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CROSS POLARIZATION RESONANT FLUORESCENCE

City University of New York PH.D. 1980

University
Microfilms
International 300 N. Zeeb Road, Ann Arbor, MI 48106 18 Bedford Row, London WC1R 4EJ, England

CROSS POLARIZATION RESONANT FLUORESCENCE

by

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A dissertation submitted to the Graduate Faculty in Physics in partial fulfillment of the requirements for the degree of Doctor of Philosophy, The City University of New York

1980

This manuscript has been read and accepted for the Graduate Faculty in Physics in satisfaction of the dissertation requirement for the degree of Doctor of Philosophy.

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Acknowledgements

I would like to thank first and foremost, Prof. M.H. Mittleman for his advice, criticism, encouragement and helpful discussions. His notes to the course on the subject of atom-laser interactions and the pre-publication copy of his book on the same topic were most helpful. I am also grateful to Prof. Mittleman for his arrangements of financial support by U.S. Office of Navel Research Contract N00014-74-C-0014. I am indebted to Ms. H.S.Weisman for the additional editorial aid that she provided.

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Introduction

Resonant fluorescence has become a subject of renewed interest due to advances in laser technology. Light nearly monochromatic and from a tunable laser can be used to excite, on resonance, a particular transition. The added capacity to produce intense radiation enables some continuous-wave lasers to pump an atomic system strongly. In such a case one has both the continual pumping of the atom and the continual production of fluorescent light quanta.

In the long time limit, one is dealing with a non-perturbative problem. The phenomena can not be thought of as disjoint one-photon processes but rather as a multiphoton phenomena in which the evolution of the joint atom-field state must be evaluated for arbitrary time. The treatment of the quantized field, in such an analysis, has been the subject of many studies[1-11].

The two-state atomic model has served as a simple example in which to discuss the essentials of the interdependent nature of the evolution of atom and field states. With a strong enough field, the rate of stimulated transitions is greater than the rate of spontaneous transitions. The resultant spectrum will be 'widened' by the creation of additional peaks. At exact resonance, these additional peaks will be symmetrically displaced from the center peak by an amount equal to the Rabi rate (the rate of stimulated transitions). Spontaneous transitions, as the source of radiative damping, are responsible for the various widths and relative heights of these peaks. On resonance, the ratio of the heights of either side peak to that of the center peak is 3:1[1-6,12-14]. The full widths of the side peaks and the center peak are 2 γ and 3 γ respectively (γ is half the Einstein A coeficient).

Initially these results were arrived at by statistical mechanical methods.[1] Various assumptions were made involving (1) the statistical factorization of atomic and field density operators and (2) an atom-field coupling in the Markoff approximation. Some of the earlier calculations treating the problem on a purely quantum electrodynamical basis failed to arrive at the aforementioned spectrum. It has been pointed out that the failure to include interference effects between competing processes has lead to erroneous results[2, 5, 6, 12]. The amplitude for the omission of the n^{th} photon with wave vector k, must be calculated by the addition of all amplitudes leading to this result. Correct quantum mechanical results and validation of previous statistical models have been established. Experimental results have been found to be in good agreement with theory[13, 14].

In addition to the two state case, the three level case has also been analyzed[15]. In such a situation two different lasers can be used to cause two different transitions. If one is relatively weak, it can be thought of as a probe of the states perturbed by the strong laser. Ionization of a strongly pumped two level atomic system has also been investigated.

The proper inclusion of radiative damping and the subsequent state of the atom are of interest to those who are concerned with processes occurring in the presence of a laser[16]. These processes could be single atom processes, two body processes, or collective effects. Resonance fluorescence is of especial interest in the study of optical bistability.[17] For great pressures, an increase of the incident intensity beyond a critical value results in a discontinuous transition from the stationary cooperative state to a stationary state characteristic of a one-atom solution (i.e. the three peak AC Stark effect).

Part I is a review of the general principles in which to analyze strong field resonance fluorescence. Section 2 includes a discussion of the radiative Hamiltonians with the laser field entered as a single mode, classical expression. Section 3 covers the standard approximations: 'Rotating Wave Approximation', dipole approximation, and the resonant approximation. Radiative damping is treated in Section 4 with a discussion of Mollow's approach[2].

In Part II there is first a calculation of the long-time limit resonant fluorescent spectrum of a particular four-state atom, strongly pumped by an intense linear polarized monochromatic laser. The atomic structure of concern involves two doubly degenerate energy levels (Fig. 1).

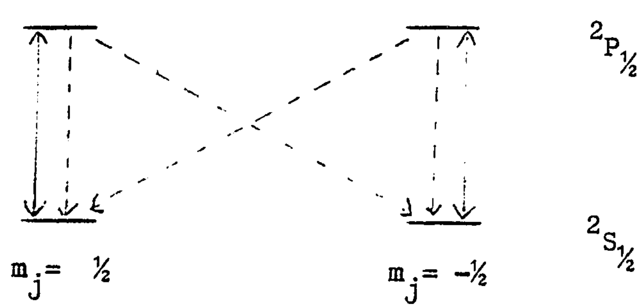

Fig. 1. Four state structure

The ratio of the height of the center peak to the height of the side peak for an on-resonance transition depends on the level structure and the polarization of the fluorescent radiation. The 3:1 ratio is peculiar to the non-degenerate two level atom. For the structure illustrated in Fig. 1, cross polarized fluorescence which results from transitions involving change in the magnetic quantum number, m, will exhibit a three peak spectrum where the ratio of heights of the center to side peak is 7:3 (on-resonance). Identical in both cases, however, is the 2:1 ratio of the area under the center peak to the area under the side peak.

While a linear-polarized laser only couples the states in pairs, a calculation of the evolution of the atomic states must include the total system as an inter-dependent coherent state. The coherent nature is inescapable for a proper analysis as will be demonstrated.

In Section 6 the degeneracy will be removed by a constant time-independent magnetic field. The shift of the atomic states depend on the strength of the magnetic field as well as the quantum numbers of each state. The unequal shifts will result in different resonant conditions for the two pairs of states of similar magnetic quantum numbers. When one of the resonance conditions is met and the magnetic field is significantly strong, optical pumbing will occur. When the laser is not on-resonance with either pair of perturbed states but instead equally detuned from both, each subsystem of a particular m will be pumbed equally. The resultant asymmetrical spectrums are discussed in Section 6.

In order to relate the theoretical finding of Sections 5 and 6, with possible experiments, an estimate of the effects of the deviations from monochromaticity is needed. In Section 7 the spectrums of previous sections are shown to be modified as the laser bandwidth becomes considerable.

The general method introduced is easily generalized to systems with greater magnetic subdegeneracy. The notation of Appendix A can be conveniently used for such a purpose. Generalization to multilevel atoms can be profitably tackled by similar methods.

PART I

2. Description of Incident and Fluorescent Fields

The various properties desired of the incident field in the situation under consideration are as follows:

polarization:	linear
spectrum:	monochromatic (provisionally)
statistical nature:	coherent
intensity:	sufficient intensity to create a greater rate of induced emissions than spontaneous emissions (i.e. saturation)

The first requirement is necessary for the problem involving degeneracy if steady state long-time limit is desired. Circular polarization will pump the atom between two states differing in magnetic quantum number (m_j) by 1. Eventually spontaneous emission will remove the electron from these two states permanently (see Fig. 2). Optical pumping in this manner will continue until the system reaches the final two states or final state (depending on the structure) (see Fig. 3).

As an example, consider the $^1S_{1/2} \rightarrow {}^2P_{3/2}$ that can arise in the Russel-Sanders coupling scheme. Fig. 2a shows the pumping between $m_j = -1/2$ of the lower level and $m_j = 1/2$ of the upper level. Eventually, due to the spontaneous decay, $m_j = 1/2$ to $m_j = 1/2$, the system will be pumped only between $m_j = 1/2$ and $m_j = 3/2$ states (Fig. 3a). This provides an example of an effective two state system. For the continual pumping among a greater number of states, linear polarization is necessary (see Fig. 1).

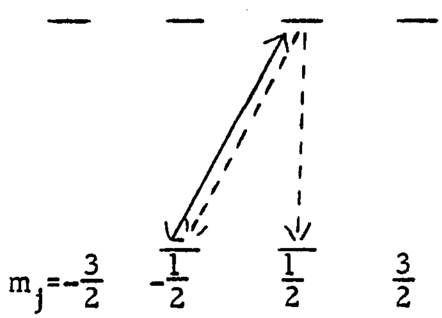

Fig. 2a.

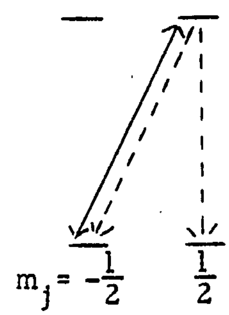

Fig. 2b.

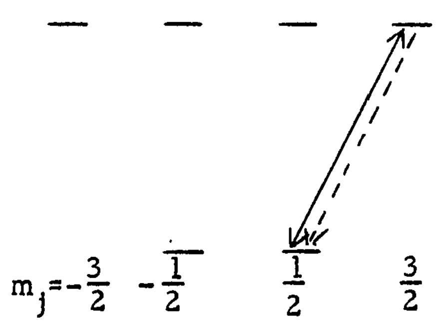

Fig. 3a.

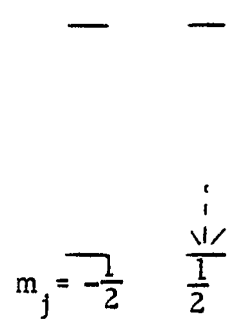

Fig. 3b.

Fig. 2 and Fig. 3. Optical Pumping.

Monochromatic radiation enables the investigation of a specific transition and allows investigations as a function of the degree of resonance (i.e. as a function of the detuning of laser frequency from atomic frequency: $\delta = \omega - \omega_a$). Lasers provide radiation that is sufficiently monochromatic (deviations from strict monochromatic incident radiation will be discussed in Section 7).

Lasers are also a source of coherent radiation. Coherence involves definitiveness of phase. The atomic sources of a laser radiate in phase to the extent that spontaneous radiation is negligible. Spontaneous radiation is an incoherent process related to vacuum fluctuations (its contribution to the laser's deviation from strict coherence will be discussed in Section 7).

Classically, a coherent state has an electric field component

$$\vec{E} \cos(\omega t - \vec{k}\cdot\vec{r} + \phi) \tag{2-1a}$$

where ϕ is time independent non-stochastic. A simple example is radio waves. For a non-cooperative radiating gas, however, ϕ is a stochastic variable which describes self-coherence. For times, τ , greater than the inverse of the decay rate

$$\left\langle e^{i\phi(t)} e^{i\phi(t+\tau)} \right\rangle \approx 0 \tag{2-1b}$$

The normalized quantum field state which has an expectation value equal to the classical coherent state Eq. (2-1a) is

$$|\alpha\rangle = e^{-\frac{1}{2}|\alpha|^2} \sum_n \frac{\alpha^n}{\sqrt{n!}} |n\rangle \tag{2-2}$$

where $|n\rangle$ is the state of n quanta in the $(\vec{k}, \frac{\vec{E}_0}{|E_0|})$ mode and

$$\alpha = -i\frac{E_0}{2} e^{-i\phi} \left(\frac{2\pi\hbar\omega}{V}\right)^{-1/2} \tag{2-3}$$

The state $|\alpha\rangle$ is an eigenfunction of the field destruction operator

$$a_{k,\lambda}|\alpha\rangle = \alpha|\alpha\rangle \tag{2-4}$$

for the appropriate k and λ. Thus,

$$\langle\alpha|A^{\perp}|\alpha\rangle = -\frac{c}{\omega}\vec{E}_0 \sin(\omega t - \vec{k}\cdot\vec{r} + \phi) \tag{2-5}$$

and

$$\langle\alpha|E^{\perp}|\alpha\rangle = \vec{E}_0 \cos(\omega t - \vec{k}\cdot\vec{r} + \phi) \tag{2-6}$$

where $A^{\perp}$ is the quantized field operator.

$$A^{\perp} = \sum_{k\lambda} \sqrt{\frac{2\pi\hbar c^2}{\omega V}} \left(\hat{\epsilon}_{k\lambda} a_{k\lambda} e^{i(\vec{k}\cdot\vec{r}-\omega t)} + \hat{\epsilon}^{\dagger}_{k\lambda} a^{\dagger}_{k\lambda} e^{-i(\vec{k}\cdot\vec{r}-\omega t)}\right) \tag{2-7}$$

and $\vec{k}$ is the wave vector

$\omega = c|k|$

λ is the polarization index

$\hat{\epsilon}_{k\lambda}$ is the polarization vector such that

$$\hat{\epsilon}_{k\lambda}\cdot\hat{\epsilon}^{\dagger}_{k'\lambda'} = \delta_k^{k'}\delta_\lambda^{\lambda'} \quad \text{and} \quad \hat{\epsilon}_{k\lambda}\cdot\hat{k} = 0 \tag{2-8}$$

$a_{k\lambda}$ $(a_{k\lambda}{}^{\dagger})$ is the radiation field destruction (creation) operator for transverse modes.

V is the normalization volume

The vector field operator satisfies the transversality condition

$$\nabla \cdot A^{\perp} = 0 \tag{2-9}$$

The transverse electric field operator is

$$E^{\perp} = -\frac{1}{c}\frac{\partial A^{\perp}}{\partial t} \tag{2-10}$$

When the intensity is great enough to ignore fluorescence into the laser mode, the operator for the laser mode can be replaced by its expectation value (i.e. Eq. (2-5)). This is true not only for the coherent state but for any state in which the average number of quanta per mode is much greater than deviations from that average:

$$\bar{N} \gg \Delta N \tag{2-11}$$

In this case the field operators, $a_{k\lambda}$ and $a_{k\lambda}^{\dagger}$ can be approximated by the number $\sqrt{\bar{N}}$. For an occupation probability distribution $|f(n)|^2$ one has

$$\begin{aligned} a_{k\lambda}\, f(n)|n\rangle &= f(n)e^{i\hat{\phi}}\sqrt{\bar{N}}\,|n\rangle \qquad \text{(See Ref. 20)} \\ &= f(n)e^{i\hat{\phi}}\sqrt{\bar{N}+n-\bar{N}}\,|n\rangle \\ &\approx f(n)e^{i\hat{\phi}}\sqrt{\bar{N}}\left(1+\frac{1}{2}\frac{n-\bar{N}}{\bar{N}}\cdots\right)|n\rangle \end{aligned}$$

The next significant term is of the order $\frac{\Delta N}{\bar{N}}$ and is small for distributions satisfying Eq. (2-11).

For a coherent state, the probability of n quanta existing is described by a Poisson distribution (from Eq. (2-2)):

$$P(n) = \frac{|\alpha|^{2n} e^{-|\alpha|^2}}{n!} \tag{2-12}$$

Eq. (2-11) becomes

$$|\alpha|^2 \gg |\alpha| \tag{2-13}$$

which is valid for large intensities. However, for a coherent state it is possible by means of a suitable transformation, to use the classical-field expression in the Hamiltonian without ignoring fluorescence into the laser mode.[2] Let Ψ' be the untransformed state.

$$\Psi = D^{-1}\Psi' \tag{2-14}$$

where D is the 'displacement operator'[19]

$$D = e^{a_{k\lambda}^{\dagger}\alpha - \alpha^* a_{k\lambda}} \tag{2-15}$$

D has the properties

$$D^{-1} a_{k\lambda} D = a_{k\lambda} + \alpha \tag{2-16}$$

$$a_{k\lambda} D \,|\, \text{vacuum}\rangle = \alpha D \,|\, \text{vacuum}\rangle \tag{2-17}$$

Thus

$$D^{-1} A^{\perp} D = -\frac{c}{\omega} E_0 \sin(\omega t - \vec{k}\cdot\vec{r} + \phi) + A^{\perp} \tag{2-18}$$

$$D^{-1} E^{\perp} D = E_0 \cos(\omega t - \vec{k}\cdot\vec{r} + \phi) + E^{\perp} \tag{2-19}$$

and

$$\Psi(t=0) = D^{-1}\psi' = |vacuum\rangle |atom\rangle_0 \tag{2-20}$$

The initial transformed state is the radiation vacuum while the Hamiltonian includes the classical-driving field. A detailed expression will be given after considerations concerning the Hamiltonian.

The total nonrelativistic Hamiltonian in the Coulomb gauge for a single particle is[19]

$$H = \frac{1}{2m}\left(p - \frac{e A_{\perp}(x)}{c}\right)^2 + e\phi + \sum_{k\lambda} \hbar\omega_{k\lambda} a^{\dagger}_{k\lambda} a_{k\lambda} \tag{2-21}$$

Since $\nabla \cdot A_{\perp} = 0$, the Hamiltonian can be expressed as

$$H = \frac{p^2}{2m} + e\phi - \frac{e A_{\perp}(x)}{mc} p + \frac{e^2 A_{\perp}^2(x)}{2mc^2} + \sum_{k\lambda} \hbar\omega_{k\lambda} a^{\dagger}_{k\lambda} a_{k\lambda} \tag{2-22}$$

where $A^{\perp}(x)$ is Eq. (2-7) at time $t = 0$.

Consider the transformation

$$\Phi = e^{i\sum \omega_{k\lambda} a^{\dagger}_{k\lambda} a_{k\lambda} t}\, \Psi \tag{2-23}$$

Given

$$e^{i\omega_{k\lambda} t\, a^{\dagger}_{k\lambda} a_{k\lambda}}\, a_{k\lambda}\, e^{-i\omega_{k\lambda} a^{\dagger}_{k\lambda} a_{k\lambda} t} = a_{k\lambda} e^{-i\omega_{k\lambda} t} \tag{2-24}$$

$$e^{i\omega_{k\lambda} a^{\dagger}_{k\lambda} a_{k\lambda} t}\, a^{\dagger}_{k\lambda}\, e^{-i\omega_{k\lambda} a^{\dagger}_{k\lambda} a_{k\lambda} t} = a^{\dagger}_{k\lambda} e^{i\omega_{k\lambda} T} \tag{2-25}$$

the wave equation for Φ is

$$i\hbar \frac{\partial}{\partial t} \Phi = \left(\frac{p^2}{2m} + e\phi - \frac{e \vec{A}_\perp (x,t)}{mc} \vec{p} + \frac{e^2 A^2_\perp (x,t)}{2mc^2} \right) \Phi \tag{2-26}$$

With the dipole approximation, which will be considered further in the next section, Eq.(2-26) can be transformed by the following gauge transformation:

$$\Psi_E = e^{-\frac{ie}{\hbar c}\vec{r}\cdot\vec{A}(t)}\ \Phi \tag{2-27}$$

$$i\hbar \frac{\partial \Psi_E}{\partial t} = \left(\frac{p^2}{2m} + e\phi - e\vec{r}\cdot\vec{E}(t) \right) \Psi_E \tag{2-28}$$

where E (t) is given by Eq. (2-10). The wave functions Ψ_E and Φ are related by Eq. (2-27), however, a perturbation calculation requires some further requirement before picking equation (2-26) or (2-28). Consider the matrix element

$$-\frac{e \vec{A}_\perp (t) \cdot \langle a | \vec{p} | b \rangle}{mc} \tag{2-29}$$

and let $A_\perp$ be the classical expression $-\frac{cE_0}{\omega} \sin\omega t$. With

$\langle a | p | b \rangle = i m \omega_{ab} \langle a | r | b \rangle$, (2-29)becomes

$$i \frac{e \, \omega_{ab}}{\omega} \langle a | \vec{r} | b \rangle \cdot \vec{E}_0 \sin \omega t \tag{2-30}$$

This is to be compared with the matrix element

$$- e \langle a | \vec{r} | b \rangle \cdot \vec{E}_0 \cos \omega t \tag{2-31}$$

Apart from phase factors these two matrix elements differ by ω_{ab}/ω. The first arises from Eq. (2-26), dropping the higher order A^2 term, this expression results for the interaction matrix. The second arises from Eq. (2-28). One cannot, however, take with equal results the first two terms of either Eq. (2-26) or (2-28) as the unperturbed Hamiltonian in light of Eq. (2-27). These two results for the transition matrix (Eq. (2-30) and (2-31) were considered by Lamb[21] who found the second expression to agree with experiment. D.-H. Yang[22] argues that it is Eq. (2-28) that should be used, with the first two terms of the Hamiltonian representing the free atom and the interaction matrix elements given by expression (2-31). The Heisenberg equations of motion from the Hamiltonian in Eq. (2-28) have classical form and expectation values which obey the correspondence principle. Consider

$$\frac{d}{dt} \left(\frac{p^2}{2m} + e\phi \right) = \frac{1}{i\hbar} \left[\frac{p^2}{2m} + e\phi , H \right] \tag{2-32}$$

$$= e \frac{\vec{p}}{m} \cdot \vec{E}(t) \tag{2-33}$$

The time rate of change of atomic energy equals the power operator. This is the classical expression.

In either case, near resonance, the term (ω_{ab}/ω) is only slightly different from 1.

In general, we want to consider an electron in a more complex atom; H_A will stand for the Hamiltonian for this complex atom:

$$i\hbar \frac{\partial \psi'}{\partial t} = (H_A - e\vec{r}\cdot\vec{E}^{\perp}(t))\psi' \tag{2-34}$$

the transformation (2-14),with Eq.(2-16) results in

$$i\hbar \frac{\partial \psi}{\partial t} = (H_A - e\vec{r}\cdot\vec{E}_0 \cos\omega t - e\vec{r}\cdot\vec{E}^{\perp}(t))\psi \tag{2-35}$$

where

$$E^{\perp} = \sum_{k\lambda} \mathcal{E}_{k\lambda} + \mathcal{E}^{\dagger}_{k\lambda} \tag{2-36}$$

$$\mathcal{E}_{k\lambda} = i\sqrt{\frac{2\pi\hbar\omega}{V}}\,\hat{\epsilon}_{k\lambda}\, a_{k\lambda}\, e^{-i\omega_{k\lambda}t} \tag{2-37}$$

and

$$\psi(0) = |\text{ vacuum }\rangle|\text{ atom }, t=0\rangle \tag{2-38}$$

For the laser mode, the number operator $a_L^{\dagger} a_L$ leads to the following expectation value:

$$\langle \psi' | a_L^{\dagger} a_L | \psi' \rangle = \langle \psi | D^{-1} a_L^{\dagger} a_L D | \psi \rangle \tag{2-39}$$

$$\langle\psi'|a_L^\dagger a_L|\psi'\rangle = |\alpha|^2 + \alpha\langle\psi|a_L^\dagger|\psi\rangle + \alpha^*\langle\psi|a_L|\psi\rangle + \langle\psi|a_L^\dagger a_L|\psi\rangle \quad (2\text{-}40)$$

All other modes

$$\langle\psi'|\, a_{k\lambda}^\dagger a_{k\lambda}\,|\psi'\rangle = \langle\psi|\, a_{k\lambda}^\dagger a_{k\lambda}\,|\psi\rangle \quad (2\text{-}41)$$

The spectrum, in the long-time limit, will be shown to be equal to

$$\frac{d}{dt}\langle\psi|\, a_{k\lambda}^\dagger a_{k\lambda}\,|\psi\rangle$$

for laser and non-laser modes.

3. Standard Approximations

The dipole approximation, resonant approximation and "rotating wave approximation" are, in some form, touched upon in a typical course in quantum mechanics. The dipole approximation, when valid, involves the omission of the spatial dependence of the incident radiation in the calculation of the evolution of the atomic state. The resonant approximation consists of the sole retention of the term involving $1/(\omega - \omega_{jk} + i\Gamma_{jk})$ when the frequency of the incident radiation (ω) is nearly equal to the frequency associated with the transition between state j and state k (ω_{jk}) and the width Γ_{jk} is less than the separation of the resonant and non-resonant states (i.e. states j and k are well defined). The rotating wave approximation (RWA) involves retention of the terms $1/(\omega - \omega_j)$ as opposed to $1/(\omega + \omega_j)$. These specific forms of the RWA and resonant approximations are one photon approximations. Both the form and validity of the RWA and resonant approximations for ongoing pumping will be reviewed in this section. However, the dipole approximation which has been used in the previous section will be discussed first.

The dipole approximation is valid when the wavelengths $(2\pi/k)$ of absorbed and emitted radiation are large compared to the linear dimensions (a) of the atomic wavefunction (i.e. $ka \ll 1$). Successive terms in the power series of e^{ikr} involve higher powers of ka, enabling one to approximate e^{ikr} as 1. (For atomic emission of optical radiation: ka $\sim 10^{-3}$). The dipole approximation cannot be made for transitions in which the dipole matrix element vanishes while the quadropole matrix element does not. The analysis, therefore, is restricted to incident frequencies which are relatively near to an allowed dipole transition and sufficiently far from a quadrupole resonance.

The resonant approximation is made by the retention of the two atomic levels in which the frequency of incident radiation is equal or nearly equal to the frequency associated with the atomic transition. The validity of such an approximation is assured when the field is weak enough so that the probability of excitations to other states is negligible as will be subsequently demonstrated.

The rotating wave approximation (RWA) is made by retaining the terms of the interaction Hamiltonian corresponding to approximate energy conservation.

These approximations can be made as follows:

Let $|U\rangle \exp(-iE_U t/\hbar)$ and $|L\rangle \exp(-iE_L t/\hbar)$ (3-1)

represent the atom upper and lower state with energies E_u and E_L respectively.

The wave function is

$$\Psi(t) = b_U(t)|U\rangle e^{-iE_U t/\hbar} + b_L(t)|L\rangle e^{-iE_L t/\hbar} \tag{3-2}$$

The atom is coupled to the laser field of frequency ω and amplitude E_o in the dipole approximation through the interaction:

$$H_{int} = -e\vec{r}\cdot\vec{E}_0 \cos \omega t \tag{3-3}$$

The wave function is governed by Schrodinger's equation:

$$i\hbar \frac{\partial \Psi}{\partial t} = (H_{atom} + H_{int})\Psi \tag{3-4}$$

The effects of spontaneous emission will be covered in Section 4 and have been for the moment omitted. Multiplying Schrodinger's equation from the left by $\langle U|$ and $\langle L|$ gives two equations for b_u and b_L

$$i\hbar \frac{db_U}{dt} = \hbar\Lambda \cos\omega t \; e^{i\omega_a t} \, b_L$$

$$i\hbar \frac{db_L}{dt} = \hbar\Lambda \cos\omega t \; e^{-i\omega_a t} \, b_U \tag{3-5a,b}$$

where $\hbar\Lambda \equiv -e\langle U|\vec{r}|L\rangle \cdot E_o$ and $\omega_a = (E_U - E_L)/\hbar$. Atomic states are defined up to a constant phase factor thereby enabling one to allow Λ to be real.

The rotating wave approximation is made by dropping terms involving $e^{\pm i(\omega+\omega_a)t}$ while retaining terms involving $e^{\pm i(\omega-\omega_a)t}$. The rapidly oscillating factor contributes only slightly as will be shown subsequently. Upon making the RWA, Eq. 3-5a, b becomes

$$i\frac{d}{dt}\begin{pmatrix} b_U \\ b_L \end{pmatrix} = \begin{pmatrix} 0 & \frac{\Lambda}{2}e^{-i\delta t} \\ \frac{\Lambda}{2}e^{i\delta t} & 0 \end{pmatrix}\begin{pmatrix} b_U \\ b_L \end{pmatrix} \tag{3-6}$$

where δ is the detuning of the laser from atomic resonance:

$$\delta = \omega - \omega_a \tag{3-7}$$

Eq. (3-6) is equivalent to

$$i\frac{d}{dt}\begin{pmatrix} b_U e^{i\delta t/2} \\ b_L e^{-i\delta t/2} \end{pmatrix} = \frac{1}{2}\begin{pmatrix} -\delta & \Lambda \\ \Lambda & \delta \end{pmatrix}\begin{pmatrix} b_U e^{i\delta t/2} \\ b_L e^{-i\delta t/2} \end{pmatrix} \tag{3-8}$$

The time independent matrix allows for simple integration

$$\begin{pmatrix} b_u e^{i\delta t/2} \\ b_L e^{-i\delta t/2} \end{pmatrix} = e^{-\frac{i}{2}(\Lambda \sigma_x - \delta \sigma_z)t} \begin{pmatrix} b_{u_o} \\ b_{L_o} \end{pmatrix} \tag{3-9a,b}$$

$$= \left(\cos \frac{\mathcal{E}}{2} t - (\Lambda \sigma_x - \delta \sigma_z) \frac{i}{\mathcal{E}} \sin \frac{\mathcal{E}}{2} t \right) \begin{pmatrix} b_{u_o} \\ b_{L_o} \end{pmatrix}$$

where σ_j are Pauli matrices: $\sigma_x = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}$ $\sigma_y = \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix}$ $\sigma_z = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}$

b_{u_o} and b_{L_o} are initial amplitudes

$\mathcal{E} = \sqrt{\Lambda^2 + \delta^2} = \sqrt{\frac{1}{\hbar} | e \langle u | \vec{r} | L \rangle \cdot E_o |^2 + (\omega - \omega_a)^2}$

is the generalized Rabi frequency for $\delta \neq 0$.

Use has been made of the anti-commutation relation

$$\{ \sigma_i , \sigma_j \} = 2 \delta_{ij}$$

in the expansion that has led from Eq.(3-9a) to Eq.(3-9b). One can replace Λ by $\int_0^t \Lambda(t') dt'$ for slowly varying electric field amplitudes such as the adiabatic switching on of the laser.

For a system initially in the lower state, the probability of being in the upper state at time t is:

$$| b_u |^2 = \left| \frac{\Lambda}{\mathcal{E}} \right|^2 \sin^2 \frac{\mathcal{E}}{2} t = \frac{1}{2} \left| \frac{\Lambda}{\mathcal{E}} \right|^2 (1 - \cos \mathcal{E} t) \tag{3-10}$$

On resonance upper and lower occupation probabilities are

$$|b_U|^2 = \frac{1}{2} - \frac{1}{2}\cos\Lambda t$$

$$|b_L|^2 = \frac{1}{2} + \frac{1}{2}\cos\Lambda t$$

Thus, on resonance actual induced transitions occur at the Rabi rate.

The diagonalization of the matrix of Eq.(3-8) yields two eigenvalues ($\pm\varepsilon/2$) and two eigenfunctions. Consequently, the two solutions of Eq. (3-4) when the rotating wave approximation has been made are:

$$\psi^{(+)} = e^{-i\frac{\varepsilon}{2}t}\frac{1}{\sqrt{2\varepsilon(\varepsilon+\delta)}}\left(\Lambda\begin{pmatrix}1\\0\end{pmatrix}e^{-i\frac{\omega}{2}t} + (\varepsilon+\delta)\begin{pmatrix}0\\1\end{pmatrix}e^{i\frac{\omega}{2}t}\right)$$

$$\psi^{(-)} = e^{i\frac{\varepsilon}{2}t}\frac{1}{\sqrt{2\varepsilon(\varepsilon+\delta)}}\left((\varepsilon+\delta)\begin{pmatrix}1\\0\end{pmatrix}e^{-i\frac{\omega}{2}t} - \Lambda\begin{pmatrix}0\\1\end{pmatrix}e^{i\frac{\omega}{2}t}\right) \qquad \text{(3-11a, b)}$$

Energy is represented by the operator $i\hbar\frac{\partial}{\partial t}$. Consequently, each solution above is a linear combination of two energy states.

Altogether, the energy eigenvalues are $\hbar\left(\frac{\omega}{2}\pm\frac{\varepsilon}{2}\right)$ and $\hbar\left(-\frac{\omega}{2}\pm\frac{\varepsilon}{2}\right)$ (apart from an overall energy of $(E_U+E_L)/2$). One can expect transitions of frequencies ω , $\omega+\varepsilon$ and $\omega-\varepsilon$ (see Fig. 4).

That the energy eigenvalues and emission frequencies are indeed these values, must be shown by an actual calculation of the fluorescent spectrum. The 'splitting' of energy levels by a strong oscillating field is referred to as the dynamical or AC stark effect. The states of equations (3-11a, b) are the

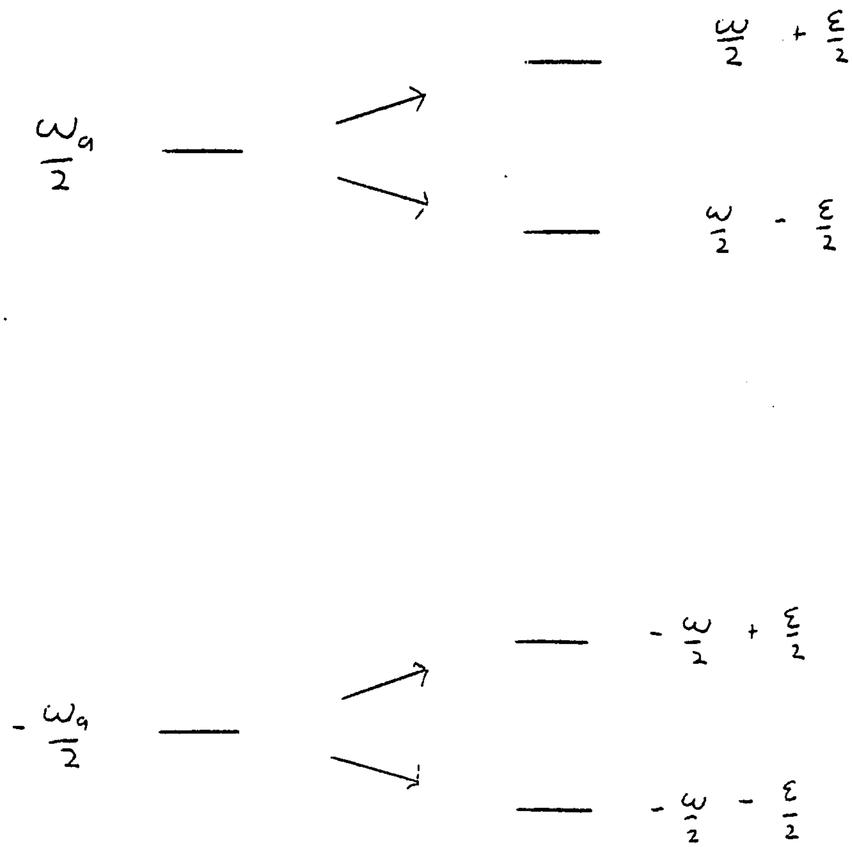

Fig. 4. AC Stark Splitting.

"dressed atom states" or combined atom-laser states.

The next higher order correction to the RWA is the Bloch-Siegert shift[24]- a shift of the atomic frequency by $\left(\frac{\Lambda}{2\omega}\right)^2\omega_a$. Thus, the RWA is a good approximation when the field is weak enough that the strength of interaction (Λ) is small compared to the atomic binding:

$$\Lambda \ll \omega \qquad (3\text{-}12)$$

There are many treatments of the RWA and its corrections[24-27]. Shirley[26] approaches the problem by the use of Floquet's Theorem[29]. Let $H(t)$ be an n x n matrix and a periodic function of time with period T. By Floquet's Theorem, the equation

$$i\frac{d}{dt}\Psi(t) = H(t)\Psi(t) \qquad (3\text{-}13a)$$

has solutions of the form:

$$\Psi_j = \phi_j e^{-iq_j t} \qquad (3\text{-}13b)$$

where $\phi_j(t+T) = \phi_j(t)$ is an n component column vector. The spatial form of Floquet's Theorem is familiar to solid state physicists as Bloch's Theorem. An equation of the form of Eq. 3-13a in classical physics is Hill's equation[27]. It is advantageous to expand H and Ψ in a Fourier series. Let $\Psi_{\alpha j}$ be the α component of the jth solution and define $\Psi^n_{\alpha j}$ and $H^n_{\alpha\beta}$ from the equations:

$$\Psi_{\alpha j}(t) = \sum_n \Psi^n_{\alpha j}\, e^{in\omega t}\, e^{-iq_j t} \tag{3-14a}$$

$$H_{\alpha\beta}(t) = \sum_n H^n_{\alpha\beta}\, e^{in\omega t} \tag{3-14b}$$

Schrodinger's equation becomes:

$$\sum_{\gamma k} \left\{ H^{n-k}_{\alpha\gamma} + n\omega\, \delta_{\alpha\gamma}\, \delta_{kn} \right\} \Psi^k_{\gamma j} = q_j\, \Psi^n_{\alpha j} \tag{3-15}$$

or

$$\begin{pmatrix} \ddots & & & & & & & \\ & E_L+3\omega-q_j & \Lambda/2 & & & & & \\ & \Lambda/2 & E_U+2\omega-q_j & & & \Lambda/2 & & \\ & & & E_L+2\omega-q_j & \Lambda/2 & & & \\ & & & \Lambda/2 & E_U+\omega-q_j & & & \Lambda/2 \\ & & \Lambda/2 & & & E_L+\omega-q_j & \Lambda/2 & \\ & & & & & \Lambda/2 & E_U-q_j & \\ & & & & \Lambda/2 & & & E_L-q_j \\ & & & & & & & & \ddots \end{pmatrix} \begin{pmatrix} \vdots \\ \Psi^3_{Lj} \\ \Psi^2_{Uj} \\ \Psi^2_{Lj} \\ \Psi^1_{Uj} \\ \Psi^1_{Lj} \\ \Psi^0_{Uj} \\ \Psi^0_{Lj} \\ \vdots \end{pmatrix} = 0$$

In lowest order consider

$$\begin{pmatrix} E_L + \omega - q & \Lambda/2 \\ \Lambda/2 & E_v - q \end{pmatrix} \begin{pmatrix} \psi_L^1 \\ \psi_v^0 \end{pmatrix} = 0 \tag{3-16}$$

The roots are $q_\pm = \frac{1}{2}(E_L + E_v + \omega \pm \mathcal{E})$. Using these eigenvalues, one can solve for ψ_v^2 and ψ_L^{-1} in terms of ψ_L^1 and ψ_v^0.

$$\psi_v^2 = \frac{\Lambda(\delta + 4\omega \mp \mathcal{E})}{-(4\omega)^2 \pm 8\omega\mathcal{E}} \psi_L^1 \quad \text{and} \quad \psi_L^{-1} = \frac{\Lambda(\delta + 4\omega \pm \mathcal{E})}{(4\omega)^2 \pm 8\omega\mathcal{E}} \psi_v^0 \tag{3-17}$$

for $\mathcal{E} \ll \omega$

$$\psi_v^2 = -\frac{\Lambda}{4\omega}\psi_L^1 \qquad \psi_L^{-1} = \frac{\Lambda}{4\omega}\psi_v^0 \tag{3-18}$$

The next order correction to Eq. (3-16) is:

$$\begin{pmatrix} E_L - \frac{\Lambda^2}{8\omega} + \omega - q & \Lambda/2 \\ \Lambda/2 & E_v + \frac{\Lambda^2}{8\omega} \end{pmatrix} \begin{pmatrix} \psi_L^1 \\ \psi_v^0 \end{pmatrix} = 0 \tag{3-19}$$

Consequently, there is a shift of atomic frequency by $\frac{\Lambda^2}{4\omega} \approx \left(\frac{\Lambda}{2\omega}\right)^2 \omega/a$

This small correction to the RWA is called the Bloch Siegert shift. Shirley has calculated correction terms up to the sixth order in $\left(\frac{\Lambda}{\omega}\right)$.

There have been several calculations of the RWA involving the two state atom interacting with one mode of the quantized field. The one mode QED interaction Hamiltonian in the dipole approximation is:

$$H_{int} = e\,i\sqrt{\frac{2\pi\hbar\omega}{V}}\langle u|\vec{r}|L\rangle\cdot\left(\hat{\epsilon}^{\dagger}_{k\lambda}\, a^{\dagger}_{k\lambda} - \hat{\epsilon}_{k\lambda}\, a_{k\lambda}\right)\left(\sigma_{+} + \sigma_{-}\right) \tag{3-20}$$

where σ_+ and σ_- are raising and lowering operators. The RWA is made by retaining only those terms which nearly conserve energy; $\sigma_+ a_{k\lambda}$ and $\sigma_- a^{\dagger}_{k\lambda}$.

Consider the wavefunction and Hamiltonian in the interaction picture:

$$\psi' = e^{iH_0 t/\hbar}\,\psi \tag{3-21}$$

$$H' = e^{+iH_0 t/\hbar}\, H_{int}\, e^{-iH_0 t/\hbar} \tag{3-22}$$

where the prime denotes interaction representation and H_0 is the non-interacting atom and field Hamiltonian

$$H_0 = \frac{E_u + E_L}{2} + \frac{\hbar\omega_a}{2}\sigma_z + \hbar\omega_k\, a^{\dagger}_{k\lambda}\, a_{k\lambda} \tag{3-23}$$

H' can be evaluated with the help of the following identities:

$$e^{i\frac{\omega_a}{2}\sigma_z t}\,\sigma_{\pm}\, e^{-i\frac{\omega_a}{2}\sigma_z t} = \sigma_{\pm}\, e^{\pm i\omega_a t} \tag{3-24}$$

$$e^{i\omega_{k\lambda}a^{\dagger}a t}\, a\, e^{-i\omega_{k\lambda}a^{\dagger}a t} = a\, e^{-i\omega_{k\lambda} t} \tag{3-25}$$

$$e^{i\omega_{k\lambda} a^\dagger a t}\, a^\dagger e^{-i\omega_{k\lambda} a^\dagger a t} = a^\dagger e^{i\omega_{k\lambda} t} \tag{3-26}$$

The energy non-conserving terms dropped from H' in the RWA are:

$$ie\sqrt{\frac{2\pi\hbar\omega}{V}}\langle U|\vec{r}|L\rangle\cdot\left(\hat{\epsilon}^{\dagger}_{k\lambda} a^{\dagger}_{k\lambda}\sigma_{+} e^{i(\omega_a+\omega_{k\lambda})t} - \hat{\epsilon}_{k\lambda} a_{k\lambda}\sigma_{-} e^{-i(\omega_a+\omega_{k\lambda})t}\right) \tag{3-27}$$

The remaining terms are those which involve the difference in frequencies:

$$H'_{RWA} = ie\sqrt{\frac{2\pi\hbar\omega}{V}}\langle U|\vec{r}|L\rangle\cdot\left(\hat{\epsilon}^{\dagger}_{k\lambda} a^{\dagger}_{k\lambda}\sigma_{-} e^{i(\omega_{k\lambda}-\omega_a)t} - \hat{\epsilon}_{k\lambda} a_{k\lambda}\sigma_{+} e^{-i(\omega_{k\lambda}-\omega_a)t}\right) \tag{3-28}$$

Thus, the problem reduces to the evaluation of the slowly varying amplitude :

$$i\hbar\frac{\partial\Psi'}{\partial t} = H'_{RWA}\Psi \tag{3-29}$$

The 'counter-rotating' terms of Eq. (3-27) are included in a determination of the Bloch-Siegert shift. Several authors have evaluated corrections to the quantized RWA up to the eighth order[25-27]. In the limit of a large number of quanta, both the semi-classical and one-mode quantized analysis of high order corrections give the same results. The corrections to the RWA from the intense radiation of the laser mode are most important. Even in this case the corrections are insignificant since the intensity necessary for the long-time limit resonant fluorescence experiments satisfies Eq. (3-12). Intensities in which $\Lambda \sim \omega$, are too strong. In this case $\hbar\Lambda$ is roughly equal to

$e r_0 E_0$, where r_0 is the Bohr radius. $\hbar\omega$ is of the order of $\frac{e^2}{r_0}$. If Λ was nearly equal to ω , the amplitude of the laser field would be as strong as the binding field. If one can still refer to the target(s) as an atom, ionization would soon occur. In various strong field transient effects, other approaches are necessary[28].

The higher atomic energy states, omitted in the resonant approximation will now be considered. By treating these states as a perturbation, our estimate of error and requirement for omission can be found.

For an atom initially in its lower state, the amplitude for upper and lower states at time t is, from Eq. (3- 9a ,b)

$$b_U = e^{-i\delta t/2}\left(-i\frac{\Lambda}{\varepsilon}\sin\frac{\varepsilon}{2}t\right)$$

$$b_L = e^{i\delta t/2}\left(\cos\frac{\varepsilon}{2}t - i\frac{\delta}{\varepsilon}\sin\frac{\varepsilon}{2}t\right) = \frac{e^{i\delta t/2}}{2}\left(\left(1-\frac{\delta}{\varepsilon}\right)e^{i\frac{\varepsilon}{2}t} + \left(1+\frac{\delta}{\varepsilon}\right)e^{-i\frac{\varepsilon}{2}t}\right) \qquad \text{(3-30a, b)}$$

For the higher states using first order perturbation theory, consider the equation for the amplitude for state j:

$$i\hbar\dot{b}_j = \sum_{k=L,U} 2\hbar\Lambda_{jk}\, e^{i(\omega_{jk}-\omega)t}\, b_k(t) \quad + \quad \omega \leftrightarrow -\omega \qquad \text{(3-31)}$$

where

$$\Lambda_{jk} = \frac{-\langle j|e\vec{r}|k\rangle\cdot\vec{E}_0}{4\hbar} \qquad \text{(3-32)}$$

and

$$\omega_{ji} = (E_j - E_i)/\hbar \qquad \text{(3-33)}$$

b_j to lowest order in coupling constant, Λ_{jk} , is

$$b_j \approx -i \sum_k \int_0^t dt' \, 2\Lambda_{jk} e^{i(\omega_{jk} - \omega)t'} b_k(t') + \omega \leftrightarrow -\omega \tag{3-34}$$

The contribution to b_j from the lower state is

$$-\Lambda_{jL} \left\{ \left(1 - \frac{\delta}{\varepsilon}\right) \frac{e^{i(\omega_{jL} - \omega + \frac{\delta+\varepsilon}{2})t} - 1}{\omega_{jL} - \omega + \frac{\delta+\varepsilon}{2}} + \left(1 + \frac{\delta}{\varepsilon}\right) \frac{e^{i(\omega_{jL} - \omega + \frac{\delta-\varepsilon}{2})t} - 1}{\omega_{jL} - \omega + \frac{\delta-\varepsilon}{2}} \right. \tag{3-35}$$

$$+ \omega \leftrightarrow -\omega$$

The contribution to b_j from the upper state is

$$\frac{\Lambda_{jU}\Lambda}{\varepsilon} \left\{ \frac{e^{i(\omega_{jU} - \omega - \frac{\delta-\varepsilon}{2})T} - 1}{\omega_{jU} - \omega + \frac{\delta-\varepsilon}{2}} + \frac{e^{i(\omega_{jU} - \omega - \frac{\delta+\varepsilon}{2})t} - 1}{\omega_{jU} - \omega + \frac{\delta+\varepsilon}{2}} \right. \tag{3-36}$$

$$+ \omega \leftrightarrow -\omega$$

As in Eq. (3-5), the interaction strengths, $\hbar \Lambda_{jk}$, are assured to be small as compared to the energy difference of the levels. The laser is nearly tuned to the transition from the lower to the upper state ($L \to U$); the detuning parameter (δ) is less than Λ or of the order of Λ . To consider the terms of expression (3-35), notice that

$$\omega_{jL} - \omega = \omega_{jU} - \delta$$

Consequently, the denominations are of the order of an energy difference divided by $\hbar$ (i.e. ω_{jU}). Contributions from these terms are of the order $\frac{\Lambda_{jL}}{\omega_{jU}} \ll 1$.

The terms in Eq. (3-36), are insignificant only if $|\omega_{jU} - \omega| >> \delta, \Lambda, \Lambda_{jL}$.
The validity of a two level resonant approximation depends on the absence of higher "accidental" resonant states (see Fig. 5). Assuming the higher states are sufficiently off resonance that the detuning of the higher states (i.e. $\omega - \omega_{jU}$) is of the order of atomic energy-level differences, the terms of Eq. (3-35) are of order $\frac{\Lambda_{jL}}{\omega - \omega_{jU}} << 1$.

The corrections due to the higher states and the counter rotating terms are of the same order and must be included together if greater accuracy is desired.(omitting, again, the case of accidental resonances). The analysis to follow will be within the limits of Eq. (3-12); hereafter the RWA and resonant approximation are assumed.

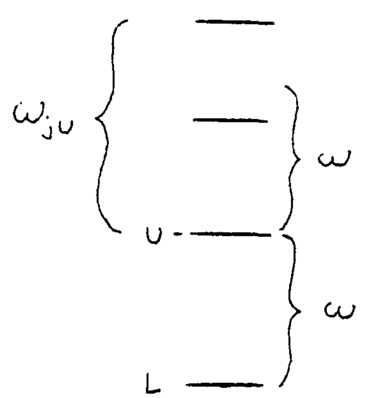

Fig. 5. Higher level structure with no 'accidental resonance'.

4. Radiative Damping

The creation of electromagnetic radiation is necessarily accompanied by an alteration of the state of the source; this is apparent from the consideration of conservation principles. However, a detailed description of the field and the state of the source as they evolve in time, requires an analysis of the equations of motion. In classical electrodynamics, there is Newton's Second Law, an equation for the particle's acceleration in response to the Lorentz Force, and Maxwell's differential equations, in which the effect of the sources upon the fields is a result of the inhomogeneous terms: $\rho(x,t)$ and $\frac{1}{c}J(x,t)$ In quantum electrodynamics, one can derive the analogous equations of motion for the 'Heisenberg operators' from the Hamiltonian and evaluate various expectation values with use of the initial state.

In both of these cases, the interdependent nature of the evolution of source and fields requires, in general, a simultaneous solution. However, in the case when the rate of energy loss due to radiation is slow, an expression for the radiation reaction force can be obtained and the equations of motion can be solved. For classical periodic motion the radiation reaction force is[42]:

$$F_{rad} = \frac{2}{3}\frac{e^2}{c^3}\ddot{v} \equiv m\tau\ddot{v} \qquad (4\text{-}1)$$

For $\tau\omega_a \ll 1$ Eq. (4-1) can be approximated by

$$F_{rad} = -m\omega_a^2\tau v \qquad (4\text{-}2)$$

Together with the external fields the problem reduces to the single second order differential equation

$$m\ddot{\vec{x}} = -m\omega_a^2 \tau \dot{\vec{x}} + \vec{E}_{ex} \tag{4-3}$$

The energy decay rate due to radiation is:

$$\Gamma = 2\omega_a^2 \tau \tag{4-4}$$

In quantum electrodynamics, when one works in the Schrodinger or interaction pictures, one starts with a single equation. However, it will be advantageous to derive a second equation that is the quantum analogy of Eq. (4-2). This will enable the elimination of the radiation operator in favor of its relation to particle operators. The treatment to be given will follow Mollow's.[2] Other treatments are given in Refs. 1, 3, 4, 30, 31.

The wave equation in the interaction picture with the rotating wave approximation is:

$$i\hbar \frac{\partial}{\partial t} |\Psi_I(t)\rangle = H_I(t) |\Psi_I(t)\rangle \tag{4-5}$$

with

$$H_I(t) = \frac{\hbar\Lambda}{2}\left(e^{-i\delta t}\sigma_+ + e^{i\delta t}\sigma_-\right) + \left(e^{i\omega_a t}\sigma_+ \vec{d}\cdot\sum_{k\lambda} \vec{\mathcal{E}}_{k\lambda} + e^{-i\omega_a t}\sigma_- \vec{d}\cdot\sum_{k\lambda} \vec{\mathcal{E}}^{\dagger}_{k\lambda}\right) \tag{4-6}$$

Formally

$$|\Psi_I(t)\rangle = T e^{\frac{1}{i\hbar}\int_0^t H_I(t')dt'} |\Psi(0)\rangle \equiv \sum_n |t\rangle_n \tag{4-7}$$

where T is the time ordering operator.

$$|t\rangle_0 = |\Psi(0)\rangle = |\Psi_{atom}(0)\rangle \prod_{k\lambda} |0_{k\lambda}\rangle_{field} \tag{4-8a}$$

and

$$|t\rangle_n = -\frac{i}{\hbar}\int_0^t H_I(t')\,dt'\,|t'\rangle_{n-1} \tag{4-8b}$$

Consider the effect of $\vec{d}\cdot\sum_{k\lambda}\vec{\mathcal{E}}_{k\lambda}(t)$ on $|t\rangle_n$. For $n > 0$, by Eq. (4-6) and (4-8b)

$$\vec{d}\cdot\sum_{k\lambda}\vec{\mathcal{E}}_{k\lambda}(t)|t\rangle_n = -\frac{i}{\hbar}\int_0^t dt'\sum_{k\lambda}\left[\vec{d}\cdot\vec{\mathcal{E}}_{k\lambda}(t),\ \vec{d}\cdot\vec{\mathcal{E}}^{\dagger}_{k\lambda}(t')\right]e^{-i\omega_a t'}\sigma_-|t'\rangle_{n-1}$$

$$-\frac{i}{\hbar}\int_0^t dt'\,H_I(t')\sum_{k\lambda}\vec{d}\cdot\vec{\mathcal{E}}_{k\lambda}(t)|t'\rangle_{n-1}$$

(4-9)

For $V\to\infty$

$$\frac{1}{V}\sum_{k\lambda}|d\cdot\epsilon_{k\lambda}|^2 \to \sum_{\lambda}\frac{1}{(2\pi)^3}\int |k|^2 d|k|\,d\Omega_k\,|\vec{d}\cdot\vec{\epsilon}_{k\lambda}|^2$$
$$= \frac{1}{(2\pi)^3}\int |k|^2 d|k|\,d\Omega_k\left(|d|^2 - |\vec{d}\cdot\hat{k}|^2\right)$$

one finds

$$\sum_{k\lambda}\left[\vec{d}\cdot\vec{\mathcal{E}}_{k\lambda}(t),\ \vec{d}\cdot\vec{\mathcal{E}}^{\dagger}(t')\right] = \frac{2}{3}\frac{d^2\hbar}{c^3\pi}\int_0^{W\simeq mc^2/\hbar} d\omega\ \omega^3 e^{-i\omega(t-t')} \tag{4-10}$$

where W is a cut-off reflecting the nonrelativistic nature of the treatment. Because Eq. (4-10) involves an expression which is highly peaked at $t = t'$, the first term of Eq. (4-9) car be approximated by:

$$-i \frac{2d^2}{3c^3\pi} \left\{ \int_0^t dt' \int_0^\infty d\omega \, \omega^3 e^{-i(\omega_a - \omega)t'} e^{-i\omega t} \right\} \sigma_- |+\rangle_{n-1} \tag{4-11}$$

For large time

$$\approx -i \frac{2d^2}{3\pi c^3} \left\{ \int_0^\infty d\omega \, \omega^3 \left(\pi \delta(\omega - \omega_a) - i \frac{P}{\omega_a - \omega} \right) e^{-i\omega t} \right\} \sigma_- |+\rangle_{n-1} \tag{4-12}$$

The second part represents the level shift. However, a proper calculation of the shift includes the upper states, renormalization and relativistic effects. A non-relativistic mass renormalization will account for the major part of the Lamb shift and can be included in the usual way (see Sakurei, Ref. 20, p. 70, for a description of Bethe's treatment of the Lamb shift). Our concern here is not with the level shift. Consequently, we will drop the latter part of Eq. (4-12) and consider ω_a to include the Lamb shift. The first term of Eq. (4-9) is, finally

$$-i \frac{2 \, d^2 \omega_a^3}{3 \, c^3} e^{-i\omega_a t} \sigma_- |+\rangle_{n-1} \equiv -i \hbar \gamma e^{-i\omega_a t} \sigma_- |+\rangle_{n-1} \tag{4-13}$$

The second term of Eq. (4-9) involves $\mathcal{E}(t) \, |t'\rangle_{n-1}$. From the analysis above we see that $\mathcal{E}(t) \, |t'\rangle_{n-1}$ is not appreciable until $t - t' \sim \frac{1}{\omega}$ (see Eq.(4-10)). A more detailed estimate will be given shortly. Consequently, the second term is of order $\frac{\Lambda}{\omega}$ smaller than the first term of $\vec{d} \cdot \sum_{k\lambda} \vec{\mathcal{E}}_{k\lambda}(+) \, |+\rangle_{n-1}$. Eq. (4-9) can now be expressed:

$$\vec{d} \cdot \sum_{k\lambda} \vec{\mathcal{E}}_{k\lambda}(+) |+\rangle_n = -i\hbar\gamma e^{-i\omega_a t} \sigma_- |+\rangle_{n-1} + O\left(\frac{\Lambda}{\omega}\right) \tag{4-14}$$

In addition

$$\vec{d} \cdot \sum_{k\lambda} \vec{\mathcal{E}}(+) |+\rangle_0 = 0 \tag{4-15}$$

A final summation gives:

$$\vec{d} \cdot \sum_{k\lambda} \vec{\mathcal{E}}_{k\lambda}(+) |+\rangle_0 + \sum_{n=1}^{\infty} \vec{d} \cdot \sum_{k\lambda} \vec{\mathcal{E}}_{k\lambda}(+) |+\rangle_n = -i\hbar\gamma e^{-i\omega_a t} \sigma_- \sum_{n=0}^{\infty} |+\rangle_{n-1}$$

$$\vec{d} \cdot \sum_{k\lambda} \vec{\mathcal{E}}_{k\lambda}(+) |+\rangle = -i\hbar\gamma\sigma_- e^{-i\omega t} |+\rangle \tag{4-16}$$

where

$$\gamma \equiv \frac{2}{3} \frac{d^2 \omega_a^3}{c^3 \hbar} \tag{4-17}$$

This relation (Eq. (4-16)) between the field operator ($\mathcal{E}$) and the atomic operator (σ_-) is the desired analogy of Eq. (4-2). Thus, the radiation absorption operator is related to the atomic current operator. The wave function must satisfy this equation as well as Schrodinger's equation. A solution involves the use of the radiative relation (Eq. (4-16)) to eliminate the absorption operator from Schrodinger's equation. We will see in detail how this operator equation is used in Section 5.

Now, let us return to the terms omitted in the above analysis to show in detail that they are indeed negligible. The first point of approximation is the replacement of the slowly varying function $|t'\rangle_{n-1}$ by $|t\rangle_{n-1}$ in Eq. (4-9) to get Eq. (4-11). $|t'\rangle$ is related to $|t\rangle$ as follows:

$$|t'\rangle = U^{-1}(t', t)|t\rangle \tag{4-18}$$

$$= \left(1 + \frac{i}{\hbar}\int_t^{t'} H_I(t'')dt'' + \cdots\right)|t\rangle \tag{4-19}$$

The next higher order correction to Eq. (4-11) is

$$-i\frac{2d^2}{3\pi c^3}\int_0^\infty d\omega\ \omega^3\int_0^t dt' e^{-i(\omega_a-\omega)t'}e^{i\omega t}\int_t^{t'}\frac{i}{\hbar}H_I(t'')dt''|t\rangle_{n-1} \tag{4-20}$$

The intensity of the laser field is much greater than the fluorescent field; thus, the dominant contribution comes from the first part of

$$H_I(t') \approx \frac{\hbar\Lambda}{2}\left(e^{-i\delta t'}\sigma_+ + e^{i\delta t'}\sigma_-\right) + \text{(non-laser terms)}$$

Since $\int_0^\infty d\omega\ \omega^3 e^{-i(\omega_a-\omega)t'}e^{i\omega t}$ is a highly peaked function at t' ~ t and δ is small compared to ω_a, the slowly varying function $H_I(t')$ can be evaluated at t' = t. Eq. (4-20) becomes:

$$-i\frac{2}{3}\frac{d^2}{c^3\pi}\frac{i}{\hbar}H_I(t)\int_0^\infty d\omega\ \omega^3\int_0^t dt'\ e^{-i(\omega_a-\omega)t'}e^{-i\omega t}(t'-t)|t\rangle_{n-1} \tag{4-21}$$

$$= \frac{2}{3}\frac{d^2}{c^3\pi\hbar}H_I(t)\int_0^\infty d\omega\ \omega^3\left(-i\frac{\partial}{\partial\omega}\right)\int_0^t dt' e^{-i(\omega_a-\omega)t'}e^{-i\omega t}|t\rangle_{n-1} \tag{4-22}$$

for long times

$$-i\,\frac{2}{3}\,\frac{d^2}{c^3\pi\hbar}\,H_I(t)\int_0^{\infty} d\omega\,\omega^3\,\frac{d}{d\omega}\left(\pi\delta(\omega-\omega_a)-iP\frac{1}{\omega_a-\omega}\right)e^{-i\omega t}\,|t\rangle_{n-1}$$

integration by parts yields

$$\frac{i\,2\,d^2}{c^3\pi\hbar}\,H_I(t)\int_0^{\infty} d\omega\;\omega^2\left(\pi\,\delta(\omega-\omega_a)-i\,P\,\frac{1}{\omega_a-\omega}\right)e^{-i\omega t}\,|t\rangle_{n-1} \qquad (4\text{-}23)$$

The added contribution to the radiative decay relation is

$$i\,\frac{2\,d^2\omega_a^3}{c^3}\;\frac{H_I(t)}{\hbar\,\omega_a}\;|t\rangle \qquad (4\text{-}24)$$

This is of an order $\frac{\Lambda}{\omega_a}$ smaller than Eq. (4-16). The shift contribution of Eq. (4-23) does not diverge as severely as that of Eq. (4-12). It also contributes an added term of higher order which can be ignored.

For the second term of Eq. (4-9), it is necessary to evaluate $\sum_{k\lambda}\vec{d}\cdot\vec{\mathcal{E}}_{k\lambda}(t)|t'\rangle$. This is just Eq. (4-9) with the dummy variable changed to t'' and the upper limit of the integrals change from t to t'. The first term of $\sum_{k\lambda}\vec{d}\cdot\vec{\mathcal{E}}_{k\lambda}(t)|t'\rangle$ is:

$$-\frac{i}{\hbar}\int_0^{t'} dt''\sum_{k\lambda}\left[\vec{d}\cdot\vec{\mathcal{E}}_{k\lambda}(t),\,\vec{d}\cdot\vec{\mathcal{E}}^{\dagger}_{k\lambda}(t'')\right]e^{-i\omega_a t''}\sigma_-|t''\rangle_{n-1} \qquad (4\text{-}25)$$

From Eq. (4-10) one finds

$$\sum_{k\lambda}\vec{d}\cdot\vec{\mathcal{E}}_{k\lambda}(t)|t'\rangle \approx -i\,\frac{2\,d^2}{3c^3\pi}\int_0^{t'} dt''\int_0^{\infty} d\omega\,\omega^3\,e^{-i(\omega_a-\omega)t''}\,e^{-i\omega t}\,\sigma_-|t''\rangle_{n-1}$$

(4-26)

The second term of Eq. (4-9) can be approximated by

$$-\frac{2}{3}\frac{d^2}{c^3\pi}\int_0^t dt'\,\frac{H_I(t')}{\hbar}\int_0^{t'} dt''\int_0^\infty d\omega\,\omega^3 e^{-i(\omega_a-\omega)t''}\,e^{-i\omega t}\,\sigma_-|t\rangle_{n-1} \qquad (4\text{-}27)$$

The ω integral produces a highly peak function of $t''-t$, thus, the t'' integration is only appreciable when t' nears t. This enables the slowly varying function $H_I(t')$ to be approximated by $H_I(t)$. Eq. (4-27) becomes

$$-\frac{2d^2}{3c^3\pi}\frac{H_I(t)}{\hbar}\int_0^t dt'\int_0^{t'} dt''\int_{-\omega_a}^\infty dW\,(W+\omega_a)^3\,e^{iWt''}e^{-i(W+\omega_a)t} \qquad (4\text{-}28)$$

where a change of integration variable has been made ($W=\omega-\omega_a$).

Upon time integrations:

$$-\frac{2d^2}{3c^3\pi}\frac{H_I(t)}{\hbar}\,e^{-i\omega_a t}\int_0^\infty dW(W+\omega_a)^3\,\frac{e^{iWt}-1-iWt}{-W^2}\,e^{-iWt}$$

$$=-\frac{2d^2}{3c^3\pi}\frac{H_I(t)}{\hbar}\,e^{-i\omega_a t}\int_0^\infty dW\,(W+\omega_a)^3\,\frac{\partial}{\partial W}\left(\frac{1-e^{-iWt}}{W}\right)$$

for long times

$$= -\frac{2 d^2 H_I(t)}{3 c^3 \pi \hbar} e^{-i\omega_a t} \int_0^\infty dW (W+\omega_a)^3 \frac{\partial}{\partial W} \left(i\pi \delta(W) - \frac{P}{W} \right)$$

integration by parts yields

$$\frac{2 i d^2 H_I(t)}{c^3 \pi \hbar} e^{-i\omega t} \int_0^\infty dW (W+\omega_a)^2 \left(\pi \delta(W) - i \frac{P}{W} \right) \tag{4-29}$$

The correction is equal to that of Eq. (4-23).

In Section 5, the four state problem will have an interaction Hamiltonian similar to that of Eq. (4-6) except for the operators $\sigma_\pm$ and $\vec{d}$. It will be shown that the operators $\frac{\vec{d}}{|d|} \sigma_\pm$ are replaced by a generalization which accounts for the various Clebsh-Gordon coefficient of the various transitions. These are the $\vec{Y}$ and $\vec{Y}^+$ operators of Eq. (5-9). The $\vec{Y}$ operator can be expressed in terms of polarization vectors $\hat{\epsilon}_j$ and lowering operators σ_-^j :

$$\vec{Y} = \sum_j \hat{\epsilon}_j \sigma_-^j \tag{4-30}$$

with $$\hat{\epsilon}_j \cdot \hat{\epsilon}_i^{\dagger} = \delta_{ij} \tag{4-31}$$

Consequently $$\left[\hat{\epsilon}_j \cdot \mathcal{E}_{k\lambda} , \hat{\epsilon}_i^{+} \cdot \mathcal{E}_{k\lambda} \right] = 0 \qquad i \neq j \tag{4-32}$$

One can now establish:

$$\forall_j : \quad |d| \, \hat{\epsilon}_j^{\,\dagger} \cdot \sum_{k\lambda} \mathcal{E}_{k\lambda}(t) = -i\hbar\gamma\sigma_-^{\,j} e^{-i\omega t} |t\rangle \tag{4-33}$$

or

$$|d| \sum_{k\lambda} \mathcal{E}_{k\lambda}(t) |t\rangle = \sum_j -i\hbar\gamma\hat{\epsilon}_j \sigma_-^{\,j} e^{-i\omega t} |t\rangle \tag{4-34}$$

and

$$|d| \sum_{k\lambda} \mathcal{E}_{k\lambda}(t) |t\rangle = -i\hbar\gamma \vec{Y} e^{-i\omega t} |t\rangle \tag{4-35}$$

This is the appropriate generalization for the four-state problems to be treated in the next section.

PART II

5. Fluorescence From an Atom with Degenerate Energy Levels

The effects of degeneracy on the spectrum can be investigated by consideration of a two level structure in which each level is doubly degenerate. An example of this type of structure within the Russell-Sanders coupling scheme is the $S_{1/2} \to P_{1/2}$ transition; in which the lower level is a $\ell = 0$ state coupled with a $S=1/2$ state and the upper level is $\ell = 1$ state coupled with a $S=1/2$ state. The latter coupling produces both $P_{3/2}$ and $P_{1/2}$ states. Because the laser pumps only to two of the upper states, the $S_{1/2} \to P_{3/2}$ transition can also be treated as a two level atom with only double degeneracy. The two cases differ only by different Clebsh-Gordon coefficients.

Another important case of a $J = 1/2$ to $J=1/2$ transition is found in the hyperfine structure consisting of a lower state resulting from the coupling of a ground state of two s electrons with a nuclear magnetic spin of 1/2. The upper state is the combination of the atomic ${}^1P_1$ and the nuclear spin.

The four states can be designated by the orbital angular momentum quantum number ℓ , and the magnetic quantum number of the total angular momentum m_j . For a $S_{1/2}$ to $P_{1/2}$ transition the four states, $|\ell, m_j\rangle$ are

$$
\begin{aligned}
|1, 1/2\rangle &= R_1(r)\left(\sqrt{2/3}\, Y_{1,1}\downarrow - \sqrt{1/3}\, Y_{1,0}\uparrow\right) \\
|0, 1/2\rangle &= R_0(r)\,(4\pi)^{-1/2}\uparrow \\
|1, -1/2\rangle &= R_1(r)\left(\sqrt{1/3}\, Y_{1,0}\downarrow - \sqrt{2/3}\, Y_{1,-1}\uparrow\right) \\
|0, -1/2\rangle &= R_0(r)\,(4\pi)^{-1/2}\downarrow
\end{aligned}
\qquad (5\text{-}1)
$$

where $\{Y_{\ell,m}\}$ are the spherical harmonics; $R_0(r)$ and $R_1(r)$ are the appropriate radial functions whose explicit form will not be considered, $\uparrow$ and $\downarrow$ are up and down spin states respectively. The four states of Eq. (5-1) are considered a complete basis set given the negligible probability of higher excitations (see Section 3). The wave functions can be expanded in these basis vectors as:

$$\psi = \sum_{\ell=0,1 \; m_j = \pm\frac{1}{2}} A_{\ell,m_j} \, e^{-iE_\ell t/\hbar} \, |\ell, m_j\rangle \tag{5-2}$$

A_{ℓ,m_j} includes the field state when the atom is in the state ℓ, m_j . Note that factorization of atom and field states is not assumed.

Schrodinger's equations can be multiplied by each $\langle \ell, m_j|$, yielding four equations for the A_{ℓ,m_j} . These four equations can be put into matrix form with the introduction of the vector

$$\psi = \begin{pmatrix} A_{1,\,1/2} \\ A_{0,\,1/2} \\ A_{1,\,-1/2} \\ A_{0,\,-1/2} \end{pmatrix} \tag{5-3}$$

The 4 x 4 matrices in the Hamiltonian can be expanded in any complete set of 16 matrices (see Appendix A for the familiar Dirac matrices). Without the interaction term, the Hamiltonian is

$$H_o = \frac{E_1 + E_o}{2} + \frac{E_1 - E_o}{2}\sigma_z + \sum_{k\lambda} \hbar\omega_{k\lambda} a^{\dagger}_{k\lambda} a_{k\lambda} \tag{5-4a}$$

where σ_z is a Dirac matrix.

The interaction Hamiltonian is (from 2-35)

$$H' = -e\vec{r}\cdot\vec{E}_o \cos\omega t - e\vec{r}\cdot(\mathcal{E} + \mathcal{E}^{\dagger}) \tag{5-4b}$$

where $\vec{E}_0 \cos\omega t$ is the applied laser field and $\mathcal{E}(\mathcal{E}^{\dagger})$ is the field destruction (creation) operator. To express the interaction Hamiltonian in matrix form, first the dipole matrix, with elements $\langle \ell\, m_j \,|\, e\vec{r} \,|\, \ell\, m_j \rangle$, is found. Using the relation

$$e\vec{r} = e\,r\sqrt{\frac{4\pi}{3}}\left(Y_{1,0}\,\hat{z} - Y_{1,1}\,\frac{\hat{x}-i\,\hat{y}}{\sqrt{2}} + Y_{1,-1}\,\frac{\hat{x}+i\hat{y}}{\sqrt{2}}\right) \tag{5-5}$$

and making use of the orthogonality of the $Y_{\ell,m}$'s

$$\int Y^{*}_{\ell m}(\theta,\phi)\, Y_{\ell' m'}(\theta',\phi')\, d\Omega = \delta_{\ell\ell'}\,\delta_{mm'}$$

the dipole matrix is:

$$\text{dipole matrix} = -d\begin{pmatrix} 0 & \hat{z} & 0 & \hat{x}-i\hat{y} \\ \hat{z} & 0 & \hat{x}-i\hat{y} & 0 \\ 0 & \hat{x}+i\hat{y} & 0 & -\hat{z} \\ \hat{x}+i\hat{y} & 0 & -\hat{z} & 0 \end{pmatrix} \tag{5-6}$$

where

$$d = \frac{1}{3} e \int_0^\infty R_0^*(r) R_1(r) r^3 \, dr \tag{5-7}$$

R_1 and R_o are each defined up to an arbitrary constant phase factor. These phase factors will be chosen to insure that d is real. The matrix of Eq. (5-6) operates on the vector of Eq. (5-3). The unit vectors in this matrix will be multiplied with the polarization vector of the field (see Eq. (5-4b) and Eq. (5-12a) below).

Eq. (5-6) can be expressed in terms of the Dirac matrices of Appendix A and written in the form

$$\text{dipole matrix} = -d\,(Y + Y^\dagger) \tag{5-8}$$

where $$Y = \hat{x}\rho\,\sigma_- - \hat{y}\,i\beta\rho\sigma_- + \hat{z}\,\beta\sigma_- \tag{5-9}$$

Y is the sum of the various lowering operators that arises in the evaluation of the dipole matrix for the states of Eqs. (5-1). As discussed in Section 3, the rotating wave approximation will be made by retention of the terms corresponding to approximate energy conservation. The matrix form of the interaction Hamiltonian after the RWA is made is

$$H' = \frac{\hbar\Delta}{2}\left(\beta\sigma_- e^{i\omega t} + \beta\sigma_+ e^{-i\omega t}\right) + d\left(Y^\dagger \cdot \mathcal{E} + \mathcal{E}^\dagger \cdot Y\right) \tag{5-10}$$

where

$$\Lambda = dE_0/\hbar \quad (\vec{E}_0 = \hat{z}E_0) \tag{5-11}$$

$\beta, \sigma_-, \sigma_+, \rho$ are Dirac matrices

$$\mathcal{E} = \sum_{k\lambda} i\, g_k\, \hat{\epsilon}_{k\lambda}\, a_{k\lambda} \tag{5-12a}$$

$$g_{k\lambda} = \sqrt{\frac{2\pi\hbar\omega_k}{V}} \tag{5-12b}$$

The first two terms of the interaction Hamiltonian express the laser coupling of the states of similar m_j. The last two terms involve operators for both linear polarized and circular polarized radiation. The dot product between Y^r and $\mathcal{E}$ is the product of the unit vectors of Eq. (5-9) with the polarization vector of the field ($\hat{\epsilon}_{k\lambda}$ of Eq. (5-12a)).

The goal is to find the spectrum in the long-time limit. The expectation value of the number operator, (i.e. $\langle\Psi(t)|a^{\dagger}_{k\lambda}a_{k\lambda}|\Psi(t)\rangle$) is related to the total number of quanta for the $k\lambda$ mode that have been emitted up to time t. To calculate the long-time limit spectrum one desires the time rate of change of $\langle\Psi(t)|\, a^{\dagger}_{k\lambda}\, a_{k\lambda}\,|\Psi(t)\rangle$ which gives the rate that light is being radiated into the $k\lambda$ mode. It will be shown that $\frac{d}{dt}\langle\Psi|\, a^{\dagger}_{k\lambda}\, a_{k\lambda}\,|\Psi\rangle$ approaches a constant for each k and λ in the long-time limit. Transient effects basically decay exponentially. (For slight deviations see Ref. 32).

The auxiliary radiative damping relation, as discussed in Section 4, is

$$d\,\vec{\mathcal{E}}\,\Psi = -i\hbar\gamma\,\vec{Y}\,\Psi \tag{5-13}$$

An evaluation of $\frac{d}{dt}\langle\Psi|\,a_{k\lambda}^{\dagger}\,a_{k\lambda}\,|\Psi\rangle$ will necessitate an evaluation of $\langle\Psi|\,a_{k\lambda}^{\dagger}\,\Gamma_i^{\mu}\,|\Psi\rangle$ (Γ_i^{μ} is one of the 16 matrices that span the space of atomic operators). With use of Eq. (5-13) and its hermitian conjugate, $\langle\Psi|\,a_{k\lambda}^{\dagger}\,\Gamma_i^{\mu}\,|\Psi\rangle$ can be related to the expectation values of atomic operators ($\langle\Psi|\Gamma_i^{\mu}|\Psi\rangle$). Reducing the problem of an evaluation of field operator matrix elements to the evaluation of atomic operator matrix elements is a crucial step in the simplification of the problem. It enables one to avoid an infinite set of coupled equations where each field matrix element for the $k\ \lambda$ mode is connected to all other modes. However, the joint effect of the continuum of field modes has been 'absorbed' in the damping relation (Eq. 5-13). These points will be illustrated in the calculation.

The 4 x 4 matrix operators can be expanded in terms of the 15 matrices given on the left hand side of Table 1 on page 50 and the unit matrix. (The unit matrix will not be explicitly written in the equations.) The operators $\frac{1 \pm \beta}{2}$ projects onto the $m_j = 1/2$ or $m_j = -1/2$ states:

$$\frac{1+\beta}{2} = \begin{pmatrix} 1 & & & \\ & 1 & & \\ & & 0 & \\ & & & 0 \end{pmatrix} \qquad \frac{1-\beta}{2} = \begin{pmatrix} 0 & & & \\ & 0 & & \\ & & 1 & \\ & & & 1 \end{pmatrix}$$

In set one of Table 1, operators involving $\sigma_\pm$ are raising or lowering operators for transitions of $\Delta m_j = 0$. It will be convenient to consider these operators with the time factor $e^{\pm i\omega t}$. Expectation values of these operators will be seen to be slowly varying. Operators with the σ_z matrix are 'probability difference' operators for the two states of particular m_j. The β operator is the 'probability difference' operator for states of $m_j = 1/2$ over states of $m_j = -1/2$. The ρ matrix is

$$\rho = \begin{pmatrix} & & & 1 \\ & & 1 & \\ & 1 & & \\ 1 & & & \end{pmatrix}$$

Operators which involve ρ are transition operators for transitions which change m_j. These transitions, referred to as cross transitions, are associated with the production of circular polarized fluorescent light $\left(\hat{\epsilon}_{k\pm} = \frac{\hat{x} \pm i \hat{y}}{\sqrt{2}} \right)$.

The operators σ_j, ρ, β obey the following rules of algebra:

$$\begin{aligned} [\sigma_i, \sigma_j] &= 2i\epsilon_{ijk}\sigma_k \\ [\rho, \sigma_i] &= 0 \\ [\beta, \sigma_i] &= 0 \\ \{\sigma_i, \sigma_j\} &= 2\delta_i^j \\ \{\rho, \beta\} &= 0 \end{aligned} \qquad (5\text{-}14)$$

where $[\ \ ]$ is the commutator and $\{\ \ \}$ is the anti-commutator.

The expectation values of field operators will be shown to depend on expectation values of the operators of Table 1; these expectation values will be found first.

Let Γ_ν^i stand for the ν th matrix of the i th set of Table 1.

$$i\hbar \frac{d}{dt}\langle\psi|\Gamma_\nu^i|\psi\rangle = \langle\psi|[\Gamma_\nu^i, H_0 + H']|\psi\rangle \tag{5-15}$$

$$i\hbar \frac{d}{dt}\langle\psi|\Gamma_\nu^i|\psi\rangle = \frac{(E_1 - E_0)}{2}\langle[\Gamma_\nu^i, \sigma_z]\rangle + \frac{\hbar\Lambda}{2}\langle[\Gamma_\nu^i, \beta\sigma_- e^{i\omega t} + \beta\sigma_+ e^{-i\omega t}]\rangle$$

$$+ d\langle[\Gamma_\nu^i, Y^\dagger\cdot\mathcal{E} + \mathcal{E}^\dagger\cdot Y]\rangle \tag{5-16}$$

The last term involves matrix elements of every field mode operator. Using Eq. (5-13) and its hermitian conjugate, the last term of Eq. (5-16) becomes:

$$i\hbar\gamma\langle Y^\dagger\cdot[\Gamma_\nu^i, Y] - [\Gamma_\nu^i, Y^\dagger]\cdot Y\rangle \tag{5-17}$$

(Y, $Y^\dagger$ and Γ_ν^i are matrices; the dot product, however, is between the unit vector $\hat{x}$, $\hat{y}$ and $\hat{z}$ of Y and $Y^\dagger$. (See Eq. (5-9).) Thus, the multimode continuum is accounted for by the introduction of the damping constant as expressed in Eq. (5-17). An infinite set of equations, index by the wave vector k and polarization λ, is thus avoided.

By use of Eq. (5-9), expression (5-17) is:

$$i\gamma\hbar\langle \rho\sigma_+[\Gamma_\mu^i, \rho\sigma_-] + i\beta\rho\sigma_+[\Gamma_\mu^i, i\beta\rho\sigma_-] + \beta\sigma_+[\Gamma_\mu^i, \beta\sigma_-]$$

$$- [\Gamma_\mu^i, \rho\sigma_+]\rho\sigma_- - [\Gamma_\mu^i, i\beta\rho\sigma_+]i\beta\rho\sigma_- - [\Gamma_\mu^i, \beta\sigma_+]\beta\sigma_- \rangle \qquad (5\text{-}18)$$

Using the rules of algebra of Eqs. (5-14), this expression is evaluated for each Γ_μ^i in Table 1 (on page 50).

Combining the results of Table 1 with the first two terms of Eq. (5-16), the following equations result:

$$i\frac{d}{dt}\langle \Gamma_\mu^1 \rangle = M_{\mu\nu}^1 \langle \Gamma_\nu^1 \rangle + A_\mu \qquad (5\text{-}19a)$$

$$i\frac{d}{dt}\langle \Gamma_\mu^2 \rangle = M_{\mu\nu}^2 \langle \Gamma_\nu^2 \rangle \qquad (5\text{-}19b)$$

$$i\frac{d}{dt}\langle \Gamma_\mu^3 \rangle = M_{\mu\nu}^3 \langle \Gamma_\nu^3 \rangle \qquad (5\text{-}19c)$$

where

$$M^1 = \begin{pmatrix} -\delta-3i\gamma & 0 & -\frac{\Lambda}{2} & 0 & 0 & 0 & 0 \\ 0 & \delta-3i\gamma & \frac{\Lambda}{2} & 0 & 0 & 0 & 0 \\ -\Lambda & \Lambda & -4i\gamma & -i\gamma & -2i\gamma & 0 & 0 \\ 0 & 0 & -4i\gamma & -4i\gamma & 4i\gamma & 0 & 0 \\ 0 & 0 & -2i\gamma & i\gamma & -4i\gamma & -\Lambda & \Lambda \\ 0 & 0 & 0 & 0 & -\frac{\Lambda}{2} & \delta-3i\gamma & 0 \\ 0 & 0 & 0 & 0 & \frac{\Lambda}{2} & 0 & -\delta-3i\gamma \end{pmatrix}$$

(5-20)

Table 1

For Γ_{μ}^{i}	The decay term is:
Set 1	
$\frac{1+\beta}{2}\sigma_{-}e^{i\omega t}$	$-3i\gamma\hbar\left\langle \frac{1+\beta}{2}\sigma_{-}e^{i\omega t}\right\rangle$
$\frac{1+\beta}{2}\sigma_{+}e^{-i\omega t}$	$-3i\gamma\hbar\left\langle \frac{1-\beta}{2}\sigma_{+}e^{-i\omega t}\right\rangle$
$\frac{1+\beta}{2}\sigma_{z}$	$-3i\gamma\hbar \quad -i\gamma\hbar\langle\beta\rangle - 4i\gamma\hbar\left\langle \frac{1+\beta}{2}\sigma_{z}\right\rangle - 2i\gamma\hbar\left\langle \frac{1-\beta}{2}\sigma_{z}\right\rangle$
β	$-4i\gamma\hbar\langle\beta\rangle - 4i\gamma\hbar\left\langle \frac{1+\beta}{2}\sigma_{z}\right\rangle + 4i\gamma\hbar\left\langle \frac{1-\beta}{2}\sigma_{z}\right\rangle$
$\frac{1-\beta}{2}\sigma_{z}$	$-3i\gamma\hbar + i\gamma\hbar\langle\beta\rangle - 4i\gamma\hbar\left\langle \frac{1-\beta}{2}\sigma_{z}\right\rangle - 2i\gamma\hbar\left\langle \frac{1+\beta}{2}\sigma_{z}\right\rangle$
$\frac{1-\beta}{2}\sigma_{+}e^{-i\omega t}$	$-3i\gamma\hbar\left\langle \frac{1-\beta}{2}\sigma_{+}e^{-i\omega t}\right\rangle$
$\frac{1-\beta}{2}\sigma_{-}e^{i\omega t}$	$-3i\gamma\hbar\left\langle \frac{1-\beta}{2}\sigma_{-}e^{i\omega t}\right\rangle$
Set 2	
$\frac{1+\beta}{2}\rho\sigma_{-}e^{i\omega t}$	$-3i\gamma\hbar\left\langle \frac{1+\beta}{2}\rho\sigma_{-}e^{i\omega t}\right\rangle$
$\frac{1+\beta}{2}\rho\sigma_{+}e^{-i\omega t}$	$-3i\gamma\hbar\left\langle \frac{1+\beta}{2}\rho\sigma_{+}e^{-i\omega t}\right\rangle$
$\frac{1+\beta}{2}\rho\sigma_{z}$	$-2i\gamma\hbar\left\langle \frac{1+\beta}{2}\rho\sigma_{z}\right\rangle - 2i\gamma\hbar\left\langle \frac{1+\beta}{2}\rho\right\rangle$
$\frac{1+\beta}{2}\rho$	$-4i\gamma\hbar\left\langle \frac{1+\beta}{2}\rho\right\rangle - 4i\gamma\hbar\left\langle \frac{1+\beta}{2}\rho\sigma_{z}\right\rangle$
Set 3	
$\frac{1-\beta}{2}\rho\sigma_{-}e^{i\omega t}$	$-3i\gamma\hbar\left\langle \frac{1-\beta}{2}\rho\sigma_{-}e^{i\omega t}\right\rangle$
$\frac{1-\beta}{2}\rho\sigma_{+}e^{-i\omega t}$	$-3i\gamma\hbar\left\langle \frac{1-\beta}{2}\rho\sigma_{+}e^{-i\omega t}\right\rangle$
$\frac{1-\beta}{2}\rho\sigma_{z}$	$-2i\gamma\hbar\left\langle \frac{1-\beta}{2}\rho\sigma_{z}\right\rangle - 2i\gamma\hbar\left\langle \frac{1-\beta}{2}\rho\right\rangle$
$\frac{1-\beta}{2}\rho$	$-4i\gamma\hbar\left\langle \frac{1-\beta}{2}\rho\right\rangle - 4i\gamma\hbar\left\langle \frac{1-\beta}{2}\rho\sigma_{z}\right\rangle$

$$M^2 = \begin{pmatrix} -\delta - 3i\gamma & 0 & 0 & -\frac{\Lambda}{2} \\ 0 & \delta - 3i\gamma & 0 & -\frac{\Lambda}{2} \\ 0 & 0 & -2i\gamma & -2i\gamma \\ -\Lambda & -\Lambda & -4i\gamma & -4i\gamma \end{pmatrix} \tag{5-21}$$

$$M^3(\Lambda) = M^2(-\Lambda) \tag{5-22}$$

$$A = -3i\gamma \begin{pmatrix} 0 \\ 0 \\ 1 \\ 0 \\ 1 \\ 0 \\ 0 \end{pmatrix} \tag{5-23}$$

$$\delta = \omega - (E_1 - E_0)/\hbar \tag{5-24a}$$

$$\Lambda = d\,E_0/\hbar \tag{5-24b}$$

The M^i are time independent; Eqs. (5-19) are Markovian differential equations. Dropping the subscripts, with the understanding that Γ^j stands for a 4 or 7 component vector, formal solutions for $\langle \Gamma^j \rangle$ are:

$$\langle \Gamma^j \rangle = e^{-i\overleftrightarrow{M}^j t} \langle \Gamma^j \rangle_0 - \delta_j^1 \frac{1}{\overleftrightarrow{M}^1} \cdot \vec{A} \tag{5-25}$$

where $\langle \Gamma^j \rangle_v$ depends on the initial conditions. To understand the nature of the solutions it is necessary to find the roots of the equation

$$\det | \lambda - M^j | = 0 \tag{5-26}$$

These roots describe the oscillatory and decay-like nature of the time dependent part of the solutions. The roots for M^2 are found from the equation:

$$\det | \lambda - M^2 | = 0$$

$$y^4 - \varepsilon^2 y^2 + \Lambda^2 i\gamma y - 9\gamma^2\delta^2 = 0 \tag{5-27}$$

where

$$y = \lambda + 3i\gamma \tag{5-28}$$

and

$$\varepsilon = \sqrt{\Lambda^2 + \delta^2} \tag{5-29}$$

(Terms of order $\frac{\gamma^2}{\varepsilon^2}$ have been dropped; interest is in the strong field case $\gamma \ll \Lambda$ $(\Rightarrow \gamma \ll \varepsilon)$).

The solutions to Eq. (5-27) in power series in γ are:

$$y_{1,2} = i\gamma \left(\frac{\Lambda^2 \pm \sqrt{\Lambda^4 + 36\delta^2\varepsilon^2}}{2\varepsilon^2} \right) + O\left(\frac{\gamma^2}{\varepsilon} \right) \tag{5-30a, b}$$

$$y_{3,4} = \pm \varepsilon - i\gamma \left(\frac{\Lambda^2}{2\varepsilon^2} \right) + O\left(\frac{\gamma^2}{\varepsilon} \right) \tag{5-30c, d}$$

For finite δ , $\mathrm{Im}\,\chi < 0$ and e^{-iM^2t} consequently decays to zero as $t \to \infty$. The initial state can be expanded in the eigenfunctions of M^2 (the four eigenfunctions are orthogonal).

$$\langle \Gamma^j \rangle_0 = \sum_\ell a_\ell Z_\ell$$

where

$$M^2 Z_\ell = \chi_\ell Z_\ell$$

and

$$\chi_\ell = -3i\gamma + y_\ell$$

Thus,

$$e^{-iM^2t} \langle \Gamma^2 \rangle_0 = \sum_\ell e^{-i\chi_\ell t} a_\ell \to 0$$

The roots of det $|\chi - M^3|$ are the same as the roots of det $|\chi - M^2|$ since the determinant is unchanged by the transformation $\Lambda \to -\Lambda$ (see Eqs. (5-21), (5-22) and (5-27)).

The roots of M^1 will be shown to be equivalent to the roots of the two matrices of Eq. (5-31). From inspection of M^1, it is clear that det $|\chi - M'|$ is independent of the sign of Λ ; hence, the sign of the Λ terms will be dropped. Next, consider the transformation, L, to the basis set $\left\{ \frac{\Gamma_1' + \Gamma_7'}{2} ; \frac{\Gamma_2' + \Gamma_6'}{2} ; \frac{\Gamma_3' + \Gamma_5'}{2} ; \Gamma_4 ; \frac{\Gamma_3' - \Gamma_5'}{2} ; \frac{\Gamma_2' - \Gamma_6'}{2} ; \frac{\Gamma_1' - \Gamma_7'}{2} \right\}$

Such a transformation has an inverse, L^{-1}. Since det $|L^{-1}(\chi - M')L| =$ det $|\chi - M'|$

the eigenvalues of the transformed matrix $L^{-1} M^{1} L$ and the eigenvalues of M^{1} are the same. By simple addition and subtraction of the equations for the $\{\Gamma'_{\mu}\}$, the transformed matrix is found to be:

$$L\, M^{1}\, L^{-1} = \begin{pmatrix} -\delta - 3i\gamma & 0 & \frac{\Lambda}{2} & & & & \\ 0 & \delta + 3i\gamma & \frac{\Lambda}{2} & & & & \\ \Lambda & \Lambda & -6i\gamma & & & & \\ & & & -4i\gamma & -4i\gamma & 0 & 0 \\ & & & -2i\gamma & -2i\gamma & \Lambda & \Lambda \\ & & & 0 & \frac{\Lambda}{2} & \delta - 3i\gamma & 0 \\ & & & 0 & \frac{\Lambda}{2} & 0 & -\delta - 3i\gamma \end{pmatrix} \tag{5-31}$$

A simple calculation of the secular equation for the eigenvalues of the 4 x 4 sub-determinant reveals that the secular equation itself and thus the roots are the same as in the case of M^2 provided the substitution $\Lambda^2 \rightarrow -\Lambda^2$ is made (except within ε).

The roots for the 3 x 3 sub-determinant are given by the equation

$$y^3 + 3\, i\, \gamma\, y^2 - \varepsilon^2 y - 3\, i\, \gamma = 0 \tag{5-32}$$

where $\quad y = \chi + 3i\gamma \qquad (\chi$ - the root)

Again, in an expansion in powers of γ, the solutions, y_j are:

$$y_1 = -i\gamma\left(\frac{3\delta^2}{\varepsilon^2}\right) \tag{5-33a}$$

$$y_{2,3} = \pm\varepsilon - i\gamma\left(\frac{3\Delta^2}{2\varepsilon^2}\right) \tag{5-33b, c}$$

All seven roots of M^1 describe decay phenomena.

As seen from Eq. (5-25), the decay-like eigenvalues of all three matrices insures the decreasing effect of the initial conditions on the state of the system as time evolves. The solution for $\langle\Gamma'\rangle$ in the long-time limit is $-\frac{1}{M'}\cdot A$

$$\langle\Gamma'\rangle_{t\to\infty} = \frac{1}{\varepsilon^2+\delta^2+18\gamma^2}\begin{pmatrix} \frac{\Delta}{2}(\delta-3i\gamma) \\ \frac{\Delta}{2}(\delta+3i\gamma) \\ -\delta^2-9\gamma^2 \\ 0 \\ -\delta^2-9\gamma^2 \\ -\frac{\Delta}{2}(\delta+3i\gamma) \\ -\frac{\Delta}{2}(\delta-3i\gamma) \end{pmatrix} \tag{5-34}$$

Thus,

$$\left|A_{1,1/2}\right|^2 - \left|A_{0,1/2}\right|^2 = \frac{-\delta^2-9\gamma^2}{\varepsilon^2+\delta^2+18\gamma^2} \tag{5-35a}$$

$$\left|A_{1,-1/2}\right|^2 - \left|A_{0,-1/2}\right|^2 = \frac{-\delta^2-9\gamma^2}{\varepsilon^2+\delta^2+18\gamma^2} \tag{5-35b}$$

$$|A_{1,1/2}|^2 + |A_{0,1/2}|^2 = |A_{1,-1/2}|^2 + |A_{0,-1/2}|^2 \tag{5-35c}$$

Equations (5-35a) and (5-35b) show how the parameters for laser pumping ($\mathcal{E}$) detuning (δ) and spontaneous emission (γ) effect the equalization of the probabilities of upper and lower levels. The probability for occupation of the upper level is diminished by detuning and equalization of probabilities is prohibited by spontaneous processes. The maximum excitation occurs for $\delta = 0$ the difference in probability is $-\left(\frac{3\gamma}{\Lambda}\right)^2$ for strong fields ($\gamma << \Lambda$). Eq. (5-35c) expresses the equal probability for the $m_j = 1/2$ states and $m_j = -1/2$ states, as expected since the two sets of equivalent states are pumped equally and spontaneous processes effect both sets equally.

Consider, now $i\frac{d}{dt}\langle\Psi(t)|a^{+}_{k\lambda}a_{k\lambda}|\Psi(t)\rangle$

$$i\frac{d}{dt}\langle\Psi(t)|a^{+}_{k\lambda}a_{k\lambda}|\Psi(t)\rangle = \frac{1}{\hbar}\langle\Psi(t)|[a^{+}_{k\lambda}a_{k\lambda}, H_0 + H']|\Psi(t)\rangle \tag{5-36}$$

$$= -\frac{2idg_k}{\hbar}\mathcal{R}e\langle\Psi(t)|a^{+}_{k\lambda}\,Y\cdot\epsilon^{+}_{k\lambda}|\Psi(t)\rangle \tag{5-37}$$

By virtue of the definition of Y (see Eq. (5-9)) the right hand side of Eq. (5-37) is a sum of terms of the form $\langle\Psi(t)|e^{-i\omega t}a^{+}_{k\lambda}\Gamma_\mu{}'|\Psi(t)\rangle$

For an evaluation of these expectation values consider

$$i\hbar\frac{d}{dt}\langle\Psi(t)|e^{-i\omega t}a^{+}_{k\lambda}\Gamma_\mu{}'|\Psi(t)\rangle =$$

$$\langle\Psi(t)|[e^{-i\omega t}a^{+}_{k\lambda}\Gamma_\mu{}', H_0 + H'] + i\omega e^{-i\omega t}a^{+}_{k\lambda}\Gamma_\mu{}'|\Psi(t)\rangle \tag{5-38a}$$

By the identity $[AB\,,\,C] = A[B,C] + [A,C]B$ one finds:

$$i\hbar \frac{d}{dt}\langle e^{-i\omega t} a^{\dagger}_{k\lambda} \Gamma^{i}_{\mu}\rangle = \langle e^{-i\omega t} a^{\dagger}_{k\lambda}[\Gamma^{i}_{\mu}, H_0 + H']$$

$$+ [e^{-i\omega t} a^{\dagger}_{k\lambda}, H_0 + H']\Gamma^{i}_{\mu} + \hbar\omega\, e^{-i\omega t} a^{\dagger}_{k\lambda} \Gamma^{i}_{\mu}\rangle \tag{5-38b}$$

where the explicit reference to the state $\Psi(t)$ is omitted. By use of the rules of algebra for the matrices and field operators, together with the expressions for H_0 (Eq. (5-4a)) and H' (Eq. (5-10)) one finds:

$$i\frac{d}{dt}\langle e^{-i\omega t} a^{\dagger}_{k\lambda} \Gamma^{i}_{\mu}\rangle = M^{i}_{\mu\nu}\langle e^{-i\omega t} a^{\dagger}_{k\lambda} \Gamma^{i}_{\mu}\rangle + \delta^{1}_{i} A_{\mu}\langle e^{-i\omega t} a^{\dagger}_{k\lambda}\rangle$$

$$+ (\omega - \omega_k)\langle e^{-i\omega t} a^{\dagger}_{k\lambda} \Gamma^{i}_{\mu}\rangle - \frac{i\, d\, g_k}{\hbar}\hat{\epsilon}_{k\lambda}\cdot\langle e^{-i\omega t}\gamma^{\dagger}\Gamma^{i}_{\mu}\rangle \tag{5-39}$$

The first two terms of Eq. (5-39) were derived in the same way as Eqs. (5-19a-c) were derived. Indeed, the commutator of the first term of Eq. (5-38b) is the same as that of Eq. (5-15).

Define the frequency of the fluorescent light relative to the laser frequency:

$$\chi_k = \omega_k - \omega \tag{5-40}$$

By integration of Eq. (5-39)

$$\langle e^{-i\omega t} a^{\dagger}_{k\lambda} \Gamma^{i}\rangle = e^{i(\chi_k - M^{i})t}\langle a^{\dagger}_{k\lambda}\Gamma^{i}\rangle_0$$

$$+ \int_0^t dt'\, e^{i(\chi_k - M^{i})(t-t')}\Big(-\frac{d g_k}{\hbar}\hat{\epsilon}_{k\lambda}\cdot\langle\Psi(t')|e^{-i\omega t'}\gamma^{\dagger}\Gamma^{i}|\Psi(0)\rangle +$$

$$\delta^{1}_{i} A\langle\Psi(t')|e^{-i\omega t'} a^{\dagger}_{k\lambda}|\Psi(0)\rangle \tag{5-41}$$

The first term depends on the initial conditions and decays to zero due to the nature of the eigenvalues of M .

The particular term, $\langle e^{-i\omega t} a_{k\lambda}^{\dagger} \Gamma_{\nu}^{i} \rangle$, necessary to evaluate Eq. (5-37) depends on the polarization. Consider first, the polarization $\epsilon_{k-} = \frac{\hat{x} - i\hat{y}}{\sqrt{2}}$

From Eq. (5-9)

$$\epsilon_{k-} \cdot \Upsilon^{\dagger} = \sqrt{2}\, \frac{1-\beta}{2}\, \rho\, \sigma_{+} \tag{5-42a}$$

$$\epsilon_{k-}^{*} \cdot \Upsilon = \sqrt{2}\, \frac{1+\beta}{2}\, \rho\, \sigma_{-} \tag{5-42b}$$

For the polarization under consideration, Eq. (5-37) becomes:

$$\frac{d}{dt} \langle \Psi(t) | a_{k-}^{\dagger} a_{k-} | \Psi(t) \rangle = -2\sqrt{2}\, \frac{d g_k}{\hbar}\, \mathcal{R}e \langle \Psi(t) | a_{k\lambda}^{\dagger} e^{-i\omega t} \Gamma_{1}^{2} | \Psi(t) \rangle \tag{5-43}$$

Define the unit vector $\hat{a}_1^{\dagger} = (1, 0, 0, 0)$ which operates on the vector Γ^2 . From Eq. (5-41), and (5-42a), for long times,

$$\frac{d}{dt} \langle \Psi(t) | a_{k-}^{\dagger} a_{k-} | \Psi(t) \rangle \approx$$

$$\frac{4 d^2 g_k^2}{\hbar^2} \mathcal{R}e\, \hat{a}_1^{\dagger} \int_0^t dt'\, e^{i(\alpha_k - M^2)(t - t')} \left\langle \Psi(t') \middle| e^{-i\omega t'} \frac{1-\beta}{2} \rho \sigma_{+} \Gamma^2 \middle| \Psi(t') \right\rangle \tag{5-44}$$

With use of Table 1

$$\left\langle \frac{1-\beta}{2} \rho \sigma_+ e^{-i\omega t'} \Gamma^2 \right\rangle = \begin{pmatrix} \frac{1}{4} - \frac{1}{4}\langle \beta \rangle + \frac{1}{2}\left\langle \frac{1-\beta}{2} \sigma_z \right\rangle \\ 0 \\ -\left\langle \frac{1-\beta}{2} \sigma_+ e^{-i\omega t'} \right\rangle \\ \left\langle \frac{1-\beta}{2} \sigma_+ e^{-i\omega t'} \right\rangle \end{pmatrix} \tag{5-45}$$

The entries of this vector are elements of set 1. Eq. (5-45) can be expressed:

$$\left\langle \frac{1-\beta}{2} \rho \sigma_+ e^{-i\omega t'} \Gamma^2 \right\rangle = N\left(e^{-iM't'} \langle \Gamma' \rangle_0 + \langle \Gamma' \rangle_{t=\infty} \right) + \begin{pmatrix} 1/4 \\ 0 \\ 0 \\ 0 \end{pmatrix} \tag{5-46}$$

where

$$N = \begin{pmatrix} 0 & 0 & 0 & -1/4 & 1/2 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & -1 & 0 \\ 0 & 0 & 0 & 0 & 0 & 1 & 0 \end{pmatrix} \tag{5-47}$$

$\langle \Gamma' \rangle_0$ depends on the initial conditions. $\langle \Gamma' \rangle_{t=\infty}$ is given by equation (5-34). The term which depends on the initial condition contains the integral:

$$\int_0^t dt' \, e^{i(\chi_k - M^2)(t-t')} N e^{-iM't'} \langle \Gamma' \rangle_0 \tag{5-48}$$

$\langle \Gamma' \rangle_0$ can be expanded in terms of the eigenfunctions of M'

$$\langle \Gamma^1 \rangle_0 = \sum_{m=1}^{7} a_m Z^1_m \tag{5-49}$$

where Z_m satisfy the equation

$$M^j Z^j_m = \chi^j_m Z^j_m \tag{5-50}$$

thus,

$$e^{-iM^1 t'} \langle \Gamma^1 \rangle_0 = \sum_m e^{-i\chi^1_m t'} a_m Z^1_m \tag{5-51}$$

Expand $N Z^1_m$ in terms of the eigenfunction of M^2:

$$N Z^1_m = \sum_{n=1}^{4} b^m_n Z^2_n \tag{5-52}$$

Eq. (5-48) is now

$$\sum_{m=1}^{7} \sum_{n=1}^{4} b^m_n a_m \int_0^t dt' \, e^{i(\chi_k - \chi_n^{(2)})(t-t')} e^{-i\chi_m^{(1)} t'} Z^2_n$$

$$= \sum_{mn} b^m_n a_m \frac{e^{-i\chi_m^{(1)} t} - e^{i(\chi_k - \chi_n^{(2)}) t}}{i[-\chi_k + (\chi_n^{(2)} - \chi_m^{(1)})]} Z_n^{(2)} \tag{5-53}$$

An examination of the eigenvalues (see Eqs. (5-30) and (5-33)) reveals that

$\mathrm{Im}(\chi_n^{(2)} - \chi_m^{(1)}) \neq 0$ for all n and m provided $\Lambda \neq 0$.

Consequently, the denominator is non-vanishing. Due to the decay-like nature of the roots, (5-53) vanishes as $t \to \infty$. One is left with

$$\frac{d}{dt}\langle a_{k-}^{+} a_{k-}\rangle = \frac{4d^2 g_k^2}{\hbar^2} \mathcal{R}e\, \hat{a}_1^{+} \int_0^t dt' e^{i(\chi_k - M)^2 (t-t')} \begin{pmatrix} \frac{1}{4} - \frac{1}{4}\langle \beta \rangle_\infty + \frac{1}{2}\left\langle \frac{1-\beta}{2}\sigma_3 \right\rangle_\infty \\ 0 \\ -\left\langle \frac{1-\beta}{2}\sigma_+ e^{-i\omega t} \right\rangle_\infty \\ \left\langle \frac{1-\beta}{2}\sigma_+ e^{-i\omega t} \right\rangle_\infty \end{pmatrix} \tag{5-54}$$

From equation (5-34) and after integration:

$$\left.\frac{d}{dt}\langle a_{k-}^{+} a_{k-}\rangle\right|_{t\to\infty} = -\frac{4d^2 g_k^2}{\hbar^2(\varepsilon^2 + \delta^2 + 18\gamma^2)} \mathcal{I}m\, a_1^{+} \cdot \frac{1}{\chi_k - M^2} \begin{pmatrix} \Lambda^2/4 \\ 0 \\ \frac{\Lambda}{2}(\delta + 3i\gamma) \\ -\frac{\Lambda}{2}(\delta + 3i\gamma) \end{pmatrix} \tag{5-55}$$

$$= -\frac{d^2 g_k^2 \Lambda^2}{\hbar^2(\varepsilon^2 + \delta^2 + 18\gamma^2)} \mathcal{I}m \frac{\chi_k^3 + 12i\gamma\chi_k^2 - (\delta^2 + \Lambda^2/2)\chi_k - i\gamma(\Lambda^2 + 6\delta^2)}{\det|\chi_k - M^2|} \tag{5-56}$$

$$= \frac{d^2 g_k^2 \Lambda^4 \gamma}{\hbar^2(\varepsilon^2 + \delta^2 + 18\gamma^2)} \frac{2\chi_k^4 + (\frac{3}{2}\Lambda^2 + \delta^2)\chi_k^2 + 6\gamma^2(\Lambda^2 + 6\delta^2)}{(\chi_k^2 + \Gamma_1^2)(\chi_k^2 + \Gamma_2^2)((\chi_k - \varepsilon)^2 + \Gamma_3^2)((\chi_k + \varepsilon)^2 + \Gamma_3^2)} \tag{5-57}$$

where

$$\Gamma_{1,2} = \gamma\left(3 - \frac{\Lambda^2 \pm \sqrt{\Lambda^4 + 36\delta^2\varepsilon^2}}{2\varepsilon^2}\right)$$

$$\Gamma_3 = \gamma\left(3 + \frac{\Lambda^2}{2\varepsilon^2}\right) \tag{5-58}$$

The ratio of either side peak to center peak is

$$\frac{3\Lambda^4}{(7\Lambda^2 + 6\delta^2)(\Lambda^2 + 6\delta^2)} \tag{5-59}$$

which on resonance is $3/7$.

For the polarization $\epsilon_{k+} = \frac{\hat{X} + i\hat{Y}}{\sqrt{2}}$ the results are trivially the same. In Eq. (5-54) M^2 is replaced by M^3 and β by $-\beta$. Both changes result in $\Lambda \to -\Lambda$, however, Eq. (5-55) is again the result. This is expected from the symmetry of the problem since initial conditions are lost.

The results (Eqs. (5-58) and (5-59)) agree with those obtained by different methods[33]. The above approach is easily generalized for greater degeneracy[34] and can be generalized to include magnetic fields as will be shown in the next section.

6. Strong Field Resonant Fluorescence from an Atom Subjected to a Constant Magnetic Field

The level structure of an atom is modified by the introduction of a constant magnetic field. Thus, one can continuously vary the structure in which to investigate the multipeak fluorescence spectrum resultant from intense radiation.

A constant magnetic field, $\vec{H}$, can be represented by the vector potential

$$\vec{A}_H = \frac{1}{2}\vec{H} \times \vec{r} \tag{6-1}$$

The total vector potential is the sum of A_H and the vector potential describing the quantized field. The terms in the Hamiltonian involving the vector potentials are

$$\frac{e}{mc}\,(\vec{A}_H + \vec{A}^{\perp})\cdot p \;+\; \frac{e^2}{2mc^2}\,(\vec{A}_H + \vec{A}^{\perp})^2 \tag{6-2}$$

With Eq. (6-1) the first term can be written:

$$\frac{e}{2mc}\,\vec{H}\cdot\vec{L} \;+\; \frac{e}{mc}\,A^{\perp}\cdot p \tag{6-3}$$

The second term of this expression has been dealt with in Section 2. With the inclusion of spin, the term

$$-\frac{e\hbar}{mc}\,S\cdot\nabla X(\vec{A}_H + \vec{A}^{\perp}) \tag{6-4}$$

is added (see p. 78 of Ref. 20). The second part of this term is of the order of

$$\left(\frac{e\hbar}{mc}\right)E_0 \sim \frac{v}{c}\,(e\,r_0\,E_0) \;=\; \frac{v}{c}\,\hbar\,\Lambda \tag{6-5}$$

This term can be dropped since velocities are small enough to warrant the non-relativistic treatment. The first part of Eq. (6-4) can be combined with the first part of Eq. (6-3) to give:

$$H_{mag} = \frac{e}{2mc}\,\vec{H}\cdot(\vec{L} + 2\vec{S}\,) \tag{6-6}$$

This completes the analysis of the first term of Eq. (6-2).

The second term of Eq. (6-2) can be written as:

$$\frac{e^2}{8mc^2}(\vec{H}\times\vec{r}\,)\cdot(\vec{H}\times\vec{R}\,) \;+\; \frac{e^2}{mc^2}A_H\,A^{\perp} \;+\; \frac{e^2}{mc^2}\,A^{\perp 2} \tag{6-7}$$

The last term has been dealt with in Section 2. The term, $\frac{e^2}{mc^2}\,A_H\,A^{\perp}$ can be shown to be small as compared to the term $\frac{e}{mc}\,A^{\perp}p$. Consider

$$\frac{\frac{e^2}{mc^2}\,\frac{1}{2}\,(\vec{H}\times\vec{r}_{ab})_j}{\frac{e}{mc}\,(p_{ab})_j} \tag{6-8}$$

P_{ab} can be related to r_{ab}.

$$p \;=\; \frac{1}{-2i\hbar}\left[p^2, r\right] = \frac{m}{-i\hbar}\left[H_{atom}, r\right]$$

Thus

$$p_{ab} \;=\; im\omega_{ab}r_{ab} \tag{6-9}$$

Eq. (6-4) can be written:

$$\frac{\mu_B(\vec{H} \times r_{ab})_j}{i\hbar\,\omega_{ab}(r_{ab})_j} \tag{6-10}$$

where μ_B is the Bohr magneton

$$\mu_B = \frac{eh}{2mc} \tag{6-11}$$

With $H = H_z$ and $|(r_{ab})_x| = |(r_{ab})_y|$ (see Eq. (5-6))

The terms of Eq. (6-5) are small provi d ed:

$$\frac{\mu_B H}{h\omega_{ab}} \ll 1 \tag{6-12}$$

This condition expresses the relation of the energy shift due to the magnetic field (i.e. $\mu_B H$) and the energy difference of an optical transition ($\hbar\omega_{ab}$). The energy shifts of concern are of the order of $2h\Lambda$, which is the total 'width' of the spectrum in energy units. Magnetic field strengths are of the order

$$H \sim \frac{2\hbar\Lambda}{\mu_B} = \frac{2\hbar e r_{ab} E_0}{\mu_B} \tag{6-13}$$

The first term of Eq. (6-7) is of the order

$$\frac{e^2}{8mc^2} H^2(x^2 + y^2) \sim 2m\Lambda^2 r_{ab}^2 \tag{6-14}$$

compared to $2h\Lambda$

$$\frac{m\Lambda r_{ab}^2}{\hbar} \sim \frac{\Lambda}{\omega_{ab}} \ll 1 \tag{6-15}$$

This concludes consideration and elimination of (6-7). The remaining magnetic terms in the Hamiltonian are given in Eq. (6-6). By evaluating the expression $\langle \ell', m_j' | H_{mag} | \ell, m_j \rangle$ with the states of Eq. (5-1) one arrives at

$$H'_{m} = \hbar\Delta(\beta - \tfrac{1}{2}\beta\sigma_z) \tag{6-16}$$

where

$$\hbar\Delta = \tfrac{2}{3}\mu_B H_z \tag{6-17}$$

Eq. (6-16) is an expression for the Hamiltonian due to a magnetic field in the $\hat{z}$ direction, the direction defined by the electric field component of the laser field. The effect of the magnetic field is to shift the atomic states. The magnetic field components in the $\hat{x}$ and $\hat{y}$ directions can be expressed

$$H_{xy} = (\hat{x} + i\hat{y})\left(\frac{H_x - iH_y}{2}\right) + (\hat{x} - i\hat{y})\left(\frac{H_x + iH_y}{2}\right)$$

The Hamiltonian is

$$H'' = \frac{\mu_B}{\hbar}\left[(J_+ + S_+)\left(\frac{H_x - iH_y}{2}\right) + (J_- + S_-)\left(\frac{H_x + iH_y}{2}\right)\right]$$

In matrix form, the Hamiltonian is

$$\begin{pmatrix} 0 & 0 & \frac{1}{3}\mu_B(H_x - iH_y) & 0 \\ 0 & 0 & 0 & \mu_B(H_x - iH_y) \\ \frac{1}{3}\mu_B(H_x + iH_y) & 0 & 0 & 0 \\ 0 & \mu_B(H_x + iH_y) & 0 & 0 \end{pmatrix}$$

or

$$H = \frac{\mu_B}{3} (2 \rho H_x - \rho \sigma_z H_x - 2i \beta \rho H_y + i \beta \rho \sigma_z H_y)$$

This Hamiltonian would couple the 15 matrices of Table 1. The enegies $\mu_B H_y$ and $\mu_B H_x$ will be restricted:

$$\mu_B H_y , \mu_B H_x << \hbar \gamma$$

With the addition of Eq. (6-16), the energy diagram is as shown in Fig. 6.

The radiative damping relationship,

$$d \vec{\mathcal{E}} \Psi = -i \hbar \gamma \vec{Y} \Psi \tag{6-18}$$

is still valid. This can be seen in two ways. In the derivation of Eq. (6-18) the wave equation in the interaction picture is considered and the formal solution is shown to satisfy the radiative damping relation (Eq. 6-18). One can transform to the interaction picture with the unperturbed atomic Hamiltonian and free field

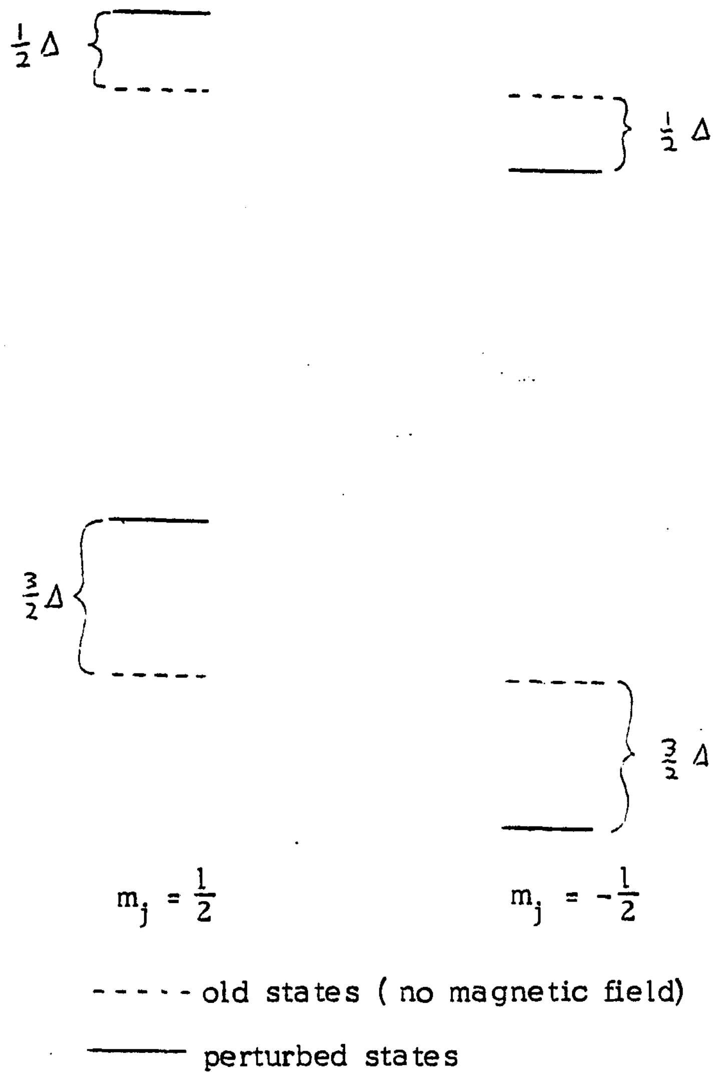

Fig. 6. Perturbed atomic states.

radiation Hamiltonian (i.e. $\sum_{k\lambda} \hbar \omega_{k\lambda} a^{\dagger}_{k\lambda} a_{k\lambda}$), as before, and obtain

$$H' + H_m \tag{6-19}$$

where H' is given in Eq. (5-10) and H_m in Eq. (6-16). Eq. (4-9) is replaced by

$$\hat{\epsilon}_j^* \cdot \sum \mathcal{E}_{k\lambda}(t)|t\rangle_n = -\frac{i}{\hbar}\int_0^t dt' \sum_{k\lambda} \left[\epsilon_j^* \cdot \mathcal{E}_{k\lambda}(t), \epsilon_j \cdot \mathcal{E}^{\dagger}_{k\lambda}(t')\right] e^{-i\omega_a t'} \sigma_-^{j} |t'\rangle_{n-1}$$

$$-\frac{i}{\hbar}\int_0^t dt' (H' + H_m) \sum_{k\lambda} \epsilon_j^* \cdot \mathcal{E}_{k\lambda}(t) |t'\rangle_{n-1} \tag{6-20}$$

H_m will be restricted to the order of H'; this insures that the last term is negligible. The next higher order correction from this term is given by Eq. (4-29) with H_I replaced by $H' + H_m$.

In the first term $|t'\rangle$ is slowly varying as before and can be taken out of the integral as in Eq. (4-11). The next higher order correction is given by Eq. (4-24) with H_I replaced by $H + H_m$. Eq. (6-20) becomes Eq. (6-18). If one retained the magnetic Hamiltonian with the atomic Hamiltonian, the transformation to the interaction picture will create exponential factors with frequencies corresponding to the structure of Fig. 4. The γ that results will have $\omega_a + O(\Delta)$ replacing ω_a . Since Δ/ω_a is small, $\gamma(\omega_a + O(\Delta)) \approx \gamma(\omega_a)$.

The addition of H_m to the Hamiltonian results in new matrices in the equations for $\langle \Gamma^1 \rangle$, $\langle \Gamma^2 \rangle$, and $\langle \Gamma^3 \rangle$. $M^1_{\mu\nu}$ becomes:

$$
\begin{pmatrix}
-\delta-\Delta-3i\gamma & 0 & -\Lambda/2 & & & & \\
0 & \delta+\Delta-3i\gamma & \Lambda/2 & & & & \\
-\Lambda & \Lambda & -4i\gamma & -i\gamma & -2i\gamma & & \\
 & & -4i\gamma & -4i\gamma & 4i\gamma & & \\
 & & -2i\gamma & i\gamma & -4i\gamma & -\Lambda & \Lambda \\
 & & & & -\Lambda/2 & \delta-\Delta-3i\gamma & 0 \\
 & & & & \Lambda/2 & 0 & -\delta+\Delta-3i\gamma
\end{pmatrix}
$$

$M^2_{\mu\nu}$ becomes:

$$
\begin{pmatrix}
-\delta-2\Delta-3i & 0 & 0 & -\Lambda/2 \\
0 & \delta-2\Delta-3i\gamma & 0 & -\Lambda/2 \\
0 & 0 & -2\Delta-2i\gamma & \Delta-2i\gamma \\
-\Lambda & -\Lambda & \Delta-4i\gamma & -2\Delta-4i\gamma
\end{pmatrix}
$$

$$
M^3_{\mu\nu}(\Delta, \Lambda) = M^2_{\mu\nu}(-\Delta, -\Lambda)
$$

(6-21a,b,c)

The roots of M^1 are found from the equation

$$x^7 + 24i\gamma x^6 - 2R^2x^5 - 4i\gamma(9R^2 - \Lambda^2)x^4 + (R^4 - 4\delta^2\Delta^2)x^3$$
$$+ 2i\gamma(6R_+^2R_-^2 - \Lambda^2R^2)x^2 - 3\gamma^2(12R_+^2R_-^2 - \Lambda^4)x$$
$$-36i\gamma^3\Lambda^2(R^2 - \Lambda^2) \quad = \quad 0$$

(6-22)

where

x - an eigenvalue

$$R^2 = \Lambda^2 + \delta^2 + \Delta^2$$
$$R_\pm^2 = \Lambda^2 + (\delta \pm \Delta)^2 \qquad (6\text{-}23a, b, c, d)$$
$$\Lambda = d\,E_0 / \hbar$$

The solution of the secular equation (6-22) can be solved as a power series in γ. The real part of x to lowest order in γ is:

$$x_{1,2} = \pm R_+$$
$$x_{3,4} = \pm R_- \qquad (6\text{-}24a, b, c, d)$$
$$x_{5,6,7} = 0$$

The roots of M^2 are found from the equation

$$y^4 + 12i\gamma y^3 - (R^2 - 6i\gamma\Delta)y^2 - i\gamma(5R^2 + \delta^2 + \Delta^2 - 36i\gamma\Delta)y$$
$$+ (\delta^2\Delta^2 - 6i\gamma\delta^2\Delta + \gamma^2(6\Lambda^2 + 9\Delta^2) - 54i\gamma^3\Delta) = 0$$

(6-25)

where

$$y = x + 2\Delta \qquad \qquad (6\text{-}26)$$

$$x = \text{an eigenvalue of } M^2$$

and terms of order γ^2/Λ^2 and γ^2/Δ^2 have been dropped. The real part of y to lowest order in γ is

$$y_{1,2} = \pm\sqrt{\tfrac{1}{2}(R^2 + R_+R_-)}$$

$$y_{3,4} = \pm\sqrt{\tfrac{1}{2}(R^2 - R_+R_-)} \qquad \qquad (6\text{-}27)$$

In dropping terms of order γ^2/Δ^2, I have restricted the analysis to strong magnetic fields (i.e. $\Delta >> \gamma$). The roots of M^2 (or M^3) determine the peaks and widths of the spectrum of light of polarization $\frac{\hat{x} - i\hat{y}}{\sqrt{2}}$ (or $\frac{\hat{x} + i\hat{y}}{\sqrt{2}}$. Aside from an overall shift of -2Δ for $\frac{\hat{x} - i\hat{y}}{\sqrt{2}}$ polarization and 2Δ for $\frac{\hat{x} + i\hat{y}}{\sqrt{2}}$ polarization, the two outer peaks are shifted to $\sqrt{\frac{1}{2}(R^2 + R_+R_-)}$ off center while the center peak is split by a frequency of $2\sqrt{\frac{1}{2}(R^2 - R_+R_-)}$. (Notice that this split disappears for $\omega = \omega_a$.)

The long time solution for $\langle \Gamma' \rangle$ is

$$\langle \Gamma' \rangle_{t\to\infty} = \frac{1}{\varepsilon^2 + \delta^2 + 2\Delta^2 + 1\ \gamma^2} \begin{pmatrix} \frac{\Delta}{2}(\delta + \Delta - 3i\gamma) \\ \frac{\Delta}{2}(\delta + \Delta + 3i\gamma) \\ -(\delta + \Delta)^2 - 9\gamma^2 \\ 4\delta\Delta \\ -(\delta - \Delta)^2 - 9\gamma^2 \\ -\frac{\Delta}{2}(\delta - \Delta + 3i\gamma) \\ -\frac{\Delta}{2}(\delta - \Delta - 3i\gamma \end{pmatrix} \qquad (6\text{-}28)$$

Thus

$$|A_{1,1/2}|^2 - |A_{0,1/2}|^2 = \frac{-|\delta+\Delta|^2 - 9\gamma^2}{\varepsilon^2+\delta^2+2\Delta^2+18\gamma^2} \tag{6-29}$$

$$|A_{1,-1/2}|^2 - |A_{0,-1/2}|^2 = \frac{-(\delta-\Delta)^2 - 9\gamma^2}{\varepsilon^2+\delta^2+2\Delta^2+18\gamma^2} \tag{6-30}$$

$$\left\{|A_{1,1/2}|^2 + |A_{0,1/2}|^2\right\} - \left\{|A_{1\,-1/2}|^2 + |A_{0\,-1/2}|^2\right\} = \frac{4\delta\Delta}{\varepsilon^2+\delta^2+2\Delta^2+18\gamma^2} \tag{6-31}$$

From equation (6-29), the $m_j=1/2$ levels are nearly equalized when the laser is detuned from the unperturbed atomic frequency, ω_a by $-\Delta$. This is consistent with Fig. 6, which illustrates the perturbed level spacings: $\omega_a \pm \Delta$. Eq. (6-31) describes the greater probability for the $m_j=1/2$ states over the $m_j=-1/2$ states. When $\delta = -\Delta$ the electron is predominantly in the $m_j=-1/2$ system, and (from Eq. (6-30)) most likely in the lower state. In this case, the laser strongly pumps the $m_j = 1/2$ system until the electron decays to the lower $m_j = -1/2$ state, which is weakly pumped by the laser.

For the spectrum itself, one evaluates

$$\frac{d}{dt}\langle a_{k\lambda}^{+} a_{k\lambda}\rangle \rightarrow -\left|\frac{d g_k}{\hbar}\right|^2 \frac{4}{\varepsilon^2+\delta^2+2\Delta^2+18\gamma^2}\, \mathrm{Im}\,\hat{a}_1 \cdot \frac{1}{x_k - M^2} \begin{pmatrix} \Lambda^2/4 \\ 0 \\ \frac{\Lambda}{2}(\delta-\Delta+3i\gamma) \\ -\frac{\Lambda}{2}(\delta-\Delta+3i\gamma) \end{pmatrix} \tag{6-32}$$

Define $X = \chi_k + 2\Delta + 3i\gamma$

$$\frac{d}{dt}\langle a_{k-}^{+} a_{k-}\rangle = -\left(\frac{dg_k}{\hbar}\right)^2 \frac{\Lambda^2}{\varepsilon^2+\delta^2+2\Delta^2} \,\mathrm{Im}\, \frac{X^3+(3i\gamma-\Delta)X^2-\left(\frac{\Lambda^2}{2}+\delta^2\right)X+i\gamma\left(\frac{\Lambda^2}{2}-3\delta^2\right)+\Delta\delta^2}{\det|\chi_k - M^2|}$$

(6-33)

Terms of order γ^2/Λ^2 and γ^2/Δ^2 have been dropped. First consider the case $\delta = 0$

$$\frac{d}{dt}\langle a_{k-}^{+} a_{k-}\rangle = -\left(\frac{dg_k}{\hbar}\right)^2 \frac{\Lambda^4}{\Lambda^2+2\Delta^2} \,\mathrm{Im}\, \frac{X^3+(3i\gamma-\Delta)X^2-\frac{\Lambda^2}{2}X+\frac{\Lambda^2}{2}i\gamma}{X^4-(\varepsilon^2+\Delta^2-6i\gamma\Delta)X^2+i\delta\Lambda^2 X}$$

(6-34)

The four roots of $\det|\chi_k - M^2|$ are

$$\chi_1 = -2\Delta - 3i\gamma$$

$$\chi_2 = -2\Delta - i\gamma\left(3 - \frac{\Lambda^2}{\Lambda^2+\Delta^2}\right)$$

$$\chi_{3,4} = -2\Delta \mp \sqrt{\Lambda^2+\Delta^2} - i\gamma\left(3 + \frac{\pm 6\Delta\sqrt{\Lambda^2+\Delta^2}+\Lambda^2}{2(\Lambda^2+\Delta^2)}\right) \qquad (6\text{-}35)$$

Define $\rho = \dfrac{\Delta}{\Lambda}$

(6-36)

For the side peak at $\gamma_k = -2\Delta + \sqrt{\Lambda^2 + \Delta^2}$ the ratio of side to center peak is

$$\left(3 + \frac{9}{2}\rho^2\right) \frac{1 + 2\rho^2 - 2\rho\sqrt{1+\rho^2}}{6\rho^2 + 7 + 6\rho\sqrt{1+\rho^2}} \tag{6-37}$$

For the side peak at $\gamma_k = -2\Delta - \sqrt{\Lambda^2 + \Delta^2}$ the ratio of side to center peak is

$$\left(3 + \frac{9}{2}\rho^2\right) \frac{1 + 2\rho^2 + 2\rho\sqrt{1+\rho^2}}{6\rho^2 + 7 - 6\rho\sqrt{1+\rho^2}} \tag{6-38}$$

Consequently, it is the lower frequency peaks that are larger for strong magnetic fields. For polarization $\hat{\epsilon}_{k+} = \frac{\hat{x} + i\hat{y}}{\sqrt{2}}$, $\rho \to -\rho$. In this case, it is the upper frequency peaks that are larger for strong magnetic fields. See Fig. 7 a, b.

Consider, now, the case $\delta = -\Delta$

$$\frac{d}{dt}\langle a_{k-}^{\dagger} a_{k-}\rangle = -\left(\frac{dg_k}{\hbar}\right)^2 \frac{\Lambda^4}{\Lambda^2 + 4\Delta^2}\, \mathrm{Im}\, \frac{x^3 + (3i\gamma - \Delta)x^2 - \left(\frac{\Lambda^2}{2} + \Delta^2\right)x + i\gamma\left(\frac{\Lambda^2}{2} - 3\Delta^2\right) + \Delta^3}{x^4 - (\Lambda^2 + 2\Delta^2 - 6i\gamma)x^2 + 4i\gamma\Lambda^2 x + \Delta^4 - 6i\gamma\Delta^3} \tag{6-39}$$

The real part of the roots of the denominator are

$$\mathrm{Re}\, x_{1,2} = \pm\frac{\Lambda}{\sqrt{2}}\sqrt{1 + 2\rho^2 - \sqrt{1 + 4\rho^2}} \tag{6-40}$$

$$\mathrm{Re}\, x_{3,4} = \pm\frac{\Lambda}{\sqrt{2}}\sqrt{1 + 2\rho^2 + \sqrt{1 + 4\rho^2}} \tag{6-41}$$

The center peak is split into two. In the limit of a large magnetic field; the two larger peaks are inappreciable (see Fig. 8a). The position of the lower peaks approach $-\Delta \mp \frac{\Lambda}{2}$ (-2Δ omitted). The double nature of the spectrum, differing by the on-resonance Rabi frequency, Λ, is to be expected; only two states are strongly pumped.

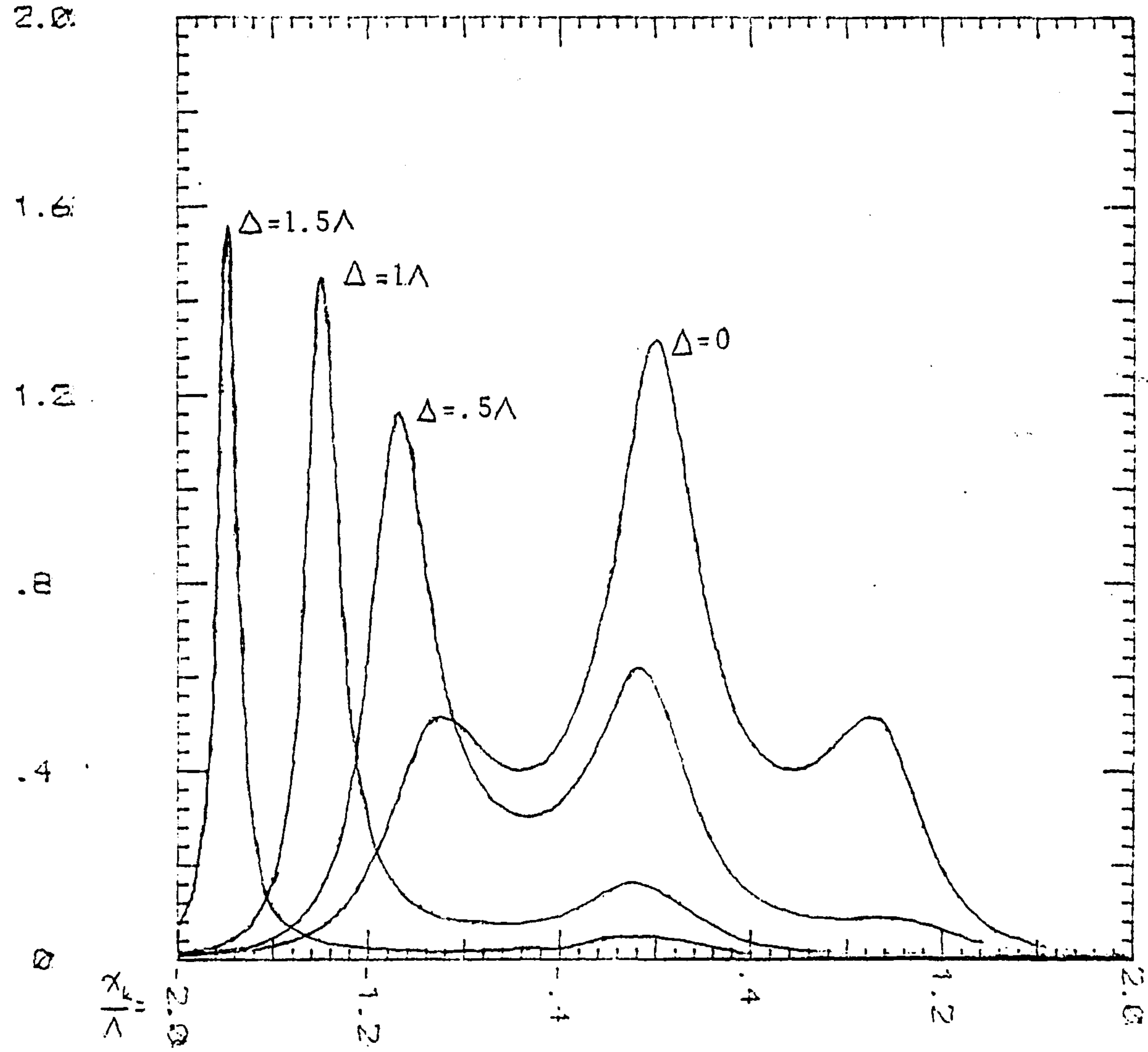

Fig. 7a. Relative spectrum for radiation of polarization ϵ_{k-}. As the external magnetic field is increased the center peak and high frequency side peak are eliminated while the low frequency peak is enhanced. The overall shift of the spectrum by 2Δ is omitted. The parameters are $\delta=0$, $\gamma=.075\Lambda$ for all four cases and $\Delta= 1.5\Lambda, 1\Lambda, .5\Lambda$, and 0 as indicated.

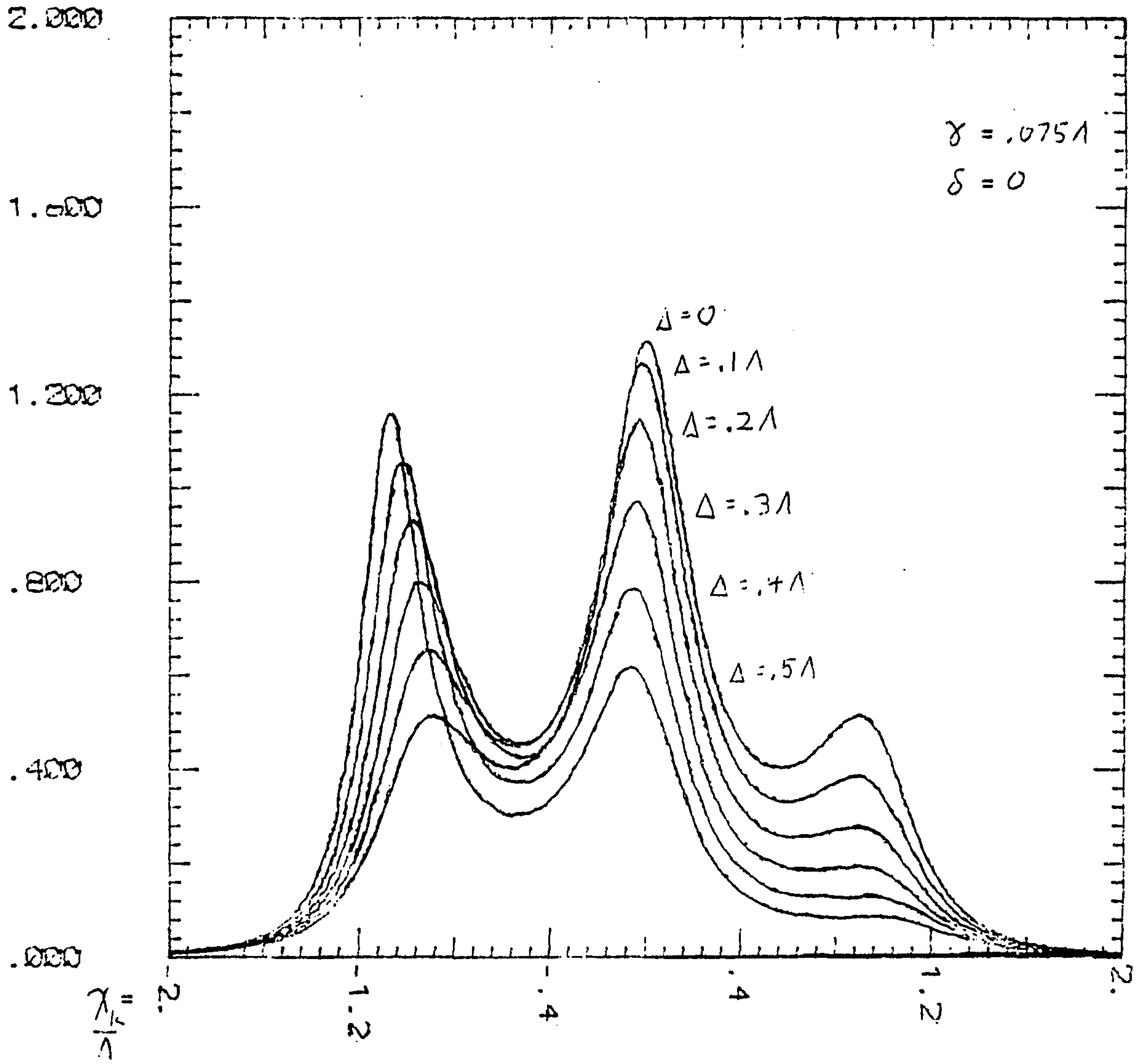

Fig. 7b. Relative spectrums for radiation of polarization ϵ_{k-} . The center peak is diminished and low frequency peak is enhanced. Note the gradual shift in the side peak in both Fig. 7a. and Fig. 7b. (See Eq. 6-35). For polarization ϵ_{k+} , $\chi_k \to -\chi_k$.

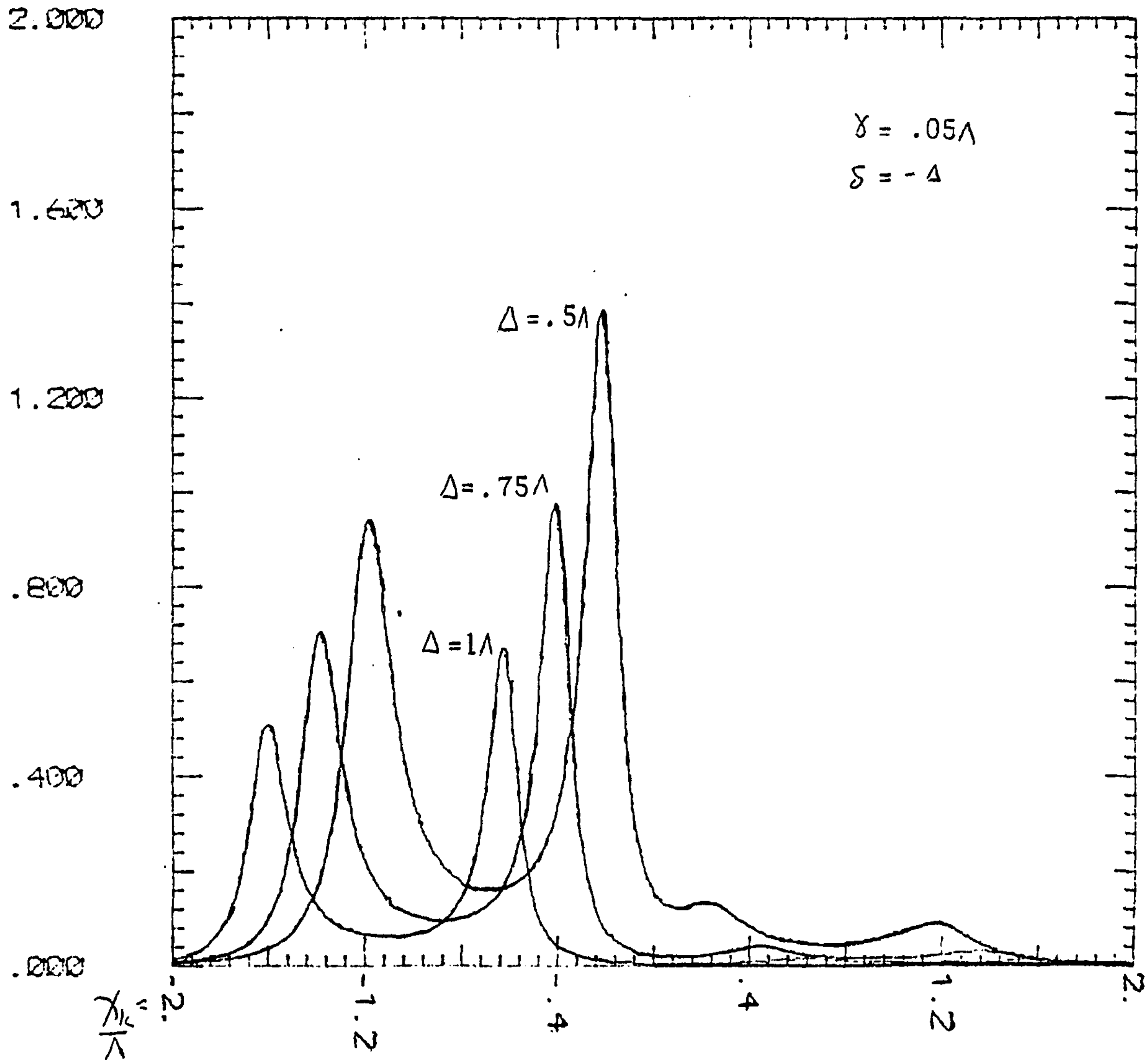

Fig. 8a. Two-peak spectrums for two-state optical pumping As the magnetic field shifts unequally the m_j = 1/2 and m_j = -1/2 states, the laser can pump only one m_j system by satisfying the new resonance condition $\delta = \pm\Delta$ (See Fig. 6.). The doublet nature of the AC Stark Shift is apparent.

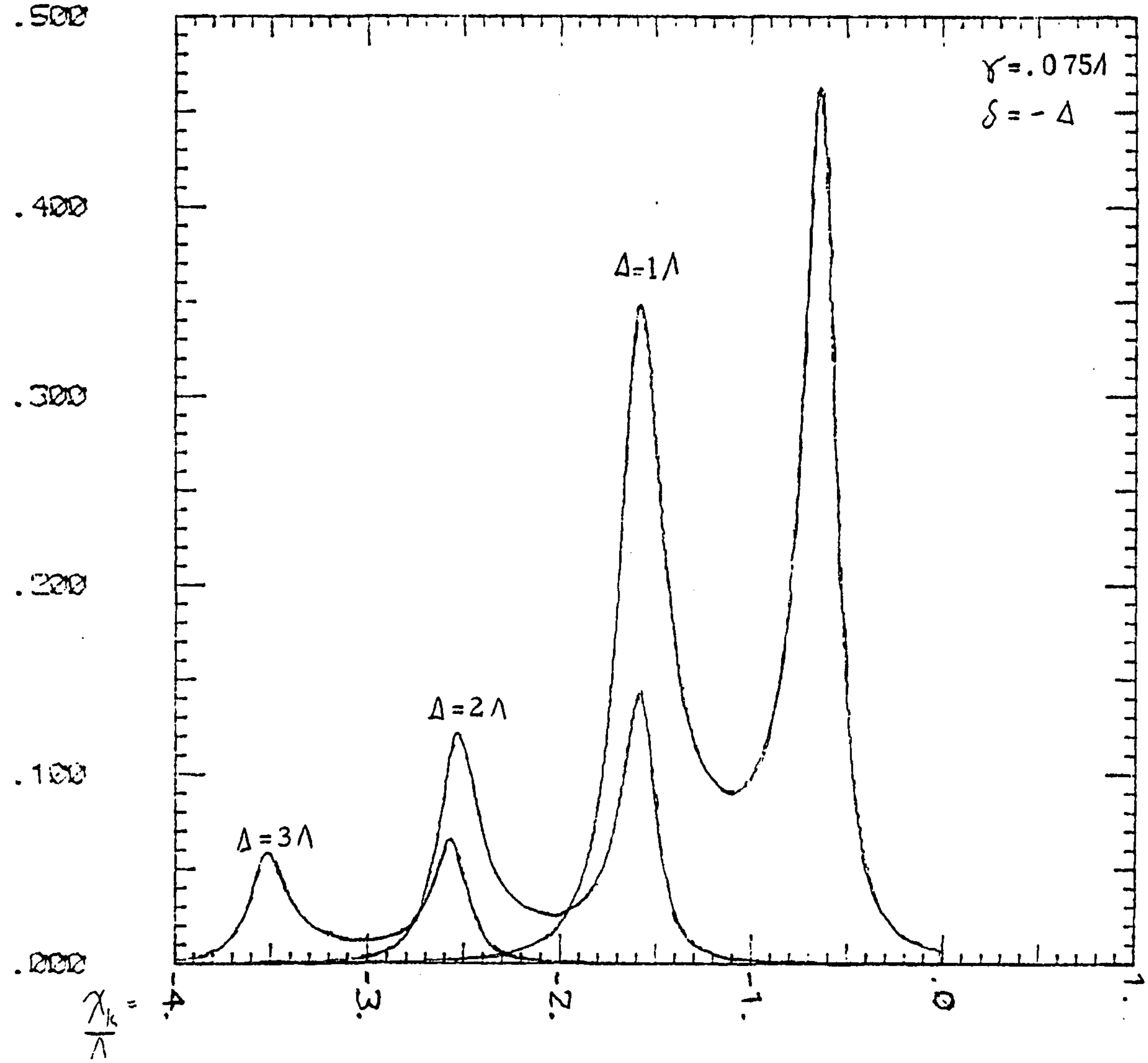

Fig. 8b. Two peak spectrums for two-state optical pumping. For larger values of Δ than those of Fig. 8a., it is apparent that 'optical pumping' to a non-pumped state is approached; the long-time limit intensity is decreased.

7. Effects of the Finite Laser Bandwidth on the Spectrum

Ample work on the inclusion of deviations from a monochromatic spectrum has been done for the two-state problem by consideration of the stochastic fluctuations involved in laser radiation [35,36,38-40]. One can also treat slowly varying drifts of amplitude and frequency by averaging the resultant expression for the spectrum (Eq. (6-15)) with respect to Λ and δ . In this section, laser bandwidth due to phase fluctuations will be considered. As is commonly done in the phase - diffusion model of the laser[41], the phase of the laser field is treated as a time dependent stochastic variable. The phase is considered to be highly fluctuating and described in the Markoff approximation by the following correlation:

$$\langle\langle \dot{\varphi}(t)\,\dot{\varphi}(t')\rangle\rangle = 2\gamma_L\,\delta(t-t') \tag{7-1}$$

The calculation for the spectrum will be redone with the substitution, $\omega t \rightarrow \omega t + \varphi$, in the elements Γ_ν' of Table 1 and in the elements $\langle e^{-i\omega t'} a_{k\lambda}^{\dagger}\,\Gamma_\nu'\rangle$. This will lead to the addition of $\dot{\varphi}(t)$ to the diagonal elements of the differential equations. Stochastic equations of this type have been dealt with by Fox[37].

Consider the general equation of the form

$$i\frac{d}{dt}\psi = \left(H_0 + \dot{\varphi}(t) H_1\right)\psi \tag{7-2}$$

where H_0 and H_1 are n x n matrices and $\dot{\varphi}(t)$ obeys Eq. (7-1).

Fox shows that Eq. (7-2) is equivalent to

$$i \frac{d}{dt} \psi = (H_0 - i \gamma_c H_1 \cdot H_1) \psi \tag{7-3}$$

To arrive at stochastic equations for the fluorescence problem, redefine the basis sets Γ_μ^i by the prescription

$$\omega t \rightarrow \omega t + \varphi \tag{7-4}$$

Equations (5-19) become

$$i \hbar \frac{d}{dt} \langle \Gamma^i \rangle = M^i \langle \Gamma_\mu^i \rangle + \dot{\varphi} \mathbb{M}^i \langle \Gamma_\mu^i \rangle + \delta_i^1 A \tag{7-5}$$

where M^I is given in Eq. (5-20), (5-21), (5-22) or Eqs. (6-21) for the magnetic case. The elements of M^1 are

for i=1 $\quad \mathbb{M}_{11}^1 = \mathbb{M}_{77}^1 = -1 \qquad \mathbb{M}_{22}^1 = \mathbb{M}_{66}^1 = 1$ (7-6)

(all other entries are zero)

for i=2 $\quad \mathbb{M}_{11}^2 = -1 \qquad \mathbb{M}_{22}^2 = 1$ (7-7)

(all other entries are zero)

for i=3 $\quad \mathbb{M}_{ij}^3 = \mathbb{M}_{ij}^2$ (7-8)

Eq. (7-5) can now be written as:

$$i\frac{d}{dt}\langle \Gamma^i\rangle = (M^i - i\gamma_c \mathbb{M}^i \cdot \mathbb{M}^i)\langle \Gamma^i\rangle + \delta_i^1 A \tag{7-9}$$

The net change of the equations for $\langle \Gamma^i \rangle$ involve the addition of $-i\gamma_c$ to each entry that has δ or $-\delta$. The matrix element $\langle a^+ e^{-i\omega t}\Gamma_\nu^i\rangle$ with the substitution (7-4) ~~are~~ governed by different matrices than the $\langle \Gamma_\nu^i \rangle$. The resultant matrices M'^j are

$$M'^1 = \begin{pmatrix} -\delta-\Delta-3i\gamma & 0 & -\Lambda/2 & & & & \\ 0 & \delta+\Delta-3i\gamma-4i\gamma_c & \Lambda/2 & & & & \\ -\Lambda & \Lambda & -4i\gamma-i\gamma_c & -i\gamma & -2i\gamma & & \\ & & -4i\gamma & -4i\gamma-i\gamma_c & 4i\gamma & & \\ & & -2i\gamma & i\gamma & -4i\gamma-i\gamma_c & -\Lambda & \Lambda \\ & & & & -\Lambda/2 & \delta-\Delta-3i\gamma-4i\gamma_c & 0 \\ & & & & \Lambda/2 & 0 & -\delta+\Delta-3i\gamma \end{pmatrix}$$

$$M'^2 = \begin{pmatrix} -\delta-2\Delta-3i\gamma & 0 & 0 & -\Lambda/2 \\ 0 & \delta-2\Delta-3i\gamma-4i\gamma_c & 0 & -\Lambda/2 \\ 0 & 0 & -2\Delta-2i\gamma-i\gamma_c & \Delta-2i\gamma \\ -\Lambda & -\Lambda & \Delta-4i\gamma & -2\Delta-4i\gamma-i\gamma_c \end{pmatrix}$$

(7-10)

$$\langle \Gamma'' \rangle_{t \to \infty} = \frac{1}{\frac{\gamma'}{\gamma}\Lambda^2 + 2(\delta^2 + \Delta^2 + 9\gamma'^2)} \begin{pmatrix} \frac{\Lambda}{2}(\delta + \Delta - 3i\gamma') \\ \frac{\Lambda}{2}(\delta + \Delta + 3i\gamma') \\ -((\delta+\Delta)^2 + (3\gamma')^2) \\ 4\delta\Delta \\ -((\delta-\Delta)^2 + (3\gamma')^2) \\ -\frac{\Lambda}{2}(\delta - \Delta + 3i\gamma') \\ -\frac{\Lambda}{2}(\delta - \Delta - 3i\gamma') \end{pmatrix} \tag{7-11}$$

where $3\gamma' \equiv 3\gamma + \gamma_c$

One now evaluates

$$-\frac{4 d^2 g_k^2}{\hbar^2} \operatorname{Im} a_1^\dagger \frac{1}{\chi_k - M'^2} \begin{pmatrix} \frac{1}{4} - \frac{1}{4}\langle \beta \rangle + \frac{1}{2}\langle \frac{1-\beta}{2}\sigma_z \rangle \\ 0 \\ -\langle \frac{1-\beta}{2} \sigma_+ e^{-i\omega t''} \rangle \\ \langle \frac{1-\beta}{2} \sigma_+ e^{-i\omega t''} \rangle \end{pmatrix} \tag{7-12}$$

where M'^2 is given in (7-5) and the elements of the column vector are given in (7-6). When $\Delta = 0$, $\delta = 0$ the ratio of side peak to center peak is

$$\frac{3\gamma + 2\gamma_c}{7\gamma + 3\gamma_c} \tag{7-13}$$

For $\delta \neq 0$ see Fig. 9. For $\Delta \neq 0$, $\delta = 0$ see Fig. 10. For $\Delta = -\delta$ see Fig. 11.

From Figure 12, the peaks for the on-resonance non-magnetic problem are shown to depend on γ_c. The peaks appear only for γ_c less than or on the order of γ.

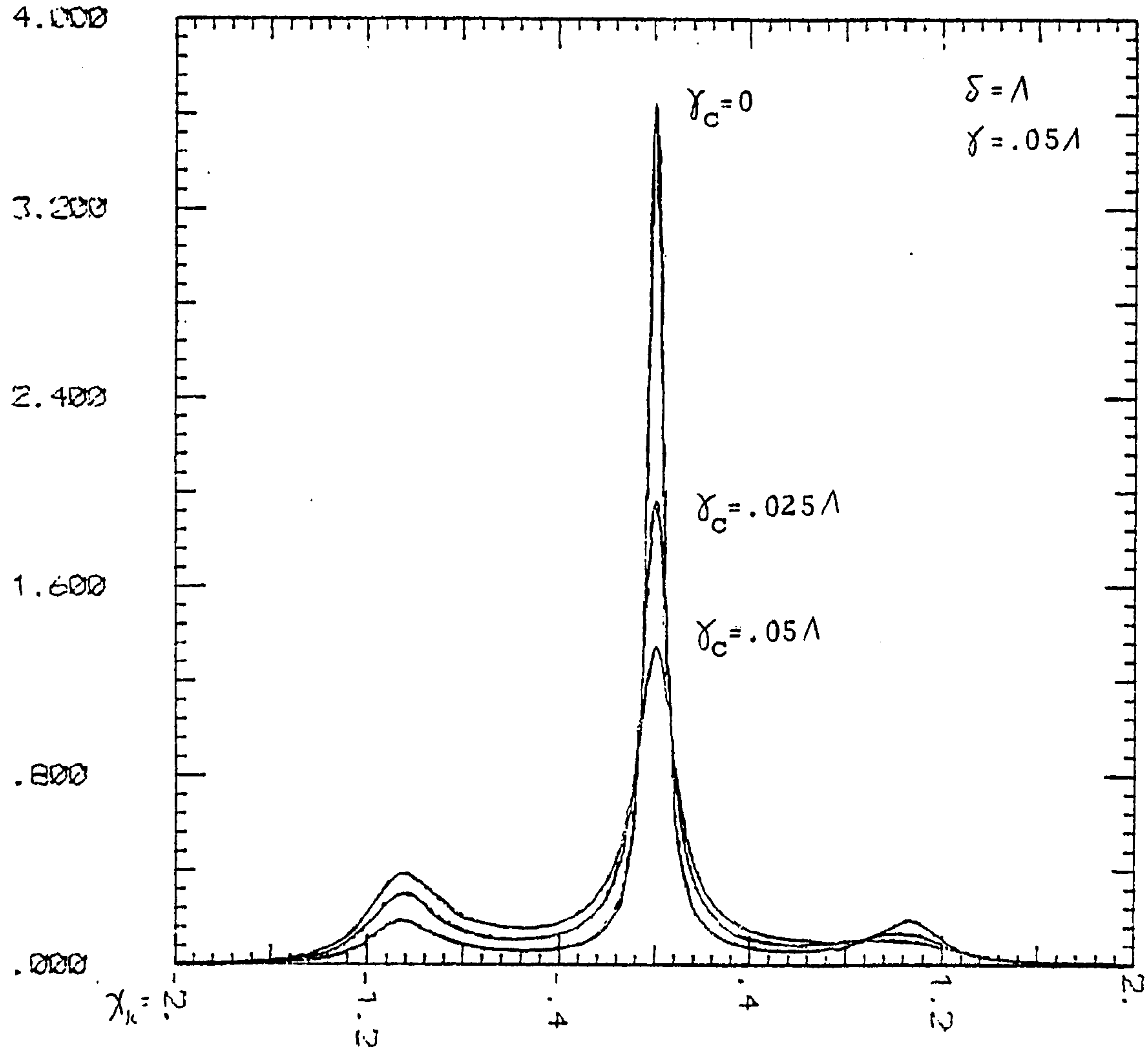

Fig. 9. Off resonance pumping. When γ_c is increased, the high frequency side peak and the center peak decrease while the low frequency peak is enhanced. γ_c is 0, $.025\Lambda$, and $.05\Lambda$ as indicated.

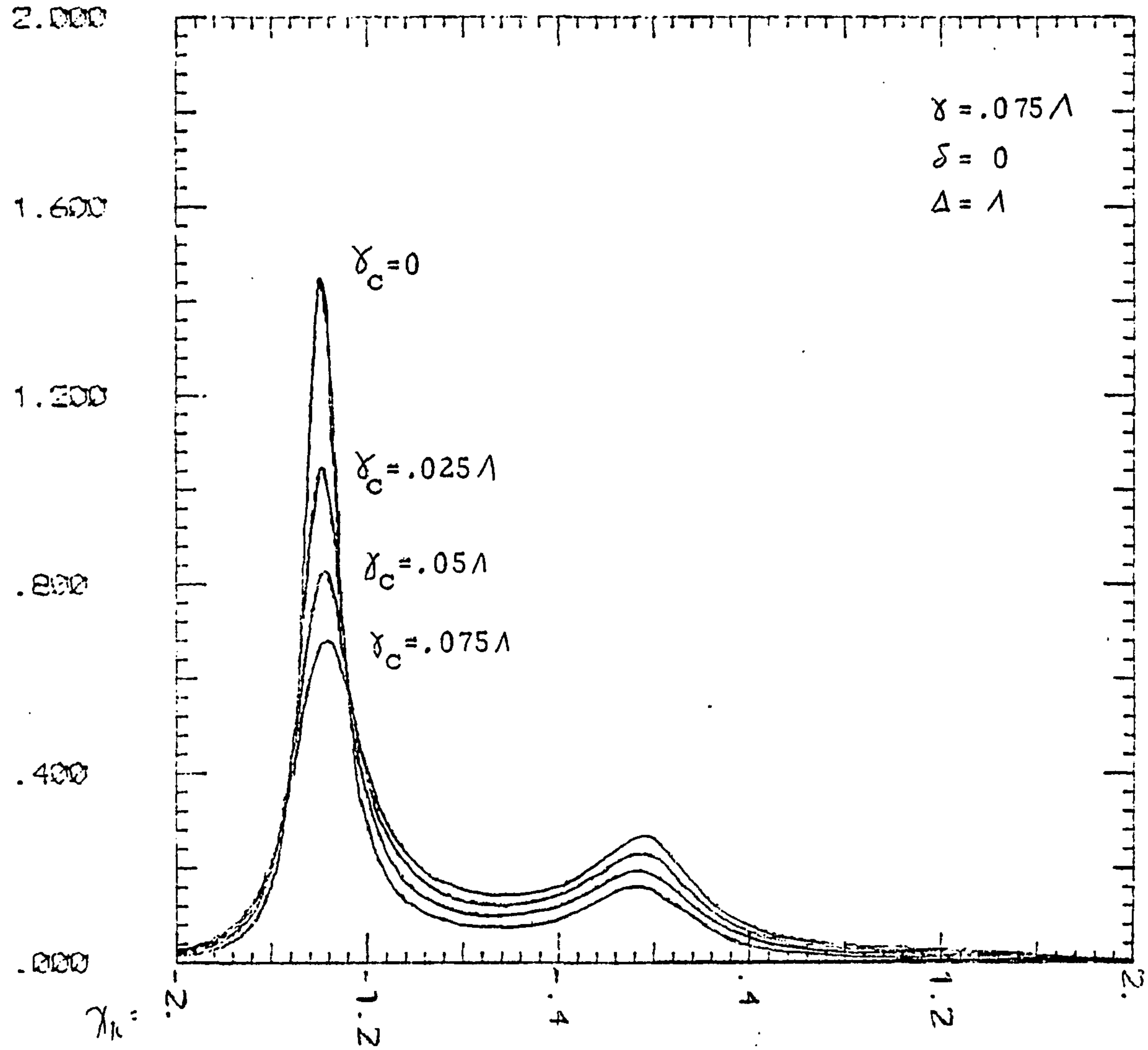

Fig. 10. Spectrum altered by a magnetic field. The one-peak spectrum obtained when a magnetic field removes the system from resonance (See Fig. 7a) is illustrated when γ_c is 0, .025Λ, .05Λ, and .075Λ.

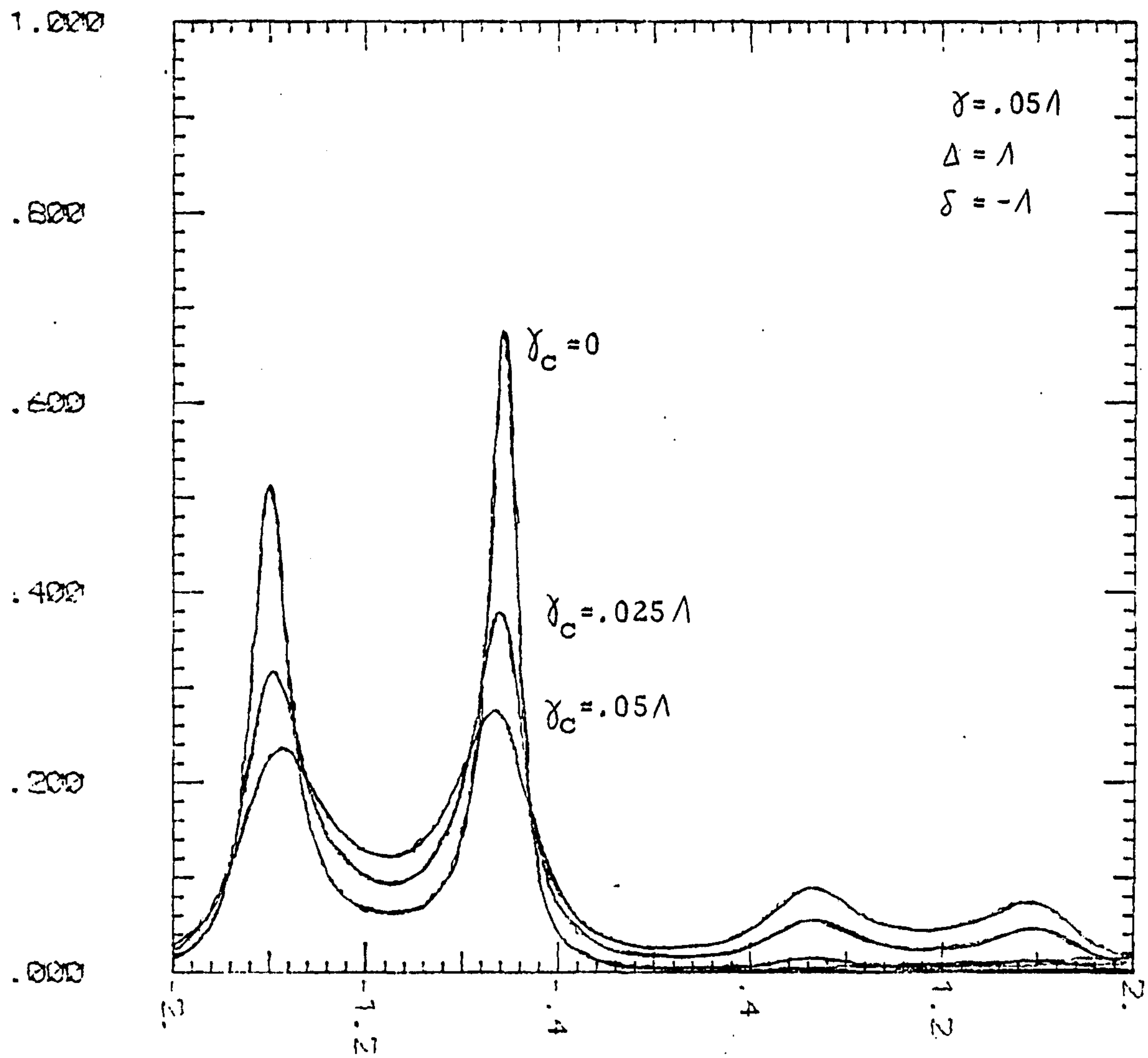

Fig. 11. Two-state optical pumping. The two peak spectrum illustrated in Fig. 8a and Fig. 8b is modified by the finite laser bandwidth. The peaks are reduced by increasing γ_c.

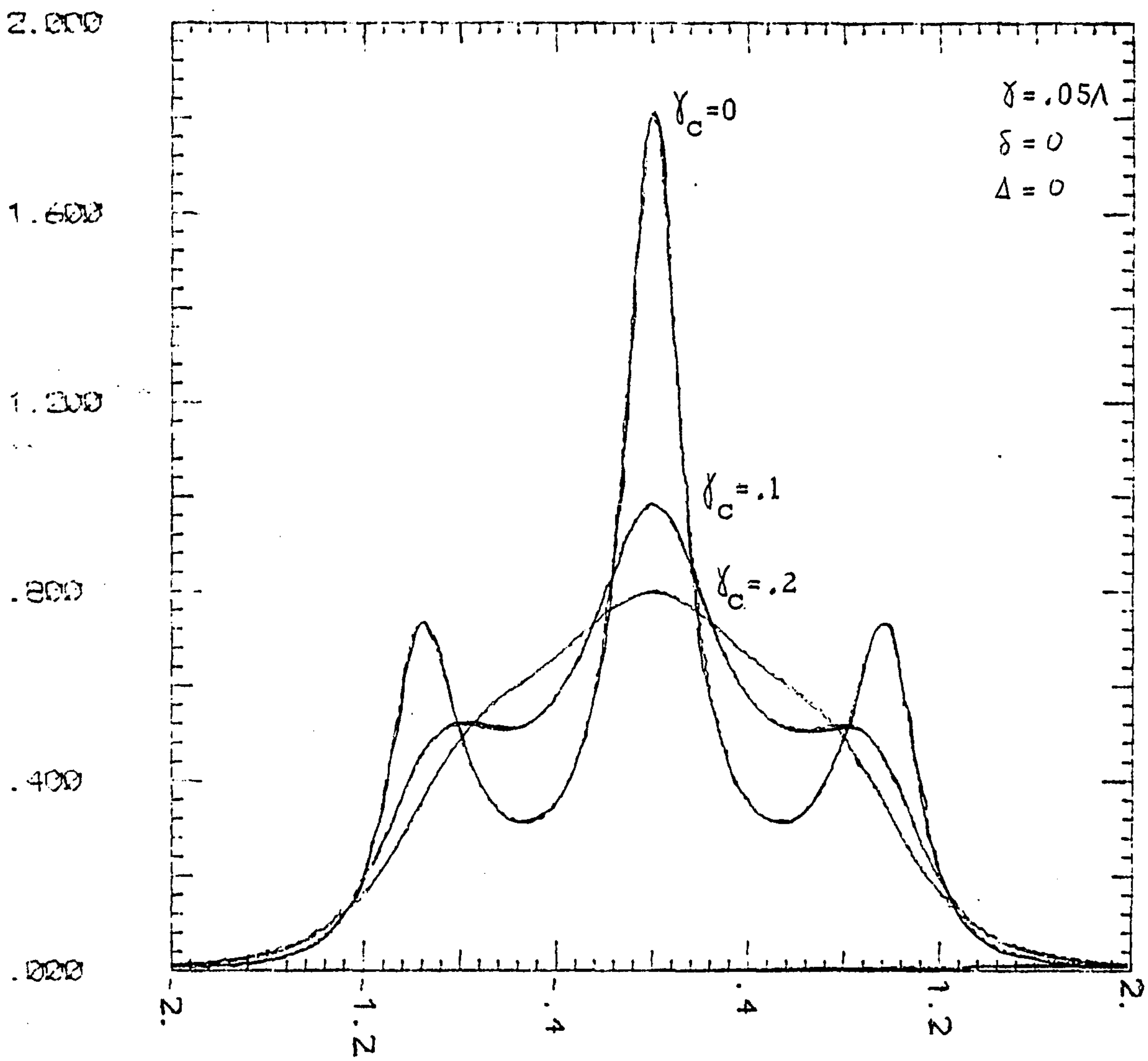

Fig. 12. Cross-polarization spectrum. The three peak spectrum is shown to reduce to a one peak spectrum as γ_c increases.

Appendix A

Pauli Matricies:

$$\sigma_1^P = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} \qquad \sigma_2^P = \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix} \qquad \sigma_3^P = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}$$

Dirac Matricies are the 4 X 4 matricies:

$$\sigma_j = \begin{pmatrix} \sigma_j^P & 0 \\ 0 & \sigma_j^P \end{pmatrix} \qquad \alpha_j = \begin{pmatrix} 0 & \sigma_j^P \\ \sigma_j^P & 0 \end{pmatrix}$$

$$\beta = \begin{pmatrix} I & 0 \\ 0 & -I \end{pmatrix} \qquad \rho = \begin{pmatrix} 0 & I \\ I & 0 \end{pmatrix}$$

Sixteen linear independent hermitian matricies are:

I (unit matrix)	ρ	
σ_j	$\rho\,\sigma_j$	$(=\alpha_j)$
β	$i\beta\rho$	
$\beta\,\sigma_j$	$i\beta\rho\,\sigma_j$	

The following relations hold:

Commutation relations:

$$[\sigma_j, \sigma_k] = 2i\epsilon_{jk\ell}\,\sigma_\ell$$

$$[\beta, \rho] = 2\beta\rho$$

$$[\beta, \sigma_j] = 0$$

$$[\rho, \sigma_j] = 0$$

Anticommutation relations:

$$\{\sigma_j, \sigma_k\} = 2\delta_{jk}$$

$$\{\beta, \rho\} = 0$$

Raising and lowering operators:

$$\sigma_+ = \sigma_x + i\sigma_y$$

$$\sigma_- = \sigma_x - i\sigma_y$$

$$\frac{1+\beta}{2}\sigma_+ = \begin{pmatrix} \sigma_+^P & 0 \\ 0 & 0 \end{pmatrix} \equiv \sigma_+^{11}$$

$$\frac{1-\beta}{2}\sigma_+ = \begin{pmatrix} 0 & 0 \\ 0 & \sigma_+^P \end{pmatrix} \equiv \sigma_+^{-1-1}$$

$$\frac{1+\beta}{2}\rho\sigma_+ = \begin{pmatrix} 0 & \sigma_+^P \\ 0 & 0 \end{pmatrix} \equiv \sigma_+^{1,-1}$$

$$\frac{1-\beta}{2}\rho\sigma_+ = \begin{pmatrix} 0 & 0 \\ \sigma_+^P & 0 \end{pmatrix} \equiv \sigma_+^{-1\,1}$$

The operator σ_+^{jk} raises the system from the lower level with $m_j = \frac{k}{2}$ to the upper level state with $m_j = \frac{j}{2}$. To treat systems of any degree of degeneracy one can introduce matricies which have Pauli matricies as elements, similiar to those above. The rules of algebra are:

$$\sigma_a^{jk}\sigma_b^{lm} = \delta_k^l\,(1^{jm} + i\varepsilon_{abc}\sigma_c^{jm}$$

where 1^{jm} is the matrix with a 2 X 2 unit matrix in the jth row and mth column.

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