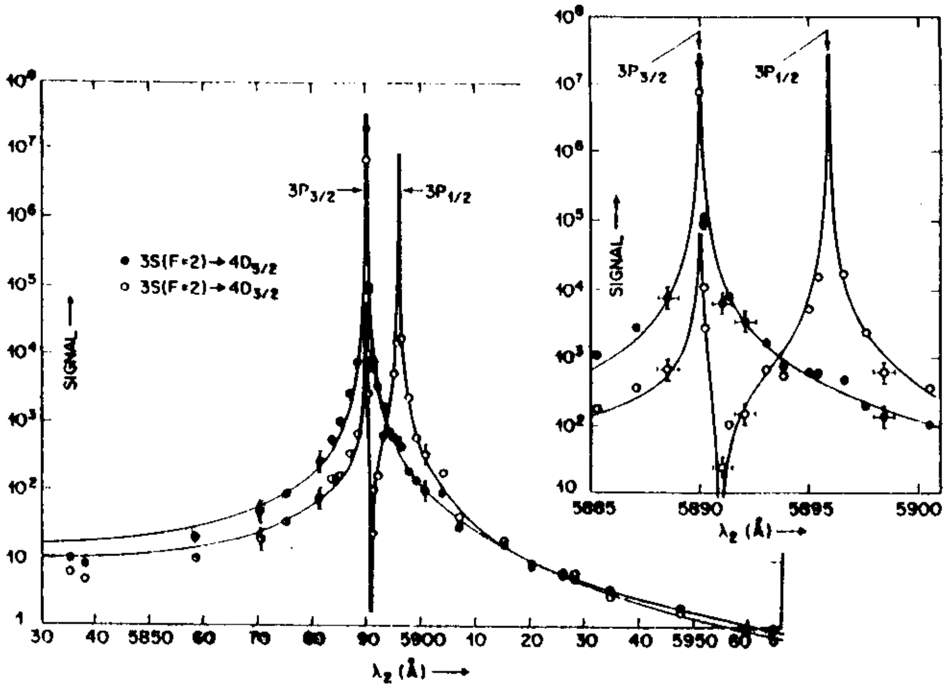


FIG. 2. Normalized two-photon transition rates for the $3S(F=2)\to 4D_{5/2}$ and $3S(F=2)\to 4D_{3/2}$ transitions as a function of the wavelength of the fixed-frequency laser, λ_2. (Note that $\nu_1=\nu_2$ for $\lambda_2=5787$ Å.) The points are experimental and the curves are theoretical. The inset shows the behavior in the region from 5885 to 5900 Å with an expanded horizontal axis.

● Single Intermediate state

○ Two Int states

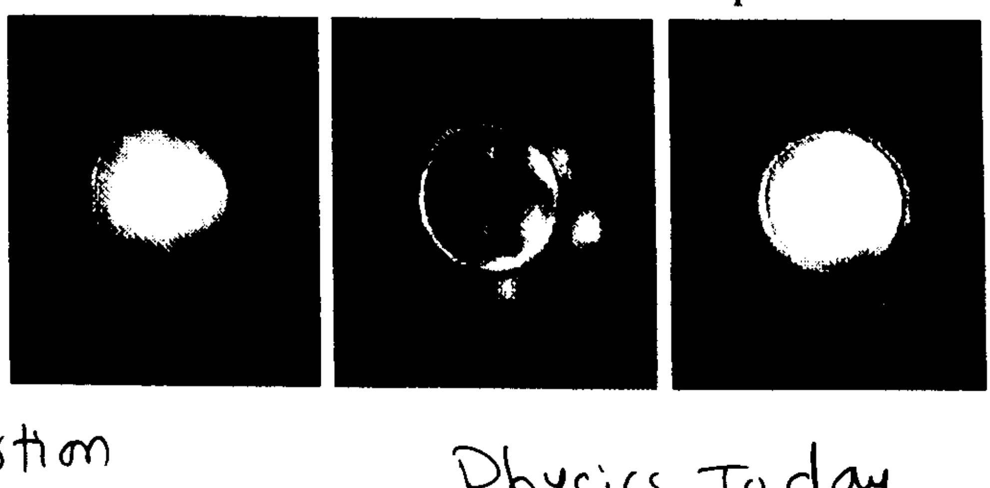
$4D_{5/2}$

$3P_{3/2}$

$3S(F=2)$

$3P_{1/2} \to 4D_{5/2}$ forbidden

Quantum Interference Used to Eliminate Optical Problem



$n_2 > 0$

Distortion correction — "Filamentation"

Physics Today — March '96

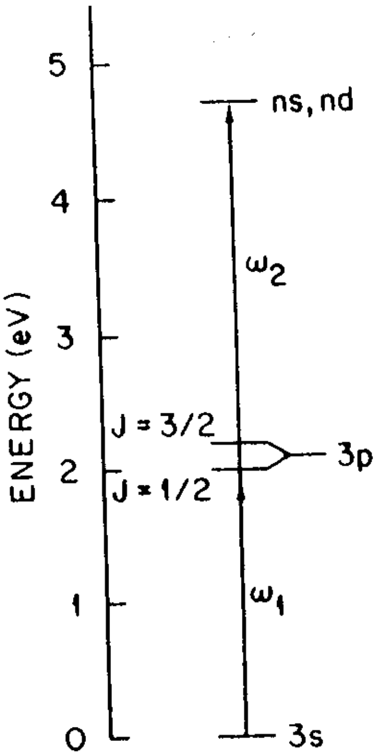


FIG. 1. Two-photon excitation scheme employed in this study. Ionization signals in the heat pipe result from collisions.

COMPTON et al.

PR A 40, 5044 (1989)

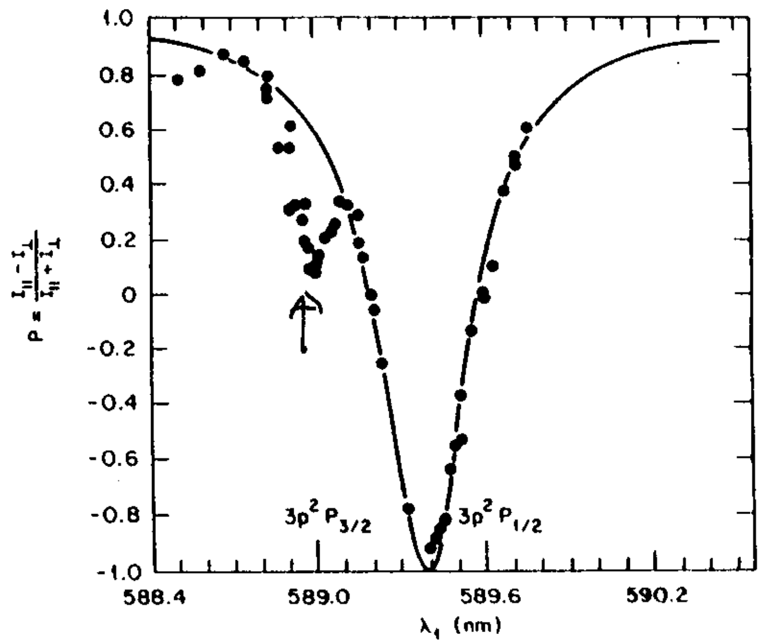


FIG. 2. Experimental measurements (●) and theoretical calculations (——) of the polarization when tuning $\omega_1(\lambda_1)$ near the $3p$ fine-structure levels for the case where $\omega_1+\omega_2$ excites the $6s$ level.

$I(\theta) \equiv A + B\cos^2\theta$

θ: Angle between pol of beams

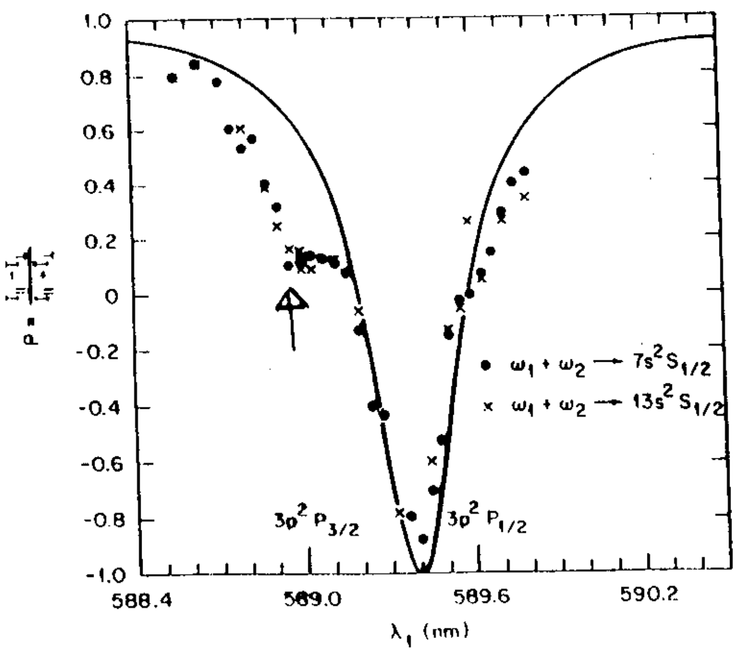


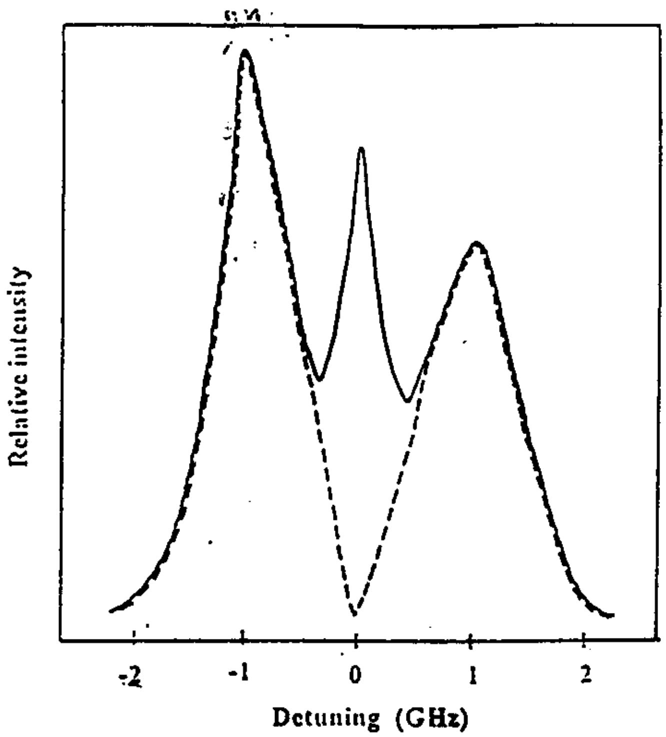
FIG. 3. Experimental measurements (●) and theoretical calculations (——) of the polarization when tuning $\omega_1(\lambda_1)$ near

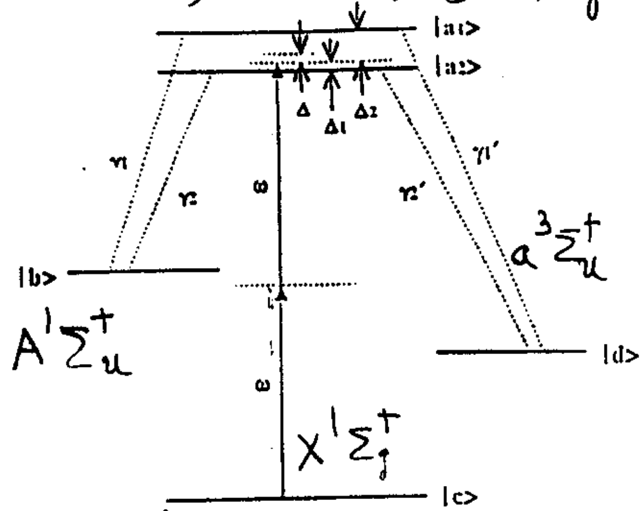
↑ on Resonance
Real state
Collisional
effects etc.

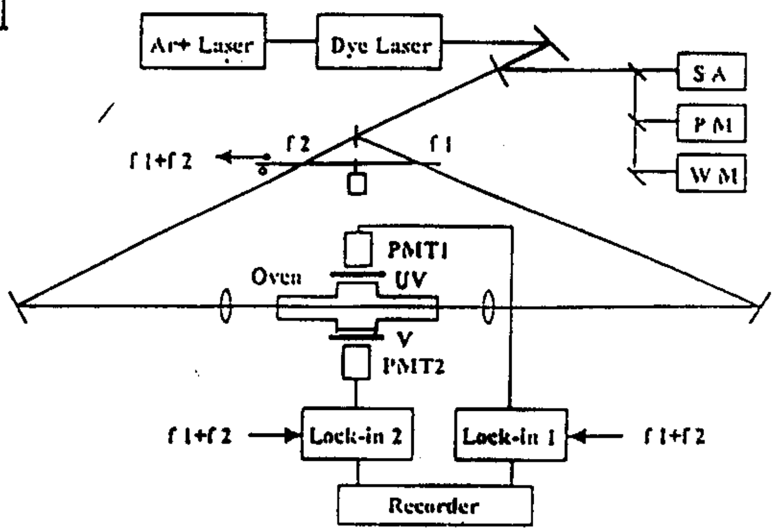
SPONTANEOUS EMISSION CANCELLATION

XIA, YE & ZHU (PRL '96)

77, 1032; 5th August







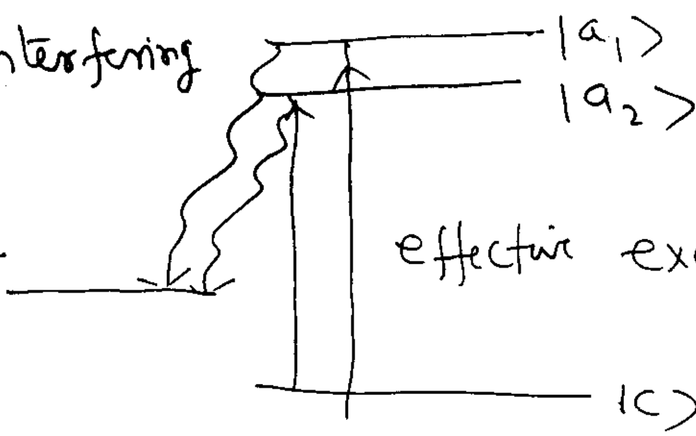
Na dimer:

$|a_1\rangle$, $|a_2\rangle$

mixing of singlet & Triplet by spin orbit int.

(Agarwal, PRA Feb 1997 to be published)

Two Interfering Pathways

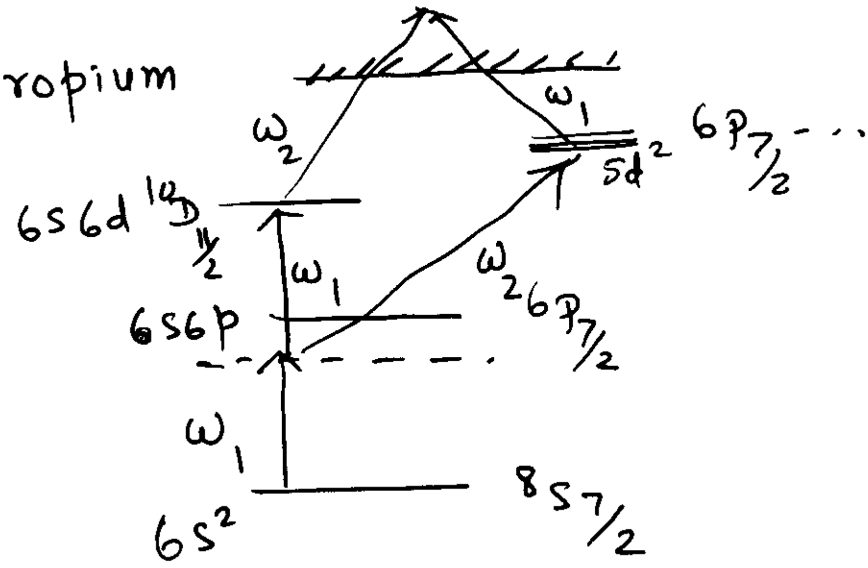


Ionization Similar Interferences

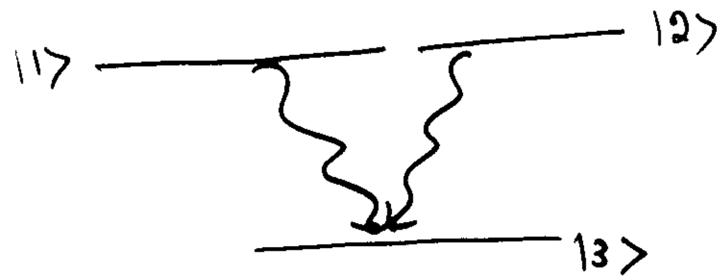
Exp in Europium

(Ahmad;

To be repeated)



EARLIEST EXAMPLE OF QUANTUM INTERFERENCE & POPULATION TRAPPING IN EXCITED STATES



$$\dot{\rho}_{11} = -2\gamma_1 \rho_{11} + (\ ) \rho_{12}$$

$$\dot{\rho}_{22} = -2\gamma_2 \rho_{22} + \underbrace{(\ ) \rho_{21}}_{\text{Interference terms}}$$

Interference terms

$\vec{d}_{13} \cdot \vec{d}_{23}^{\,*}$ (Zeeman sublevels zero)

Coherence & population Trapping $\rho_{12} \neq 0$, $\rho_{11} \neq 0$ | $\rho_{22} \neq 0$

Pump |1> initially

COHERENT POPULATION TRAPPING

(Agarwal, Quantum Optics 1974)

SEARCH FOR SYSTEMS WHERE $d_{13} \cdot d_{23} \neq 0$

NATURAL OCCURENCE IN SYSTEMS

MAN MADE — LABORATORY CREATION — EM FIELDS

The absorption of the probe is proportional to the imaginary part of the Induced Polarization

From the Maxwell equation for the field

$$E \sim e^{i\frac{\omega}{c}\sqrt{1+4\pi\chi(\omega)}L} \tag{1}$$

Intensity is

$$|E|^2 \sim |exp\{\frac{i\omega}{c}\sqrt{1+4\pi\chi(\omega)}L\}|^2 \tag{2}$$

The Intensity attenuation αL is given by

$$\alpha L = \frac{4\pi\omega L}{c} Im(\chi(\omega)) \tag{3}$$

Induced Polarization is $nd_{ij}\rho_{ji}$

$$\alpha L = \frac{4\pi\omega Ln|d_{ij}|^2}{\hbar c} Im(\frac{\rho_{ji}}{(d_{ji}.E/\hbar)}) \tag{4}$$

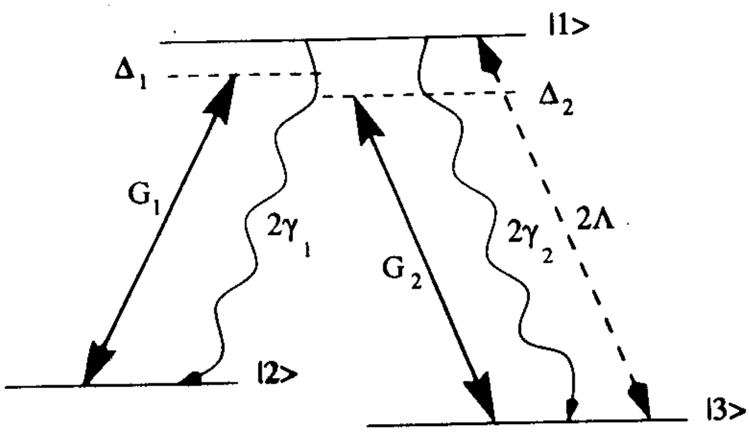
Which reduces to

$$\alpha L = \alpha_0 L \quad Im(\frac{\rho_{ji}\Gamma_{ji}}{(d_{ji}.E/\hbar)}) \equiv \alpha_0 L \ A \tag{5}$$

↓ net absorption

↓ usual absorption at line center

LAMBDA SYSTEM



$$\dot{\rho}_{11} = -2(\gamma_1+\gamma_2)\rho_{11} + 2\Lambda\rho_{33} + iG_1\rho_{21} - iG_1^*\rho_{12} + iG_2\rho_{31} - iG_2^*\rho_{13}$$

$$\dot{\rho}_{12} = -(\gamma_1+\gamma_2+\Gamma_{12}^{ph}+i\Delta_1)\rho_{12} - iG_1(\rho_{11}-\rho_{22}) + iG_2\rho_{32}$$

$$\dot{\rho}_{13} = -(\gamma_1+\gamma_2+\Lambda+\Gamma_{13}^{ph}+i\Delta_2)\rho_{13} + iG_1\rho_{23} - iG_2(\rho_{11}-\rho_{33})$$

$$\dot{\rho}_{22} = 2\gamma_1\rho_{11} - iG_1\rho_{21} + iG_1^*\rho_{12}$$

$$\dot{\rho}_{23} = -(\Gamma_{23}^{ph} - i(\Delta_1-\Delta_2))\rho_{23} + iG_1^*\rho_{13} - iG_2\rho_{21}$$

$$\dot{\rho}_{33} = 2\gamma_2\rho_{11} - 2\Lambda\rho_{33} - iG_2\rho_{31} + iG_2^*\rho_{13}$$

$$A = Real\frac{(\Gamma_{23}+i\Delta_2)\Gamma_{13}}{G_1^2 + (\Gamma_{13}+i\Delta_2)(\Gamma_{23}+i\Delta_2)}, \qquad (1)$$

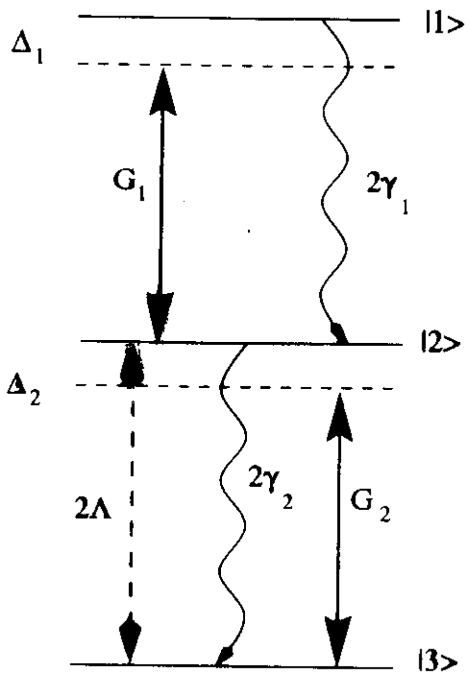
for, $G_1 >> G_2$, $\Delta_1 = 0$ and $\Lambda = 0$.

Line center $\Delta_2 = 0$

$$A = \frac{\Gamma_{23}\Gamma_{13}}{G_1^2} \rightarrow 0 \text{ if } \Gamma_{23} = 0$$

WHY 'A' $\propto$ to Γ_{23} ($|2\rangle \leftrightarrow |3\rangle$ electric dibole forbidden)

LADDER SYSTEM



$$\dot{\rho}_{11} = -2\gamma_1\rho_{11} + iG_1\rho_{21} - iG_1^*\rho_{12}$$

$$\dot{\rho}_{12} = -(\gamma_1 + \gamma_2 + \Gamma_{12}^{ph} + i\Delta_1)\rho_{12} + iG_1(\rho_{22} - \rho_{11}) - iG_2^*\rho_{13}$$

$$\dot{\rho}_{13} = -(\gamma_1 + \Gamma_{13}^{ph} + i(\Delta_1 + \Delta_2))\rho_{13} + iG_1\rho_{23} - iG_2\rho_{12}$$

$$\dot{\rho}_{22} = 2\gamma_1\rho_{11} - 2\gamma_2\rho_{22} + 2\Lambda\rho_{33} - iG_1\rho_{21} + iG_1^*\rho_{12} + iG_2\rho_{32} - iG_2^*\rho_{23}$$

$$\dot{\rho}_{23} = -(\gamma_2 + \Lambda + \Gamma_{23}^{ph} + i\Delta_2)\rho_{23} + iG_1^*\rho_{13} + iG_2(\rho_{33} - \rho_{22})$$

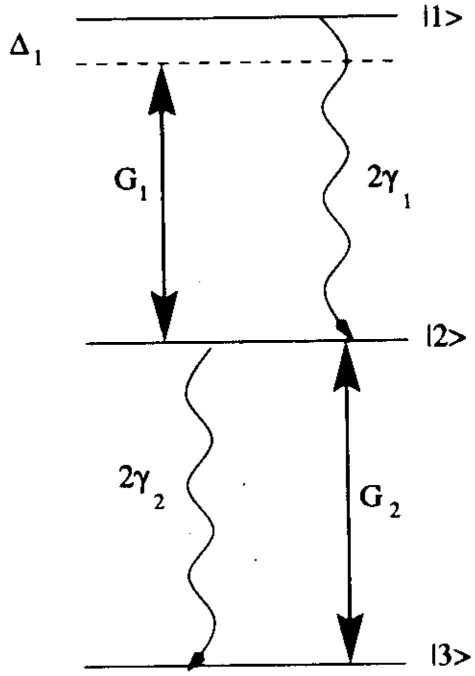
$$\dot{\rho}_{33} = 2\gamma_2\rho_{22} - 2\Lambda\rho_{33} - iG_2\rho_{32} + iG_2^*\rho_{23}$$

$$A = Real\frac{(\Gamma_{13} + i\Delta_2)\Gamma_{23}}{G_1^2 + (\Gamma_{23} + i\Delta_2)(\Gamma_{13} + i\Delta_2)}, \qquad (1)$$

for, $G_1 >> G_2$, $\Delta_1 = 0$ and $\Lambda = 0$

$A \neq 0$ as Γ_{13} typically $\neq 0$

LADDER SYSTEM



$$\dot{\rho}_{11} = -2\gamma_1\rho_{11} + iG_1\rho_{21} - iG_1^*\rho_{12}$$

$$\dot{\rho}_{12} = -(\gamma_1 + \gamma_2 + \Gamma_{12}^{ph} + i\Delta_1)\rho_{12} + iG_1(\rho_{22} - \rho_{11}) - iG_2^*\rho_{13}$$

$$\dot{\rho}_{13} = -(\gamma_1 + \Gamma_{13}^{ph} + i(\Delta_1 + \Delta_2))\rho_{13} + iG_1\rho_{23} - iG_2\rho_{12}$$

$$\dot{\rho}_{22} = 2\gamma_1\rho_{11} - 2\gamma_2\rho_{22} + 2\Lambda\rho_{33} - iG_1\rho_{21} + iG_1^*\rho_{12} + iG_2\rho_{32} - iG_2^*\rho_{23}$$

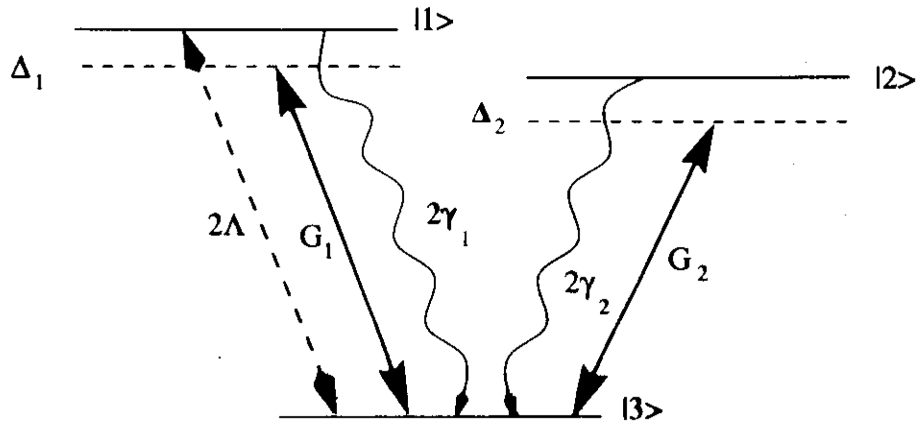
$$\dot{\rho}_{23} = -(\gamma_2 + \Lambda + \Gamma_{23}^{ph} + i\Delta_2)\rho_{23} + iG_1^*\rho_{13} + iG_2(\rho_{33} - \rho_{22})$$

$$\dot{\rho}_{33} = 2\gamma_2\rho_{22} - 2\Lambda\rho_{33} - iG_2\rho_{32} + iG_2^*\rho_{23}$$

$$A \equiv \frac{G_2^2}{(\Gamma_{23}\gamma_2 + 2G_2^2)} Real \frac{(\Gamma_{13} + \gamma_2 + i\Delta_1)\Gamma_{12}}{G_2^2 + (i\Delta_1 + \Gamma_{13})(i\Delta_1 + \Gamma_{12})}, \qquad (1)$$

for, $G_2 >> G_1$, $\Delta_2 = 0$ and $\Lambda = 0$.

V - SYSTEM



$$\dot{\rho}_{11} = -2\gamma_1\rho_{11} + 2\Lambda\rho_{33} + iG_1\rho_{31} - iG_1^*\rho_{13}$$

$$\dot{\rho}_{12} = -(\gamma_1 + \gamma_2 + \Gamma_{12}^{ph} + i(\Delta_1 - \Delta_2))\rho_{12} + iG_1\rho_{32} - iG_2^*\rho_{13}$$

$$\dot{\rho}_{13} = -(\gamma_1 + \Lambda + \Gamma_{13}^{ph} + i\Delta_1)\rho_{13} - iG_2\rho_{12} - iG_1(\rho_{11} - \rho_{33})$$

$$\dot{\rho}_{22} = -2\gamma_2\rho_{22} + iG_2\rho_{32} - iG_2^*\rho_{23}$$

$$\dot{\rho}_{23} = -(\gamma_2 + \Gamma_{23}^{ph} + i\Delta_2)\rho_{23} - iG_1\rho_{21} - iG_2(\rho_{22} - \rho_{33})$$

$$\dot{\rho}_{33} = 2\gamma_1\rho_{11} + 2\gamma_2\rho_{22} - 2\Lambda\rho_{33} - iG_1\rho_{31} + iG_1^*\rho_{13} - iG_2\rho_{32} + iG_2^*\rho_{23}$$

$$A \equiv \frac{(G_2^2 + \gamma_2\Gamma_{23})}{(2G_2^2 + \gamma_2\Gamma_{23})} Real \frac{(\Gamma_{12} + \gamma_2(\frac{G_2^2}{G_2^2 + \gamma_2\Gamma_{23}}) + i\Delta_1)\Gamma_{13}}{G_2^2 + (i\Delta_1 + \Gamma_{12})(i\Delta_1 + \Gamma_{13})} \quad (1)$$

for, $G_2 >> G_1$, $\Delta_2 = 0$ and $\Lambda = 0$.

Nature of the Quantum Interference in Electromagnetic Field Induced Control of Absorption

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(August 31, 1996)

PRA Feb 1997

Abstract

Various three level schemes for electromagnetic field induced control of absorption are analysed to isolate the precise nature of the quantum interference. Such interference manifests through the dispersive contributions to the absorption line shapes. Depending on the excitation scheme the dispersive terms can be either constructive or destructive. The collisional dephasing in some cases can change the nature of interference.

PACS Nos. 42.50Gy, 42.50Hz

Single optical Transition

$$\mathrm{Im}\ \chi(\omega) = L_{W_0}(\Delta_1)$$

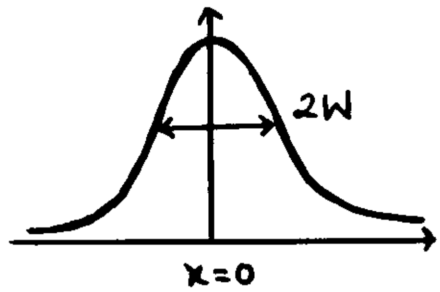
L

Large G

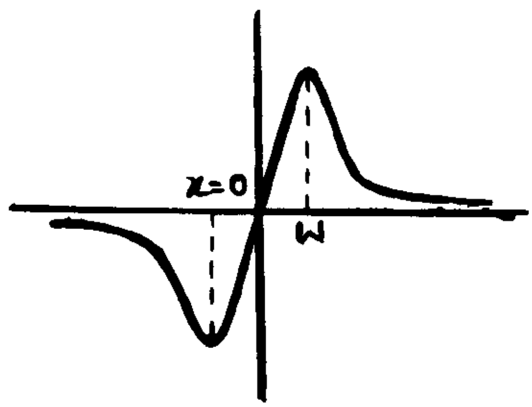
$$Im\chi(\omega) = \frac{1}{2}\left[L_W(\Delta_1 - G) + L_W(\Delta_1 + G)\right] + \frac{\beta}{G}\cdot\frac{1}{2}\left[D_W(\Delta_1 - G) - D_W(\Delta_1 + G)\right]$$

$2G$ is Rabi frequency, Δ_1 is probe detuning, β is the interference parameter.

$$L_W(x) = \frac{W/\pi}{x^2+W^2} \longrightarrow \frac{W}{\pi x^2}$$



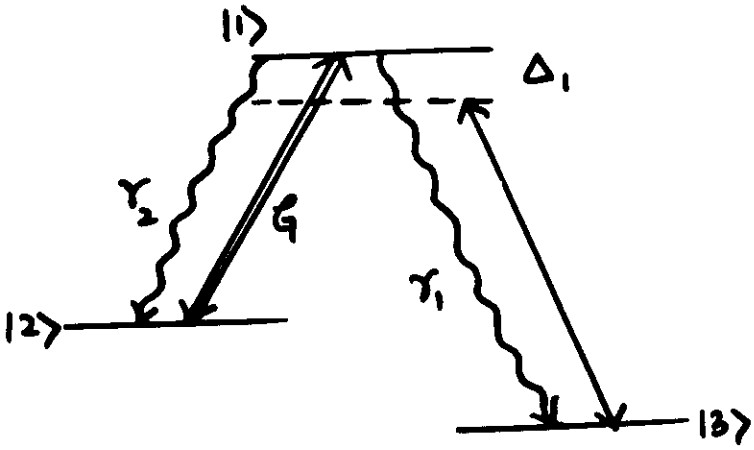
$$D_W(x) = \frac{x/\pi}{x^2+W^2} \longrightarrow \frac{1}{\pi x}$$



$$Im\chi(\omega) \sim \tfrac{1}{2}L_W(\Delta_1 - G)$$

$$Im\chi(\omega) = \frac{W-\beta}{G^2}$$

at line center in the absence of Control Laser



$$W = \frac{1}{2}(\Gamma_{13} + \Gamma_{23})$$

$$\beta = \frac{(\Gamma_{13} - \Gamma_{23})}{2}$$

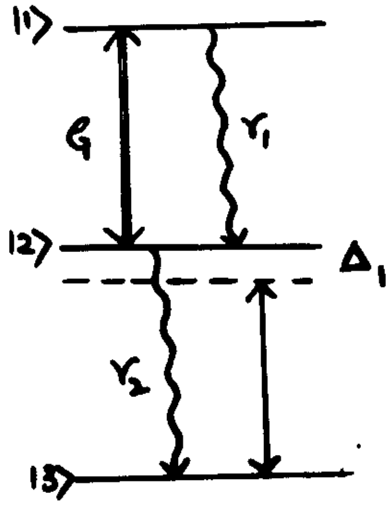
NO DEPHASING
i.e. $\Gamma_{23} = 0$

ABSOLUTE ZERO
IN ABSORPTION

DESTRUCTIVE
INTERFERENCE

NO INTERFERENCE
IF $\Gamma_{13} = \Gamma_{23}$

EVEN IF $\Gamma_{13} >> \Gamma_{23}$,
NEAR ZERO
ABSORTION



$$W = \frac{1}{2}(\Gamma_{13} + \Gamma_{23})$$

$$\beta = \frac{(\Gamma_{23} - \Gamma_{13})}{2}$$

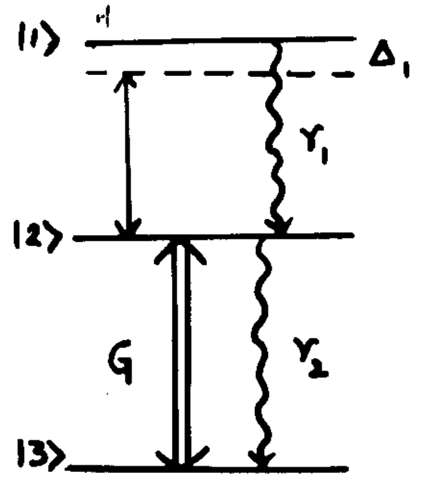
EVEN IF $\Gamma_{23} = 0$,
DEPHASING
DEPENDS ON γ_1

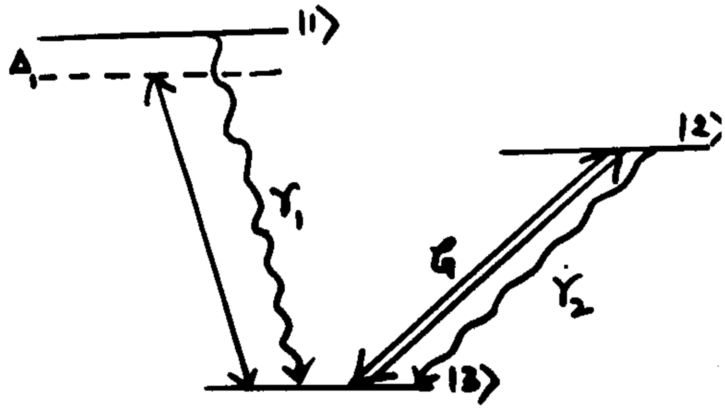
NEAR ZERO
ABSORPTION IF
γ_1 IS SMALL

DESTRUCTIVE
INTERFERENCE

NO INTERFERENCE
IF $\Gamma_{13} = \Gamma_{23}$

β DEPENDS ON
RELATIVE
MAGNITUDES
OF γ_1 AND γ_2.





$$W = \frac{1}{2}(\Gamma_{13} + \Gamma_{23}) \rightarrow \gamma_1 + \frac{\gamma_2}{2}$$

$$\beta = \frac{(\Gamma_{12} - \Gamma_{13} - 2\gamma_2)}{2} \rightarrow -\frac{\gamma_2}{2}$$

WITHOUT
DEPHASING

ABSORPTION
PROFILE DEPENDS
ON γ_2

CONSTUCTIVE
INTERFERENCE

β IS NEGATIVE

NO INTERFERENCE
IF $\gamma_2 \rightarrow 0$

$$W = \frac{1}{2}(\Gamma_{13} + \Gamma_{23}) \rightarrow \gamma_1 + \frac{\gamma_2}{2}$$

$$\beta = \frac{(\Gamma_{13} - \Gamma_{12} - 2\gamma_2)}{2} \rightarrow -\frac{3\gamma_2}{2}$$

WITHOUT
DEPHASING

ABSORPTION
PROFILE DEPENDS
ON $\gamma_2\left(\frac{G^2}{G^2 + \gamma_2\Gamma_{23}}\right)$

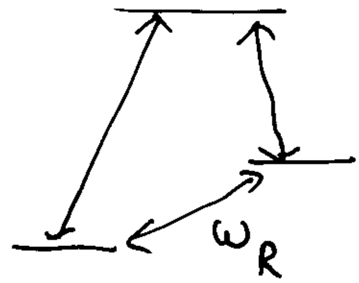
CONSTRUCTIVE
INTERFERENCE

β IS NEGATIVE

NO INTERFERENCE
IF $\gamma_2 \rightarrow 0$

Λ system

NORMAL CASE



$\omega_R \gg$ Any relevant Frequencies, γ's, Rabi Freq

what if ω_R is comparable to "Rabi"

Assumption that each field interacts with just one transition : breaks down

NEWER TYPES OF INTERFERENCES!

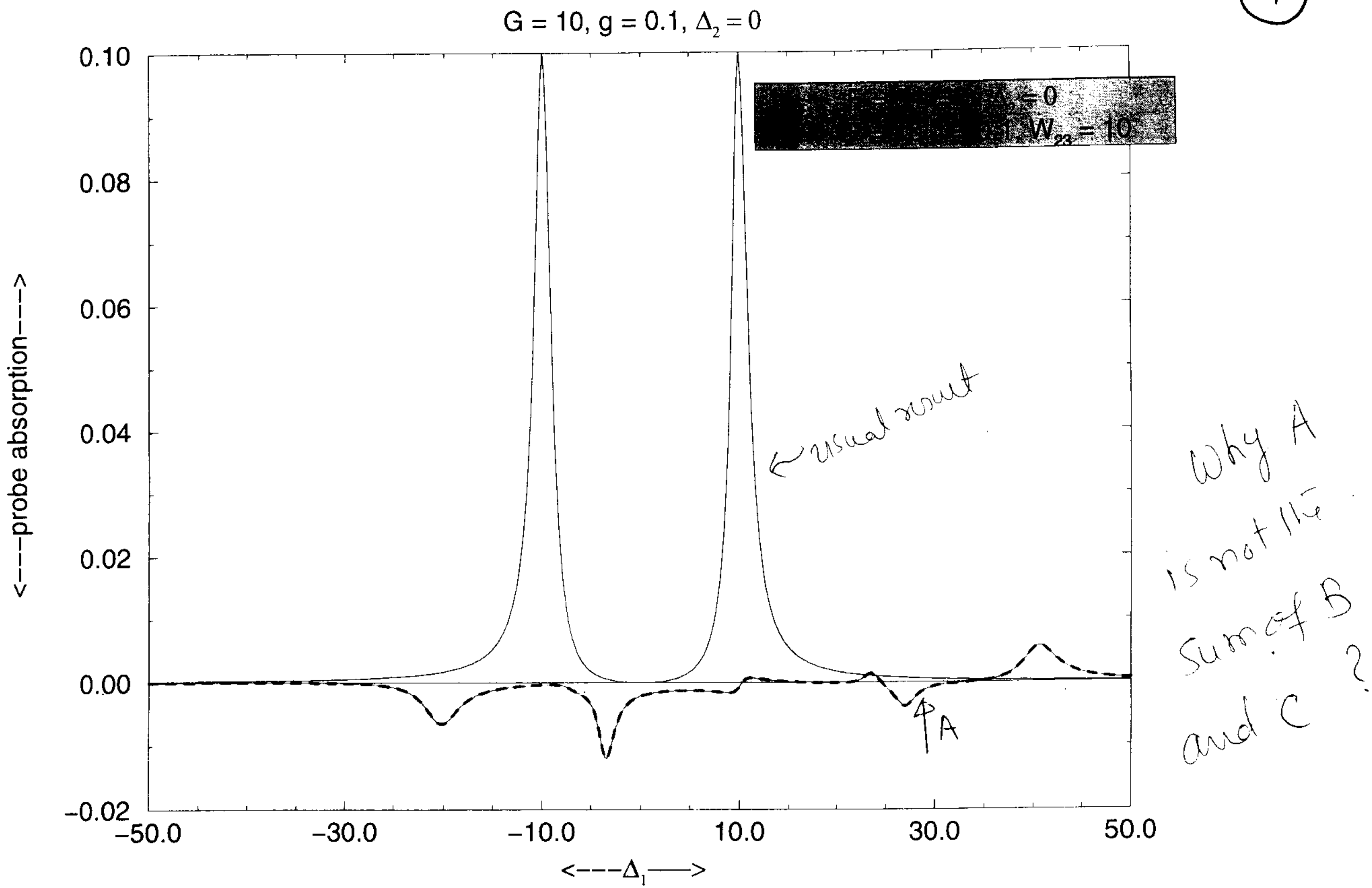
In K : Ground state Hyperfine $\sim$ 450 MHz

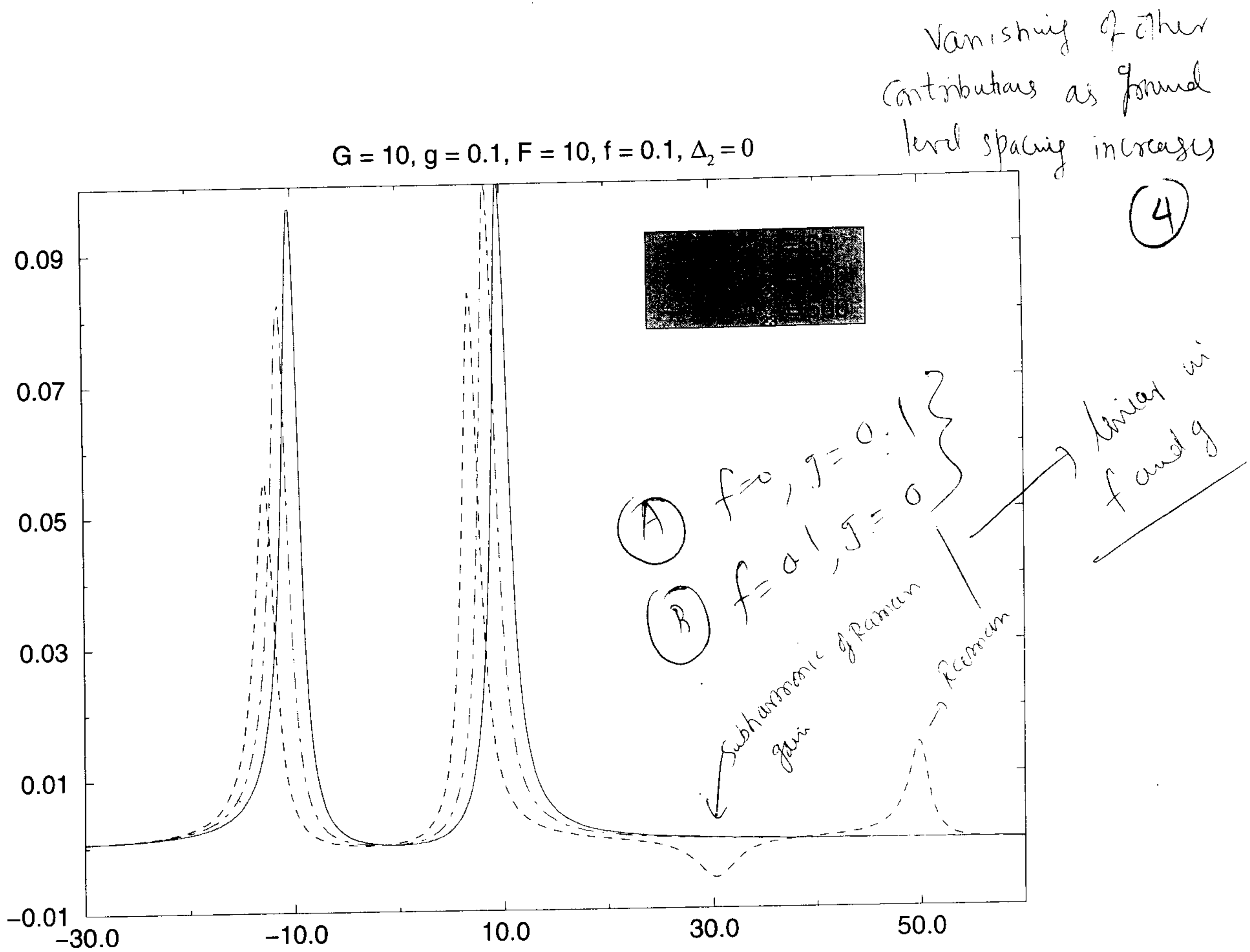
which can be easily comparable to the Rabi Freqs —

(S. MENON)

can get even "gain" in regions where normally one will have absorption — OPENS UP

(1)


G = 10, g = 0.1, $\Delta_2 = 0$
<---probe absorption--->
0.10
0.08
0.06
0.04
0.02
0.00
-0.02
-50.0
-30.0
-10.0
10.0
30.0
50.0
<---Δ_1--->
← usual result
A
Why A is not the sum of B and C ?



Vanishing of other contributions as formed level spacing increases
(4)
G = 10, g = 0.1, F = 10, f = 0.1, $\Delta_2 = 0$
[redacted]
(A) f=0, g=0.1
(B) f=0.1, g=0
linear in f and g
Subharmonic of Raman gain
Raman
0.09
0.07
0.05
0.03
0.01
−0.01
−30.0
−10.0
10.0
30.0
50.0

Inhibition and Enhancement of Two Photon Absorption

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The possibility of transparency against two photon absorption is predicted. Detailed absorption profiles under different conditions of the control laser are given. A novel explanation of the absorption minimum is given in terms of the two photon Fermi golden rule and the dressed states. Possibility of considerable enhancement of two photon absorption even in the presence of Doppler broadening is demonstrated. [S0031-9007(96)00798-3]

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It is well known [1] from second order perturbation theory [2] that the two photon absorption can exhibit interference minimum depending on the location of the intermediate states. The occurrence of such interferences depends on the existence of at least two intermediate states, and on a special relationship between the dipole matrix elements and detunings. Thus the interference minimum in two photon absorption is determined by the intrinsic properties of the medium. In this Letter we propose a method whereby this interference minimum can be *induced as well as controlled* by changing intensity and frequency of the electromagnetic field especially applied to achieve such an objective. One thus has the possibility of making the medium transparent against two photon absorption. We further demonstrate the enhancement of two photon absorption. Our analysis also includes Doppler broadening. Clearly this control of two photon absorption should be of importance in the context of related issues like two photon lasing and pulse propagation, which will be discussed elsewhere. Further, one has the possibility of *enhancing* $\chi^{(2)}$ of media possessing small permanent dipole moment or media with strong magnetic dipole transitions [3]. Here $\chi^{(2)}$ would essentially be proportional to two photon coherence.

Consider the situation shown in Fig. 1. The probability of absorption of two photons according to the perturbation theory is given by

$$P_2 = \frac{2\pi}{\hbar}\left|\frac{\langle 1|\vec{d}\cdot\vec{\mathcal{E}}_1|2\rangle\langle 2|\vec{d}\cdot\vec{\mathcal{E}}_2|3\rangle}{E_2 - E_3 - \hbar\omega_2} + \frac{\langle 1|\vec{d}\cdot\vec{\mathcal{E}}_1|2'\rangle\langle 2'|\vec{d}\cdot\vec{\mathcal{E}}_2|3\rangle}{E_{2'} - E_3 - \hbar\omega_2}\right|^2 \times \delta(E_1 - E_3 - \hbar\omega_1 - \hbar\omega_2). \quad (1)$$

The absorption probability vanishes if the corresponding two photon matrix element vanishes. This will happen for a value of ω_2 determined by the dipole matrix elements and the location of the intermediate states. Clearly, if there is only *one* intermediate state, then the two photon absorption always occurs. In this case we can use a control laser to couple the intermediate state with some other state to make the medium transparent against two photon absorption. The idea of using control lasers in modifying the optical properties [4] has met with tremendous success since the early proposals. Harris and co-workers [5] introduced the idea of electromagnetic field induced transparency which in the meantime has been the subject of several experiments [6,7]. It has also been shown that control field induced quantum interference and field induced transparency effects can be utilized for enhancing nonlinear optical cross sections [8], for decreasing the threshold of switching in optical bistability [9], for lasing without inversion [10], and for enhancing refractive index [11]. These ideas have also been used for quenching spontaneous emission noise [12].

We present a model scheme to show how the idea of two photon transparency will work. We illustrate the idea by considering the example of the relevant energy levels of Rb. The transitions are shown in Fig. 1. The 2γ's represent the rates of spontaneous decay. The field on the transition $|2\rangle \leftrightarrow |4\rangle$ is the control field and will be shown

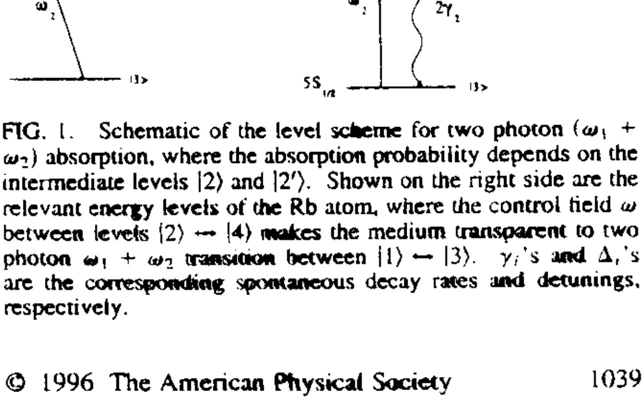


FIG. 1. Schematic of the level scheme for two photon ($\omega_1 + \omega_2$) absorption, where the absorption probability depends on the intermediate levels $|2\rangle$ and $|2'\rangle$. Shown on the right side are the relevant energy levels of the Rb atom, where the control field ω between levels $|2\rangle \leftrightarrow |4\rangle$ makes the medium transparent to two photon $\omega_1 + \omega_2$ transition between $|1\rangle \leftrightarrow |3\rangle$. γ_i's and Δ_i's are the corresponding spontaneous decay rates and detunings, respectively.

to create transparency against two photon absorption. Let Δ's represent various detunings: $\Delta_1 = \omega_{12} - \omega_1$, $\Delta_2 = \omega_{23} - \omega_2$, $\Delta = \omega_{42} - \omega$, and G's denote the coupling coefficients $G_1 = \vec{d}_{12} \cdot \vec{\mathcal{E}}_1/\hbar$, $G_2 = \vec{d}_{23} \cdot \vec{\mathcal{E}}_2/\hbar$, $G = \vec{d}_{42} \cdot \vec{\mathcal{E}}/\hbar$. After a canonical transformation the effective Hamiltonian is

$$\begin{aligned} H = {} & \hbar\Delta_2|2\rangle\langle 2| + \hbar(\Delta_1 + \Delta_2)|1\rangle\langle 1| \\ & + \hbar(\Delta + \Delta_2)|4\rangle\langle 4| - \hbar(G|2\rangle\langle 4| \\ & + G_1|1\rangle\langle 2| + G_2|2\rangle\langle 3| + \text{H.c.}) . \end{aligned} \tag{2}$$

The probability for two photon absorption is given by the population in the state $|1\rangle$. We will assume, as usual, that G_1 and G_2 are weak, whereas the control field G can be of *arbitrary* magnitude. Thus ρ_{11} needs to be calculated to the order $G_1^2G_2^2$, but to all orders in G. This can be done using density matrix equations with *spontaneous emission* terms properly included

$$\begin{aligned} \dot{\rho} = {} & -\frac{i}{\hbar}[H,\rho] - \gamma_1\{|1\rangle\langle 1|,\rho\} - \gamma_2\{|2\rangle\langle 2|,\rho\} \\ & - \gamma\{|4\rangle\langle 4|,\rho\} + 2\gamma_1\rho_{11}|2\rangle\langle 2| \\ & + 2\gamma_2\rho_{22}|3\rangle\langle 3| + 2\gamma\rho_{44}|2\rangle\langle 2| . \end{aligned} \tag{3}$$

It should be borne in mind that we are dealing with a *four level* system. Calculations show that the two photon excitation of the level $|1\rangle$ consists of *two* terms—one containing the two photon resonant denominator $(\gamma_1 + i\Delta_1 + i\Delta_2)$ and the other corresponding to stepwise excitation. Assuming large intermediate state detuning, the dominant term in two photon excitation is the one containing the two photon resonant denominator, i.e.,

$$\begin{aligned} I_c = {} & \frac{G_1^2G_2^2}{\gamma_1}\,\mathrm{Im}\frac{i(\gamma + i\Delta + i\Delta_2)}{(\gamma_1 + i\Delta_1 + i\Delta_2)[(\gamma_2 + i\Delta_2)(\gamma + i\Delta + i\Delta_2) + G^2]} \\ & \times \frac{\gamma_1 + \gamma + i\Delta_1 - i\Delta}{(\gamma_1 + \gamma_2 + i\Delta_1)(\gamma_1 + \gamma + i\Delta_1 - i\Delta) + G^2} . \end{aligned} \tag{4}$$

In the limit $G \to 0$, $\Delta_i \gg \gamma_i$,

$$I_c \Rightarrow \frac{G_1^2G_2^2}{(-\Delta_1\Delta_2)[\gamma_1^2(\Delta_1 + \Delta_2)^2]} . \tag{5}$$

For large Δ_i's and $G \neq 0$, Eq. (4) leads to the possibility of a *minimum* in two photon absorption corresponding to

$$\Delta_1 = -\Delta_2 = \Delta . \tag{6}$$

Before analyzing the origin of the minimum we present a number of two photon absorption profiles obtained from the numerical solution of the density matrix equations (3). Unlike (4) the numerical work includes *all* the contributions. We will present the results both *with* and without *Doppler* broadening. We will assume that the fields ω_1 and ω_2 are counterpropagating but the *control field is copropagating* with ω_1. The Doppler averaging is done in the usual manner by replacing ω_i by $\omega_i - \vec{k}_i \cdot \vec{v}$, etc. We show the *peak* of the two photon absorption, i.e., ρ_{11} at $\Delta_1 + \Delta_2 = 0$, as a function of the detuning Δ_1. Figure 2 gives the behavior for different values of intensity and frequency of the control laser. We scale all detunings and field Rabi frequencies in terms of γ's. For the purpose of argument the actual values of individual γ's are irrelevant. For comparison results in the absence of a control laser are also shown. Note the minima in two photon absorption at $\Delta_1 = 20$ for $G = 50$. The linewidth of the D_2 line in Rb is about 6 MHz, and thus $\Delta_1 = 20$ corresponds to 60 MHz. Further a pump Rabi frequency $2G$ of about 100γ will correspond roughly to a power level of 1 W assuming a beam diameter of order 0.5 mm (intensity ≈ 500 W/cm^2) and thus providing one with adequate power. A variety of lasers such as Ti:sapphire can be used as coupling lasers [7]. For control laser strength $G > \Delta\omega_D$ we have a similar behavior with a minima at Δ (not shown). Notice the asymmetries in the two photon excitation spectra. These arise from the finite detuning of the control laser. Note further that one member of the doublet is *narrower* [13] than the Doppler width. We show in Fig. 3 the behavior of the minimum

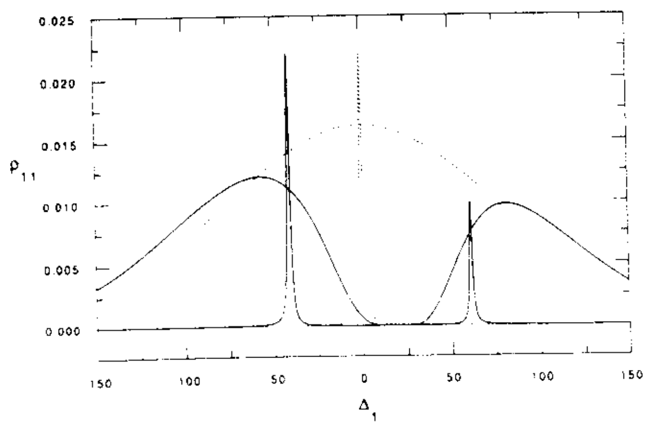
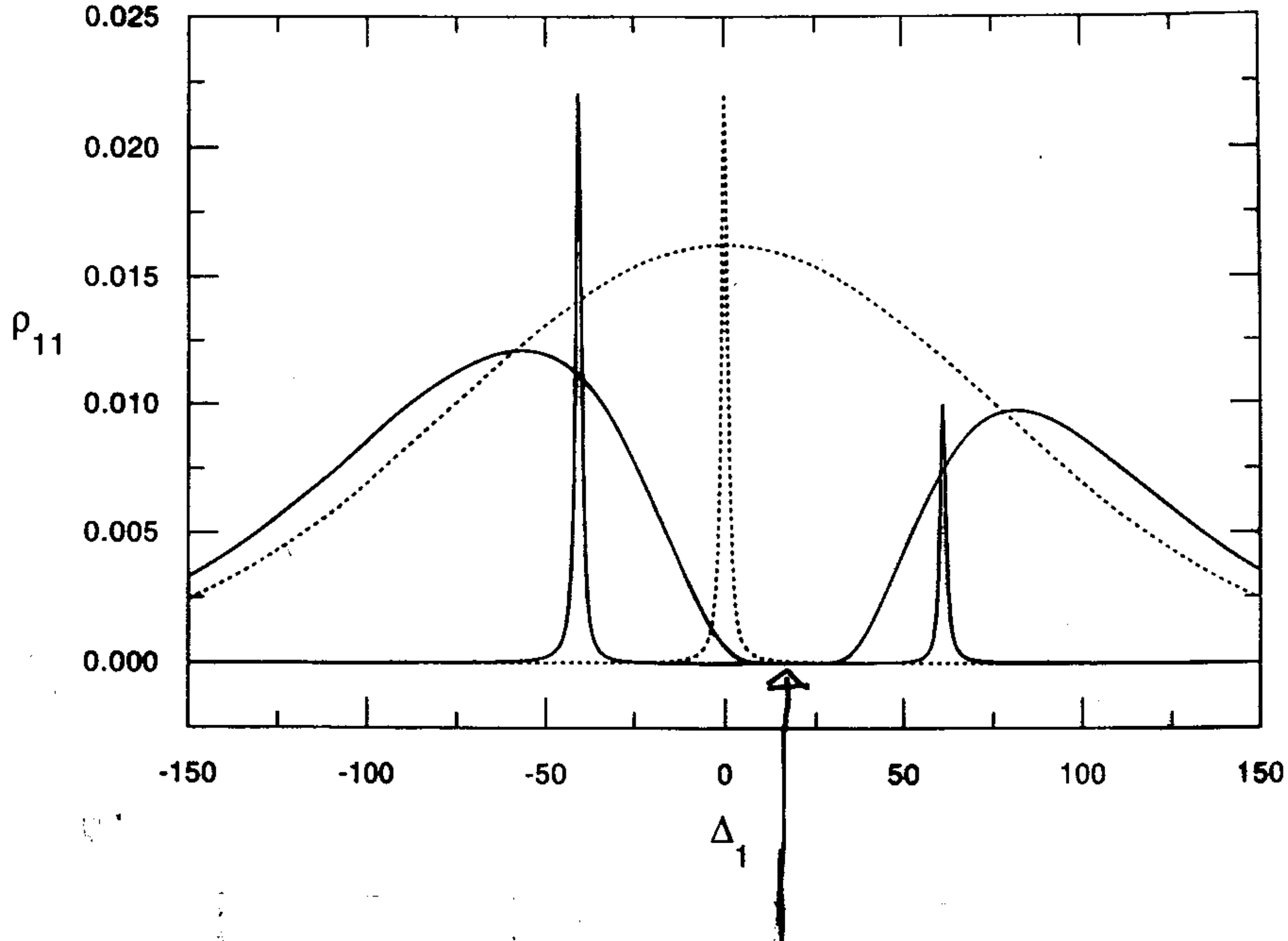
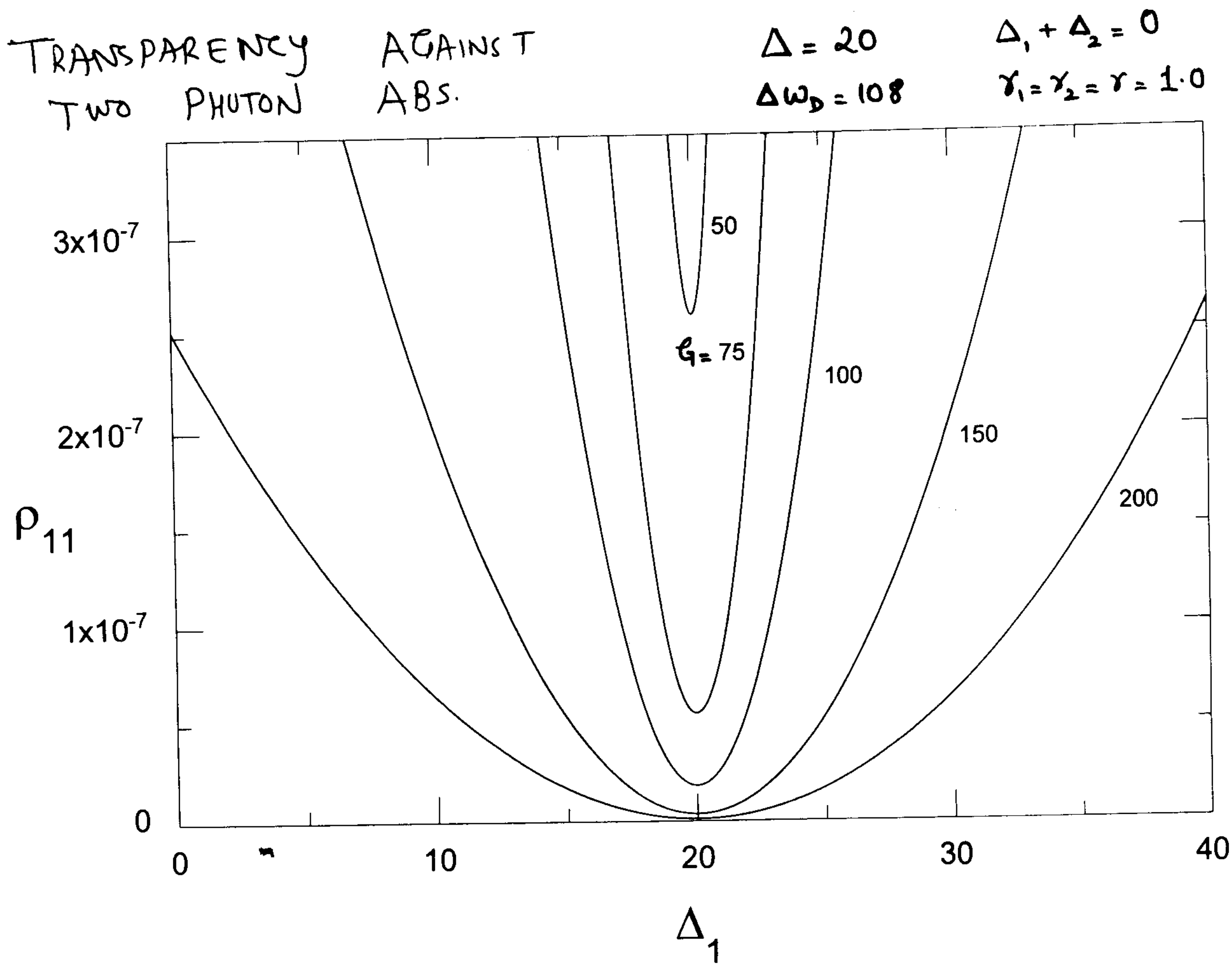
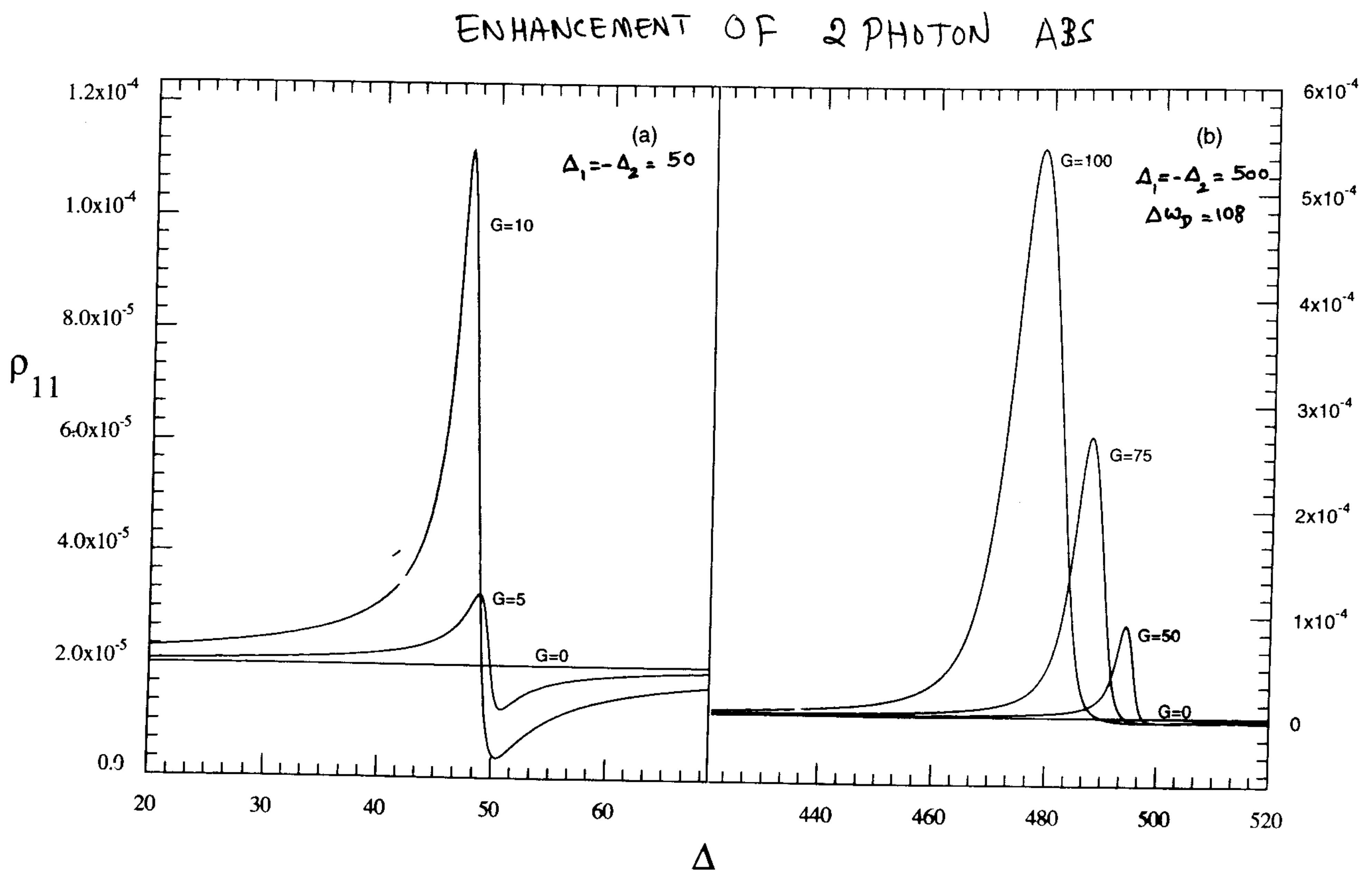


FIG. 2. The two photon absorption spectra at $\Delta_1 + \Delta_2 = 0$. The dashed curves are in the absence of the control field, the one with much larger width is in the presence of Doppler width $\Delta\omega_D/2\gamma\sqrt{ln 2} = 108$ (the actual number is 45 times less). The solid curves are in the presence of a control field $G = 50$, $\Delta = 20$. The Doppler result in this case is 20 times less than shown. All γ_i's have been set to unity.

TWO PHOTON ABS WITH COUPLING LASER APPLIED ON INTERMEDIATE STATE




TRANSPARENCY AGAINST
TWO PHOTON ABS.
Δ = 20
Δω_D = 108
Δ_1 + Δ_2 = 0
γ_1 = γ_2 = γ = 1.0
ρ_11
3x10^-7
2x10^-7
1x10^-7
0
50
G = 75
100
150
200
0
10
20
30
40
Δ_1

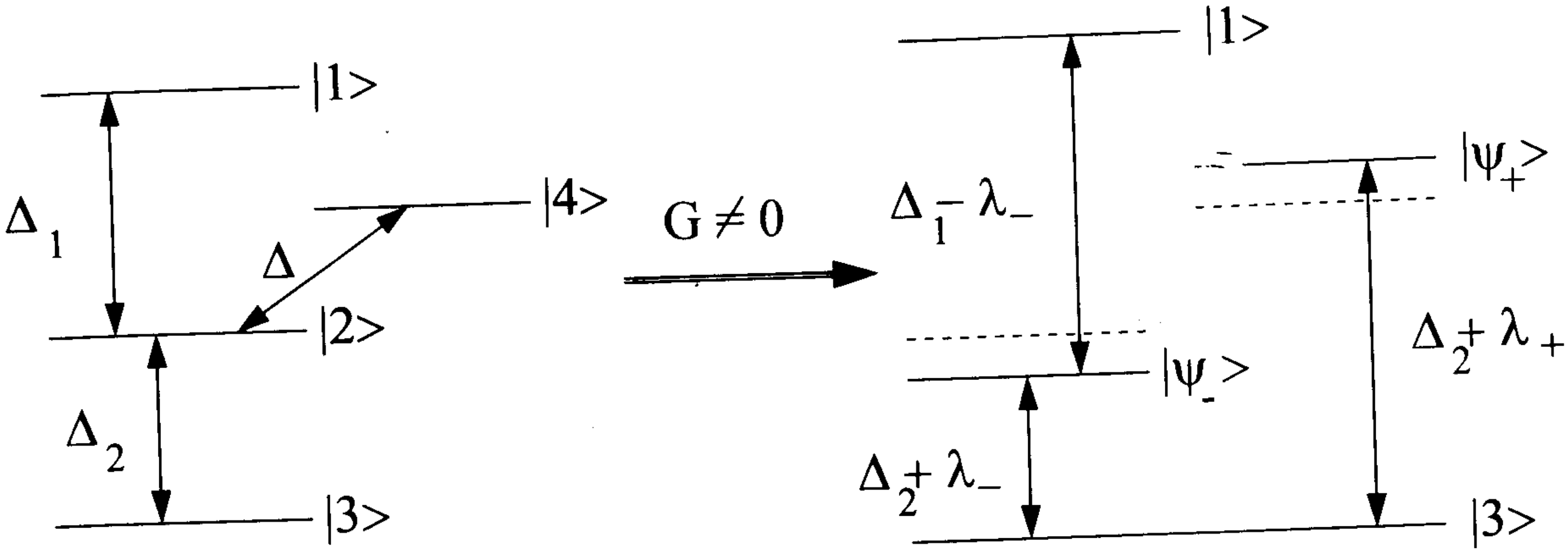

ENHANCEMENT OF 2 PHOTON ABS
(a)
Δ1=-Δ2 = 50
(b)
Δ1=-Δ2 = 500
Δωp =108
G=10
G=5
G=0
G=100
G=75
G=50
G=0
ρ11
Δ
1.2x10-4
1.0x10-4
8.0x10-5
6.0x10-5
4.0x10-5
2.0x10-5
0.0
6x10-4
5x10-4
4x10-4
3x10-4
2x10-4
1x10-4
0
20
30
40
50
60
440
460
480
500
520

$$H = \hbar\,\Delta_2 |2\rangle\langle 2| + \hbar(\Delta_1+\Delta_2)|1\rangle\langle 1| + \hbar(\Delta+\Delta_2)|4\rangle\langle 4|$$

(BARE STATES)

$$H_{Total} = \hbar\,\Delta_2|2\rangle\langle 2| + \hbar(\Delta_1+\Delta_2)|1\rangle\langle 1| + \hbar(\Delta+\Delta_2)|4\rangle\langle 4| - \hbar\left(G|2\rangle\langle 4| + G_1|1\rangle\langle 2| + G_2|2\rangle\langle 3| + H.C.\right)$$

$G_1, G_2 \ll G$



$$\langle 1|\vec{d}|\psi_\pm\rangle\langle\psi_\pm|\vec{d}|3\rangle \Rightarrow \frac{\langle 1|\vec{d}|2\rangle\langle 2|\vec{d}|3\rangle}{2\sqrt{1+x^2}\left(\sqrt{1+x^2}\pm x\right)}$$

$$x = \frac{\Delta}{2G}$$

CONTROL LASER INDUCED SUB DOPPLER SPECTROSCOPY

PHYSICAL REVIEW A VOLUME 53, NUMBER 4 APRIL 1996

Sub-Doppler resolution in inhomogeneously broadened media using intense control fields

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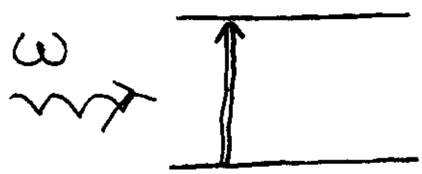
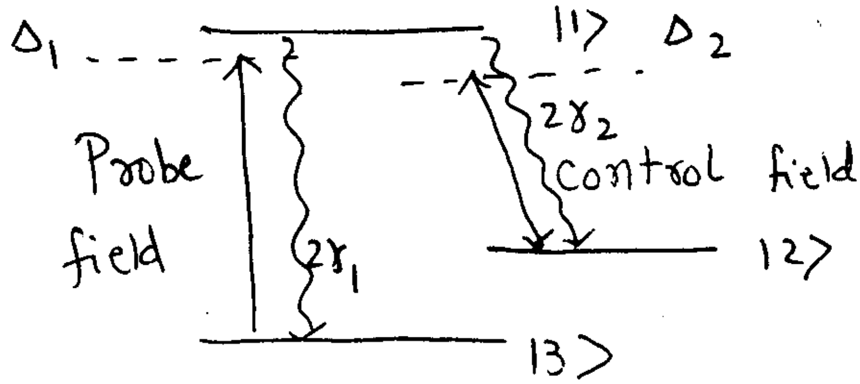
We propose a scheme for obtaining sub-Doppler resolution for one transition of an inhomogeneously broadened, three-level atomic system, by using an intense control field at the other transition. Analytical and numerical calculations are presented to delineate the mechanism responsible for this sub-Doppler resolution, and quantify the extent to which Doppler broadening can be reduced.



Weak field abs. Γ

Lorentzian → Gaussian

$$\exp\left\{-(\delta-\Delta)^2/2D^2\right\}$$



Line Profile $\propto \mathrm{Im}\left\{ (\Delta_1-\Delta_2) \left[|G|^2 - i(\gamma_1+\gamma_2 - i\Delta_1)(\Delta_1-\Delta_2) \right]^{-1} \right\}$

$\Delta_1 \to \Delta_1 + x$

$\Delta_2 \to \Delta_2 + x$ $\quad x = kv$

⟶ copropagating fields
⟶

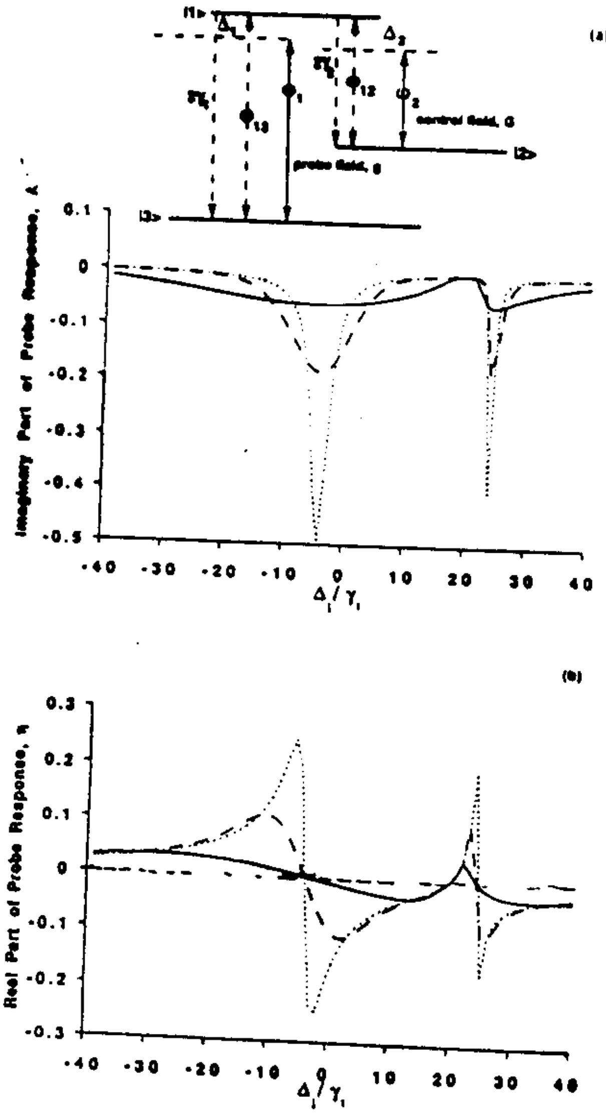


FIG. 1. (a) Probe absorption spectrum in a Λ system for Doppler widths D of 0.01 (dot), 5 (dash), and 20 (solid). Other parameters are $G=10\gamma_1$, $\Delta_2=20\gamma_1$, and $\gamma_2=\gamma_1$. Inset: Schematic representation of a three-level Λ system. The spontaneous decay rates from $|1\rangle$ to $|3\rangle$ and $|1\rangle$ to $|2\rangle$ are $2\gamma_1$ and $2\gamma_2$, respectively. ω_{12} and ω_{13} are the resonance frequencies of the two allowed transitions. (b) Real part of probe response in Λ system for parameters identical to (a).

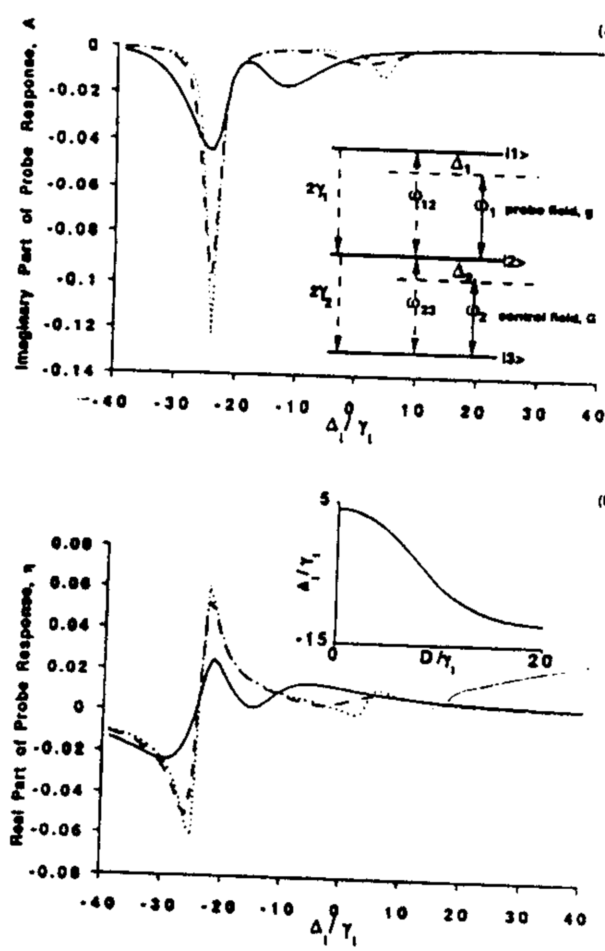


FIG. 2. (a) Probe absorption spectrum in a ladder system for Doppler widths D of 0.01 (dot), 5 (dash), and 20 (solid). Other parameters are $G=10\gamma_1$, $\Delta_2=20\gamma_1$, and $\gamma_2=\gamma_1$. Inset: Schematic representation of a three-level ladder system. The spontaneous decay rates from $|1\rangle$ to $|2\rangle$ and $|2\rangle$ to $|3\rangle$ are $2\gamma_1$ and $2\gamma_2$, respectively. ω_{12} and ω_{23} are the resonance frequencies of the upper and lower transitions, respectively. (b) Real part of the probe response in ladder system for parameters identical to (a). Inset: Value of Δ_1 at which maximum absorption occurs for the line at $\Delta_1=(-\Delta_2/2)+\frac{1}{2}\sqrt{\Delta_2^2+4|G|^2}$, as a function of D.

Doppler width affects
Two components differently

Pole structure ? x dependent

'x' random variable

sensitive to applied

Maxwellian distribution

field strength

Let $\Delta_2^2 + 4|G|^2 > D^2$

$$\Delta_1 = \frac{\Delta_2 - i(\gamma_1+\gamma_2) - x}{2} \pm \frac{\sqrt{\Delta_2^2+4|G|^2}}{2}\left\{1 + \frac{i\Delta_2(\gamma_1+\gamma_2 - ix)}{\Delta_2^2+4|G|^2}\right\}$$

"Fluctuating Poles" (random matrix)

$\langle x \rangle = 0$, $\langle x^2 \rangle = \langle (kv)^2 \rangle = D^2$

Fluctuation in $\Delta_1 \Rightarrow$ line width.

$$\frac{\gamma_1+\gamma_2+D}{2}\left(1 \mp \frac{\Delta_2}{\sqrt{\Delta_2^2+4|G|^2}}\right)$$

RESOLUTION WITHIN HOMO-- LINEWIDTH

TRAP EXPS

$\Delta_2 = 0$; identical $D/2$

$\neq 0$ $(\ ) \sim \begin{matrix}1.7 \\ 0.3\end{matrix}$ etc

HYSICAL REVIEW A VOLUME 54, NUMBER 4 OCTOBER 1996

Sub-Doppler linewidth with electromagnetically induced transparency in rubidium atoms

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We report the experimental observation of steady-state electromagnetically induced transparency and sub-Doppler linewidth in a Doppler-broadened Λ-type Rb atomic system formed on the ^{87}Rb D_2 transitions. The Λ system is coupled on one transition by a strong laser and probed on the other transition by a weak laser. The observed sub-Doppler linewidth decreases with the increasing detuning of the strong-coupling laser from the atomic transition frequency and is in good agreement with the theoretical calculations of G. Vemuri, G. S. Agarwal, and B. D. N. Rao [Phys. Rev. A 53, 2842 (1996)]. [S1050-2947(96)10810-6]

PACS number(s): 42.50.Gy, 42.50.Fx, 42.50.Md

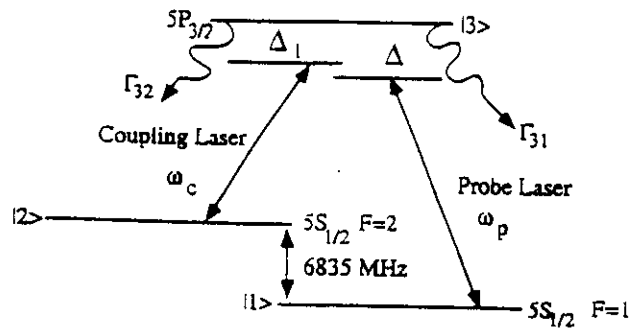


FIG. 1. The energy level structure of ^{87}Rb D_1 transition and the laser coupling scheme, which forms an effective three-level Λ-type sytem. $\Delta_1 = \omega_c - \omega_{32}$ ($\Delta = \omega_p - \omega_{31}$) is the coupling (probe) laser detuning. Γ_{31} (Γ_{32}) is the spontaneous decay rate from state $|3\rangle$ to state $|1\rangle$ ($|2\rangle$).

our theoretical prediction

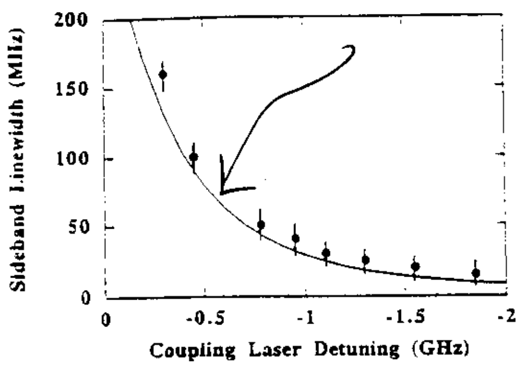


FIG. 5. Measured (dots) and calculated [from Eq. (2), solid line] linewidths of the probe absorption line at the probe detuning $\Delta = \Delta_+ = \Delta_1/2 + \frac{1}{2}\sqrt{\Delta_1^2 + 4\Omega^2}$ versus the probing detuning Δ_1. For the experimental measurements, the coupling laser power was about 600 mW. The parameters used in Eq. (2) are $\Gamma_{31} + \Gamma_{32} = 6$ MHz, $D = 540$ MHz, and $\Omega = 250$ MHz.

our amplifier result
Vemuri, Agarwal + Rao, PR A54,

Y. ZHU + J. LIN

PR A 53, 1768 (1996)

$\Delta = -\Omega' \approx \Delta_1 = -1.3$ GHz exhibits the sub-Doppler line widths that approach the Rb natural linewidth. For example, the measured linewidth for the gain feature in curves 1 and 2 is ~ 10 MHz (about equal to the Rb natural linewidth of 6 MHz after convolution of the laser linewidths of a few MHz for the Ti:sapphire laser and the probe laser).

was kept at $\Delta_1 = -1.3$ GHz. The probe gain was observed when the pump power was greater than 2 mW (the estimated pump Rabi frequency is about 30 MHz). As the pump power increases, the probe gain increases accordingly and is maximized at the pump power of about 75 mW (the estimated Rabi frequency is about 150 MHz) as shown by curves 1–3. The probe gain decreases for further increases of the pump power, and the gain profile changes into a dispersive shape as shown by curve 4. At even higher pump intensities, the probe spectral feature at $\Delta \approx \Delta_1$ exhibits a broadened absorption line profile (see curve 5). Overall, the gain spectral feature at

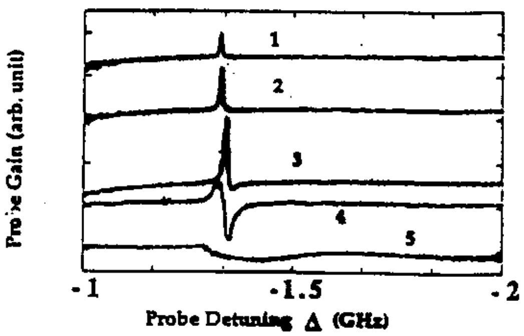
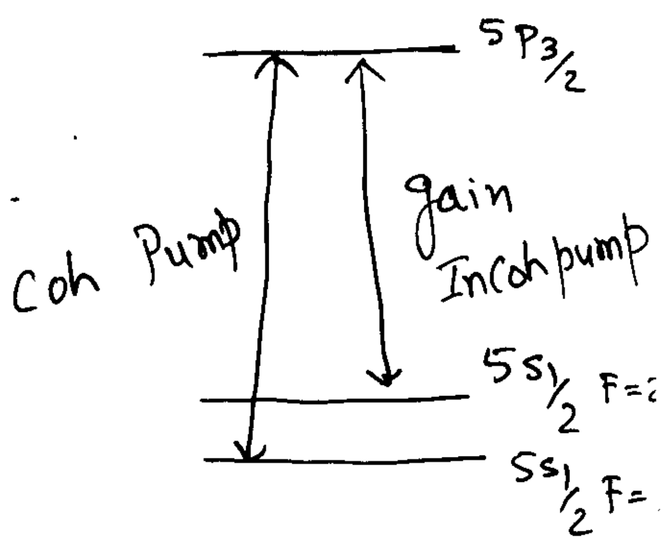


FIG. 8. The measured probe gain spectra near the ^{87}Rb $5S_{1/2}$ $(F=2) \leftrightarrow 5P_{3/2}$ transition with several different coupling-laser intensities (for clarity, different curves have been vertically displaced). The pump laser was tuned to the center of the ^{87}Rb $5S_{1/2}$ $(F=2) \leftrightarrow 5P_{3/2}$ transition and the coupling laser was detuned from the ^{87}Rb $5S_{1/2}$ $(F=1) \leftrightarrow 5P_{3/2}$ transition by $\Delta_1 = -1.3$ GHz. Curves 1–5 correspond to the pump powers 7, 23, 75, 235, and 750 mW, respectively. As the pump intensity increases, the probe spectral line shape changes from amplification to dispersion, then to absorption. The linewidth increases from near the natural linewidth (10 MHz) for the gain feature at lower intensities to a Doppler-broadened width for the absorption feature at higher intensities.



LWI Case : Include Incoh pumping Λ

Γ_p = probe laser linewidth
Γ_c = control

$$\text{width} = \frac{\gamma_1+\gamma_2+\Gamma_p+2\Lambda+D}{2}\left(1-\frac{\Delta_2}{\sqrt{\Delta_2^2+4G^2}}\right)$$

< 2 MHz

$$+\ \frac{\Gamma_c+\Gamma_p+\Lambda}{2}\left(1+\frac{\Delta_2}{\sqrt{\Delta_2^2+4G^2}}\right) +$$

$\hookrightarrow \sim(\Gamma_c+\Gamma_p+\Lambda)$

$\sim$ 10 MHz observed line width.

Λ = 1 MHz

Rabi = 120 MHz

Δ_2 = 1.4 GHz

G. VEMURI, G.S. AGARWAL, BDN RAO,
Phys Rev. A54 ~~in press~~
3695 Oct 1996

QUANTUM INTERFERENCES AND SUB-NATURAL LINEWIDTHS IN SPONTANEOUS EMISSION

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OUTLINE

- EARLY EXAMPLES OF QUANTUM INTERFERENCES IN EMISSION
- SUBNATURAL LINE WIDTHS – SIZE OF DETUNING & STRENGTH OF CONTROL LASER
- QUENCHING OF SPONTANEOUS EMISSION AT LINE CENTER – ORIGIN – DISPERSIVE CONTRIBUTION
- APPLICATION OF TWO PHOTON FERMI-GOLDEN RULE FOR SE QUENCHING (MULTIPHOTON, IONIZATION PROCESSES)
- LINE WIDTH SUM RULE
- ELECTROMAGNETIC FIELD INDUCED DOUBLE POLES (etc) in RESONANT SCATTERING – NEW LINE SHAPES

SPRINGER TRACTS IN MODERN PHYSICS

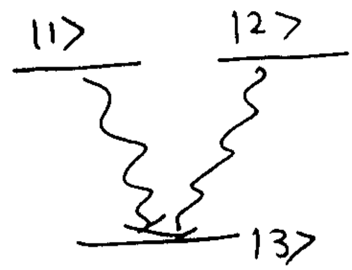
Quantum Statistical Theories of Spontaneous Emission and their Relation to Other Approaches

G. S. Agarwal

Ergebnisse der exakten Naturwissenschaften

Volume 70

Springer-Verlag Berlin Heidelberg New York 1974



B) Three-Level Atom with Degenerate Spectrum

Consider now a three-level atom whose levels $|1\rangle$ and $|2\rangle$ are degenerate so that the possible transitions are $|1\rangle \to |3\rangle$, $|2\rangle \to |3\rangle$. The matrix elements of the density operator are found to satisfy equations (ignoring the Lamb shift terms)

$$\partial\varrho_{11}/\partial t = -\kappa\{2\mu_1\varrho_{11} + \mu\varrho_{21} + \mu^*\varrho_{12}\}, \tag{15.30a}$$

$$\partial\varrho_{22}/\partial t = -\kappa\{2\mu_2\varrho_{22} + \mu\varrho_{21} + \mu^*\varrho_{12}\}, \tag{15.30b}$$

$$\partial\varrho_{21}/\partial t = -\kappa(\mu_1 + \mu_2)\varrho_{21} - \kappa\mu^*(\varrho_{11} + \varrho_{22}), \tag{15.30c}$$

$$\begin{aligned} \partial\varrho_{31}/\partial t &= -\kappa\{\mu_1\varrho_{31} + \mu^*\varrho_{32}\}, \\ \partial\varrho_{32}/\partial t &= -\kappa\{\mu_2\varrho_{32} + \mu\varrho_{31}\}, \end{aligned} \tag{15.30d}$$

$\dot{\varrho}_{11} = -(\ )\,\varrho_{11}$

where

$$\kappa = \tfrac{2}{3}(\omega/c)^3, \quad \mu_1 = |d_{13}|^2, \quad \mu_2 = |d_{23}|^2, \quad \mu = d_{13}\cdot d_{23}^*. \tag{15.30e}$$

Equation (15.30) are linear equations and can be solved easily. We consider here only the simplified situation when $\mu = \mu_1 = \mu_2$ and then, defining $\gamma = \kappa\mu$, we obtain the equations

$$\begin{aligned} \partial\varrho_{11}/\partial t &= -\gamma(2\varrho_{11} + \varrho_{12} + \varrho_{21}), \\ \partial\varrho_{22}/\partial t &= -\gamma(2\varrho_{22} + \varrho_{21} + \varrho_{12}), \\ \partial\varrho_{31}/\partial t &= -\gamma(\varrho_{31} + \varrho_{32}) \\ \partial\varrho_{32}/\partial t &= -\gamma(\varrho_{32} + \varrho_{31}), \\ \partial\varrho_{21}/\partial t &= -2\gamma\varrho_{21} - \gamma(\varrho_{11} + \varrho_{22}), \end{aligned} \tag{15.31}$$

which admit a constant of integration

$$\varrho_{11} + \varrho_{22} - \varrho_{12} - \varrho_{21} = \alpha. \tag{15.32}$$

$$\varrho_{11}(t) = \tfrac{1}{4}(1 + e^{-4\gamma t}) + \tfrac{1}{2}e^{-2\gamma t},$$
$$\varrho_{22}(t) = \tfrac{1}{4}(1 + e^{-4\gamma t}) - \tfrac{1}{2}e^{-2\gamma t},$$
$$\varrho_{12}(t) = \varrho_{21}(t) = -\tfrac{1}{4}(1 - e^{-4\gamma t}),$$

implying that in the steady state

$$\varrho_{11}(\infty) = \varrho_{22}(\infty) = \tfrac{1}{4},$$
$$\varrho_{33}(\infty) = \tfrac{1}{2},$$
$$\varrho_{12}(\infty) = \varrho_{21}(\infty) = -\tfrac{1}{4}. \qquad (15.34)$$

Coherence (in ρ_{11}) effects not seen to $O(\gamma t)$

This behavior (15.34) may seem somewhat surprising because one would expect that in the steady state the atom would remain in the ground state. On the other hand, for the initially symmetric excitation

$$\varrho(0) = |\psi\rangle\langle\psi|, \quad |\psi\rangle = 2^{-\frac{1}{2}}(|1\rangle + |2\rangle),$$
$$\varrho_{11}(0) = \varrho_{22}(0) = \varrho_{12}(0) = \varrho_{21}(0) = \tfrac{1}{2},$$

we find

$$\varrho_{11}(t) = \varrho_{22}(t) = \varrho_{12}(t) = \varrho_{21}(t) = \tfrac{1}{2}e^{-4\gamma t},$$

and hence in the steady state

$$\varrho_{11} = \varrho_{22} = \varrho_{12} = \varrho_{21} = 0,$$
$$\varrho_{33} = 1, \qquad (15.35)$$

and the atom is left in the ground state. The steady state behavior can also be discussed in rather general terms (see also Appendix C). For the case we are discussing the interaction with the radiation field is through a combination of the atomic operators:

$$S^+ = 2^{-\frac{1}{2}}(A_{13} + A_{23}), \quad S^- = 2^{-\frac{1}{2}}(A_{31} + A_{32}),$$
$$S^z = \tfrac{1}{4}\{A_{22} + A_{11} - 2A_{33} + A_{12} + A_{21}\}. \qquad (15.36)$$

It can be shown that these operators satisfy the same commutation relations as the spin-$\frac{1}{2}$ operators, i.e.

$$S^+S^+ = S^-S^- = 0, \quad S^+S^z = -S^zS^+ = -\tfrac{1}{2}S^+, \quad S^zS^z = \tfrac{1}{4} \text{ etc.}$$

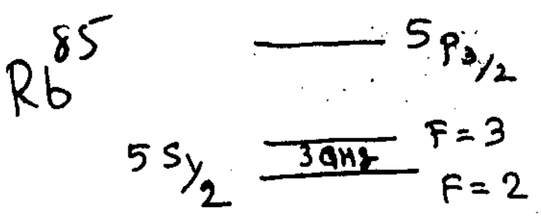
The eigenstates of S^z are given by

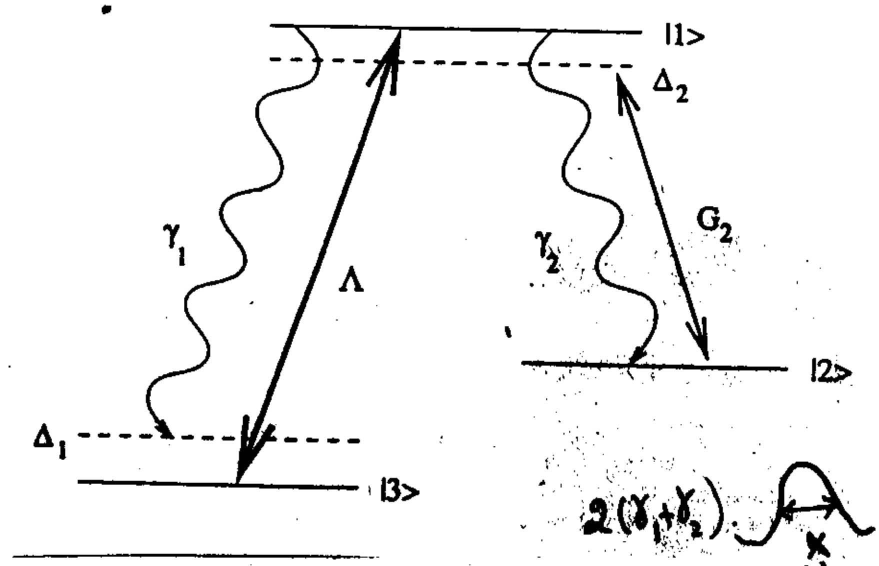
$$S^z|\pm\rangle = \pm\tfrac{1}{2}|\pm\rangle, \quad |+\rangle = 2^{-\frac{1}{2}}(|1\rangle + |2\rangle), \quad |-\rangle = |3\rangle. \qquad (15.37)$$

Since the coupling in the interaction Hamiltonian is via the operators defined by (15.36), one would expect the system to be found in the ground state of S^z only if it is prepared at time $t = 0$ in a state which is the linear combination of the states $|\pm\rangle$ as defined by (15.37). From this we see again that the symmetric state will decay to the ground state $|3\rangle$, whereas the unsymmetrized state (15.33) could result in different behavior as it cannot be expressed as a linear combination of $|\pm\rangle$.

Analog of what is currently called coupled and uncoupled states.

MODEL





$2G_2$ is the Rabi frequency of the control laser of frequency ω_l, 2Λ is the pumping, $2\gamma_1$ and $2\gamma_2$ are respectively the rates of spontaneous emission on the two transitions. The spectrum of spontaneous emission on the transition $|1\rangle \leftrightarrow |3\rangle$ is known to be related to the two time correlation function

$$S(\omega) \equiv \int_0^\infty d\tau \langle A_{13}(t+\tau)A_{31}(t)\rangle e^{i\Delta_1\tau} + c.c., \tag{1}$$

where $\Delta_1 = \omega_{13} - \omega$ and $A_{\alpha\beta}(t) \equiv |\alpha\rangle\langle\beta|$ is the dipole moment operator for the transition $|\alpha\rangle \leftrightarrow |\beta\rangle$. The correlation function in (1) can be calculated using the usual density matrix equations and the quantum regression theorem. After a canonical transformation so as to eliminate the optical frequencies, the density matrix equations read

$$\begin{aligned}
\dot\rho_{11} &= -2(\gamma_1+\gamma_2+\Lambda)\rho_{11} + 2\Lambda\rho_{33} + iG_2\rho_{21} - iG_2^*\rho_{12} \\
\dot\rho_{22} &= 2\gamma_2\rho_{11} - iG_2\rho_{21} + iG_2^*\rho_{12} \\
\dot\rho_{21} &= -(\Gamma_{21} - i\Delta_2)\rho_{21} - iG_2^*\rho_{22} + iG_2^*\rho_{11} \\
\dot\rho_{31} &= -\Gamma_{31}\rho_{31} - iG_2^*\rho_{32} \\
\dot\rho_{32} &= -(\Gamma_{32} + i\Delta_2)\rho_{32} - iG_2\rho_{31}.
\end{aligned} \tag{2}$$

where $\Gamma_{\alpha\beta}$ denotes the total decay rate of the off diagonal element $\rho_{\alpha\beta}$. For our model

$$\Gamma_{31} \equiv \gamma_1 + \gamma_2 + 2\Lambda,\ \Gamma_{32} = \Lambda,\ \Gamma_{21} = (\gamma_1 + \gamma_2 + \Lambda),\ \Delta_2 = \omega_{12} - \omega_l. \tag{3}$$

The relevant equations for the calculation of (1) are

$$\left\{ \frac{d}{d\tau} + \begin{pmatrix} \Gamma_{31} & iG_2^* \\ iG_2 & \Gamma_{32} + i\Delta_2 \end{pmatrix} \right\} \begin{pmatrix} \langle A_{13}(t+\tau)A_{31}(t)\rangle \\ \langle A_{23}(t+\tau)A_{31}(t)\rangle \end{pmatrix} = 0. \tag{4}$$

which are to be solved subject to initial conditions

$$\langle A_{13}A_{31}\rangle = \rho_{11}, \langle A_{23}A_{31}\rangle = \rho_{12}. \tag{5}$$

Clearly the spectral characteristics of $S(\omega)$ will be determined by the zeros of the polynomial

$$P(z) = (z + \Gamma_{31})(z + \Gamma_{32} + i\Delta_2) + |G_2|^2. \tag{6}$$

i.e., by

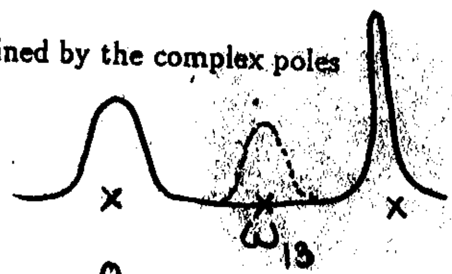
$$\begin{aligned} z_\pm = &-\frac{1}{2}(\Gamma_{31} + \Gamma_{32} + i\Delta_2) \\ &\pm \frac{1}{2}\sqrt{(\Gamma_{32} + i\Delta_2 - \Gamma_{31})^2 - 4|G_2|^2}. \end{aligned} \tag{7}$$

We analyse the roots in the limit of large Rabi frequency $\sqrt{4|G_2|^2 + \Delta_2^2} \equiv \Omega$. To leading order in Γ/Ω we can write the two roots of (6) as

$$\begin{aligned} z_\pm &\simeq -\frac{1}{2}(\Gamma_{31} + \Gamma_{32} + i\Delta_2) \pm \frac{1}{2}i\Omega \mp \frac{1}{2}(\Gamma_{31} - \Gamma_{32})\frac{\Delta_2}{\Omega} \\ &= \pm\frac{1}{2}i(\Omega \mp \Delta_2) - \frac{1}{2}\Gamma_{31}\left(1 \pm \frac{\Delta_2}{\Omega}\right) - \frac{1}{2}\Gamma_{32}\left(1 \mp \frac{\Delta_2}{\Omega}\right). \end{aligned} \tag{8}$$

Thus according to equation (1) the spectrum will be determined by the complex poles

$$-i\Delta_1 = z_\pm.$$



The resonances in the spectrum occur at

$\omega_{13} - \omega =$ $\Delta_1 = \mp\frac{1}{2}(\Omega \mp \Delta_2)$, → 0, $G \to 0$; → Δ_2 (Raman)

with half widths

$$\frac{\Gamma_{31}}{2}\left(1 \pm \frac{\Delta_2}{\Omega}\right) + \frac{\Gamma_{32}}{2}\left(1 \mp \frac{\Delta_2}{\Omega}\right); \tag{11}$$

Thus the peak at $\Delta_1 = (\Omega + \Delta_2)/2$ will exhibit a width which could be *much smaller than the natural line width if $\Delta_2 \gg 2G_2$ and if Λ is very small.* On the contrary, the peak at $\Delta_1 = -(\Omega - \Delta_2)/2$ is broadened beyond the homogeneous line width. For $\Delta_2 = 0$, both the peaks have identical widths $(\Gamma_{31} + \Gamma_{32})/2$ which for small rate of pumping reduces to *half* the width in the absence of the control laser [13].

$0.15\ \Gamma_{31}$ if $\Delta_2 = 20$, $G_2 = 10$

SUM RULE

$$\sum (\text{Half widths}) = (\Gamma_{31} + \Gamma_{32})$$

$$= (\gamma_1 + \gamma_2) \quad \text{for Spont Emission}$$

$$= \text{Half width of the Line for } G \to 0.$$

$$|\psi_1\rangle = N_1 \left[|2\rangle - \frac{G_2}{G_1} |3\rangle \right]$$

$$|\psi_2\rangle = N_2 \left[|1\rangle + \frac{G_2^*}{\lambda_2} |2\rangle + \frac{G_1^*}{\lambda_2} |3\rangle \right]$$

$$|\psi_3\rangle = N_3 \left[|1\rangle + \frac{G_2^*}{\lambda_3} |2\rangle + \frac{G_1^*}{\lambda_3} |3\rangle \right]$$

N's normalizations

$$N_\ell \equiv \left(1 + \frac{|G_1|^2 + |G_2|^2}{\lambda_\ell^2} \right)^{-1/2}; \quad \ell = 2, 3$$

$$N_2^2 + N_3^2 = 1$$

$|\psi_2\rangle \to |1\rangle$ decay $2(\gamma_1 + \gamma_2) N_2^2$

$|\psi_3\rangle \to |1\rangle$ $2(\gamma_1 + \gamma_2) N_3^2$

Net $\underline{2(\gamma_1 + \gamma_2)}$ O.K.

B. Quantum Interferences— Quenching by Dispersive Contributions

After having discussed the possibilities of the subnatural widths, we discuss the actual spectrum of emission. We then demonstrate quenching of spontaneous emission in the region $\Delta_1 \sim 0$. For simplicity, we consider the case Δ_2=0. The calculation shows that

$$S(\omega) = 2\rho_{11}\mathrm{Re}\{\frac{(-i\Delta_1 + \Gamma_{32} + \gamma_2)}{(\Gamma_{31} - i\Delta_1)(\Gamma_{32} - i\Delta_1) + |G_2|^2}\}; \tag{12}$$

The total spectrum thus consists of a *sum of Lorentzian and dispersive contributions*. On using (3) in (13) we get

$G_2 \gg \Gamma_{ij}$

$$S(\omega)/\rho_{11} = \{\frac{\frac{1}{2}(\gamma_1 + \gamma_2 + 3\Lambda)}{(\Delta_1 + G_2)^2 + \left(\frac{\gamma_1+\gamma_2+3\Lambda}{2}\right)^2} + ...\}$$
$$+ \frac{(\gamma_2 - \gamma_1 - \Lambda)}{2G_2}\{\frac{\Delta_1 + G_2}{(\Delta_1 + G_2)^2 + \left(\frac{\gamma_1+\gamma_2+3\Lambda}{2}\right)^2} - ...\}, \tag{14}$$

where '...' represents terms with $G_2 \to -G_2$ and where

$$\rho_{11} \equiv \Lambda\,|\,G_2\,|^2\,\{3\Lambda\,|\,G_2\,|^2 + \gamma_1\,|\,G_2\,|^2 + \gamma_2\Lambda\,(\gamma_1 + \gamma_2 + \Lambda)\}^{-1}. \tag{15}$$

Note that in the region $\Delta_1 = G_2$, the dispersive contribution is unimportant and the spectrum is well approximated by a single line with width $(\gamma_1 + \gamma_2 + 3\Lambda)$. However for the region $\Delta_1 \sim 0$, all four contributions in (14) are equally important. The last two dispersive contributions are the interferences. At the line center Δ_1=0, the contribution of two Lorentzians is

$$L = \frac{(\gamma_1 + \gamma_2 + 3\Lambda)}{G_2^2}. \tag{16}$$

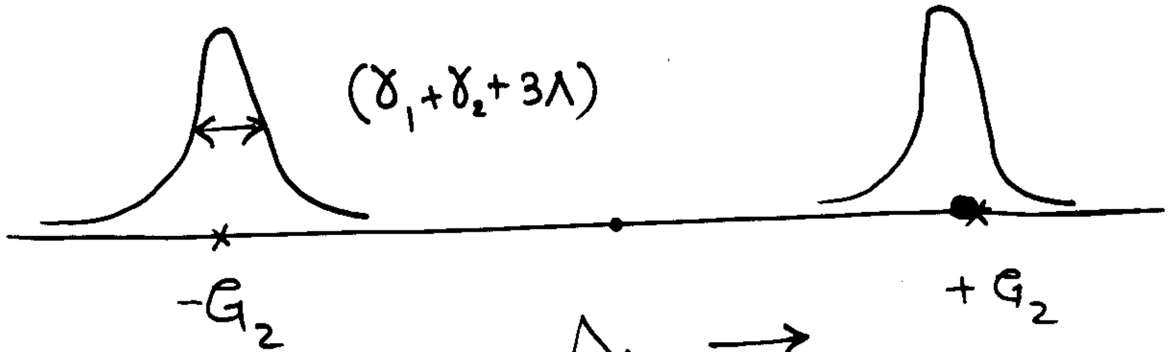
The interference terms from dispersive contributions lead to

$$D = \frac{(\gamma_2 - \gamma_1 - \Lambda)}{G_2^2}. \tag{17}$$

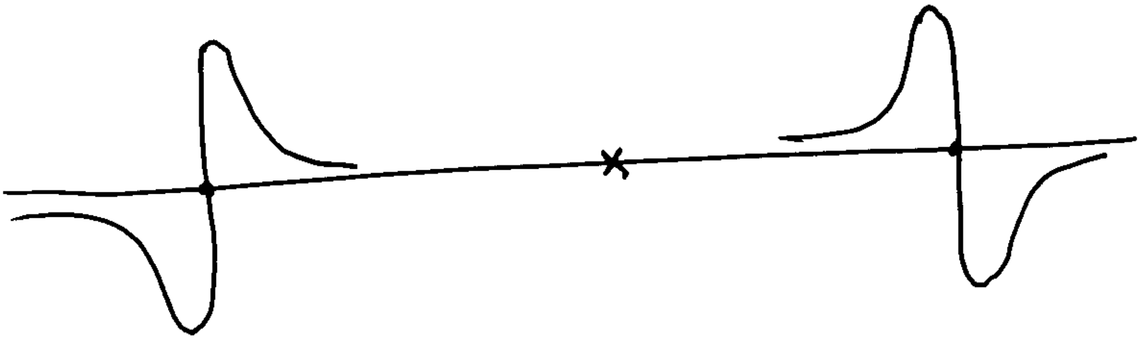
This can be constructive or destructive depending on the sign of $(\gamma_2 - \gamma_1 - \Lambda)$. The quenching of noise occurs if

$$\gamma_1 + \Lambda > \gamma_2. \tag{18}$$

The contribution (16) is what one would expect from a simple argument based on detuning *i.e.*, on the line wings.



weight $\boxed{1}$



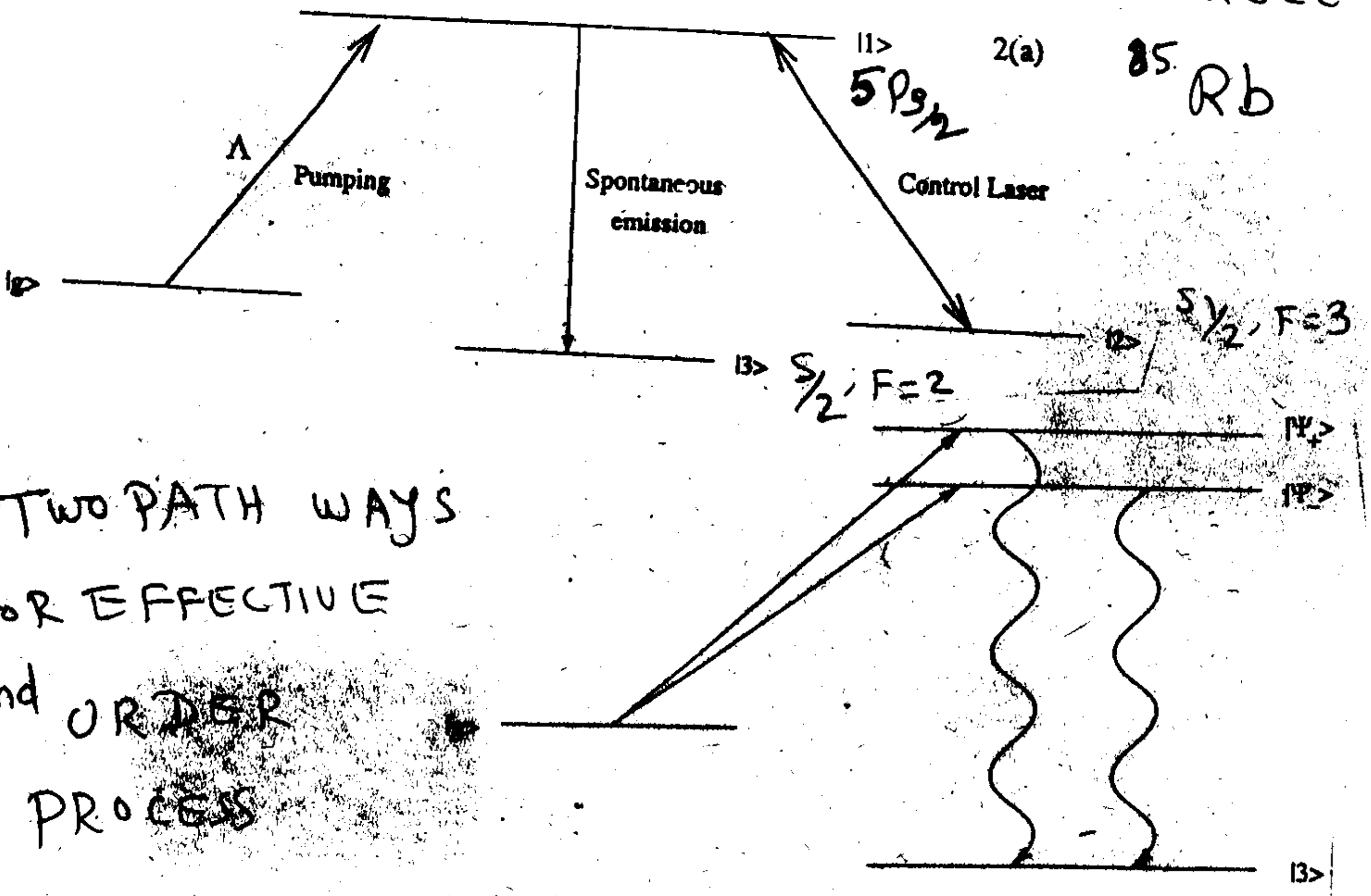
weight $\left(\frac{\gamma_2-\gamma_1-\Lambda}{2G_2}\right)$ $\Rightarrow$

Region $\underline{\Delta_1 \sim -G_2}$: only Lorentzian centered at $-G_2$ significant

Region $\Delta_1 \sim 0$ <u>all four</u> contributions imp.

Net $\dfrac{(\gamma_1+\gamma_2+3\Lambda) + (\gamma_2-\gamma_1-\Lambda)}{G_2^2}$ ← Interference Term

DRESSED STATES + FERMI GOLDEN RULE



TWO PATH WAYS FOR EFFECTIVE 2nd ORDER PROCESS

Consider the somewhat simpler model shown in Fig. 2(a). Let the pump be a broad band source. In the dressed states created by the control laser the various pumping and emission processes become as shown in Fig. 2(b). The net Hamiltonian can be written in the form (all energies being measured from $|1\rangle$)

$$
\begin{aligned}
H &= -\hbar G_2\left(|1\rangle\langle 2|e^{-i\omega_l t} + |2\rangle\langle 1|e^{i\omega_l t}\right) + E_2|2\rangle\langle 2| \\
&\quad + E_g|g\rangle\langle g| + E_3|3\rangle\langle 3| + H_{0V} + H_{1V} + H_p, \\
H_p &= -\hbar g\left(|1\rangle\langle g|e^{-i\omega_g t} + h.c.\right) \\
H_{1V} &= -\left(|1\rangle\langle 3|\left(\vec{\mathcal{E}}_V.\vec{d}_{13}\right) + c.c.\right),
\end{aligned}
\tag{19}
$$

where H_{OV} and $\mathcal{E}_V$ are respectively the unperturbed energy and amplitude of the vacuum of electromagnetic field. The fast optical frequency ω_l can be eliminated by a rotating frame transformation and we can also introduce the dressed states $|\psi_\pm\rangle$. For simplicity, let us assume that $|E_2| = \hbar\omega_l$. In terms of dressed states ($|1\rangle = \alpha|\psi_+\rangle + \beta|\psi_-\rangle$) we get

$$
\begin{aligned}
H &= E_+|\psi_+\rangle\langle\psi_+| + E_-|\psi_-\rangle\langle\psi_-| + E_g|g\rangle\langle g| \\
&\quad + E_3|3\rangle\langle 3| + H_{OV} - \hbar g\left(|\alpha\psi_+ + \beta\psi_-\rangle\langle g|e^{-i\omega_g t} + h.c.\right) \\
&\quad - \left(|\alpha\psi_+ + \beta\psi_-\rangle\langle 3|\left(\vec{\mathcal{E}}_V.\vec{d}_{13}\right) + h.c.\right).
\end{aligned}
\tag{20}
$$

We can now calculate the rate R for absorption of a photon of frequency ω_g and emission of a photon of frequency ω_{ks}, and momentum $\vec{k}$, polarization s. This is a second order process and we use second order Fermi-Golden rule [12]

$$R = \frac{2\pi}{\hbar}\sum_{ks} \Big| \frac{\langle 1_{ks}, 3|H_{1V}|0,\psi_+\rangle\langle 0,\psi_+|H_p|g;0\rangle}{(E_+ - E_g - \hbar\omega_g)} + \frac{\langle 1_{ks}, 3|H_{1V}|0,\psi_-\rangle\langle 0,\psi_-|H_p|g;0\rangle}{(E_- - E_g - \hbar\omega_g)} \Big|^2 \times \delta(E_3 - \hbar\omega_g + \hbar\omega_{ks} - E_g). \quad (21)$$

where $|0\rangle$ and $|1_{ks}\rangle$ represent the states of electromagnetic field with zero and one photon respectively. The matrix element in (21) is to be modified in the resonance region by taking into account appropriate damping effects. For a broad band pump with pump energy spread $\mathcal{E}$ we have to average over all pump energies $\hbar\omega_g$

$$R = \frac{2\pi}{\hbar\mathcal{E}}\sum_{ks} \Big| \frac{\langle 1_{ks}, 3|H_V|0,\psi_+\rangle\langle \psi+,0|H_p|g,0\rangle}{E_+ - (E_3 + \hbar\omega_{ks})} + \frac{\langle 1_{ks}, 3|H_V|0,\psi_-\rangle\langle \psi-,0|H_p|g,0\rangle}{E_- - (E_3 + \hbar\omega_{ks})} \Big|^2, \quad (22)$$

spectrum

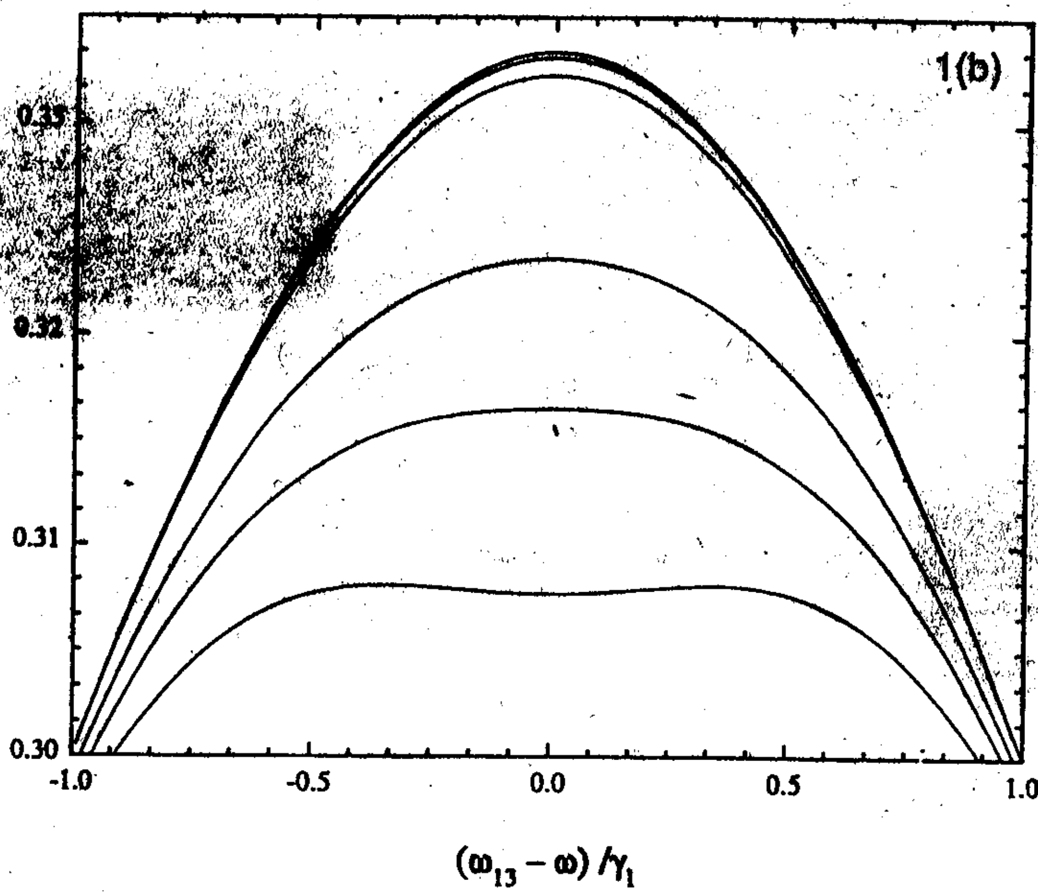
where various matrix elements are easily read from (20). Clearly the transition rate can be zero if the matrix element in (22) can *vanish*. This clearly can happen for appropriate frequency of the spontaneously emitted photon. Thus for a value of ω_{ks} which in general depends on the matrix elements, R can become zero. For the example under consideration, interference occurs if $E_3 = \hbar\omega_{ks}$ since $E_+ = -E_-$. Thus the flourescence as a function of ω_k will exhibit a minimum which comes about from the interference between two paths as shown in Fig. 2(b). Note further that the matrix element in (22) is just the Raman matrix element. The *cancellation arises* from the energy dependence of the Raman polarizability or the dispersive nature of the interaction. Thus the simple physical picture based on Fermi-Golden rule for second order process enables us to understand control laser induced interference effects and quenching of spontaneous emission.

Agarwal, Phys. Rev. A54, R3734 (1996)

EXPERIMENTS OF SUCKEWER ET AL

(J. Phys. B26, 4057 (1993))

We next examine the form of (12) in the limit $\gamma_2 \to 0$ and for *moderate values of G_2*. The results are shown in Fig. 1(b). We see that with increase in G_2 there is reduction in the peak height at $\Delta_1 = 0$. This trend is consistent with the observation of Suckewer and coworkers [10] on the quenching of spontaneous emission. The reduction further depends on relative values of G_2 and γ_1. Thus for a fix G_2 the transition with smaller γ_1 will be most affected. This is again consistent with the experimental observation [10,14].



$\gamma_2 \to 0$; $\frac{\Lambda}{\gamma_1} = 1.0$,

$G_2/\gamma_1 = 0,\ 0.05,\ 0.1,\ 0.3,$
0.4 and 0.5

Quenching *A*-coefficients by photons in a short discharge tube

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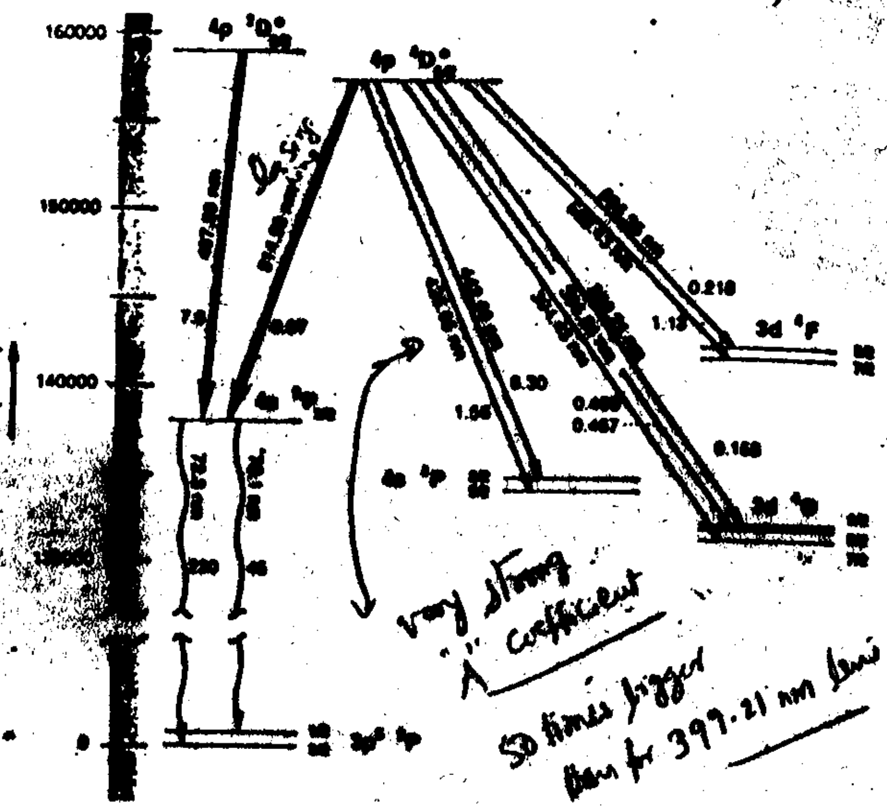


Figure 2. Partial Ar II energy-level diagram with transition probabilities in units of 10^7 s^{-1}.

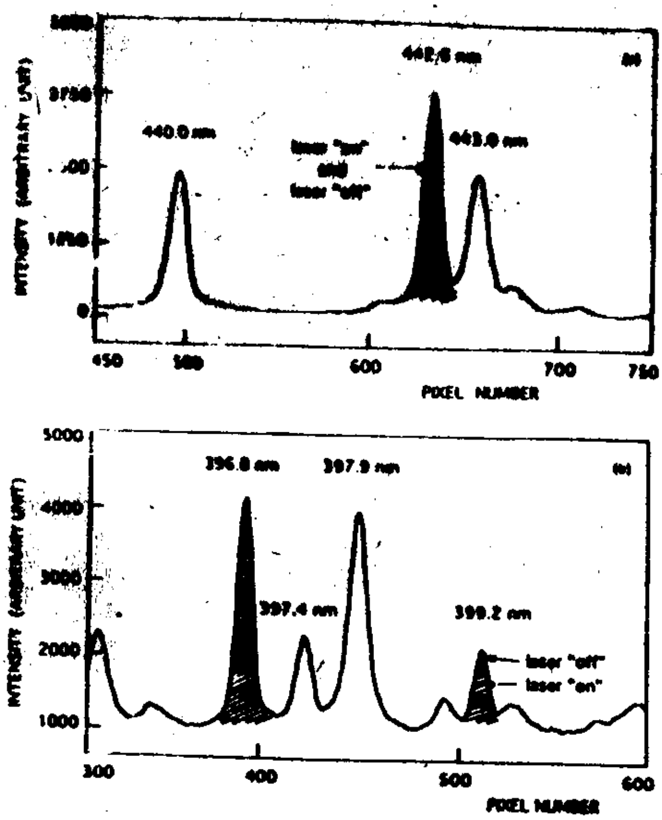


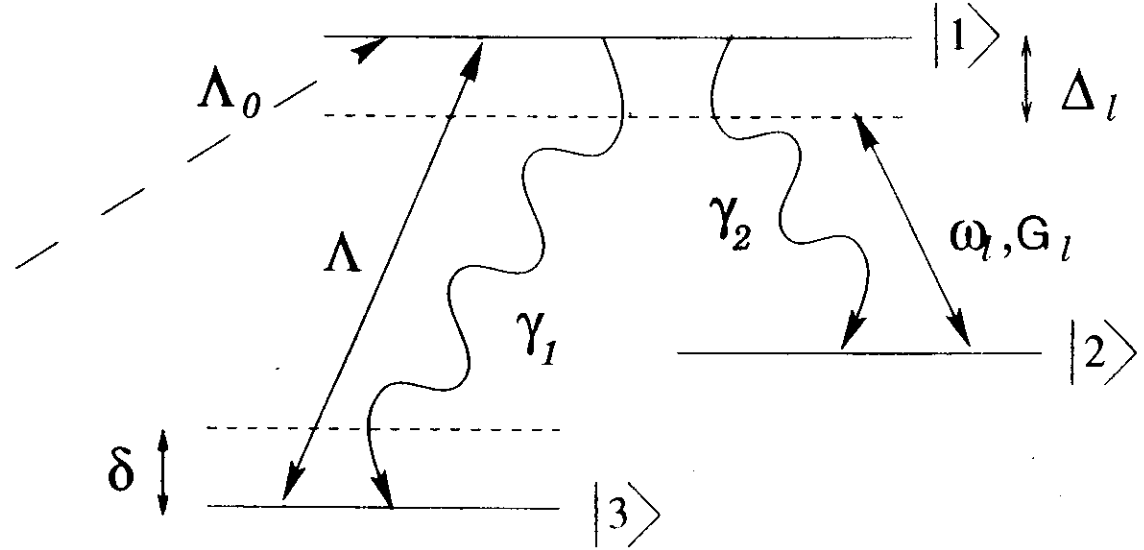
Figure 3. Spectra of spontaneous emission from the short discharge tube in the vicinity of the 399.2 nm line (*a*) and in the vicinity of the 442.6 nm line (*b*). Shaded areas of the lines are those related to the quenching effect when ...

GOLDBERGER, WATSON, KROLL, DRELL, BELL : DECAY OF UNSTABLE PARTICLES : HOW GENERAL IS EXPONENTIAL LAW IN "EXP" REALISABLE TIME DOMAIN

POLES OF S-MATRIX NOT NECESSARILY SIMPLE - MODELS WITH HIGHER ORDER POLES

SEVERAL EXACTLY SOLUBLE MODELS IN QUANTUM FIELD THEORY

MODIFICATIONS OF BREIT-WIGNER FORMULA ?

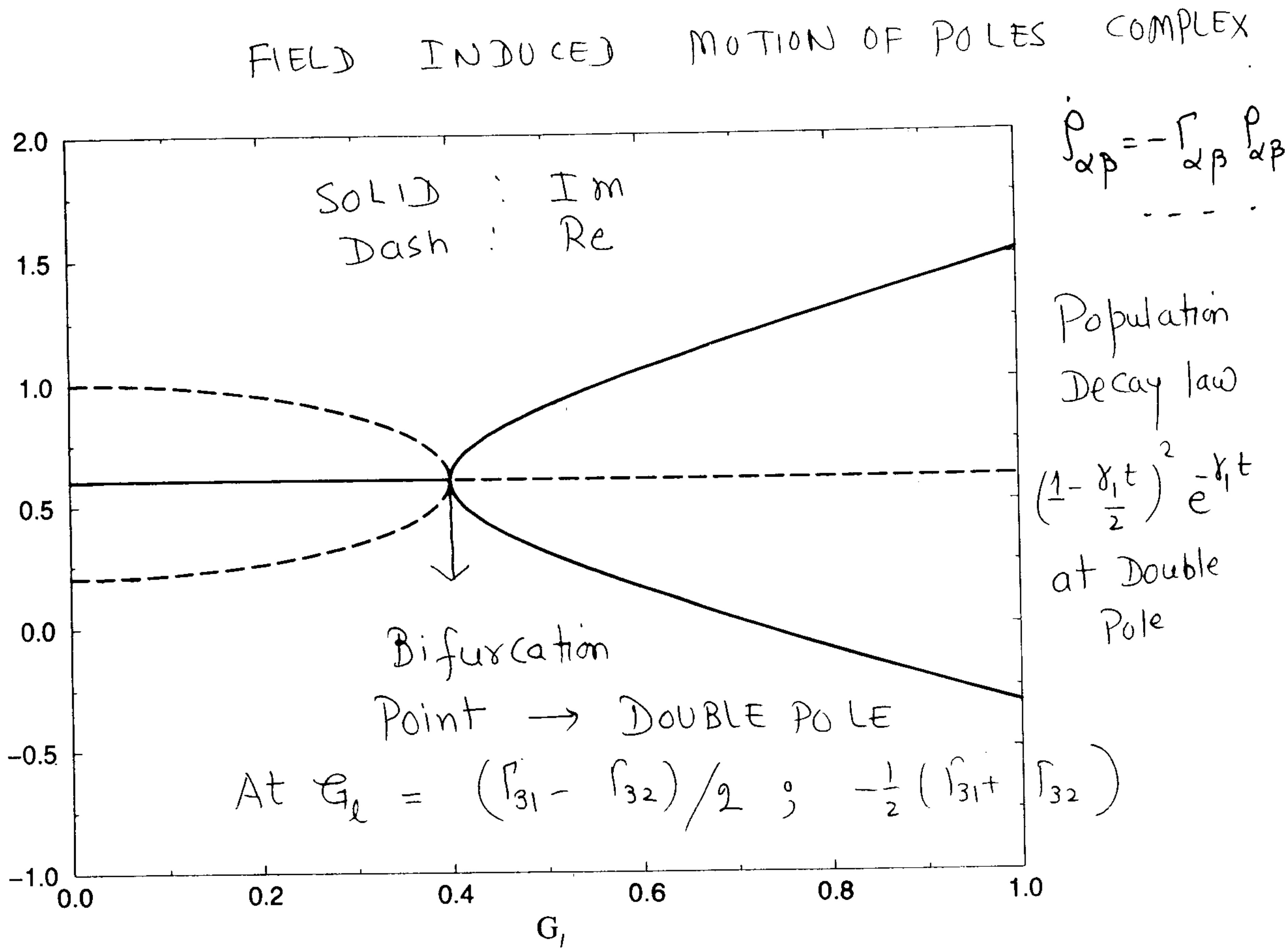


$\gamma_2 = \Lambda = 0$, OPTICAL REALIZATION OF

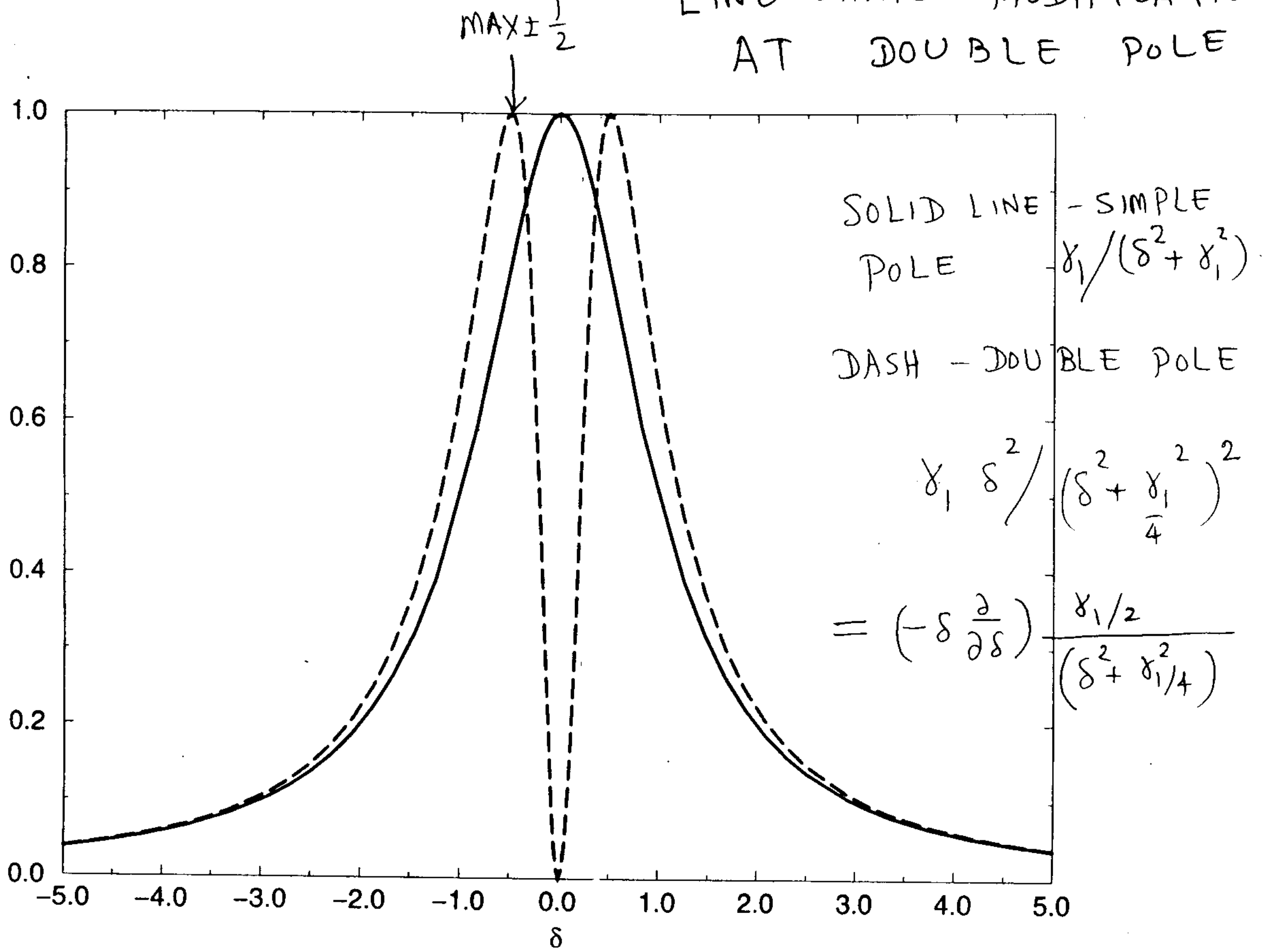
<u>EXTENDED FRIEDRICHS-LEE MODEL</u>

$B_1 \leftrightarrow B_2 \leftrightarrow$ [CCP] Continuum Coupling

DENSITY MATRIX - LIOUVILLE SPACE - SEARCH FOR COMPLEX POLES


FIELD INDUCED MOTION OF POLES COMPLEX
$\dot{\rho}_{\alpha\beta} = -\Gamma_{\alpha\beta}\,\rho_{\alpha\beta}$ - - - .
SOLID : Im
Dash : Re
Population Decay law
$\left(1 - \frac{\gamma_1 t}{2}\right)^2 e^{-\gamma_1 t}$
at Double Pole
Bifurcation Point → DOUBLE POLE
At $G_\ell = (\Gamma_{31} - \Gamma_{32})/2$; $-\frac{1}{2}(\Gamma_{31} + \Gamma_{32})$
2.0
1.5
1.0
0.5
0.0
−0.5
−1.0
0.0
0.2
0.4
0.6
0.8
1.0
G_1

LINE SHAPE MODIFICATION AT DOUBLE POLE



OPTICAL REALIZATION OF CASCADE MODEL –

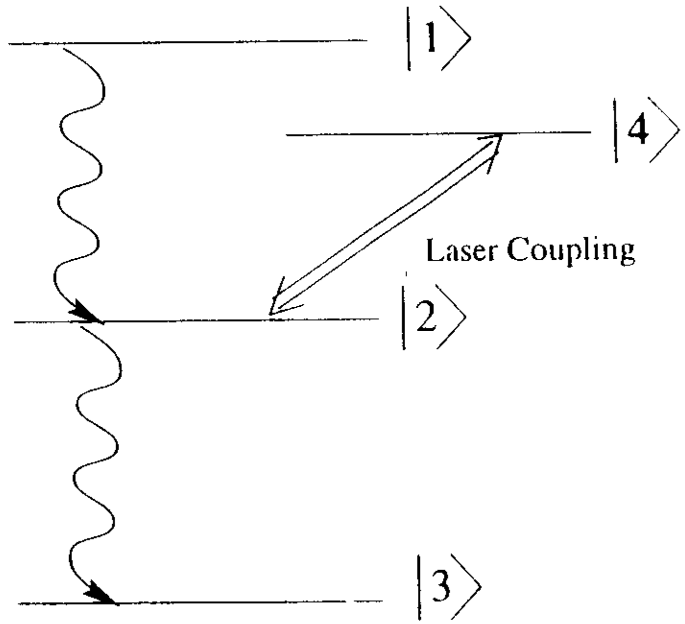
SINGLE PARTICLE ⟶ TWO PARTICLES ⟶ 3 PARTICLES

⟹ Coupling in this <u>Channel</u>

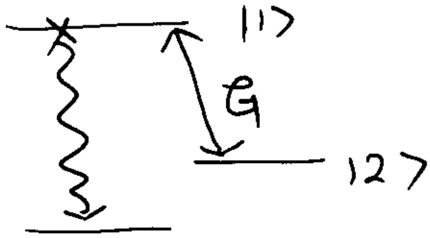
$A \rightarrow B_1\theta$

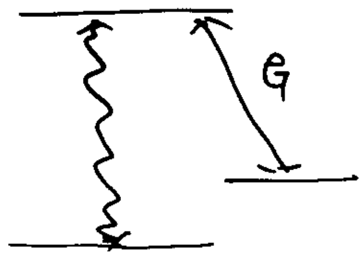
$\rightarrow C\theta\varphi$

$B_1\theta \rightleftarrows B_2\theta$



Energy Transfer studies – dd interaction

A. 

B. 

$(\Psi_\pm, \Psi_0)$ $(\Phi_\pm, \Phi_0)$

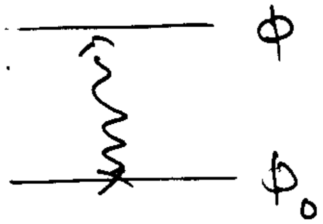
Resonant Transitions (large G)

$|\Psi_\pm, \Phi_0\rangle \rightarrow |\Psi_0, \Phi_\pm\rangle$

$|\Psi_\pm\rangle \rightarrow \frac{1}{\sqrt{2}}(\cos\theta\,|1\rangle + \sin\theta\,|2\rangle)$ etc

$(\cos^4\theta + \sin^4\theta)$

B. distinct Atoms



Φ could be resonant with Ψ_+ or Ψ_-

or even $|1\rangle$

Very many Possibilities : Φ line width large Compared to G.