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### THE QUANTUM THEORY OF OPTICAL BISTABILITY

IN NONLINEAR SYSTEMS

A thesis

submitted in partial fulfilment

of the requirements for the Degree

of

Doctor of Philosophy

at the

University of Waikato

by

MOIRA LETCHME STEYN-ROSS

University of Waikato, Hamilton, New Zealand. December 1981 To the memory of my brother

CHARLES LOUIS STEYN

#### ABSTRACT

This thesis is concerned with the investigation of the behaviour of various nonlinear systems when excited by a coherent light-field.

Of particular interest to us is the possibility of observing optical bistability in these systems.

In Chapter One we introduce the general theoretical methods we employ to perform all investigations.

In Chapters Two and Three we develop a quantum theory of the interaction of light with a variety of different systems, i.e. Raman active media, the parametric oscillator, the two-photon absorber and a system of three-level atoms.

In each case, a master equation, containing all statistical information about the system, is derived. This enables the systematic inclusion of a damping mechanism into each model.

Discussion is limited to the steady state behaviour of the systems, and in general we assume the deterministic limit in which we ignore quantum fluctuations in the field variables. It is then possible to factorise the steady state expectation values of these variables.

Steady state calculations indicate that each system may exhibit optical bistability in output field/intensity dependent on input field/ intensity.

To determine whether a system will display optical bistability, it is necessary to perform a stability analysis. Where possible, such analytical calculations are performed. However, in certain cases the complexity of the highly nonlinear systems results in these calculations becoming extremely difficult. In such cases, conclusions relating to the stability of the system are drawn from graphical plots of its steady state behaviour. Chapters Four to Eight are devoted to a study of the intracavity interaction of coherent radiation with semiconductors.

As explained in Chapter Four (of an introductory nature), this work was prompted by two recent experiments which indicated the existence of optical bistability in semiconductors. Although both experiments used semiconductors as the nonlinear material, the mechanism proposed to produce the observed bistability in each case was vastly different.

In Chapter Eight we discuss one of these mechanisms - interband excitation. We present a very simple theory of this effect and show that in essence it is equivalent to the theory of optical bistability in two-level systems. However, we stress that our theory is only approximate and relies on the validity of several simplifying assumptions.

Chapters Five and Six concern the other form of semiconductor optical bistability - arising from the interaction between a light field and excitons comprising the semiconductor.

In order to develop a quantum theory of this system using master equation techniques, it is necessary to transform the fermion system to a boson system. Bosonisation transformations required to effect this are developed in Chapter Five.

In Chapter Six we present a fully quantum mechanical theory of optical bistability in excitonic systems. Two types of bistability are found: bistability in output intensity and also exciton number, dependent on input intensity.

In Chapter Seven we investigate the effects of quantum fluctuations in a low density exciton system, by considering its Fokker-Planck equation. We discuss in detail the adiabatic elimination of stochastic variables with regard to the system's Fokker-Planck equation

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#### ACKNOWLEDGEMENTS

Firstly, I wish to acknowledge the help and guidance I received from my supervisors, Professor D.F. Walls and Dr C.W. Gardiner.

My research work was also aided by useful discussions with members of our theoretical group: Drs K.J. McNeil and P.D. Drummond and Mr G. Milburn.

I also wish to thank my husband for his constant encouragement and support.

Finally, I thank Mrs Christine Coates for the consistently high standard of typing she maintained throughout the production of this thesis.

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### Note:

Although certain symbols are used to define more than one quantity, in all cases the text provides a clear and unambiguous indication of the desired definition.

۵.	Bohr radius			
ai,ãi	Constants (i = integer)			
âi	System variable (i = integer)			
at <sub>rj</sub>	Fermion creation operator for an electron in band j, of momentum k			
А	System operator			
Ai (g,t)	Drift matrix in Fokker-Planck Equation			
A	Vector potential			
bi, Đi	Arbitrary constant (i = integer)			
bi,bap,b	Boson operator			
BĻ	Creation operator for exciton at discrete lattice site L			
B <sup>+</sup> αβ	Operator generating permutations of boson operators when acting on the ground state			
С	Velocity of light; cooperativity parameter			
C <sup>†</sup> <sub>K</sub>	Boson creation operator for exciton of momentum K			
di	Arbitrary constant (i = integer)			

d <sup>†</sup> <sub>Kj</sub>	Fermion creation operator for hole, momentum $\kappa_j$			
Dej(a,t)	Diffusion matrix in Fokker-Planck equation			
e	Electric charge			
ei	Constants (i = integer)			
ê	Polarisation vector			
Е, Еа, Еь, Е, , Е2 <i>.</i>	Amplitude of classical external driving field			
Ei	Eigenvalue of Schrodinger equation			
$E_v(R_v)$ $E_c(R_c)$	Energy of valence and conduction band states			
Ē	Electric field			
f(x)	Arbitrary function of $\underline{X}$			
$f_{\nu}(\underline{k})$	Fourier transform of $\gamma$ -th state of Hydrogen-like state			
F	Arbitrary fermion operator			
F., F., F2	Arbitrary variables			
Ŧ	Boson image of fermion operator			
9,9i,9ij	Light-matter coupling constants			
g <sup>(2)</sup> (0)	Second order correlation function			
G,,G,,G,	Variables			
$h = \hbar 2\pi$	Planck's constant			

```
H, Hi, Hamiltonian operator
Hs.c., Hex,
Ho, He.m. (etc.)
```

H	Magnetic field			
L	Imaginary unit			
I	Steady state input intensity			
Im	Imaginary part			
<u>K</u> I	Wavevector			
K∟	Raman coupling constant as defined by Lugovoi (1977)			
Κp	Coupling constant associated with the parametric oscillator			
Ki	Strength of damping due to coupling to various reservoirs (i = integer)			
Li	Constant (i = integer)			
L1, L2, L3	Operators associated with a Fokker-Planck equation			
m	Mass of particle			
mi	Constant (i = integer)			
Μ	System operator			
M1, M2	Exciton-exciton interaction coefficients			

- $\eta$  Integer; density of particles
- $\gamma_1$  Steady state exciton number
- N<sub>2</sub> Steady state output intensity arising from exciton-light interaction
- **η**κ Occupation number, refractive index
- $ar{n}$ : Thermal occupation number of reservoir
- N, N Number of particles in system
- Nv Number of particles per unit volume
- N<sup>h</sup>, N<sup>e</sup>, Exciton inversion operators
- $\widehat{N}$  Particle number operator in many body system
- $\mathcal{N}$  Normalisation constant
- O Arbitrary operator

NL

- Q Normal mode coordinate; constant
- $\hat{q}$  Coordinate operator
- Qi Reservoir operator
- $\tilde{\rho}i$  Constant (i = integer)
- $p_{m{arphi}}$  Probability of system being in state  $~m{arphi}$  .
- **p**; Momentum of jth particle
- P, P' Permutation operators

<u>P</u>	Polarisation vector		
P, P,	Operators which project out the non-antisymmetrised components from boson states		
P(2)	Quasi-probability distribution function		
P, K	Projection operator (defined in adiabatic elimination procedure)		
r, r <sub>2</sub>	Exciton-light interaction coefficients		
ĩ	Position vector		
Re	Real part		
S	Stokes intensity (Chapter Two); Laplace transform variable (Chapter Seven)		
Sı	Saturation parameter		
S(t)	Reduced density operator		
<b>S(ω)</b>	Fluorescent spectrum		
t	Time		
Т	Temperature		
$T_1, T_2$	Atomic damping rates		
Tr	Trace		
U.( <u>r</u> ),U2( <u>r</u> )	Mode functions		
Ur; ( <u>x</u> )	Bloch function of particle with wavevector $\kappa_j$ .		

- ${\cal V}$  Projection of probability distribution function
- $v_{\mu}$  Electron-hole interaction coefficient ( $\mu$  = integer)
- **υ(q)** Fourier transform of Coulomb interaction potential
- V System volume; Reservoir-System interaction Hamiltonian
- $V_{\beta}, V_{\beta}, V_{F}$  Boson and fermion vector spaces
- V(Ix-y) Coulomb interaction potential
- ${oldsymbol w}$  Projection of probability distribution function
- X System variable
- <u>X</u> Displacement operator
- Y, Y System variables
  - d mh/me+mh)
  - di Complex numbers corresponding to system operators
     (i = integer)
  - αij Electronic polarisability tensor of ith molecule
    β: Complex numbers corresponding to system operators
    (i = integer)
  - Dimensionless adiabatic elimination parameter

Xa, Xc, Xi XAi, Xo	Damping coefficients (i = integer)			
$f_{a_i}(t)$	Stochastic fluctuating term			
δΑ,δΒ,δι,δ2	Detuning parameters			
δι <u>j</u>	Kronecker delta			
δ(x-x')	Dirac delta-function			
٤.	Vacuum permittivity			
ει	Expansion coefficients (i = integer)			
Eb,n	Binding energy of nth excitonic state			
η	Exciton-exciton damping coefficient; system parameter			
X	Raman coupling constant (Chapter Two); exciton-light coupling constant in high exciton density limit (Chapter Six))			
χı	Two-level system coupling constant			
Xa	Two-photon absorber coupling constant			
xj	Reservoir-system coupling constant (j = integer)			
λί	Eigenvalue; constant (i = integer)			
Дo	Vacuum permeability			
<u>ب</u> ر	Electric dipole moment			
V	Quantum number; resonant frequency			

Ei (t)	Gaussian Stochastic process
Πι	Product
p <sup>(t)</sup>	Density operator
σ <sup>+</sup> ,σ <sup>-</sup> ,σ <sub>x</sub> } σ <sub>y</sub> ,σ <sub>y</sub>	Pauli spin matrices
Ê	Sum
T1, T2	Collisional damping rates
( <i>422</i> '	Wavefunction describing relative motion of electrons and holes
<i>Φμ</i> ( <u>×</u> )	Eigenfunction of single particle Schrodinger equation; arbitrary function of $\underline{X}$
X	Exciton-exciton interaction coefficient; Raman coupling constant
<u>X</u> (n)	nth order susceptibility tensor
$\Psi(\underline{r})$	Field operator
<i>\</i> \$\( <u>\$</u> )}	State vector
ωί	Frequency of ith state
$-\Omega_{\nu,\kappa}$	Resonance frequency of exciton (quantum number $ u$ , momentum K )

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#### CHAPTER 1

#### INTRODUCTION

#### ∮1.1 General Background

Nonlinear optics involves the study of the physical phenomena associated with the nonlinear response of a medium to applied electromagnetic fields.

For example, in certain substances, the electric polarisation may be a quadratic or cubic function of the applied electric field amplitudes. Dielectric and magnetic susceptibilities and indices of refraction can also be functions of applied field intensities.

Such behaviour of materials has led to the observation of many nonlinear optical processes, for example: optical harmonic generation, optical rectification and production of combination frequencies (Bloembergen, 1965).

When a nonlinear optical interaction is coupled to itself via a feedback mechanism, the phenomenon of optical bistability may occur. This is the ability of a system to exist in two stable states; thus exhibiting two distinct light transmission regimes for a given input field intensity.

Such an idea was first suggested by Seidel (1969) and Szoke et.al. (1969), with regard to an optical resonator filled with a medium displaying an absorption dependent on light intensity.

The first theoretical treatment of the problem was presented by McCall (1974), and the experimental observation of the effect followed soon after (Gibbs et.al., 1976).

To date, over two hundred published papers bear testiment to the vast interest in the field. Although much of this research concerns the investigation of collective behaviour, about forty percent of these papers are devoted to experimental considerations and possible technological applications. Suggested applications of optically bistable devices include: differential amplifiers, pulse-shapers and limiters, memory elements and optical triodes.

Of greatest technological importance has been the development of miniaturised devices, requiring low switching powers. These are seen to provide a gate-way to an entirely new field of communication systems: one utilising optical switching and optical signal processing. The ultrahigh switching times attainable lead to the exciting possibility of an alloptical computer.

The most commonly considered example of an optically bistable device consists of a Fabry-Perot interferometer filled with a medium in which the refractive index is intensity dependent. This is shown schematically in Fig. 1.1



FABRY - PEROT INTERFEROMETER

Fig. 1.1. Simple Optically Bistable Device

These are known as intrinsic devices. Bistability occurs as follows: Increasing the input power to the resonator changes the intensity within the material causing a change in the refractive index. This change has the effect of shifting the Fabry-Perot resonance closer to the exciting field resonance. This results in an increase in the field within the resonator, again changing the refractive index. As can be seen, this feedback leads to a runaway effect, which will eventually (under certain conditions) result in a switching from negligible transmission to complete transmission.

In the pioneering experiments of Gibbs et.al., the Fabry-Perot interferometer was filled with sodium vapour. These atoms were chosen because they exhibit a saturable absorption (an absorption dependent on light intensity). Although important, this absorption was not the dominant effect, but instead the intensity dependence of the refractive index gave rise to the optical bistability. (In fact, optical bistability due to absorption alone is difficult to observe).

In such a sodium vapour-filled device however, bistability occurs only over a narrow wavelength region. Thus, highly frequency stabilised lasers must be used to observe the effect. Also, the device is relatively large ( $^{0}$  10 cm) and displays relatively slow switching times ( $^{0}$  microseconds) compared to electronic switches ( $^{0}$  nanoseconds).

Thus, the actual practical use of these sodium-vapour devices seems limited.

Another approach, developed by P. Smith and E. Turner of Bell Laboratories, (Murray Hill, New Jersey), makes use of a hybrid device to generate optical bistability (Smith, 1980). In this case, the Fabry-Perot interferometer is filled with an electro-optic crystal. A beam splitter sends part of the output beam to a detector which converts it to an electrical signal. After amplification, the signal is again applied to the crystal, through electrodes on the crystal end-faces. The electric field, which varies with light intensity, thus modulates the crystal's refractive index.

This so-called 'artificial non-linearity' is much greater than the non-linearity produced in any known intrinsic device. Experiments can thus be performed using low power continuous wave lasers. A further advantage of such a device is that it can be switched electrically as well as optically.

Another promising development in the quest for practically useful optically bistable devices was discovered independently by two groups; one at Bell Laboratories, Murray Hill (Gibbs et.al., 1979a); and the other at Heriot-Watt University in Edinburgh (Miller, et.al., 1979).

Both these groups conducted low temperature experiments (120 K or lower) and used semiconductor materials as the nonlinear medium. In the experiments of Gibbs et.al., GaAs was the active medium, whilst Miller et.al., used InSb.

It is interesting to note that the mechanism responsible for bistability is quite different in the two experiments. In GaAs, excitonic interaction provides the nonlinearity; whereas in InSb, interband excitation, modelled as a two-level system gives rise to the necessary nonlinear effect.

Although the observation of bistability still depends on wavelength in such semiconductor materials, the dependence is not as critical as in sodium vapour.

Also, the observed nonlinearities are large enough to allow for the use of very thin samples; and as the materials involved are solids a resonator can be simply constructed by preparing a flat sample with parallel faces. The large nonlinearities that are generated result in only a small optical path length being necessary to observe bistability. Thus the device exhibits relatively small response times ( $\sim$  picoseconds). Table I indicates the merits of the various devices discussed in this section. Clearly, semiconductor devices seem the most promising of the intrinsic type. The operating conditions of these are comparable to those of the hybrid type.

# Table I: Bistable Optical Devices: Operating Characteristics

(After Smith, 1980)

DEVICE	SWITCHING POWER (W)	SWITCHING TIME (s)	SWITCHING ENERGY (J)
INTRINSIC:			
CS <sub>2</sub>	$3 \times 10^{5}$	$5 \times 10^{-10}$	$1.5 \times 10^{-4}$
Na vapour	l x 10 <sup>-2</sup>	$1 \times 10^{-5}$	$1 \times 10^{-7}$
GaAs	$2 \times 10^{-1}$	$4 \times 10^{-8}$	$8 \times 10^{-9}$
InSb	1 x 10 <sup>-2</sup>	$<5 \times 10^{-7}$	<5 x 10 <sup>-9</sup>
HYBRID:			
LiNbO <sub>3</sub>	$1 \times 10^{-5}$	$5 \times 10^{-8}$	$5 \times 10^{-13}$
(Fabry-Perot Int.)			
Liquid-crystal matrix	5 x 10 <sup>-7</sup>	$4 \times 10^{-2}$	$2 \times 10^{-8}$

#### $\phi$ 1.2 Summary of Results

The exciting developments discussed in the previous section provided the motivation for this thesis, which involves an investigation of bistability in various systems.

In all cases, we will consider the intracavity behaviour of a nonlinear medium when driven by laser fields. Of greatest importance to us will be the determination of such properties as:

- Output intensities: describing any possible bistable behaviour (or any other interesting features), and
- 2) Fluorescent Spectra: as these can be experimentally observed, they provide us with a possible test of our theoretical model. Such a comparison will indicate the relative merit of our theory and the accuracy of predicted macroscopic properties.

The remainder of this chapter  $\oint$ 1.3 concerns the general methods and approximations we will utilize to obtain this information.

To systematically define many-body systems, our approach is within the framework of the second quantisation. We discuss the general form of the system's Hamiltonian in  $\oint 1.3(b)(i)$ .

Such a second quantised theory is characterised by an N-particle wavefunction, (N = number of particles in the system). When dealing with an interacting gas, N is typically of the order  $10^{23}$ , thus a full description of the entire system is clearly impossible. Fortunately, the wavefunction contains a lot of irrelevant information not required in the calculation of important physical properties.

We reduce the complexity of the system by adopting a statistical approach in which we specify only the expected probability of finding a particle in a given state. We then define the density operator of the system as an expansion in terms of the many-body wavefunction. Such an operator is analogous to a probability distribution function. It provides all information about the system, yielding such properties as expectation values. Discussion of all systems in this thesis is thus in terms of the master equation: an equation of motion for the density operator – described in detail in  $\oint 1.3(c)$  and (d).

Although the master equation contains all statistical information about the system, this information is not easily extracted. Instead, we find it necessary to transform our system to a complex phase-space in order to calculate required physical properties. We discuss such phasespace distributions in  $\oint 1.3(f)$ .

Finally in Chapter One, we discuss the basis of an approximation used throughout this thesis: adiabatic elimination of fast variables. This procedure enables the replacement of certain dynamical variables with their stationary solutions in all calculations, provided they relax on a time scale very much faster than other variables.

Chapter Two determines the conditions under which optical bistability can be observed in various non-linear systems. Discussion in this chapter is restricted to a deterministic theory only. In such an approach, quantum fluctuations are neglected. However, as we need only calculate steady state expectation values to determine expected output intensities, this approximation will not produce a significant deviation from reality.

We firstly consider optical bistability arising in Raman active media, and review a semiclassical theory due to Lugovoi (1977). This predicts bistability in Stokes intensity, dependent on input intensity. We then present a quantum mechanical theory based on a model similar to Lugovoi's: quantised lattice vibrations, which give rise to the observed Raman frequency shift, are adiabatically eliminated. Analysis shows that only dispersive bistability (non-zero detuning) will be observed in such a system. Further discussion reveals the possibility of absorptive bistability on inclusion of a non-resonant susceptibility into the system. However, as these effects are relatively small, such behaviour is unlikely.

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In contrast to the rigorous approach followed in the investigation of optical bistability in the Raman effect, we utilize an approximate formalism in the remainder of Chapter Two: Effective Hamiltonian approach.

We firstly discuss the optical behaviour of an effective two-level atomic model in this manner. On adiabatic elimination of atomic variables and in the dispersive limit (large detuning), this system exhibits identical behaviour to the previously discussed Raman system.

In  $\oint 2.2(b)$  we use an effective Hamiltonian first employed by Graham (1970), to consider optical bistability arising from a parametric oscillator resonantly driven by two input fields. The possibility of bistability in output intensity varying as one of the input intensities is predicted. This bistability is also shown to depend critically on the other input intensity.

Finally in Chapter Two, we consider the behaviour of a driven two-photon absorber, utilizing an effective Hamiltonian. On adiabatic elimination of atomic variables, this system behaves exactly like a parametric oscillator.

Thus Chapter Two reveals the occurrence of optical bistability in a wide variety of non-linear systems. It also shows how apparently different systems can exhibit the same macroscopic properties, in certain limits. The following equivalences were established:

Raman System ↔ Effective Two-level System (on adiabatic elimination of atoms and in dispersive limit)

Parametric Oscillator ↔ Two-photon absorber (on adiabatic elimination of atomic variables). Such a correspondence is not surprising, as although the respective systems describe different processes, the form of the nonlinearity responsible for optical bistability is the same.

In Chapter Three, we investigate a rather different bistable system - one involving three-level atoms.

We note that a fully quantum mechanical theory of optical bistability in two-level systems, containing quantum fluctuations, is an extremely complicated problem (Drummond and Walls, 1980b). The complexity generated by introducing a further atomic level into the theory renders all calculations intractable.

We thus again restrict discussion to the development of a deterministic theory, in which we ignore the effects of quantum fluctuations.

Calculations performed on optical-Bloch equations for such a system shows bistability arises, due to the phenomenon of coherent population trapping. Bistable behaviour is shown to vanish when atomic collisions become dominant, as the coherence necessary for population trapping is destroyed.

To date, such a bistability has not been experimentally observed.

Chapter Four introduces the concept of optical bistability in semiconductors.

Although both the experiments of Miller et.al. (1979) and Gibbs. et.al. (1979a) are discussed, we focus most attention on the latter experiment; in which light-exciton interaction was seen to provide the mechanism for bistability. The interband excitation model, proposed to describe the experiments of Miller et.al. is discussed in Chapter Eight.

Chapters Five and Six are devoted to the development of a microscopic theory of the intracavity interaction of coherent light with a medium consisting of excitons. To formulate the system Hamiltonian in terms of exciton operators, we make use of a unitary operator which transforms pairs of fermion operators to boson operators. We are then able to use a master equation approach and systematically include such effects as exciton-lattice and exciton-exciton interactions into our model.

In the two cases of high and low exciton densities, steady state analysis revealed bistability and hysteresis in the system. Bistability of a dispersive and absorptive nature in both exciton number and output intensity, dependent on input intensity was found.

As yet, only high exciton density dispersive bistability has been experimentally observed. The bistability curves we predicted in the high exciton density case agrees qualitatively with the experimental curves obtained by Gibbs et.al., for GaAs.

In Chapter Seven we discuss quantum fluctuations in the low exciton density semiconductor system, with reference to the Fokker-Planck equation. To calculate expectation values it is necessary to reduce the dimensionality of the system. We thus introduce the method of adiabatic elimination of variables in stochastic systems.

#### \$1.3 Theoretical Considerations

## ∮1.3(a) System Model

This thesis is devoted to a study of the interaction of light with various nonlinear systems. As we will employ the general methods of quantum optics, the approach to each problem will have several features in common.

In each case, the model for our system will involve the intracavity interaction of coherent radiation (laser fields) with a nonlinear medium. This is represented in the schematic diagram:



Fig. 1.2. Intracavity Interaction of Coherent Light with a Nonlinear Medium

The cavity (indicated by the two vertical bars in the figure) will, in general, be a Fabry-Perot interferometer.

For each problem, we will assume the incident field strongly couples to a single mode within the resonating cavity. This mode will in future be referred to as the cavity (or pump) mode.

Experimentally, such an assumption relies on the ease with which the cavity can be tuned. Clearly, the single mode approximation becomes physically unreasonable when the cavity mode frequencies are not widely separated.

We also assume that the cavity mode of interest couples, in turn, to a single mode comprising the non-linear substance. These 'system' modes will be of various forms: e.g. molecular vibrations, atomic levels, excitonic states.

We will be mostly interested in determining expressions for the output intensity, as optical bistability concerns the behaviour of output intensity, dependent on input intensity.

We now briefly review the general theoretical methods we will employ to study each problem.

#### §1.3(b) Second Quantised Theory of a Loss-less System

## $\oint$ 1.3(b)(i) Hamiltonian

Each of the systems we will be considering will involve the intracavity interaction of radiation with a medium comprised of atoms, (in gaseous or crystalline phase).

The non-relativistic classical Hamiltonian describing such a system neglecting losses, consists of three major parts, (Heitler, 1960),

$$H = H_{e.m.} + H_{e.m.-matter} + H_{I} \qquad (1.1)$$

where

$$H_{e.m.} = \frac{1}{2} \int_{cavity} \left( \varepsilon_0 |\underline{E}|^2 + \mu_0 |\underline{H}|^2 \right) dV \quad (1.1a)$$

is the Hamiltonian for the pure radiation field within the cavity. (  $\underline{E}$  and  $\underline{H}$  are the electric and magnetic fields, obeying Maxwell's equations;  $\mathcal{E}_o$ ,  $\mathcal{M}_o$  are the permittivity and permeability of the vacuum).

The term  $H_{e.m.-matter}$  describes the energy associated with the interaction of the radiation with charged material particles. It is obtained from the Hamiltonian for a free particle system, via the substitution:

(where P is the momentum of the particle, A is the vector potential of the electro-magnetic field).

Thus,

$$H_{e.m.-matter} = \mathcal{L}_{j} \left[ \frac{1}{2m} \left( p_{j} - e \underline{A}(\underline{r}_{j}) \right) + V(\underline{r}_{j}) \right] - (1.1b)$$

( $\underline{r}_j$  refers to the position of the jth particle and  $\bigvee(\underline{r}_j)$  is the external Coulomb potential: nuclear attraction in the case of bound electrons;  $\gamma n$  is the mass of the particle).

In the context of the first quantisation, we express the momentum operator as,

$$p \rightarrow \frac{\pi}{i} \Sigma$$

yielding,

$$H_{e.m.-matter} = \mathcal{E}_{j} \left[ \frac{1}{2m} \left( \frac{\pi \nabla}{i} - e \underline{A} \right) + V(\underline{r}_{j}) \right] \qquad (1.1c)$$

Finally, the term  $H_{I}$  is due to the interaction between particles. For example, for two electrons, this becomes a Coulomb interaction,

$$H_{I} = \frac{e^{2}}{|\Upsilon_{1} - \Upsilon_{2}|}$$
(1.1d)

As the systems described in this thesis will be comprised of many particles, discussion will be within the framework of the second quantisation. Such an approach illustrates the particle nature of the light and matter fields.

To this end, we define a quantum vector space for a many-body system by assuming that any complete set of operators describing a single particle can also be used to describe n such particles.

Thus we no longer specify the properties of each individual particle. Instead, the system is described in terms of the number of particles of eigenvalue  $\lambda_i$ , say, of the operator A. This is referred to as the occupation number,  $\eta_i$ , of the ith state.

That is, in the state vector space (Fock space) of the system, with basis vectors

$$|n_{1}, n_{2}, n_{3}, \dots, n_{i}, \dots \rangle$$
 (1.2)

for a given operator A, we associate an eigenvalue  $\lambda_1$  to  $n_1$  particles,  $\lambda_2$  to  $n_2$  particles, and  $\lambda_i$  to  $n_i$  particles, etc.

To use such a state vector space we must express the Hamiltonian in terms of operators which can properly act on the vectors described by Eq.(1.2). This is achieved by introducing boson and fermion operators.

## \$1.3(b)(ii) Bosons and Fermions

The boson annihilation and creation operators,  $b_i$  and  $b_i$ , are defined by,

$$b_i | n_i, \dots, n_i, \dots \rangle = \sqrt{n_i} | n_i, \dots, n_{i-1}, \dots \rangle$$

and

$$b_i \mid n_i, \dots, n_i, \dots \rangle = \sqrt{1 + n_i} \mid n_i, \dots, n_i + 1, \dots \rangle$$

Such operators satisfy the commutation relations:

$$[b_i, b_j] = [b_i, b_j] = 0$$
;  $[b_i, b_j] = \delta_{ij}$ 

The fermion annihilation and creation operators,  $\Delta_i$  and  $\alpha_i^{\dagger}$  are defined similarly; except that the occupation numbers can only take the values 0 and 1, and,

 $a: |0\rangle_{F} = 0$  where  $|0\rangle_{F} = vacuum state$ .

Also, fermions obey the anticommutation relations,

$$[a_{\mu}, a_{\nu}]_{+} = [a_{\mu}^{\dagger}, a_{\nu}^{\dagger}]_{+} = 0 ; [a_{\mu}, a_{\nu}^{\dagger}]_{+} = \delta_{\mu\nu}$$

which is sufficient to ensure that any state can only be occupied by one particle at a time: the Pauli Exclusion Principle.

Of particular interest in later chapters, will be the behaviour of a two-level atom in the presence of an exciting field.

Haken (1970) proves the formal equivalence between a system of twolevel atoms and a system of  $\frac{1}{2}$ -spins:

> SPIN OPERATORS  $\sigma^{+} = \sigma_{x} + i \sigma_{y} \iff a_{2}^{+} a_{1}$   $\sigma^{-} = \sigma_{x} - i \sigma_{y} \iff a_{1}^{+} a_{2}$  $\sigma_{3}^{+} a_{2} = a_{1}^{+} a_{1}$

where the components of the spin operator,  $\sigma_x$ ,  $\sigma_y$  and  $\sigma_y$  can be represented by Pauli matrices; and  $\sigma^+$ ,  $\sigma^-$  are spin-flip operators. The fermion operators  $\Omega_1$ ,  $\Omega_2$  refer to the two atomic levels

117, 127.

Using the fermion commutation relations and the fact that

$$a_{1}^{+}a_{1} + a_{2}^{+}a_{2} = 1$$

in a two-level system, we find the relations:

$$\begin{bmatrix} \sigma^+, \sigma^- \end{bmatrix} = 2 \sigma_3$$
$$\begin{bmatrix} \sigma^\pm, \sigma_3 \end{bmatrix} = -\frac{1}{7} \sigma^\pm$$
$$\begin{bmatrix} \sigma^+, \sigma^- \end{bmatrix}_+ = 1$$
$$(\sigma^+)^2 = (\sigma^-)^2 = 0$$

### (1.3(b)) (iii) The Pure Radiation Field

We quantise the pure radiation field in the cavity by expanding the vector potential in terms of plane waves and boson operators: (ignoring spin),

$$A(\mathbf{r},t) = \mathcal{E}_{\mathfrak{g}} \left( \frac{\pi}{2\omega_{\mathfrak{g}} \epsilon_{\mathfrak{o}} \mathsf{V}} \right)^{\frac{1}{2}} \widehat{\mathcal{E}}_{\mathfrak{g}} \left( \exp\left[ i\left( \underline{\mathsf{R}}_{\mathfrak{g}}, \underline{\mathsf{r}} - \omega_{\mathfrak{g}} t \right) \right] \right) + b_{\mathfrak{g}}^{\dagger} \exp\left[ -i\left( \underline{\mathsf{R}}_{\mathfrak{g}}, \underline{\mathsf{r}} - \omega_{\mathfrak{g}} t \right) \right] \right)$$

$$(1.3)$$

The boson operators have the time dependence in the Heisenberg picture:

$$b_{\ell}(t) = b_{\ell} \exp(-i\omega_{\ell}t)$$
;  $b_{\ell}^{\dagger}(t) = b_{\ell}^{\dagger} \exp(i\omega_{\ell}t)$ 

The propagation vector  $\underline{k}_{l}$ , satisfies,

$$\frac{\omega^2 \iota}{c^2} = |\underline{R}_{\ell}|^2$$

and the  $\hat{\underline{e}}_{\ell}$  are polarisation vectors, assumed perpendicular to each other

$$\underline{\underline{e}} \ \underline{k} \ \underline{\underline{e}} \ \underline{k'} = 0$$
  
and satisfying the transversality condition:  
Using the relations  
$$\mu_{\bullet} \ \underline{\underline{H}} = \nabla \times \underline{\underline{A}}$$
$$\underline{\underline{E}} = -\frac{\partial \underline{\underline{A}}}{\partial t}$$

and Eq. (1.3) in He.m., we find the Hamiltonian for the pure radiation field, (Louisell, 1973),

$$H_{e.m.} = \mathcal{E} \pi \omega_{\ell} \left( b_{\ell} b_{\ell} + \frac{1}{2} \right) \qquad -----(1.4)$$

Thus after second quantisation, the field is described in terms of separate energy quanta, of energy  $\hbar\omega_{l}$  .

### $\oint$ 1.3(b)(iv) Interaction between Field and Matter

We introduce the particle nature of the electrons comprising the matter by defining the field operators,

$$\Psi(\mathbf{r}) = \hat{\xi}_{\mu} (\mathbf{r}) \mathbf{a}_{\mu}$$

 $Q_{\mu}$  are solutions  $Q_{\mu}$  are fermion operators and the where the of the single particle Schrodinger equation:

$$\begin{bmatrix} -\frac{\hbar^2 \nabla^2}{2m} + V(\underline{r}) \end{bmatrix} (\varphi_u(\underline{r}) = E_{\mu}(\varphi_u(\underline{r})) = \hbar \omega_{\mu}(\varphi_u(\underline{r}))$$

We then formulate a second quantised theory by expressing the interaction Hamiltonian as, (Heitler, 1960),

$$H_{e.m.-matter} = \int \Psi^{+}(\underline{r}) \left\{ \frac{1}{2m} \left( \frac{\hbar}{i} \underline{\nabla} - e A(\underline{r}) \right)^{2} + V(\underline{r}) \right\} \Psi(\underline{r}) dV \quad (1.5)$$
  
and

$$H_{I} = \frac{1}{2} \int \Psi^{\dagger}(\underline{r}) \Psi^{\dagger}(\underline{r}') \frac{e^{2}}{|\underline{r} - \underline{r}'|} \Psi(\underline{r}') \Psi(\underline{r}) dV dV' \qquad (1.6)$$

Thus, the Hamiltonian for the loss-less system is given by Eqs. (1.4), (1.5) and (1.6).

We also include a term in the Hamiltonian to describe the interaction between the incident field and cavity mode,

Hp

The explicit form of this term is discussed in later chapters.

## $\oint$ 1.3(b)(v) Electric Dipole Approximation

Louisell (1973) shows that under the dipole approximation, we may replace the interaction Hamiltonian,

He.m.-matter = 
$$\frac{1}{2m} (p - eA)^2 + V(r)$$

by

He.m.-matter = 
$$\frac{1}{2m}P^2 + V(\underline{r}) - \mu \cdot \underline{E}(\underline{r}, t)$$
 - (1.7)

where  $\mu = e \underline{r}$  is the atomic dipole moment. We stress that this is only an approximation, the limitations of which are discussed by Power and Zienau (1959).

Despite its approximate form, however, we make frequent use of the interaction Hamiltonian Eq. (1.7) in further chapters.
### $\oint$ 1.3(c) Density Operator Equation of Motion

All statistical information about the system is obtained from the density operator, defined in the Schrodinger picture as,

where the state vector |arphi(t)
angle obeys the Schrodinger equation,

and  $\mathbb{P}_{\psi}$  is the probability of the system being in the state  $|\psi(t)\rangle$ .

Using Eq. (1.9) and its adjoint, we find, from Eq. (1.8), in the Schrodinger picture,

$$i\hbar \frac{\partial}{\partial t} \rho = \mathcal{E}_{\gamma} \rho_{\gamma} \left\{ i\hbar \frac{\partial}{\partial t} |\gamma\rangle \langle \gamma| + |\gamma\rangle i\hbar \frac{\partial}{\partial t} \langle \gamma| \right\}$$
$$= \mathcal{E}_{\gamma} \rho_{\gamma} \left\{ H_{1}\gamma \rangle \langle \gamma| - |\gamma\rangle \langle \gamma| H \right\}$$

$$\Rightarrow i \hbar \frac{\partial \rho}{\partial t} = [H, \rho] \qquad -----(1.10)$$

Which is the equation of motion for the density operator.

We note that the average value of any system operator,  ${\sf A}$  , is given by,

 $\langle A \rangle = \text{Trace}(A_{\rho})$  (1.11)

# ∮1.3(d) Quantum Theory of Damping: Master Equation

In order to present a realistic model of a physical system we must include a damping mechanism, to account for losses.

With this in mind, we utilize the quantum theory of damping, as presented by Louisell (1973).

We assume all damping arises from a weak coupling between the system and a large collection of other systems, having many degrees of freedom and in thermal equilibrium. Such a collection of systems is commonly referred to as a reservoir. For example, a reservoir could consist of modes of the radiation field, vibrational modes in a solid, or electron or hole states in an energy band of a solid. It is assumed that the system-reservoir interaction has little effect on the reservoir.

Following Louisell (1973) we write the total Hamiltonian for the system and reservoir as,

$$H_T = H + H_R + V$$

where H is the system Hamiltonian, described in  $\oint 1.3(b)$ ; H<sub>R</sub> is the reservoir Hamiltonian and  $\bigvee$  is the Hamiltonian describing the interaction between reservoir and system.

The system-reservoir interaction can be described as a scattering process from the system mode, of frequency  $\omega_{\circ}$ , to a reservoir mode of frequency  $\omega_{j}$ . The effective interaction energy takes the form:

$$V = \pi \mathcal{E}_{j} (X_{j} Q_{j}^{\dagger} M + h.c.)$$
 (1.12)

where  ${\sf M}$  is a system operator (fermion, boson) and  ${\sf Q}_j$  is a reservoir operator.

The coupling constant  $\chi_j$  denotes the strength of the interaction and will depend on the particular system we are considering.

Although the sum in Eq. (1.12) includes all modes of the reservoir, only energy conserving modes, satisfying  $\omega_j \approx \omega_o$  will interact strongly. As these terms evolve in time according to  $\exp \pm i(\omega_j - \omega_o) t$ which is approximately unity at  $\omega_j \approx \omega_o$ , we expect such processes to dominate in the interaction. However, terms of the form,  $Q_j M$ ,  $Q_j^{\dagger} M^{\dagger}$  have been neglected in the expansion of V. These terms vary as  $e \times p \pm i (\omega_j + \omega_0) t$  which is rapidly varying and will average out to zero over long times.

Such neglect of energy non-conserving terms is known as the rotating wave approximation (R.W.A.).

In general, the behaviour of the reservoir variables will be of little interest to us and we will be mainly concerned with the behaviour of the system.

However, the density operator corresponding to the total system contains reservoir information. Thus, the density operator equation of motion (defined by Eq. (1.10)), from which we obtain statistical properties of the system, also contains this reservoir information.

We wish to remove all explicit reference to the reservoir from the equation of motion of the density operator and thus obtain an equation in terms of system operators only. This is achieved by tracing  $\rho(t)$  over the reservoir variables, yielding a reduced density operator,

$$S(t) = Tr_{R} \rho(t)$$

( Tr<sub>R</sub> = trace over reservoir variables).

The statistical properties of any system operator is then defined in terms of the reduced density operator S(t). For example, the average value of M is given by,

$$\langle M(t) \rangle = Tr_{R,S} [M\rho(t)]$$
 (Tr<sub>R,S</sub> = trace over reservoir, system)  
= Tr<sub>R</sub> M Tr<sub>S</sub>  $\rho(t)$   
 $\Rightarrow \langle M(t) \rangle = Tr_{S} [M S(t)]$ 

The equation of motion for S(t) is known as the master equation, describing the system's evolution under coupling to a reservoir.

Louisell (1973) derives such a master equation, in the Markoff approximation, via an iterative procedure. From this very comprehensive work we now only quote the form of the damping master equation we will be using in further chapters.

## 1) Light mode interacting with a reservoir (Intracavity)

The interaction Hamiltonian takes the form:

$$V = \mathcal{E}_j X_j b_j^{\dagger} b + h.c.$$

where  $b_j$  is a reservoir operator; b is the boson operator describing the light mode.

Damping is described by the master equation:

$$\frac{\partial s}{\partial t} = \sqrt[3]{n} \left( [b^{\dagger}s, b] + [b^{\dagger}, sb] \right) + \sqrt[3]{(1+n)} \left( [bs, b^{\dagger}] + [b, sb^{\dagger}] \right)$$
(1.13a)

where  $\delta = \pi |\chi(\omega_0)|^2 g(\omega_0)$ ;  $\omega_0$  is the resonant frequency of the light mode and  $g(\omega_0)$  is the density of reservoir modes.

Physically,  $\delta$  represents the cavity half-width.

 $ar{n}$  is the thermal occupation number of the reservoir.

$$\bar{n} = [exp(hw_{kT}) - 1]^{-1}$$
 (1.13b)

We may also write Eq. (1.13a) in the more compact form:

$$\frac{\partial S}{\partial t} = \Im \{ [bs, b] + [b, sb] \} + 2 \Im [[b, s], b] \___(1.13c)$$

2) <u>Two-level atom interacting with a reservoir</u> (Neglecting atomic collisions) This is represented as follows:  $2 - \frac{1}{5} \hbar \omega_{Res}$ .

and the interaction Hamiltonian takes the form;

$$V = \mathcal{E}_j \, \chi_j \, \dot{b}_j \, \sigma^- + h.c.$$

where  $\sigma$ ,  $\sigma$ <sup>+</sup> are spin flip operators describing the two-level system. If we neglect the dephasing effect of collisions, damping is described by;

$$\frac{\partial S}{\partial t} = \frac{\delta_0}{2} \left\{ (1+\bar{n})([\sigma^-S,\sigma^+] + [\sigma^-, S\sigma^+]) + \bar{n}([\sigma^+S,\sigma^-] + [\sigma^+, S\sigma^-]) \right\} \quad ----(1.14)$$

where  $\delta_{\mathbf{o}}$  is the Einstein A-coefficient for spontaneous emission from a two-level atom.

We note that expressions such as Eqs. (1.13) and (1.14) have also been derived by Haken (1970).

The total time evolution of the system is thus described by the master equation:

$$\frac{\partial \rho}{\partial t} = -\frac{i}{\hbar} \left[ H, \rho \right] + \frac{\partial \rho}{\partial t} \bigg|_{damping}$$
where H is as given in  $\oint 1.3(b)$  and  $\frac{\partial \rho}{\partial t} \bigg|_{damping}$  is given by Eqs. (1.13),  
(1.14).

Before we consider in detail the methods used to obtain information about the system from the master equation, let us briefly review another fundamental concept: coherent states.

# ∮1.3(e) Coherent States (Louisell, 1973)

The number state representation,  $|n\rangle$  introduced in  $\oint 1.3(b)$  provides an orthogonal set of basis vectors, which describe a system of harmonic oscillators.

An alternative representation, more useful in the description of the radiation field, involves the coherent states  $|\alpha\rangle$ .

These minimal uncertainty states satisfy, (for a boson operator b),  $b | \alpha \rangle = \alpha | \alpha \rangle$ 

They are constructed from the number states as follows:

 $|\alpha\rangle = e^{-\frac{1}{2}|\alpha|^2} \sum_{n=0}^{\infty} \frac{\alpha^n}{\sqrt{n!}} |n\rangle$ 

Such states are not orthogonal:

$$\langle \beta | \alpha \rangle = e^{-\frac{1}{2}(|\alpha|^2 + |\beta|^2) + \alpha \beta^*}$$

but do form an over-complete set, obeying the completeness relation:

$$\int |\alpha\rangle \langle \alpha| \frac{d^2 \alpha}{\pi} = \xi_0^{\infty} \ln \langle \alpha| = 1$$

We now review methods used in later chapters to obtain required information about the system.

#### 

The master equation derived in  $\oint 1.3(d)$  contains all the statistical information about the system. The total behaviour of the system: matter-field interactions, behaviour of matter and fields alone and damping effects - is described by this equation.

An exact solution of the master equation would yield a great deal of knowledge about the system.

In certain problems, time dependent matrix elements of the density operator / have been obtained by use of Laplace transforms (Lambropolous, 1967; McNeil and Walls, 1974). Such matrix elements describe atomic populations and polarisations. However, in general, extraction of information from the master equation in this manner is an arduous task (see for example Saxena and Agarwal, 1980).

We note that the density operator plays the same role as a probability distribution defined in a phase space for the system.

We may define the system variables in a complex phase space by constructing a suitable probability distribution to which we may map the density operator. By adopting such a phase space representation, we can calculate expectation values and time ordered correlation functions of variables by performing classical averages, and yet still retain all quantum mechanical information. This is because the transformed master equation can commonly be expressed as a Fokker-Planck equation, which may be often exactly solved.

More importantly, the stochastic differential equations corresponding to these Fokker-Planck equations can always be solved in a linearised theory (Chaturvedi et.al., 1977). Thus, utilization of such techniques provides a general solution for systems close to equilibrium, in which the linearisation approximation is valid.

The first phase space quasi-probability function was proposed by Wigner (1932); which related the density operator to a c-number distribution of classical variables:

$$W(p,q) = (\frac{1}{(2\pi)^2} \iint e^{-ip\mu - iq\nu} Tr(e^{i\mu\hat{p} + i\nu\hat{q}}p) d\mu d\nu - (1.15)$$

where  $\hat{q}$  and  $\hat{p}$  are coordinate and momentum operators of a harmonic oscillator; q and p are classical variables.

Thus, W(p,q) (the Wigner function) establishes a correspondence between classical variables and quantum mechanical operators:

$$\begin{array}{c} q \rightarrow \hat{q} \\ p \rightarrow \hat{p} \end{array}$$

Further development of such transformations were given independently by Glauber (1963) and Sudarshan (1963).

# $\oint 1.3(f)(i)$ Glauber-Sudarshan P-representation

This transformation expresses the density operator as a diagonal expansion of coherent states:

$$\rho = \int P(\underline{\alpha}) |\alpha\rangle \langle \alpha | d^2 \alpha \qquad (1.16)$$

where  $P(\underline{\alpha})$  is the quasi-probability distribution.  $[(\underline{\alpha}) = (\alpha, \alpha^*)]$ 

From the operator identities: ( b - boson operator).

$$b|\alpha\rangle\langle\alpha| = \alpha|\alpha\rangle\langle\alpha|$$

$$b^{\dagger}|\alpha\rangle\langle\alpha| = \left[\alpha^{*} + \frac{\partial}{\partial\alpha}\right]|\alpha\rangle\langle\alpha|$$

$$|\alpha\rangle\langle\alpha|b^{\dagger} = |\alpha\rangle\langle\alpha|\alpha^{*}$$

$$|\alpha\rangle\langle\alpha|b = \left[\frac{\partial}{\partial\alpha^{*}} + \alpha\right]|\alpha\rangle\langle\alpha|$$
(1.17)

the transformations follow:

$$b\rho = \int \left[ \alpha P(\underline{\alpha}) \right] |\alpha\rangle \langle \alpha | d^{2}\alpha$$

$$b' = \int \left[ \left( \alpha^{*} - \frac{\partial}{\partial \alpha} \right) P(\underline{\alpha}) \right] |\alpha\rangle \langle \alpha | d^{2}\alpha$$

$$\rho b' = \int P(\underline{\alpha}) \alpha^{*} |\alpha\rangle \langle \alpha | d^{2}\alpha$$

$$\rho b = \int \left[ \left( \alpha - \frac{\partial}{\partial \alpha^{*}} \right) P(\underline{\alpha}) \right] |\alpha\rangle \langle \alpha | d^{2}\alpha$$
(1.18)

By applying transformations (1.16), and (1.17), we can derive a Fokker-Planck equation from the master equation.

The Glauber-Sudarshan P-representation is, however, not applicable to a certain class of problems arising in quantum optics. For example, use of this P-representation in the case of a driven single mode interferometer filled with a non-linear absorber, yields a Fokker-Planck equation with a non-positive-definite diffusion term (Drummond and Gardiner, 1980). Thus a smooth, normalisable distribution function cannot be defined for this system; and we cannot form stochastic differential equations from the Fokker-Planck equation.

Problems related to singularities in the P-function can be removed by using instead the Glauber R-representation (Glauber, 1963):

$$\rho = \frac{1}{\pi^2} \iint d^2 \alpha \ d^2 \beta \ R(\alpha^*, \beta) \exp[-(|\alpha|^2 + |\beta|^2)/2] \ |\alpha\rangle < \beta |\beta|^2$$

which always exists. Unfortunately, the R-representation is not easily applied to the type of problem arising in quantum optics.

A more useful approach is to employ the generalised P-representations developed by Drummond and Gardiner (1980).

### $\oint$ 1.3(f)(ii) Complex P-representation

In this representation, we expand the density operator in terms of a non-diagonal projection operator:

$$\rho = \iint_{c,c'} P(\alpha,\beta) \Lambda(\alpha,\beta) d\alpha d\beta \qquad (1.19)$$

where  $P(\alpha,\beta)$  is the complex-P function;  $\Lambda(\alpha,\beta)$  is a projection operator; and  $\alpha$  and  $\beta$  are complex variables to be integrated on separate contours C and C'.

Systems which exhibited non-normalisable distribution functions under application of the Glauber-Sudarshan P-representation can now be solved via the complex-P representation; as appropriate choice of the contours Cand C' in the complex phase space of  $\prec$  and  $\beta$  always allows the exact solution of such Fokker-Planck equations.

The operator identities for this transformation are:

$$b \Lambda = \alpha \Lambda$$

$$b^{\dagger} \Lambda = \left[\beta + \frac{\partial}{\partial \alpha}\right] \Lambda$$

$$\Lambda b^{\dagger} = \Lambda \beta$$

$$\Lambda b^{\dagger} = \left[\frac{\partial}{\partial \beta} + \alpha\right] \Lambda$$
(1.20)

and the following transformations exist

$$b \rho \longleftrightarrow \alpha P(\alpha, \beta)$$

$$b \rho \longleftrightarrow (\beta - \partial_{\beta \alpha}) P(\alpha, \beta)$$

$$\rho b^{\dagger} \longleftrightarrow P(\alpha, \beta) \beta$$

$$\rho b \longleftrightarrow (-\frac{2}{2\beta} + \alpha) P(\alpha, \beta)$$
(1.21)

Again, we generate a Fokker-Planck equation from the master equation on application of the transformations, Eqs. (1.19), (1.20), (1.21).

To justify the existence of the corresponding stochastic differential equations, however, we must employ the positive P-representation.

#### $\phi$ 1.3(f)(iii) Positive P-representation

This defines the density operator through the transformation,

$$\rho = \int P(\alpha, \beta) \Lambda d^2 \alpha d^2 \beta \qquad (1.22)$$

where  $\alpha$  and  $\beta$  now vary over the whole complex plane.

Drummond and Gardiner (1980) show that for any Fokker-Planck equation derived via the Glauber-Sudarshan P-representation, a corresponding equation with positive semi-definite drift coefficient exists in the positive P-representation. Also, from such a Fokker-Planck equation we may correctly define stochastic differential equations.

The identities of the complex P-representation (Eqs. (1.20), (1.21)) also apply to the positive P-representation. In addition, there exist the further properties,

$$\begin{array}{c} b^{\dagger}\rho \iff \left[\beta - \frac{\partial}{\partial \alpha_{x}}\right]^{P(\alpha,\beta)} \iff \left[\beta + i\frac{\partial}{\partial \alpha_{y}}\right]^{P(\alpha,\beta)} \\ \text{and} \\ \rho b \iff \left[-\frac{\partial}{\partial \beta_{x}} + \alpha\right]^{P(\alpha,\beta)} \iff \left[i\frac{\partial}{\partial \beta_{y}} + \alpha\right]^{P(\alpha,\beta)} \\ \end{array}$$

where

$$\alpha = \alpha_{x} + i \alpha_{y}$$
  
$$\beta = \beta_{x} + i \beta_{y}$$

Thus, by application of the Drummond-Gardiner generalised Prepresentations, construction of a Fokker-Planck equation with a normalisable distribution function is always possible; and we may also form the corresponding stochastic differential equations.

For completeness, we note the general form of a Fokker-Planck equation for a set of C - number variables  $(\alpha_1, \ldots, \alpha_n) = (\underline{\alpha})$ :

$$\frac{\partial P}{\partial t}(\underline{a},t) = \sum_{l} \frac{\partial}{\partial a_{l}} A_{l}(\underline{a},t) P(\underline{a},t) + \frac{1}{2} \sum_{l} \frac{\partial}{\partial a_{l}} \frac{\partial}{\partial a_{j}} D_{lj}(\underline{a},t) P(\underline{a},t)$$
(1.23)

In the positive-P representation, this becomes, (Drummond, 1979)

$$\frac{\partial P(a,t)}{\partial t} = \mathcal{E}_{I} \left\{ \frac{\partial}{\partial a_{I}^{x}} A_{I}^{x} + \frac{\partial}{\partial a_{I}^{y}} A_{I}^{y} \right\} P(a,t) + \mathcal{E}_{Ij} \frac{\partial}{\partial a_{I}^{y}} \frac{\partial}{\partial a_{J}^{y}} P(a,t)$$

$$+\frac{1}{2} \sum_{j} \left\{ \frac{\partial}{\partial a_{j}^{x}} \frac{\partial}{\partial a_{j}^{x}} d_{k}^{x} d_{j}^{x} + \frac{\partial}{\partial a_{k}^{x}} \frac{\partial}{\partial a_{j}^{y}} d_{k}^{y} d_{j}^{y} \right\} P(\underline{\alpha}, t)$$
(1.24)

where  $\alpha_{I} = \alpha_{I}^{\times} + i \alpha_{I}^{\vee}$ ,  $A_{I} = A_{I}^{\times} + i A_{I}^{\vee}$ and Γ

$$d_{\underline{j}} d_{\underline{j}}^{\mathsf{T}} ]_{I_j} = \mathsf{D}_{I_j}$$

As Eq. (1.24) has an explicitly positive-semi-definite diffusion coefficient, we may form the corresponding Ito (Arnold, 1974) stochastic differential equation:

$$\frac{d}{dt} dt = -A_{\ell}(d,t) + d_{\ell}(d) \mathcal{E}_{\mu}(t) \qquad (1.25)$$

where the  $\mathcal{J}_{\mu}(t)$  comprise a Gaussian process,

$$\langle \xi_i(t) \xi_j(t') \rangle = \delta_{ij} \delta(t-t')$$

In most cases, it will be more convenient to write Eq. (1.25) as,

$$\frac{\partial}{\partial t} d t = -A t (d) + f t (t) \qquad ----(1.26)$$

 $\uparrow$  's are fluctuating terms describing noise in the system. where the They have the property

$$\langle \Gamma_{j}(t) \rangle = 0$$
 (1.27a)

and obey the correlation relations:

$$\langle T_i(t) T_j(t') \rangle = \delta(t-t') D_{ij}$$
 (1.27b)

where the  $D_{ij}$  are diffusion coefficients, defined in Eq. (1.23). Finally in this introductory chapter, we review the basis and importance of an approximation frequently used in quantum optics: adiabatic elimination of variables.

#### \$1.3(g) Adiabatic Elimination of Variables: an introduction

We can describe a general multi-dimensional system in terms of a set of variables,  $\mathcal{A}(\widehat{a}_1, \widehat{a}_2, .., \widehat{a}_n)$ .

In some cases potential conditions are satisfied and we may exactly solve the Fokker-Planck equation for such systems, obtaining the quasiprobability distribution function. Average values and correlation functions of system variables may then be calculated. These represent the most important calculations we can perform, as they yield expressions for emitted intensities and fluorescent spectra - experimentally observable features.

However, such exact solutions to the Fokker-Planck equation exist only in special cases.

If it is possible to assume the linearisation approximation, there exist techniques enabling the derivation of statistical averages of variables for two-dimensional systems, (Chaturvedi et.al., 1977).

For systems of higher dimensionality, however, similar calculations become extremely complicated.

A common approach to simplify the system is to reduce the dimensionality of the system by considering physical conditions under which certain variables may be eliminated. This method, known as adiabatic elimination of (fast) variables, relies on the fact that certain variables relax at a much faster rate than others: they possess larger damping coefficients than other variables. They will thus reach their steady state before the other variables have significantly developed in time. We may then disregard the time dependence of these so-called fast variables, which are said to adiabatically follow the other system variables. Mathematically, this amounts to replacing the fast variables with their steady state values in all system equations.

Firstly, let us consider how such a procedure is implemented in a deterministic theory, where quantum fluctuations (noise terms) are neglected. Our system  $\mathcal{A}(\widehat{\alpha}_1, \widehat{\alpha}_2, \dots, \widehat{\alpha}_n)$  can then be described by the equations,

$$\dot{a}_{1} = f_{1} (\hat{a}_{1}, ..., \hat{a}_{n})$$

$$\dot{a}_{2} = f_{2} (\hat{a}_{1}, ..., \hat{a}_{n}) \qquad (1.28)$$

$$\vdots \qquad \vdots$$

$$\dot{a}_{n} = f_{n} (\hat{a}_{1}, ..., \hat{a}_{n})$$

We eliminate variables 1 and 2 say, if they are more heavily damped than the other variables. The system is then described by the set,  $\mathcal{A}'(\widehat{a}_{3,..}\widehat{a}_{\cdot})$  and the equations,

$$\hat{a}_3 = f_3 \left( (\hat{a}_1)_{ss}, (\hat{a}_2)_{ss}, \hat{a}_3, \dots \hat{a}_n \right)$$

$$\vdots \qquad (1.29)$$

$$\hat{a}_n = f_n \left( (\hat{a}_1)_{ss}, (\hat{a}_2)_{ss}, \hat{a}_3, \dots \hat{a}_n \right)$$

where  $(\widehat{\alpha}_{\iota})_{ss}$  and  $(\widehat{\alpha}_{\iota})_{ss}$  are formed by setting  $\dot{\widehat{\alpha}}_{\iota} = 0$  and  $\dot{\widehat{\alpha}}_{\iota} = 0$ in Eqs. (1.28).

We have thus reduced the dimensionality by two.

This method was used in the theory of the laser, (Lamb, 1964), where it was assumed the atomic linewidths were much larger than the cavity linewidth. The atomic variables thus follow the electric field adiabatically. The approximation is quite justified in the case of the gas laser, where atomic decay rates are  $\sim 10^8 \ {\rm sec}^{-1}$  and radiative decay rates  $\sim 10^7 \ {\rm sec}^{-1}$ .

The procedure of adiabatic elimination of variables is thus well defined when we are dealing with deterministic equations. However, as a complete description of the system requires the inclusion of quantum fluctuations, we need to consider the system's Fokker-Planck and stochastic differential equations.

Great care must be taken when we attempt adiabatic elimination on stochastic differential equations, because of the presence of the fluctuating noise terms. The necessary procedures to achieve this end are discussed in detail in Chapter Seven.

#### CHAPTER 2

#### OPTICAL BISTABILITY IN NONLINEAR SYSTEMS

The theoretical foundations of nonlinear optics were firmly established by the early 1930's.

However, at that time, the experimental observation of the associated effects was not possible because the high field intensities, necessary to produce the nonlinearities, were not attainable.

That is, propagation of light through a medium generates a polarising effect on the outer electrons of the atoms comprising the medium. If the electric field amplitude of the incident light wave is of the same order as the electric field binding the electrons to the atoms  $(\sim 3 \times 10^8 \text{ V/cm})$ , a nonlinear relationship between the polarisation and the electric field will be clearly observed.

The classical light sources available in the 1930's produced electric fields much smaller than the atomic Coulomb fields. These classical radiation fields, acting only as a small perturbation to atomic fields, thus induced a polarisation directly proportional to the applied electric field.

However, with the advent of the laser in the 1960's higher power flux densities became available for use in optics experiments.

As lasers exhibit light fields in excess of 10<sup>8</sup> V/cm the nonlinear relationship between electric field and polarisation is readily observed experimentally. In crystalline media, which are in general anisotropic, the polarisation is expressed as,

$$\underline{P} = \mathcal{E}_{\bullet} \left( \underline{\chi}^{(i)} \underline{E} + \underline{\chi}^{(2)} \underline{E} \underline{E} + \underline{\chi}^{(3)} \underline{E} \underline{E} \underline{E} \underline{E} + \dots \right)$$

$$re \qquad \mathcal{E}_{\bullet} \qquad \text{is the permittivity of the vacuum and} \qquad \gamma^{(n)}$$

where  $\xi_o$  is the permittivity of the vacuum and  $\chi^{c}$  is the nth order susceptibility tensor. The first term represents the usual, linear polarisation.

As mentioned in Chapter One,  $\oint$ l.l, such a non-linear response of a medium to applied fields provides the mechanism for optical bistability.

Various nonlinear systems have been investigated in the study of optical bistability. The most well known of these is the system of a Fabry-Perot interferometer filled with a gas of two-level atoms, first discussed by McCall (1974). A semiclassical theory based on this model was presented by Bonifacio and Lugiato (1976), and a fully quantum mechanical theory has been proposed by Drummond and Walls (1980b).

An alternative approach in the investigation of optical bistability was adopted by Marburger and Felber (1978) and Drummond and Walls (1980a). These theories are based on a non-linear polarisability model, which involves the inclusion of a cubic nonlinearity in the polarisation.

In the first section of this chapter, we investigate the conditions necessary to observe optical bistability in Raman active materials. We show that such materials also exhibit a cubic nonlinearity in the polarisation.

We later consider materials displaying parametric oscillation and finally investigate optical bistability arising from two-photon absorption.

Throughout this chapter we adopt a quantum mechanical approach; however, quantum fluctuations are not properly included, as we assume complete factorisation of expectation values. Thus we develop only a deterministic theory.

### \$2.1 Raman Processes

When a light beam traverses a crystalline medium, scattering from electrons comprising the substance occurs. Molecular vibrations, however can noticably affect this scattering, resulting in the emitted light being shifted by a frequency characteristic of these vibrations. This is known as Raman scattering. Quantum mechanically, Raman scattering is the process in which a system absorbs a photon of frequency  $\omega_i$  and emits a photon of frequency  $\omega_{\mathfrak{F}}$ , while undergoing a transition to an excited state  $\mathfrak{t}(\omega_i - \omega_{\mathfrak{F}})$ . In a molecular system, the final state is a vibrational level.

The general theory of Raman scattering has been well documented, for example, see, Bloembergen (1965), Penzkofer et.al. (1980), Bloembergen and Shen (1965), Giordmaine and Kaiser (1966), Wang (1969), to quote only a few.

We present now only the salient features of the process, before considering in detail the mechanism for optical bistability.

# $\oint 2.1(a)$ Interaction Hamiltonian and Nonlinear Polarisation

We consider the interaction of radiation with a system of  $\, \, \mathfrak{N} \,$  molecules, each consisting of two or more atoms.

Radiation interacting with such a molecule induces an electric dipole moment  $\mu$ , directly proportional to the electric field  $\underline{E}$ ,

$$\mu = \mathcal{E}_{\mu i} = \mathcal{E}_{\lambda i j} E_{j} \qquad -----(2.1)$$

where  $d_{ij}$  is the electronic polarisability tensor of the i'th molecule.

Physically, the dipole moment is due to the displacement of the electrons with respect to the nucleus of a particular atom. Thus, if the nuclei always remain stationary,  $d_{ij}$  is a constant at any given frequency. However, the nuclei are not stationary and oscillate about their equilibrium positions. This clearly results in a departure from linearity. Placzek (1934) described this distortion mathematically by expanding  $d_{ij}$  in a Taylor series of normal mode vibrational coordinates,  $Q_{ij}$ , (For a discussion of normal mode coordinates see Pantell and Puthoff, 1969, Chapter 7):

$$\alpha_{ij} = \alpha_{ij} + \left(\frac{\partial_{\alpha_{ij}}}{\partial q}\right)_{equ.} \qquad (2.2)$$

where  $\alpha_{ij}^{\circ}$  is the polarisability evaluated at the equilibrium positions of the nuclei.

Introducing an explicit time dependence into E and q of the form,

$$\tilde{q} = q \cos \omega_r t$$
,  $E_j = \tilde{E}_j \cos \omega_j t$ 

Eq. (2.1) becomes,

$$\mu_{i} = \alpha_{ij} \tilde{E}_{j} \cos \omega_{j} t + \frac{1}{2} \left( \frac{\partial \alpha_{ij}}{\partial q} \right)_{equ} \tilde{Q} \tilde{E}_{j} \left[ \cos(\omega_{j} + \omega_{v})t + \cos(\omega_{j} - \omega_{v})t \right]^{(2.3)}$$

We see then that molecular vibrations add corrections to the dipole moment, producing two new emission lines, one above and one below the excitation frequency  $\omega_j$ . This is, of course, the observed Raman effect.

The term  $\alpha_{ij} \tilde{E}_j \cos \omega_j t$  yields the usual dipole transition spectrum. The emission lines corresponding to the frequencies  $\omega_j - \omega_{\sigma}$ and  $\omega_j + \omega_{\sigma}$  represent Stokes and Anti-Stokes emission, respectively.

From Eq. (2.3) we see that the term responsible for the Raman effect in Eq. (2.2) is,

$$(\alpha_{ij})_{Raman} = \left(\frac{\partial \alpha_{ij}}{\partial q}\right)_{equ.}$$

$$\Rightarrow \mu_i = \left(\frac{\partial \alpha_{ij}}{\partial q}\right) q E_j \qquad -----(2.4)$$

- we assume all spatial derivatives of  $\alpha_{ij}$  are evaluated at the equilibrium nuclei position.

Thus, from Eq. (1.7) derived in  $\oint 1.3(b)(v)$ , we find the interaction energy corresponding to the Raman effect is, i.e.

$$H_{INT} = -\mu \cdot \underline{E}$$

$$H_{INT} = -\frac{1}{2} \mathcal{E}_{ij} \left( \frac{\partial \alpha_{ij}}{\partial q} \right) q E_{j} E_{i} \qquad (2.5)$$

Also, if there are  $N_v = \frac{n}{\sqrt{v}}$  identical molecules per unit volume in the substance, then the nonlinear polarisation is given by

$$\underline{P} = \frac{1}{\sqrt{2}} \left\langle \begin{array}{c} \mu_i \right\rangle = N_{\sqrt{2}} \left\langle \mu_i \right\rangle = N_{\sqrt{2}} \left\langle \begin{array}{c} \mu_i \right\rangle = N_{\sqrt{2}} \left\langle \begin{array}{c} \frac{\partial \alpha}{\partial q} \right\rangle \left\langle q \right\rangle E \qquad ----(2.6)$$
(where  $\langle \rangle$  denotes statistical average).

We will show in  $\oint 2.1(c)(ii)$  that in general,

$$\langle q \rangle = f(E^2)$$

Thus,

$$P \propto E_i E_j E_k$$

showing that the Raman effect is associated with a polarisation cubic in electric field amplitudes.

We now turn to a discussion of optical bistability in Raman systems.

# ∮2.1(b) Semiclassical Approach

A semiclassical theory of optical bistability arising from the coherent excitation of a Fabry-Perot interferometer filled with a Ramanactive substance was first presented by Lugovoi (1977).

In this semiclassical approach, the molecular system is treated quantum mechanically, whereas the fields are assumed to be classical.

He assumes the polarisation takes the form given by Eq. (2.6) and obtains an equation of motion for the vibrational mode:

$$\ddot{X} + 2h\dot{x} + \omega_r x = \frac{1}{m} \frac{\partial d}{\partial x} E^2 \qquad (2.7)$$
(where  $X = \langle q \rangle$ )

As expected, Eq. (2.7) greatly resembles the equation of motion for a driven damped harmonic oscillator: (m = reduced mass; 2h = width of spontaneous Raman scattering line;  $\omega_r$  = resonance frequency of vibrational mode). In this theory, the electromagnetic field obeys Maxwell's equations. Thus the propagation of the light field is governed by the wave equation:

$$\underline{\nabla} \times \left( \underline{\nabla} \times \underline{E} \right) + \frac{1}{C^2} \frac{\partial^2 \underline{E}}{\partial t^2} = -\frac{4\pi}{C^2} \frac{\partial^2 \underline{P}}{\partial t^2}$$

where the electric field is a sum of Stokes and applied (cavity) field components. (Anti-Stokes emission is neglected in this theory).

One of Lugovoi's major assumptions is that the vibrational modes (phonons) are damped on a timescale much faster than that of the Stokes or laser modes. It is then possible to eliminate the phonons adiabatically. This amounts to replacing the variable X with its steady state value in the field equations (for fixed E ).

In this limit, he derives the following equations of motion:

$$\dot{\alpha}_{p} = -(\lambda_{p} + i \delta_{1}) \alpha_{p} - K_{L} \alpha_{p} |\alpha_{s}|^{2} + i E_{L}$$
 (2.8)  
 $|\dot{\alpha}_{s}|^{2} = -2\lambda_{s} + 2\omega_{s} K_{L} |\alpha_{p}|^{2} |\alpha_{s}|^{2}$  (2.9)

where

 $\alpha_p$  = complex amplitude of exciting (cavity field);

- $|\alpha_s|^2$  Stokes intensity;
- $\delta_1$  = detuning of cavity mode from external driving field,  $E_L$
- $\omega_s$  = Stokes frequency; ( $\lambda_p$ ,  $\lambda_s$  : cavity and Stokes damping respectively)
- $K_{L}$  = Raman coupling constant, as defined by Lugovoi =  $K'_{L} + i K''_{L}$

By solving these equations in the steady state  $(\dot{\alpha}_p = |\dot{\alpha}_s|^2 = 0)$ Lugovoi predicts bistability in Stokes intensity, dependent on the laser intensity  $|E_L|^2$ .

We may obtain greater insight into the system by adopting a quantum mechanical approach.

#### $\oint 2.1(c)$ Adiabatic Elimination of Phonons: Quantum Theory

As in the previous section, we consider the intracavity interaction of a coherent driving field with a Raman-active medium.

### $\oint$ 2.1(c)(i) Hamiltonian

The total physical system is described by the Hamiltonian H , discussed in  $\oint$ 1.3(b),

Herm is as described in Eq. (1.4) in  $\oint 1.3(b)(ii)$ .

The term  $H_{matter}$  describes the energy of the uncoupled molecular system and takes the general form,

The discussion of  $\oint 2.1(a)$  showed that the interaction term is given by,

In general, the electric field is the sum of the exciting (cavity) field,  $\underline{E}\rho$  and the scattered Stokes and Anti-Stokes fields,  $\underline{E}s$ ,  $\underline{E}_{AS}$ Let us restrict further discussion to the behaviour of the Stokes mode alone, and set,

$$E = E_p + E_s$$

The Hamiltonian assumes its proper second quantised form if we express the system variables as,

$$q_{j} = (mN)^{-\frac{1}{2}} \sum_{Rr} (expi \underline{Rr}_{j}) (b_{Rr}(t) + b_{-Rr}(t)) (\frac{\pi}{2\omega})^{\frac{1}{2}} \qquad ----(2.13a)$$

$$\underline{E} = \mathcal{E}_{\lambda} \left( \frac{2\pi \hbar \omega_{\lambda}}{\varepsilon} \right)^{1/2} \hat{\underline{e}}_{\lambda} i \left[ b_{R_{\lambda}}(t) \exp(-i\underline{R}_{\lambda} \underline{r}_{j}) - b_{R_{\lambda}}(t) \exp(i\underline{R}_{\lambda} \underline{r}_{j}) \right]$$

where N is the number of molecules within the interaction volume V;  $\hat{\underline{e}}_{\lambda}$  is the unit polarisation vector for the light field;  $\underline{r}_{j}$  denotes the position of the jth molecule and  $\underline{k}_{r}$  is the wave vector of the vth mode.

Substituting Eqs. (2.13a) and (2.13b) in Eqs. (2.11), (2.12), we find, in the rotating wave approximation (defined in  $\oint 1.3(d)$ )

( n = refractive index).

We now adopt a single mode approximation, in which we assume the incident field strongly couples to a single cavity mode (described by the operator  $b_{
m 
ho}$  ), which in turn couples strongly to a single Stokes mode. Thus we may write;

$$H_{matter} = \hbar w b b \qquad ----(2.14b)$$

$$H_{e.m.} = \pi w_p b_p b_p + \pi w_s b_s b_s \qquad -----(2.15b)$$

$$H_{INT} = \frac{1}{2} \chi \left[ \frac{1}{2} \frac{1}{$$

where

$$X = \left(\frac{\partial \alpha}{\partial q}\right) \frac{N^{\frac{1}{2}} \pi h^{\frac{3}{2}}}{n_{s} n_{p} V m^{\frac{1}{2}}} \left(\frac{2 \omega_{p} \omega_{s}}{\omega}\right) \stackrel{\text{ep}}{=} \stackrel{\text{es}}{=} \delta(\underline{R}_{p} - \underline{R}_{s} - \underline{R}_{v})$$

(Raman coupling constant).

and  ${\sf b}$  is the boson operator for the phonon mode, frequency  $\omega$  .

Assuming the applied laser field can be treated classically the Hamiltonian describing the interaction between incident field and cavity mode is;

where  $E_L$  is the amplitude of the driving field of frequency  $\omega_L$ . Finally, we must include damping of phonon, cavity and Stokes modes.

The discussion of  $\oint 1.3(d)$  showed that a valid description of damping is to assume coupling of the mode in question to a reservoir in thermal equilibrium.

The Hamiltonian takes the form;

 $H_{damping} = b_p Q_1^{\dagger} + b_s Q_2^{\dagger} + b Q_3^{\dagger} + h.c. \qquad ----(2.18)$ where  $Q_1, Q_2, Q_3$  are reservoir operators.

### $\oint 2.1(c)(ii)$ Master Equation and Average Values

We now form a master equation (equation of motion for density operator ho ) from the Hamiltonian, in a frame rotating at the laser frequency  $\omega_{
m L}$  ,

$$\frac{\partial \rho}{\partial t} = -\frac{i}{\hbar} \left[ \left\{ H_{\text{matter}} + H_{\text{e.m.}} + H_{\text{INF}} + H_{\text{D}} \right\}_{\text{I}}, \rho \right] + \frac{\partial \rho}{\partial t} \left| \frac{\partial \rho}{\partial t} \right|_{\text{damping}}$$
(2.19)

[ ] denotes interaction picture]

where 
$$\frac{\partial \rho}{\partial t} \Big|_{damping} = \hat{\Sigma}_i \, \forall i \{ [bi \rho, b_i] + [bi, \rho b_i] + 2 \bar{n}_i [[bi, \rho], b_i] \} + \forall v \{ [b \rho, b] + [b, \rho b^{\dagger}] + 2 \bar{n}_v [[b, \rho], b] \}$$

where  $\forall \sigma$  is decay rate of the phonon mode, ( the index i = s, p)  $\forall \rho$ ,  $\forall s$  are the rates of the Stokes and pump modes respectively and the  $\overline{n}_i$  are thermal occupation numbers of the reservoirs as discussed in  $\oint 1.3(d)$ . We obtain average values of the operators from the master equation as for any arbitrary operator 0,

$$\langle \dot{0} \rangle = \text{Trace}(\dot{\rho} 0)$$
 (2.20)

where  $\langle \rangle$  denotes expectation value and the dot indicates a derivative with respect to time. As mentioned in the beginning of this chapter, we are presenting only a deterministic theory, in which quantum fluctuations are ignored. We may thus assume complete factorisation of expectation values (e.g.  $\langle b_s b_p \rangle = \langle b_s \rangle \langle b_p \rangle$ , etc.). Using Eqs. (2.19) and (2.20), we then find, (for a cold reservoir,  $\bar{n} = 0$ ):

$$\dot{\alpha}_p = -(\vartheta_p + i\delta_i)\alpha_p + E_L - i\chi\alpha_s\beta$$
 (2.21a)

$$\alpha_{\rm s} = -\delta_{\rm s} \alpha_{\rm s} - \ell_{\rm s} \alpha_{\rm p} \rho \qquad (2.21b)$$

$$\dot{\beta} = -(\lambda_{r} + i\delta_2)\beta - i\lambda_{r} + i\lambda_{2}\beta^{*} \qquad (2.21c)$$

where  $\alpha_p = \langle b_p \rangle$ ,  $\alpha_s = \langle b_s \rangle$ ,  $\beta = \langle b \rangle$  and we note  $\alpha_p$ ,  $\alpha_s$ ,  $\beta$  are all complex numbers.

Also,  $\delta_1 = \omega_p - \omega_L$  is the relative detuning of the laser field with respect to the cavity mode we are considering; and  $\delta_2 = \omega - (\omega_p - \omega_s)$ is the frequency mismatch between the natural frequency of the phonon mode and that of the Stokes transition.

We now wish to adiabatically eliminate the phonon modes, following Lugovoi's approach.

Physically, this approximation is based on the assumption that the phonons are damped at a much faster rate than either the Stokes or pump modes, i.e.

As discussed in Chapter One, mathematically this is equivalent to setting  $\dot{\beta} = 0$  in Eq. (2.21c), yielding

We can now appreciate the cubic nonlinearity of the polarisation, by setting  $\langle q \rangle = \beta$  and  $E = \alpha p$  in Eq. (2.6),

$$\Rightarrow Equ. 2.6 \Rightarrow \underline{P} = -Nv \frac{\partial d}{\partial q} \frac{i \chi ds |dp|^2}{\partial q}$$

We may thus identify a third order nonlinear susceptibility, by forming the definition:

RAMAN NONLINEAR POLARISATION  

$$\frac{P}{P} = \chi_{RAMAN}^{(3)} ds |dp|^{2}$$
where  $\chi_{RAMAN}^{(3)} = \chi_{Nv} (\frac{\partial d}{\partial q}) [\delta_{2} - i \delta_{v}]$ 
(2.23)

Hence,  $\chi^{(3)}$  is a complex quantity, which becomes purely imaginary and negative on resonance  $(\delta_2 = 0)$ . This agrees with the expected behaviour of the third order Raman susceptibility, as discussed by Bloembergen (Bloembergen, 1970).

Returning to the field equations, Eq. (2.21), by substituting Eq. (2.22), we find the equations governing the system in the adiabatic limit:

$$\dot{\alpha}_{p} = -(\aleph_{p} + i\delta_{1})\alpha_{p} + E_{L} - \chi^{2} (\aleph_{r} + i\delta_{2})\alpha_{p}|\alpha_{s}|^{2} - (2.24a)$$

$$\dot{\alpha}_{s} = -\aleph_{s}\alpha_{s} + \chi^{2} (\aleph_{r} - i\delta_{2})\alpha_{s}|\alpha_{p}|^{2} - (2.24b)$$

$$\dot{\alpha}_{s} = -\aleph_{s}\alpha_{s} + \chi^{2} (\aleph_{r} - i\delta_{2})\alpha_{s}|\alpha_{p}|^{2} - (2.24b)$$

In such a deterministic theory, we can form an equation for the time development of the Stokes intensity,  $|\alpha_s|^2$ , from Eq. (2.24b);

$$|\dot{\alpha}_{s}|^{2} = \dot{\alpha}_{s} d_{s}^{*} + \dot{\alpha}_{s}^{*} d_{s}$$

We thus find the following field equations for this Raman system:

$$\dot{\alpha}_{p} = -(\lambda_{p} + i\delta_{1})\alpha_{p} + E_{L} - \kappa_{\alpha_{p}} |\alpha_{s}|^{2} - (2.25a)$$

$$|\alpha s|^2 = -2 \forall s |\alpha s|^2 + 2 K' |\alpha s|^2 |\alpha p|^2$$
 (2.25b)

where the Raman coupling constant is defined by;

$$K = K' + iK'', \quad K' = \frac{\chi^2 \delta_{\tau}}{\delta_{\tau}^2 + \delta_{\tau}^2}, \quad K'' = \frac{\chi^2 \delta_{\tau}}{\delta_{\tau}^2 + \delta_{\tau}^2}$$

We note that Eqs. (2.25) are equivalent to those derived by Lugovoi, Eqs. (2.8), (2.9).

# $\oint 2.1(c)$ (iii) Bistability Conditions

In the following discussion we will denote the Stokes intensity as,

and,

$$5 \leftrightarrow |\alpha_{s}|^{2}$$
  
I  $\leftrightarrow |E_{L}|^{2}$ 

Solving Eqs. (2.25) in the steady state  $(\dot{\alpha} \rho = |\dot{\alpha}_{S}|^{2} = 0)$  predicts two stationary regimes:

1) 
$$S = O \Rightarrow |\alpha p|^2 = \underline{I}$$
 (2.26a)  
and  $\vartheta_p^2 + \delta_1^2$ 

<sup>2)</sup> 
$$S \neq 0 \Rightarrow S^{2} (K^{12} + K^{\prime\prime 2}) + S(2K^{\prime} \aleph_{p} + 2\delta_{1} K^{\prime\prime}) + \aleph_{p}^{2} + \delta_{1}^{2} - \frac{K^{\prime}}{\aleph_{s}} I = 0$$
 (2.26b)

Thus the system has the following behaviour:



Fig. 2.1. Steady State Stokes Intensity vs Input Intensity

The system can exist in either of the two stationary states,

- (a) described by Eq. (2.26a)
- (b) described by Eq. (2.26b)

We now wish to determine the conditions under which the system will be found in each of these states; and the conditions required to observe a transition between stable states. We obtain this information by performing a linearised stability analysis.

### Stability Analysis

We investigate the behaviour of the system close to a stable state by considering the behaviour of small fluctuations about the steady state: i.e. set

$$dp = d_0 + \tilde{d}_p$$

$$S = S_0 + \tilde{S} \qquad (2.27)$$

where  $\alpha_0$ , So are the steady state values of  $\alpha_p$ , S; and  $\widetilde{\alpha}_p$ and  $\widetilde{S}$  are the time dependent fluctuations about these values.

$$\begin{pmatrix} \dot{\alpha}_{p} \\ \dot{\alpha}_{p} \\ \dot{\alpha}_{p} \\ \dot{\varsigma} \end{pmatrix} = \begin{pmatrix} -(\aleph_{p}+i\delta_{1})-\kappa s_{\circ} & 0 & -\kappa a_{\circ} \\ 0 & -(\aleph_{p}+i\delta_{1})-\kappa s_{\circ} & -\kappa a_{\circ} \\ 2\kappa' s_{\circ} a_{\circ} & 2\kappa' s_{\circ} a_{\circ} & 0 \end{pmatrix} \begin{pmatrix} \tilde{\alpha}_{p} \\ \tilde{\alpha}_{p} \\ \tilde{\varsigma} \end{pmatrix}$$
(2.28a)

 $\Rightarrow \dot{X} = A \times ; X = (\tilde{\alpha}_{p}, \tilde{\alpha}_{p}^{*}, \tilde{s}) ; A \text{ is the matrix}$ 

defined in Eq. (2.28a).

To find the eigenvalues corresponding to normal modes, we form,

$$det (I\lambda - A) = 0$$

$$\Rightarrow \quad \lambda^3 + \tilde{a}, \lambda^2 + \tilde{a}_2 \lambda + \tilde{a}_3 = 0 \qquad ----(2.28b)$$

where

$$\tilde{a}_{1} = 2(\lambda_{p} + \kappa's_{o}) ; \quad \tilde{a}_{2} = (\lambda_{p} + \kappa's_{o})^{2} + (\lambda_{1} + \kappa''s_{o})^{2} + 4\kappa'^{2}s_{o}|a_{o}|^{2}$$

$$\tilde{a}_{3} = 4\kappa's_{o}|a_{o}|^{2}(\lambda_{p}\kappa' + \kappa''\delta_{1} + 1\kappa|^{2}s_{o})$$

Equation (2.28b) is known as the dispersion equation and its roots determine the time dependence of the variables  $\widetilde{\alpha}_{p}, \widetilde{\alpha}_{p}^{*}, \widetilde{\delta}$ . For example,  $\widetilde{\alpha}_{p}$  will have the following time dependence,

$$\tilde{\alpha}_{p}(t) = \mathcal{E}_{i} f(\alpha_{o}, s_{o}) e^{\lambda i t}$$

Clearly, we require negative real parts of the eigenvalues  $\lambda$ : for stability in the system. This is known as the Hurwitz stability criterion and is equivalent to the conditions, Glansdorff and Prigogine (1974).  $\Delta_1 > 0$ ,  $\Delta_2 > 0$ ,  $\Delta_3 > 0$ 

where

$$\Delta_{1} = \tilde{\alpha}_{1} , \Delta_{2} = \begin{vmatrix} \tilde{\alpha}_{1} & 1 \\ \tilde{\alpha}_{3} & \tilde{\alpha}_{2} \end{vmatrix} , \Delta_{3} = \begin{vmatrix} \tilde{\alpha}_{1} & 1 & 0 \\ \tilde{\alpha}_{3} & \tilde{\alpha}_{2} & \tilde{\alpha}_{1} \\ 0 & 0 & \tilde{\alpha}_{3} \end{vmatrix}$$

Thus, for stability, we require

(i) 
$$\tilde{a}_1 > 0$$
 and (ii)  $\tilde{a}_3 > 0$ 

The first condition is always satisfied as  $\[mathcal{P}\]$ , K<sup>1</sup> and S<sub>o</sub> are all necessarily positive. However, condition (ii) is not always met, allowing for bistability in the system. That is, there will be a transition between stable states (a), b) in Fig. 2.1) when

$$\widetilde{\Omega}_{3} = O \implies K'' \delta_{1} = -(\delta_{p}K' + |K|^{2}S_{0}) \qquad (2.29)$$

Thus, a transition will only be observed if K'' and  $\delta_1$  are both non-zero. As this amounts to the presence of non-zero detuning in the system, we see that transitions will occur only in the dispersive regime.

We note that Eq. (2.29) is equivalent to the expression,

$$\partial I = O$$
, from Eq. (2.26b)

Hence, the system is stable with respect to small fluctuations when  $\frac{\partial I}{\partial 5} > O$ , which corresponds to a positive slope of the curve in Fig. 2.1; and the system becomes unstable when  $\frac{\partial I}{\partial S} < O$ , corresponding to a negative slope of the curve.

#### Transitions in the System

We may thus determine the behaviour of the system by examining the slope of the curve described by Eq. (2.26b). We note however, that Eq. (2.26b) predicts two types of behaviour.



Fig. 2.2. Stokes Intensity vs. Input Intensity

The arrows indicate transitions predicted by the stability analysis. Fig. 2.2a describes bistability and hysteresis, and is akin to a first order phase transition. Fig. 2.2b however, predicts a smooth transition between stable states and can be thought of as a second order phase transition.

The conditions necessary for the observation of case A or case B are determined by the coefficients in Eq. (2.26b):

1) Case A (analogous 1st order phase transition) will be observed if,

$$\frac{K' \delta_{p} + \delta_{1} K''}{K'^{2} + K''^{2}} < 0 \qquad ----(2.30a)$$

2) Case B requires

$$\frac{K' \delta_{p} + \delta_{1} K''}{K'^{2} + K''^{2}} > O \qquad (2.30b)$$

As, in a physically realistic system we have K' and  $\delta_p$  non-zero; bistability (analogous 1st order phase transition) will only be observed if  $\delta_1$  or K'' are negative.

Thus a necessary condition for bistability in the system is the inclusion of a non-zero detuning.

### $\oint$ 2.1(c)(iv) Non-Resonant Susceptibilities

Fundamental to our treatment of Raman interactions was the assumption that the exciting field was strongly coupled to a particular molecular transition. Transitions far off resonance were assumed negligible.

However, the effect of such non-resonant transitions can be included into the theory: as shown by Bloembergen and Shen (1965), these transitions result in a contribution to the third order susceptibility.

i.e.

$$\chi^{(3)} = \chi^{(3)}_{\text{Res.}} + \chi^{(3)}_{\text{NR}} \qquad \begin{bmatrix} \text{Res: Resonant} \\ \text{NR: Non-Resonant} \end{bmatrix}$$

The  $\chi_{NR}^{(3)}$  terms involve only pure electric-dipole matrix elements, with no resonant denominator and thus are real quantities. It is shown (Bloembergen & Shen, 1965) that the ratio of  $|\chi_{Res}^{(3)}|^2$  to  $|\chi_{NR}^{(3)}|^2$ is about 10 to 1.

Inclusion of such a non-resonant susceptibility into our theory would lead to the imaginary part of the constant K having a frequency independent part. Thus,

$$\mathsf{K}'' \; \Rightarrow \; \mathsf{K}'' + \mathsf{C}_1 \, \chi_{\mathsf{NR}}^{(3)}$$

where  $C_1$  is a constant depending on system parameters.

Hence K'' has a non-zero imaginary part, even at resonance. This allows the possibility of observing bistability at resonance, if  $\chi^{(3)}_{NR}$  is large enough.

However, as the magnitude of  $\chi_{NR}^{(3)}$  is relatively small, this situation would probably not occur.

Thus, in general, we conclude that bistability will only be observed in such a Raman system in the dispersive limit  $(\delta, \neq 0, \delta_1 \neq 0)$ .

### $\oint$ 2.1(d) Phonon Reservoir Theory

The discussion of  $\oint 2.1(c)$  assumed the Stokes and pump modes coupled strongly to a single phonon mode.

We consider now the system in which the phonons form a reservoir for the Stokes and pump modes. By treating the phonons as a reservoir in thermal equilibrium, we have essentially eliminated them; and we thus expect the behaviour of such a system to be similar to that described in  $\oint 2.1(c)$ .

The Hamiltonian is the same as before: Eq. (2.10), except the interaction term is now viewed as,

 $H_{INT} = \pi \chi \left[ b^{\dagger} b^{\dagger} s b p + h.c. \right]$ 

 $\Rightarrow H_{INT} = b_{s}^{+} b_{p} Q^{+} + h.c. \qquad -----(2.31)$ 

where Q is a reservoir phonon operator.

The master equation associated with the Hamiltonian, Eq. (2.31), is derived using the standard methods of the quantum theory of damping.

We thus find the master equation for the total system (in a frame rotating at the laser frequency  $\omega_{L}$  ),

The term  $\frac{\partial \rho}{\partial t} |_{damping}$  describes damping of Stokes and pump modes and assumes the usual form:

$$\frac{\partial \rho}{\partial t} = \mathcal{E}_i \, \forall i \left\{ \mathbb{E} b_i \rho, b_i^{\dagger} \right\} + \mathbb{E} b_i, \rho b_i^{\dagger} + 2 \, \bar{n} i \left[ \mathbb{E} b_i, \rho \right], b_i^{\dagger} \right\}$$

$$(2.33)$$
(*i=S,p*)
(*i=S,p*)
(*i=S,p*)
(*i=S,p*)
(*i=S,p*)
(*i=S,p*)
(*i=S,p)*
(*i=S*

The final term of Eq. (2.32) stems from the interaction term, Eq. (2.31). Treating the phonons as reservoir operators, we obtain the equation,

$$\frac{\partial \rho}{\partial t} = \chi \left\{ (1+\bar{n}) \left( \mathbb{E} b_{p} b_{s}^{\dagger} \rho, b_{p}^{\dagger} b_{s} \right] + \mathbb{E} b_{p} b_{s}^{\dagger}, \rho b_{p} b_{s} \right] \right\}$$

$$\frac{\partial \rho}{\partial t} \left| \underset{\text{Interaction}}{\text{Raman}} + \bar{n} \left( \mathbb{E} b_{p}^{\dagger} b_{s} \rho, b_{p} b_{s}^{\dagger} \right] + \mathbb{E} b_{p}^{\dagger} b_{s}, \rho b_{p} b_{s} \right] \right\} (2.34)$$

where  $\chi$  is the coupling constant and  $\overline{n}$  represents the thermal occupation number of a reservoir of phonons.

## ∮2.1(d)(i) Deterministic Theory

In the deterministic limit, we neglect quantum fluctuations and assume complete decorrelation of expectation values. We thus obtain the equations of motion from Eq. (2.32)  $(\vec{n} = 0)$ :

$$\dot{\alpha}_{p} = E_{L} - (\forall p + i\delta_{1}) \alpha p - \alpha_{p} \chi (|\alpha_{s}|^{2} + 1) \qquad (2.35)$$

$$\dot{\alpha}_{p}^{*} = E_{L}^{*} - (\forall p - i\delta_{1}) \alpha_{p}^{*} - \alpha_{p}^{*} \chi (|\alpha_{s}|^{2} + 1) \qquad (2.36)$$

$$\dot{\alpha}_{s} = - \forall_{s} \alpha_{s} + \chi \alpha_{s} |\alpha_{p}|^{2} \qquad (2.37)$$

$$\dot{\alpha}_{s}^{*} = - \forall_{s} \alpha_{s}^{*} + \chi \alpha_{s}^{*} |\alpha_{p}|^{2} \qquad (2.38)$$

where  $\langle b_p \rangle = d_p$ ,  $\langle b_s \rangle = d_s$ 

We obtain an equation of motion for the Stokes intensity  $|\alpha_s|^2$ , from Eqs. (2.37) and (2.38),

$$|\alpha_{s}|^{2} = 2 \chi |\alpha_{p}|^{2} |\alpha_{s}|^{2} - 2 \chi s |\alpha_{s}|^{2}$$
 (2.39)

Equations (2.35), (2.36) and (2.39) are equivalent to Eqs. (2.25a) and (2.25b) derived in  $\oint 2.1(c)$ ; except now the Raman coupling constant  $\chi$ is real. Hence all the results of  $\oint 2.1(c)$  are applicable to this case if we set K''=O in Eqs. (2.25) We conclude from such a comparison that bistability will not be observed in the phonon reservoir system as,

$$\frac{\chi_p}{\chi} > 0$$
 (2.40)

for all  $\aleph_p$ ,  $\chi$  and Eqs. (2.30) indicate that this system will exhibit an anologous second order phase transition only.

The marked difference between the behaviour of this system and the one of  $\oint 2.1(c)$  is due to the corresponding real and complex nature of the respective coupling constants  $~\chi~$  and ~K~.

That is, in the theory of  $\oint 2.1(c)$ , frequency mismatch between Stokes, pump and phonon modes generated a complex coupling constant K Such frequency shifts occur in the reservoir theory of this section but their effect is only to slightly shift the cavity resonance frequency, thus these terms are generally neglected (Louisell, 1973).

Hence, bistability will not be observed in the phonon reservoir system.

### $\oint$ 2.2 Effective Hamiltonian Formalism

In the final sections of this chapter, we consider bistability in three further systems: the effective two-level system, the parametric oscillator and the two-photon absorber.

We model each system in terms of an effective Hamiltonian, which we state without proof. Such Hamiltonians have been widely used in quantum optics, for example the work of Shen (1967) and Walls and McNeil (1974) on nonlinear optics; and the work of Graham (1970) on the parametric oscillator.

To derive an effective Hamiltonian, it is often necessary to eliminate certain field operators, resulting in an effective interaction between remaining variables. This is usually achieved by performing a unitary transformation on the system. In general, however, this is only possible if we may sum over virtual excitations of the eliminated variables; for example, virtual atomic levels (Power, 1974).

We now use the effective Hamiltonian approach to view the Raman effect from a different perspective to that of the previous section, and consider electronic excitation within the atom.

#### \$2.2(a) Atomic System: Effective Two-Level Model

We again assume an intracavity interaction between light and matter; in which two driven cavity modes are coupled to two atomic transitions, as indicated in Fig. 2.3:



Fig. 2.3. Effective Two-Level Model ( $|1\rangle$  and  $|2\rangle$  are atomic energy levels).

The most general case involves two incident fields,  $E_1$  and  $E_2$ , driving the two cavity modes of frequency  $\omega_1$  and  $\omega_2$ . These cavity modes are coupled to the transitions; first, from  $|1\rangle$  to several intermediate virtual levels,  $|i\rangle$ , and secondly, from these intermediate levels to  $|2\rangle$ .

Thus, a transition from  $|1\rangle$  to  $|2\rangle$  proceeds as a two-photon process, via the virtual levels,  $|i\rangle$ . The fields are not on resonance with the  $|1\rangle \longrightarrow |2\rangle$  transition.

Such a process can be described by the effective interaction Hamiltonian:

$$H_{eff. INT} = f_{h}(\chi_{1}b_{1}b_{2}^{\dagger}\sigma^{+} + h.c.) \qquad ----(2.41)$$

where  $b_1$  and  $b_2$  are boson operators describing cavity modes;  $\sigma^+$ ,  $\sigma^-$  are spin-flip operators for the  $|1\rangle \rightarrow |2\rangle$  transition and  $\chi_1$  denotes the strength of the interaction.

The total Hamiltonian of the system in the single mode approximation and rotating wave approximation then becomes,

$$H = H_0 + H_{Eff.INT} + H_D + H_{damping}$$

where

$$H_{o} = \frac{\hbar}{2} \omega_{o} \sigma_{z} + \hbar \omega_{1} b_{0}^{\dagger} b_{1} + \hbar \omega_{2} b_{2}^{\dagger} b_{2} - ---(2.42a)$$

(  $\omega_o$  = resonance frequency of two-level system).

$$H_{b} = i\hbar (E, b, e^{-i\omega_{1}'t} + E_{2}b_{2}^{\dagger}e^{-i\omega_{2}'t} - h.c.) - (2.42b)$$

( $\omega_1', \omega_2'$  are the frequencies of the two applied fields, of amplitude E, and E<sub>2</sub> respectively).

The cavity damping takes the same form as in the previous section

Heavity damping = 
$$Q_1 b_1^{\dagger} + Q_2 b_2^{\dagger} + h.c.$$
 (2.42c)

 $(Q_1,Q_2 = reservoir operators, for example, modes of the radiation field)$ and we assume atomic damping of the form,

Hatomic damping =  $Q_A \sigma^+ + h.c.$  (2.42d) ( $Q_A$  = reservoir operator).

Thus, although there is no direct coupling between  $|1\rangle$  and  $|2\rangle$  we assume radiative damping occurs via these levels, as indicated by Eq. (2.42d).
From the Hamiltonian given by Eqs. (2.42), (2.42a), (2.42b), (2.42c) and (2.42d), we form the master equation, in the interaction picture.  $\frac{\partial \rho}{\partial t} = -\frac{i}{\hbar} \left[ \left\{ H_0 + H_{Eff.INT} + H_D \right\}_{I}, \rho \right] + \frac{\partial \rho}{\partial t} \left| damping \right|$ 

(subscript I denotes interaction picture).

where 
$$\frac{\partial \rho}{\partial t} = \hat{\mathcal{E}}_{i} \quad \forall i \{ [b_{i}\rho, b_{i}^{\dagger}] + [b_{i}, \rho b_{i}^{\dagger}] + 2 \bar{n}_{i} [[b_{i}\rho], b_{i}^{\dagger}] \}$$
  
 $\frac{\partial f}{\partial t} = \frac{\partial \rho}{\partial t} + \frac{\partial A}{2} \{ (1+\bar{n})([\sigma^{-}\rho, \sigma^{+}] + [\sigma^{-}, \rho\sigma^{+}] + \bar{n}([\sigma^{+}\rho, \sigma^{-}] + [\sigma^{+}, \rho\sigma^{-}]) \}$ 

 $(\delta_1, \delta_2 = \text{damping coefficient of modes 1 and 2;} \delta_A = \text{radiative}$ damping constant of two-level atom;  $\overline{n} = \text{number of thermal quanta}$ .

As before, we obtain equations of motion for system variables from the master equation, assuming complete decorrelation of operators. We also assume a zero temperature reservoir and set  $\tilde{n} = 0$ .

We find the following semiclassical equations.

Field equations:

$$\dot{\alpha}_{1} = E_{1} - i \chi_{1} \alpha_{2} \rho_{21} - (\chi_{1} + i \Delta) \alpha_{1} - (2.43a)$$

$$\dot{\alpha}_2 = E_2 - i \chi_1 \alpha_1 \rho_{12} - (\chi_2 + i \Delta') \alpha_2 \qquad ----(2.43b)$$

Atomic equations:

$$\dot{\rho}_{11} = -i \chi_1 d_2 d_1^* \rho_{21} + i \chi_1 d_1 d_2^* \rho_{12} + \delta_A \rho_{22} - (2.44a)$$

$$\dot{\rho}_{22} = -i\chi_1 d_1 d_2^* \rho_{12} + i\chi_1 d_2 d_1^* \rho_{21} - \chi_A \rho_{22} - (2.44b)$$

$$\dot{\rho}_{12} = -i\chi_1 d_2 d_1^* (\rho_{22} - \rho_{11}) - (\frac{\lambda_A}{2} + i\delta) \rho_{12} - (2.44c)$$

$$\rho_{11} + \rho_{22} = 1$$
 (2.44d)

- and the complex conjugate equations.

In Eqs. (2.43), (2.44), 
$$\alpha_1 = \langle b_1 \rangle$$
,  $\alpha_2 = \langle b_2 \rangle$ ,  $\rho_{ij} = \langle i | \rho | j \rangle$   
 $\Delta, \Delta' = \text{detunings between cavity modes and applied fields,}$   
 $\Delta = \omega_1 - \omega_1'$ ;  $\Delta' = \omega_2 - \omega_2'$ ;  $\delta$  = atomic detuning =  $\omega_1 - \omega_2 - \omega_0$ .

In this model, we assume the atom relaxes to its steady state rapidly compared with the cavity modes. The discussion of  $\oint 1.3(g)$  in the introduction showed that this approximation is justified if our intracavity interaction model is similar to a typical gas laser.

Thus, if 
$$\lambda_A \gg \lambda_1, \lambda_2$$

we may adiabatically eliminate the atoms, by setting

$$\dot{\rho}_{11} = \dot{\rho}_{22} = \dot{\rho}_{12} = 0$$
 in Eqs. (2.44)

This yields, (for real  $X_1$  )

$$(\rho_{11})_{55} = 1 - (\rho_{22})_{55}$$
 -----(2.45b)

Substituting these steady state values back into the field equations, Eqs. (2.43), we find,

$$\dot{\alpha}_{1} = E_{1} - (\delta_{1} + i\Delta)\alpha_{1} - \frac{\chi_{1}^{2}\alpha_{1}|\alpha_{2}|^{2}}{(\frac{\delta_{A}}{2} - i\delta)\left[1 + |\alpha_{1}|^{2}|\alpha_{2}|^{2} \cdot 2\chi_{1}^{2}(\frac{\delta_{A}^{2}}{4} + \delta^{2})^{-1}\right]}$$
(2.46a)

$$\dot{\alpha}_{2} = E_{2} - (\aleph_{2} + i\Delta')\alpha_{2} + \chi_{1}^{2}\alpha_{2}|\alpha_{1}|^{2} \qquad (2.46b)$$

$$\left(\frac{\aleph_{A}}{2} + i\delta\right)\left[1 + |\alpha_{1}|^{2}|\alpha_{2}|^{2} \cdot 2\chi_{1}^{2}\left(\frac{\aleph_{A}^{2}}{4} + \delta^{2}\right)^{-1}\right]$$
In the limit of large detuning (dimensional limit) we can expand

In the limit of large detuning (dispersive limit), we can expand the denominators in Eqs. (2.46) to first order. That is, we assume

$$\frac{2\chi_1^2}{\left[\frac{\chi_A^2}{4} + \delta^2\right]} |\alpha_1|^2 |\alpha_2|^2 \ll 1$$

and find the field equations:

$$\dot{\alpha}_{1} = E_{1} - (\aleph_{1} + i\Delta)\alpha_{1} - \frac{\chi_{1}^{2} - \alpha_{1} |\alpha_{2}|^{2}}{\left(\frac{\aleph_{A}}{2} - i\delta\right)} - (2.47a)$$

$$\dot{\alpha}_{2} = E_{2} - (\aleph_{2} + i\Delta')\alpha_{2} + \frac{\chi_{1}^{2} - \alpha_{2} |\alpha_{1}|^{2}}{\left(\frac{\aleph_{A}}{2} + i\delta\right)} - (2.47b)$$

These equations describe the behaviour of an effective two-level system, in the dispersive limit on adiabatic elimination of the atomic variables.

We note that if we set the input fields on resonance with the cavity modes  $(\Delta = \Delta' = 0)$  and consider one driving field only  $(E_2 = 0)$ , the Eqs. (2.47) become,

where

$$\chi_{2} = \rho_{2}' + i \rho_{2}'' ; \rho_{2}' = \frac{\chi_{1}^{2} \chi_{A}}{2 \left[ \frac{\chi_{1}^{2}}{4} + \delta^{2} \right]} ; \rho_{2}'' = \frac{\chi_{1}^{2} \delta}{\left[ \frac{\chi_{A}^{2}}{4} + \delta^{2} \right]}$$

Equations (2.48) are equivalent to the Raman equations, previously derived in  $\oint 2.1(c)$ . Thus, all results obtained in  $\oint 2.1$  pertaining to bistability and hysteresis are also applicable to such an atomic system.

The major result, we may quote, is that this system will exhibit bistability in the intensity  $|\alpha_2|^2$ , dependent on the input intensity  $|E_1|^2$ .

# $\oint 2.2(b)$ The Parametric Oscillator

Parametric oscillation occurs when a nonlinear crystal is driven by light of frequency  $\omega_i$ , producing two output light fields of different frequencies,  $\omega_{f_1}$  and  $\omega_{f_2}$ .

If excitation proceeds via virtual states, we can write the effective Hamiltonian as, (Graham, 1970),

Heff. INT = 
$$i\hbar K_p (b_1^{\dagger} b_2^{\dagger} b_3 - b_1 b_2 b_3^{\dagger})$$
 -----(2.49)  
where  $b_1$ ,  $b_2$  and  $b_3$  are boson operators for the three cavity  
modes; and  $K_p$  is the coupling constant, chosen real.

Equation (2.49) describes the process,



Assuming that it is possible to drive the cavity modes of frequency  $\omega_1$  and  $\omega_2$  on resonance, we find the total system Hamiltonian:

 $H = H_0 + H_{Eff. INT} + H_0 + H_{damping}$ 

where

$$H_{o} = \hat{\mathcal{E}}_{i} \dagger \omega_{i} \dot{b}_{i} \dot{b}_{i} ; \quad H_{Eff.INT} \text{ is as given in Eq. (2.49),}$$
$$H_{o} = i \dagger (E_{i} \dot{b}_{i} e^{-i\omega_{i}t} + E_{2} \dot{b}_{2} e^{-i\omega_{2}t} - h.c.)$$
$$H_{damping} = \hat{\mathcal{E}}_{i} \dot{b}_{i} \dot{Q}_{i} + h.c.$$
(where  $Q_{i}$  are reservoir operators).

and

The master equation follows as in previous sections and again we obtain the deterministic equations: (cold reservoir, n = 0)

$$\dot{\beta}_1 = - \delta_1 \beta_1 + K_P \beta_2^* \beta_3 + E_1$$
 (2.50a)

$$\dot{\beta}_{2} = E_{2} - \delta_{2}\beta_{2} + \kappa_{p}\beta_{1}\beta_{3} - K_{p}\beta_{1}\beta_{2} - (2.50c)$$

where  $\langle b_1 \rangle = \beta_1$ ,  $\langle b_2 \rangle = \beta_2$ ,  $\langle b_3 \rangle = \beta_3$ ;  $\langle b_1, \rangle = \langle b_3, \rangle$  are the respective damping rates of modes 1, 2, and 3.

We now solve the system in the steady state, by setting  $\dot{\beta}_1 = \dot{\beta}_2 = \dot{\beta}_3 = o$ in Eqs. (2.50).

Equation (2.50a) yields,

$$(\beta_3)_{SS} = \frac{-\kappa_P \frac{\beta_2 \beta_1}{\gamma_3}}{\gamma_3}$$

which when substituted in Eqs. (2.50b) and (2.50c) results in the equation of state for the system:

We have thus derived an equation of state in terms of the two input driving fields and one cavity mode  $(\beta_2)$  only.

We can write Eq. (2.51) as a real equation,

$$|E_{2}|^{2} = |\beta_{2}|^{2} \left[ \begin{cases} \lambda_{2} + \frac{\chi_{p} |E_{1}|^{2}}{(\lambda_{1} + \chi_{p} |\beta_{2}|^{2})^{2}} \end{cases} \right]^{2} - --(2.52)$$

The behaviour of the system, as described by Eq. (2.52) is plotted in Fig. 2.5, for various values of system parameters. The form of these curves suggest a bistability in the intensity  $|\beta_2|^2$ , dependent on  $|E_2|^2$  - this is indicated in Fig. 2.2.



Fig. 2.4. Expected Bistability in Parametric Oscillator System (arrows indicate bistable transitions).

To determine the conditions necessary for bistability, a stability analysis must be performed. However, the complexity of this six variable system  $(\beta_1, \beta_1^*, \beta_2, \beta_2^*, \beta_3, \beta_3^*)$  does not allow a straightforward analytical approach.

Nevertheless, Fig. 2.5 clearly indicates instability in the system and we can expect bistability to occur as shown in Fig. 2.4.

We can obtain the sufficient conditions for such a bistability: this requires the solutions of the equation,

$$\frac{d |E_2|^2}{d |\beta_1|^2} = C$$

to be real and positive.



On performing this, we find bistability can only be expected when,

This behaviour is clearly shown in Fig. 2.5:  $|\beta_2|^2$  becomes double-valued for a given  $|E_2|^2$  when condition (2.53) is satisfied.

Thus the parametric oscillator is an example of a system exhibiting bistability in output intensity, varying in accordance with an input field intensity, and critically dependent on a second input field intensity.

Finally in this chapter, we consider bistability in a further system, the two-photon absorber.

## $\oint 2.2(c)$ Two Photon Absorber

As in previous sections, we model the system as the intracavity interaction of two driven cavity modes with two atomic levels. We indicate this as follows:



Fig. 2.6. Two Photon Absorber

We assume there exist a number of virtual intermediate levels  $|\dot{\iota}\rangle$ , between levels  $|l\rangle$  and  $|2\rangle$ , over which we can perform a sum. This enables construction of an effective Hamiltonian,

Heff. INT =  $\hbar (X_a b_1 b_2 \sigma^+ + h.c.)$  (2.54) where  $b_1$ ,  $b_2$  are boson operators for the cavity modes of frequency  $\omega_1$ ,  $\omega_2$ ;  $\sigma^+$  is the spin-flip operator for the  $|1\rangle \rightarrow |2\rangle$  transition. Including the effects of cavity and atomic damping, and two driving fields on resonance with the cavity modes; we find the deterministic equations, following the same procedure as in previous sections,

### Field Equations:

$$\dot{a}_1 = E_1 - \delta_1 a_1 - i X_a a_2^* \rho_{21}$$
 -----(2.55a)

$$\dot{\alpha}_2 = E_2 - \delta_2 \alpha_2 - i \lambda_a \alpha_1^* \rho_{21}$$
 -----(2.55b)

Atomic Equations:

$$\dot{p}_{11} = +i \lambda a d_1 d_2 p_{12} - i \lambda a d_1 d_2^* p_{21} + \delta_A p_{22} - (2.56a)$$

$$\dot{\rho}_{12} = -i \chi_{ad_{1}} d_{2}^{*} (\rho_{22} - \rho_{11}) - \frac{\aleph_{A}}{2} \rho_{12}$$
 ----(2.56c)  
=  $\langle b_{1} \rangle$ ;  $d_{2} = \langle b_{2} \rangle$ ;  $\rho_{ij} = \langle i|\rho|j \rangle$ 

where  $\alpha_1 = \langle b_1 \rangle$ ;  $\alpha_2 = \langle b_2 \rangle$ ;  $\rho_{ij} = \langle i|\rho|j \rangle$ and  $E_1$ ,  $E_2$  are amplitudes of the incident fields driving cavity modes  $\alpha_1$ ,  $\alpha_2$ , respectively;  $\lambda_1$ ,  $\lambda_2$ ,  $\lambda_A$  are the cavity mode and atomic damping rates. (We assume  $\bar{n} = 0$ )

As in the effective two-level system, we adiabatically eliminate the atomic variables, in the limit,

$$\forall_A \gg \forall_1, \forall_2$$

This results in the field equations,

$$\dot{\alpha}_{1} = E_{1} - \aleph_{1} \alpha_{1} - \frac{2 \aleph_{A} \chi_{a}^{2} \alpha_{1} |d_{a}|^{2}}{\aleph_{A}^{2} + 8 \chi_{a}^{2} |\alpha_{1}|^{2} |\alpha_{2}|^{2}} - (2.57a)$$

$$\dot{\alpha}_{2} = E_{2} - \aleph_{2} \alpha_{2} - \frac{2 \aleph_{A} \chi_{a}^{2} \alpha_{2} |\alpha_{1}|^{2} |\alpha_{2}|^{2}}{\aleph_{A}^{2} + 8 \chi_{a}^{2} |\alpha_{1}|^{2} |\alpha_{2}|^{2}} - (2.57b)$$

We now adopt the absorptive limit, in which the damping coefficient  $X_A$  becomes large with respect to other system parameters. This is valid in such an adiabatic limit. Assuming

$$\frac{8 \chi_{a}^{2} |d_{1}|^{2} |d_{2}|^{2}}{\chi_{A}^{2}} << 1$$

we expand the denominators in Eqs. (2.57) to first order, resulting in the equations,

$$\dot{d}_{1} = E_{1} - \chi_{1} d_{1} - \chi_{a} d_{1} |d_{2}|^{2} \qquad (2.58a)$$

$$\dot{d}_{2} = E_{2} - \chi_{2} d_{2} - \chi_{a} d_{2} |d_{1}|^{2} \qquad (2.58b)$$

$$\chi_{a} = \frac{2 \chi_{a}^{2}}{\chi_{A}}$$

where

Equations (2.58) describe the behaviour of a two-photon absorber on adiabatic elimination of atomic variables.

It is interesting to note that in the steady state,  $\dot{\alpha}_1 = \dot{\alpha}_2 = O$ Eqs. (2.58) predict the equation of state,

$$|E_2|^2 = |d_2|^2 \left[ \begin{cases} \chi_2 + \frac{\chi_a |E_1|^2}{(\chi_1 + \chi_a |d_2|^2)^2} \end{bmatrix} - (2.59) \end{cases}$$

which is equivalent to the equation of state for the parametric oscillator, given by Eq. (2.52).

Thus, in the absorptive limit (  $\aleph_A$  large) the two-photon absorber behaves like a parametric oscillator (resonance case). As before, we expect the system to display bistability when; (c.f. Eq. (2.53))

$$|E_1|^2 > 4 \frac{y_1^2 y_2}{\chi_a}$$

as indicated in Fig. 2.7.



Fig. 2.7. Expected Bistability in the Two-Photon Absorber -- in Output Intensity, Dependent on Input Intensity.

Again, we have shown the equivalence of two nonlinear systems.

#### CHAPTER 3

## OPTICAL BISTABILITY FROM A SYSTEM OF THREE-LEVEL ATOMS

# 

Semiclassical and fully quantum mechanical theories of optical bistability, arising from the intracavity interaction of coherent light with a system of two-level atoms, have been discussed by many authors (for example, McCall (1974), Bonifacio and Lugiato (1976), and Drummond and Walls (1980b)).

These theories describe two distinct regimes, referred to as absorptive and dispersive bistability.

Absorptive bistability is a resonance effect, and occurs due to the feedback introduced into the system from the interplay of the following conditions.

- An increase in intensity results in saturation of the atoms and thus less absorption of radiation by the atoms, and
- ii) the intensity of the radiation within the cavity increases as absorption by the atoms decreases.

Dispersive bistability, associated with a system exhibiting an intensity dependent refractive index, arises because of the optical feedback generated when the following conditions are met:

- An increase in intensity changes the refractive index, consequently shifting the cavity resonance frequency towards the exciting frequency, and
- ii) the intensity of the cavity radiation increases as the cavity frequency shifts closer to the exciting frequency.

In practice, however, purely absorptive bistability is difficult to observe; and the first experimental observation of optical bistability relied on the intensity dependence of the refractive index. In this chapter, we consider an unusual effect displayed by threelevel atoms under the action of two driving fields, which provides a mechanism for optical bistability in such a system.

This phenomenon, known as coherent population trapping occurs when the driving fields are on resonance with the atoms and the atomic damping is of a certain radiative nature. It results in a transfer of the entire atomic population to a linear combination of the two lower levels of the atom, if the system is initially prepared in a statistical mixture of all three levels. This final linear combination of levels does not interact with the driving fields, thus the system evolves to a non-absorbing state.

The theory of this effect is discussed at length by Arimondo and Orriols, (1976), Orriols (1979) and Whitley (1977).

Experimentally, these so-called narrow non-absorption resonances have been observed by Gray et.al. (1978) and Alzetta et.al. (1976, 1979).

In  $\oint 3.2$  and  $\oint 3.3$  we discuss the interaction of a coherent driving field with an 'inverted-V' and 'V' configuration respectively. In both systems, we find the following steady state behaviour.



# INPUT

Fig. 3A. Steady State Behaviour in Three-Level Systems

A form of such a curve suggests bistability in the system: branches of positive slope are assumed to describe stable states, and the branch having negative slope is assumed unstable. Bistable transitions will then occur at points A and B in Fig. 3A. To rigorously determine the occurrence of bistability, a linearised stability analysis must be performed. However, the complexity of the three-level system renders any such analytical calculation intractable. Hence, in this chapter the stability of the system will not be explicitly investigated.

The behaviour displayed by Fig. 3A does however indicate an instability in the system. Thus whenever such behaviour is found in our investigations in this chapter, we will assume there is a possibility of observing bistability in the system.

# $\phi$ 3.2 'Inverted-V' Configuration

## 

We consider the intracavity behaviour of a system of N three-level atoms ('inverted-V' configuration), subject to two coherent driving fields.

This is indicated in Fig. 3.1.



Fig. 3.1. 'Inverted-V' Configuration Interacting with Two Fields

 $E_a$  and  $E_b$  represent the amplitudes of the two input fields. These drive two cavity modes (assumed to be on resonance with the fields) of frequency  $\omega_a$ ,  $\omega_b$ . The atomic levels  $|1\rangle$ ,  $|2\rangle$  and  $|3\rangle$  have energies  $\hbar\omega_1$ ,  $\hbar\omega_2$  and  $\hbar\omega_3$  respectively. Atomic transitions are as indicated in Fig. 3.1, which we note, is equivalent to a Raman system.

We construct a Hamiltonian for such a system, in terms of the operators,  $\sigma_1^+$ ,  $\sigma_2^+$ ,  $\sigma_1^-$ ,  $\sigma_2^-$  where  $\sigma_1^+$ ,  $\sigma_1^-$  are spin-flip operators for the  $|1\rangle \rightarrow |3\rangle$  transition, and  $\sigma_2^+$ ,  $\sigma_2^-$  are spin-flip operators for the  $|2\rangle \rightarrow |3\rangle$  transition,

i.e. 
$$\sigma_1 = \{3\}, \sigma_1 = \{3\}, \sigma_1 = \{1\}, \sigma_2 = \{2\}, \sigma_2 = \{3\}, \sigma_2 = \{2\}, \sigma_1 = \{2\}, \sigma_1 = \{2\}, \sigma_1 = \{2\}, \sigma_2 = \{3\}, \sigma_2 = \{2\}, \sigma_1 = \{2\}, \sigma_2 = \{3\}, \sigma_2 = \{2\}, \sigma_1 = \{2\}, \sigma_2 = \{3\}, \sigma_2 = \{2\}, \sigma_1 = \{2\}, \sigma_2 = \{3\}, \sigma_2 = \{2\}, \sigma_1 = \{2\}, \sigma_2 = \{3\}, \sigma_2 = \{2\}, \sigma_1 = \{3\}, \sigma_2 = \{3\}, \sigma_2 = \{2\}, \sigma_1 = \{3\}, \sigma_2 = \{3\}, \sigma_1 = \{2\}, \sigma_1 = \{3\}, \sigma_2 = \{3\}, \sigma_2 = \{3\}, \sigma_2 = \{3\}, \sigma_2 = \{3\}, \sigma_1 = \{3\}, \sigma_2 = \{3\}, \sigma_2 = \{3\}, \sigma_2 = \{3\}, \sigma_1 = \{3\}, \sigma_2 = \{3\}, \sigma_1 = \{3\}, \sigma_1 = \{3\}, \sigma_2 = \{3\}, \sigma_2 = \{3\}, \sigma_1 = \{3\}, \sigma_2 = \{3\}, \sigma_1 = \{3\}, \sigma_2 = \{3\}, \sigma_1 = \{3\}, \sigma_2 = \{3\}, \sigma_2$$

common upper level, 3.

We assume an electric-dipole interaction between cavity modes and atoms:

Following the second quantised formalism, we expand the dipole operator  $\mu$  as, (Agarwal, 1974),

 $\underline{\mathcal{M}} = \underline{\mathcal{M}}_{13} \ \sigma_1^- + \underline{\mathcal{M}}_{23} \ \sigma_2^- + \text{h.c.} \qquad (3.2)$ and the electric field  $\underline{E}$  as,  $\underline{E} = \underline{E}_1 + \underline{E}_2$ 

where

$$\underline{E}_{1} = i \left( \frac{\hbar \omega_{a}}{2\epsilon_{o}} \right)^{V_{2}} \left[ \underline{\hat{e}}_{1} u_{1}(\underline{r}) \mathbf{b}_{1} - \underline{\hat{e}}_{1}^{*} u_{1}^{*}(\underline{r}) \mathbf{b}_{1}^{*} \right] 
\underline{E}_{2} = i \left( \frac{\hbar \omega_{b}}{2\epsilon_{o}} \right)^{V_{2}} \left[ \underline{\hat{e}}_{2} u_{2}(\underline{r}) \mathbf{b}_{2} - \underline{\hat{e}}_{2}^{*} u_{2}^{*}(\underline{r}) \mathbf{b}_{2}^{*} \right]$$
(3.3)

where b, and  $b_2$  are boson operators for the two cavity modes. Again we utilize the single mode approximation.  $U(\underline{r})$  is a mode function, chosen to describe the cavity and  $\underline{\hat{e}}$  is the polarisation vector.

Substituting Eqs. (3.3) and (3.2) in Eq. (3.1), we find, in the rotating wave approximation,

$$H_{INT} = \hbar \left[ g_{13} b_1^{\dagger} \sigma_1^{-} + g_{23} b_2^{\dagger} \sigma_2^{-} + h.c. \right] \qquad -----(3.4)$$

where

$$g_{13} = \underline{\mu}_{13}^{*} i u_{1}^{*}(\underline{r}) \left(\frac{\omega_{a}}{2\kappa\epsilon_{o}}\right)^{\frac{1}{2}}; \quad g_{23} = \underline{\mu}_{23}^{*} i u_{2}^{*}(\underline{r}) \left(\frac{\omega_{b}}{2\kappa\epsilon_{o}}\right)^{\frac{1}{2}}$$

We write the total Hamiltonian for the system as,

where the free Hamiltonian is,

 $H_{0} = \hbar \omega_{a} b_{1}^{\dagger} b_{1} + \hbar \omega_{b} b_{2}^{\dagger} b_{2} + \hbar (\omega_{3} - \omega_{1}) \sigma_{1}^{2} + \hbar (\omega_{3} - \omega_{2}) \sigma_{2}^{2} \quad (3.5)$ and  $H_{INT}$  is given by Eq. (3.4). The interaction between driving fields and cavity modes is given by,

again, we have assumed classical driving fields. The cavity and atomic damping is described by the Hamiltonian terms,

H cavity damping =  $Q_1b_1^{\dagger} + Q_2b_2^{\dagger} + h.c.$  (3.7) where  $Q_1$  and  $Q_2$  are reservoir operators (e.g. modes of the radiation field);

Hatomic damping = 
$$Q_A \sigma_i^+ + Q_B \sigma_2^+ + h.c.$$
 (3.8)

(  $Q_A$  and  $Q_B$  are reservoir operators).

The form of the damping expressed by Eq. (3.8) describes spontaneous emission from the two pseudo two-level systems. We do not include radiative coupling between levels  $|1\rangle$  and  $|2\rangle$ .

Such a damping scheme assumes relaxation to levels other than the three we are considering is on a much slower time scale.

The reverse case is considered by Brewer and Hahn (1975), who derive steady state and transient solutions of such a three-level system. They assume relaxation between levels  $|1\rangle$ ,  $|2\rangle$  and  $|3\rangle$  is slow compared to decay from these states to other nearby levels. We form the master equation (in the interaction picture),

$$\frac{\partial \rho}{\partial t} = -\frac{i}{\hbar} \begin{bmatrix} \{H_0 + H_{INT} + H_D\}_{I}, \rho \end{bmatrix} + \frac{\partial \rho}{\partial t} \begin{vmatrix} \varphi \\ \partial z \end{vmatrix} + \frac{\partial \rho}{\partial t} \begin{vmatrix} Atomic \\ damp. \end{vmatrix}$$
Atomic damp. (3.9)

(subscript I denotes interaction picture) where

$$\frac{\partial p}{\partial t}\Big|_{\substack{\text{cavity}\\\text{domp.}}} = \sum_{i=1}^{\infty} \kappa_i \{ [b_i \rho, b_i] \} + [b_i, \rho b_i] \} + 2 \overline{n}_i [[b_i, \rho], b_i] \}$$
(3.10)

and

where the K: are cavity damping coefficients;  $\mathcal{Y}_{A_i}$  are spontaneous emission rates and the  $\overline{n}_i$  are the number of thermal quanta present in the reservoir.

In all following discussion, we assume a zero temperature reservoir, and set  $\bar{n}_i = 0$  .

The equation describing atomic damping, Eq. (3.11), has been derived under the Markoff assumption (Louisell, 1973). Such an approach cannot properly describe the effect of atomic collisions.

However, in any intracavity experiment, we expect the atomic vapour density to be such that collisions cannot be ignored. We include these effects in phenomenological manner, discussed presently.

We derive the equations describing the time evolution of system variables, from the master equation, by assuming complete decorrelation of expectation values:

Field Equations:

$$\dot{a}_{1} = E_{a} - K_{1} a_{1} - i g_{13} N \rho_{13}$$
 (3.12a)

$$\dot{a}_2 = E_b - K_2 a_2 - i g_{23} N \rho_{32}$$
 -----(3.12b)

(and hermitian conjugate equations)

where  $\langle b_1 \rangle = \alpha_1$ ,  $\langle b_2 \rangle = \alpha_2$ ,  $\langle i|\rho|j \rangle = \rho_{ij}$ 

The factor N appears in Eqs. (3.12) as we are assuming the cavity modes couple to N identical non-interacting atoms. Note, there are no cavity detuning terms in Eqs. (3.12) as we assume cavity modes are on resonance with the applied fields.

## Atomic Equations:

$$\dot{\rho}_{11} = -ig_{13}^* d_1^* \rho_{31} + ig_{13} d_1 \rho_{13} + \delta_{A1} \rho_{33} - ....(3.13a)$$

$$\dot{\rho}_{22} = -ig_{23}^* d_2^* \rho_{32} + ig_{23} d_2 \rho_{23} + \delta_{A_2} \rho_{33} \qquad (3.13b)$$
  
$$\dot{\rho}_{33} = ig_{13}^* d_1^* \rho_{31} - ig_{13} d_1 \rho_{13} + ig_{23}^* d_2^* \rho_{32} - ig_{23} d_2 \rho_{23}$$

$$\rho_{33} = \iota g_{13} \alpha_1 \rho_{31} - \iota g_{13} \alpha_1 \rho_{13} + \iota g_{23} \alpha_2 \rho_{32} - \iota g_{23} \alpha_2 \rho_{23} \rho_{33} - (\lambda_{A_1} + \lambda_{A_2}) \rho_{33}$$

$$\dot{\rho}_{12} = -i(\Delta - \Delta')\rho_{12} - ig_{13}^* \alpha_1^* \rho_{32} + ig_{23} \alpha_2 \rho_{13} - (3.13d)$$

$$\dot{\rho}_{13} = -(i\Delta + \frac{\lambda_{A_1} + \lambda_{A_2}}{2})\rho_{13} - ig_{13}^* \alpha_1^* n_{31} + ig_{23}^* \alpha_2^* \rho_{12} - (3.13e)$$

$$\dot{\rho}_{23} = -(i\Delta' + \frac{\delta_{A_1} + \delta_{A_2}}{2})\rho_{23} + ig_{13}^* \alpha_1^* \rho_{21} - ig_{23}^* \alpha_2^* n_{32} - (3.13f)$$

(and hermitian conjugate equations), where  $\operatorname{Nij} = \rho_{ii} - \rho_{jj}$ ;  $\Delta$ ,  $\Delta'$ are atomic detunings:  $\Delta = \omega_3 - \omega_1 - \omega_2$ ,  $\Delta' = \omega_3 - \omega_2 - \omega_5$ .

We mentioned previously the necessity to include collisional effects. As collisions tend to result in a leak of atomic population from the system, we expect  $\rho_{\mu}$  and  $\rho_{22}$  to decay at some given rate. Also, the dephasing effect of collisions could give rise to a coupling between levels  $|1\rangle$  and  $|2\rangle$ .

Following the phenomenological approach of Orriols (1979), we introduce the collisional damping rates,  $\tau_i$ ,  $\tau_2$  associated with the following equations:

$$\dot{\rho}_{11} = \frac{1}{\tau_{1}} \left( \rho_{22} - \rho_{11} \right)$$
 (3.14a)

$$\dot{\rho}_{22} = \bar{\tau}_{1} (\rho_{11} - \rho_{22}) \qquad -----(3.14b)$$
  
$$\dot{\rho}_{12} = -\frac{1}{\tau}_{2} \rho_{12} \qquad -----(3.14c)$$

We do not consider velocity-changing collisions. The approach we adopt is by no means rigorous, but adequately describes the expected behaviour.

Equations (3.13) and (3.14) describe the behaviour of the atomic variables.

## $\oint$ 3.2(b)(i) Steady State Behaviour of Atomic Variables

Let us now turn our attention to the atomic system alone, and consider Eqs. (3.13) and (3.14).

As an interesting and most simple case, we assume the fields are on resonance with the two atomic transitions  $(\Delta = \Delta' = 0)$ . Also, it is assumed that the atomic vapour is dilute enough to enable the neglect of collisional effects,  $(\tau_1^{-1} = \tau_2^{-1} = 0)$ .

Then, in the steady state  $(\dot{
ho}_{ij}=0)$ , we find,

$$\rho_{33} = \rho_{13} = \rho_{23} = 0$$

$$\rho_{22} = \frac{|\alpha_1|^2}{|\alpha_1|^2 + |\alpha_2|^2} ; \quad \rho_{11} = \frac{|\alpha_2|^2}{|\alpha_1|^2 + |\alpha_2|^2} ; \quad \rho_{12} = \frac{-\alpha_1^* \alpha_2}{|\alpha_1|^2 + |\alpha_2|^2} \quad (3.15)$$

Thus, we see there is no steady state population in the upper level; and the system resembles a two-level atom, with dipole matrix element  $ho_{12}$  .

As  $\rho_{33} = 0$  there is no emitted fluorescence. Hence, we may use the system as a switch: at a certain value of the detuning the cavity becomes non-absorbing and thus transparent, so by varying the detuning, the system may switch from one state to another (absorbing to transparent).

The behaviour described by Eqs. (3.15) is, of course, the manifestation of coherent population trapping, discussed in the introduction,  $\oint$ 3.1.

# $\oint$ 3.2(b)(ii) Steady State Behaviour of Total System

Having considered the atomic system, we now discuss how this behaviour gives rise to optical bistability.

To completely describe the general system  $(\Delta \neq 0, \Delta' \neq 0, \tau_1^{-1} \neq 0, \tau_2^{-1} = 0)$ in the steady state, we must solve Eqs. (3.12), (3.13), and (3.14) with  $\dot{\rho}_{ij} = 0$ ,  $\dot{\alpha}_1 = \dot{\alpha}_2 = 0$ . However, the solution of such atomic equations proves a formidable task.

In practice, one must solve the system numerically, as done by Orriols (1979). He finds the following steady state behaviour for the population of the upper level.



Fig. 3.2: Steady State Atomic Population as a Function of Detuning (After Orriols (1979)) (Schematic)

The narrow resonance at zero detuning is indicated by the solid curve in Fig. 3.2., which describes the system in which  $\mathcal{T}_1^{-1} = \mathcal{T}_2^{-1} = \mathcal{O}$ (no collisional damping). Such curves have also been obtained by Walls and Zoller (1980), and the norrow resonance dip is also shown to have an intensity dependent width.

The dotted curve in Fig. 3.2 describes the system in which collisional effects have been included. We note that the dip vanishes, thus collisions tend to wash out this interesting effect.

Numerical solutions of the system as given by Orriols (1979), and Walls and Zoller (1980) provide a useful picture of the system's atomic behaviour. However, to investigate the possible occurrence of optical bistability in the system, we need an analytic solution to the atomic and cavity mode equations. As such a solution is not possible in the general case, described by Eqs. (3.12), (3.13) and (3.14), we must impose some simplifying conditions on the system. Fortunately, an analytical solution to the steady state atomic equations is possible in the special case,

i)  $\alpha_1 = \alpha_2 \quad (g_{13} = g_{23} = g)$ : We assume the same cavity mode coupled to both atomic transitions, and we consider one driving field  $E_{\alpha}$ , only.

ii)  $\Gamma_1^{-1} = \Gamma_2^{-1} = O$  : We assume a dilute atomic vapour and ignore collisional effects.

iii) We assume symmetrical detuning:  $\Delta = -\Delta' = \frac{\omega_{21}}{2}$ 

Thus the field equation now becomes,

 $\dot{\alpha}_1 = -K\alpha_1 - iNg^*(\rho_{31} + \rho_{32}) + KE$  (3.16) where  $K = K_1 = K_2$  and KE = Ea.

The atomic Eqs. (3.13) and (3.14) are now soluble, as shown by P. Zoller, yielding, (Walls and Zoller, 1980)

$$\rho_{11} = \rho_{22} = \frac{1}{2} \left( 1 - \rho_{33} \right)$$

$$\rho_{33} = \frac{S_1}{2 + 3S_1}$$
(3.17)

where  $S_1$  is the saturation parameter,

Also,

$$g_{\alpha_1} \rho_{13} = -(\alpha_1 g \rho_{23})^* = -\frac{\omega_{21}}{2} \rho_{12}$$

thus,

$$\rho_{31} + \rho_{32} = \frac{1}{4T_1ga_1}\rho_{33}$$
(3.19)

We see from Eqs. (3.16) and (3.19) that the atom-cavity mode interaction is determined by the population of the upper level,  $\rho_{33}$ . We have seen that  $\rho_{33}$  as a function of detuning exhibits a distinct dip at resonance. It is the intensity dependence of the width of this dip that gives rise to bistability (output field  $|\alpha_1|^2$ , dependent on input field  $|\mathsf{E}|^2$ ), (Walls and Zoller, 1980; Walls et.al, 1981).

# ∮3.2(c) Bistability

Following the approach adopted in the study of two-level systems (Drummond and Walls, 1980b), we scale the system variables as follows:

$$X = 2 |g_{d_1}| (T_1 T_2)^{1/2} , \quad Y = 2 |g_E| (T_1 T_2)^{1/2}$$

$$C = \frac{N |g|^2 T_2}{2 K} , \quad \delta = \omega_{21} T_2$$
(3.20)

and find the state equation,

$$Y = X \left[ 1 + \frac{8C\delta^2}{\delta^4 + 4\delta^2 + 2\chi^2\delta^2 + 4\chi^4} \right]$$
(3.21)

For C > 1 the possibility of observing bistability in the system is indicated, as shown in the schematic diagram, Fig. 3.3



Fig. 3.3. Schematic Diagram of Bistability Arising from a System of Three-Level Atoms

- (a) Output field vs. Input field.
- (b) Output field vs. detuning.

(arrows indicate expected bistable transitions)

We note the condition C > 1, to observe bistability, is four times less than that required to observe bistability in two-level systems (Bonifacio and Lugiato, 1976).

The bistability discussed in this section has relied on the fulfillment of certain specialised conditions, for example, the choice of the damping.

Fig. 3.2 shows that the behaviour of  $\rho_{33}$  changes dramatically when collisional damping becomes significant. We thus expect the onset of bistability to be partly determined by the effects of collisions on the system.

To investigate the relationship between bistability and collisional damping rates, we must again solve Eqs. (3.13) and (3.14). These can only be solved numerically on inclusion of collisional damping rates  $\tau_i, \tau_2$ .

Fig. 3.4 shows the variation of output field (X) with input field (Y) as the ratio  $\tau_2 / \tau_1$  is varied. The disappearance of bistable behaviour is indicated, as the ratio  $\tau_2 / \tau_1$  increases: as collisional damping rates become comparable with radiative damping rates.

Finally in this section, we discuss the importance of the condition  $\alpha_1 = \alpha_2$  , required for bistability.

We expect that any departure from this simplification will drastically affect the observed bistability.

Again, the system equations were solved numerically, and Fig. 3.5 shows the behaviour of the system as the ratio  $\alpha_1/\alpha_2$  is varied.

Bistability is shown to vanish as the system becomes more asymmetrical and the ratio  $d_2/d_1$  increases.

We now investigate the possibility of observing optical bistability in a slightly different system, the 'V' configuration.





Fig. 3.5. Output Field (x) vs. Input Field (Y) for Different Values of  $\alpha_1/\alpha_2$  for the Inverted-V Configuration (C = 4,  $\delta$  = 1)

### $\phi$ 3.3 'V' Configuration

As in the previous section, we consider the intracavity interaction of two driven cavity modes with a system of N three-level atoms.

We indicate this in Fig. 3.6.



Fig. 3.6. 'V' Configuration - Interaction Between Atom and Fields

Ea and Eb are the amplitudes of the input fields, resonantly driving the two cavity modes of frequency  $\omega_a$ ,  $\omega_b$ . These modes couple to the two atomic transitions  $|1\rangle \leftrightarrow |3\rangle$  and  $|2\rangle \leftrightarrow |3\rangle$ .

This system can be described in terms of two driven two-level atoms, with a common lower level 13. This is similar to the 'inverted-V' configuration described in the previous section, where the upper level was common to both transitions.

The Hamiltonian for the 'inverted-V' configuration, Eqs. (3.4), (3.5), (3.6), (3.7) and (3.8) will be equivalent to one describing this 'V' configuration; the only difference is in the definition of the spin-flip operators,  $\sigma_1^{\pm}$  and  $\sigma_2^{\pm}$ .

That is, in the 'V' configuration,  $\sigma_1^+$  and  $\sigma_1^-$  are spin-flip operators for the  $|1\rangle \leftrightarrow |3\rangle$  transition, where

 $\sigma_1^+|3\rangle = |1\rangle$ ,  $\sigma_1^-|1\rangle = |3\rangle$ ,  $\sigma_1^+|2\rangle = 0$  (3.22a) and  $\sigma_2^+$ ,  $\sigma_2^-$  are spin-flip operators for the  $|3\rangle \leftrightarrow |2\rangle$  transition, where,

$$\sigma_2^+|3\rangle = |2\rangle, \quad \sigma_2^-|2\rangle = |3\rangle, \quad \sigma_2^\pm|1\rangle = 0$$

$$(3.22b)$$

. . . . . .

Bearing in mind the definitions Eqs. (3.22a), (3.22b), the Hamiltonian for the 'V' configuration is identical to that of the 'inverted-V' configuration Eqs. (3.4) to (3.8).

We may thus derive equations for the system variables from the master equation, Eq. (3.9).

## Field Equations:

$$\dot{\alpha}_{1} = E_{a} - K_{1}\dot{\alpha}_{1} - ig_{13}N\rho_{13}$$
 (3.23a)

$$\dot{\alpha}_2 = E_b - K_2 \alpha_2 - i g_{23} N \rho_{23}$$
 (3.23b)

where  $\alpha_1$  and  $\alpha_2$  are amplitudes of the two cavity modes. Damping of these modes is described by the constants  $K_1$  and  $K_2$ ; and  $\rho_{ij}$ are matrix elements of the density operator.

## Atomic Equations:

$$\dot{\rho}_{11} = -ig_{13} d_1 \rho_{31} + i d_1^* g_{13}^* \rho_{13} - \delta_{A1} \rho_{11}$$
 (3.24a)

$$\dot{\rho}_{22} = -ig_{23}\alpha_2\rho_{32} + ig_{23}^*\alpha_2^*\rho_{23} - \partial_{A_2}\rho_{22}$$
 (3.24b)

$$\dot{\rho}_{33} = ig_{13} d_1 \rho_{31} - ig_{13}^* d_1^* \rho_{13} + ig_{23} d_2 \rho_{32} - ig_{23}^* d_2^* \rho_{23} \quad (3.24c) + \forall A_1 \rho_{11} + \forall A_2 \rho_{22}$$

$$\dot{\rho}_{12} = -ig_{13} d_1 \rho_{32} + ig_{23}^* d_2^* \rho_{13} - \left\{ \frac{\chi_{A_1 + \chi_{A_2}}}{2} + i(\Delta - \Delta') \right\} \rho_{12} \quad (3.24d)$$

$$\dot{\rho}_{13} = -ig_{13} \alpha_1 (\rho_{33} - \rho_{11}) + ig_{23} \alpha_2 \rho_{12} - \left\{ \frac{\aleph_{A_1}}{2} + i\Delta \right\} \rho_{13}$$
 (3.24e)

$$\dot{\rho}_{23} = -ig_{23}d_2(\rho_{33}-\rho_{22}) + ig_{13}d_1\rho_{21} - \left\{\frac{\aleph_{A_2}}{2} + i\Delta'\right\}\rho_{23} \quad (3.24f)$$

$$\rho_{11} + \rho_{22} + \rho_{33} = 1 \tag{3.24g}$$

where  $\Delta = \omega_1 - \omega_3 - \omega_a$ ,  $\Delta' = \omega_2 - \omega_3 - \omega_b$  are the relative atomic-cavity mode detunings;  $\lambda_{A_1}$ ,  $\lambda_{A_2}$  are the spontaneous emission rates of the two atomic transitions  $|1\rangle \leftrightarrow |3\rangle$  and  $|2\rangle \leftrightarrow |3\rangle$ . We have neglected collisional effects in this case.

As in the 'inverted-V' configuration, we attempt an analytic solution of the atomic equation in the specialised case,

1)  $\alpha_1 = \alpha_2 (K_1 = K_2 = K; g_{13} = g_{23} = g)$ , and we consider one driving field,  $E_a$  only.

2) Symmetrical detuning,  $\Delta = -\Delta' = -\frac{\omega_{21}}{2}$ 

and we define the damping coefficients,

$$\aleph_{A_1} = \aleph_{A_2} = \frac{1}{T_1}$$
,  $\frac{\aleph_{A_1}}{2} = \frac{1}{T_2}$ 

We then find the field equation

$$\dot{\alpha}_{1} = KE - K\alpha_{1} - igN(\rho_{23} + \rho_{13})$$
 ----(3.25)

(where KE = Ea ), and the steady state atomic variables,

 $\rho_{11} = \rho_{22} = \frac{A_1}{\frac{1}{T_1} + 3A_1}$ 

where

$$A_{1} = \frac{4 |g_{\alpha_{1}}|^{2} \left[ \left( \frac{1}{T_{1}^{2}} - \omega_{2_{1}}^{2} + 4 |g_{\alpha_{1}}|^{2} \right) + 2 \omega_{2_{1}}^{2} \right]}{T_{1} \left( \frac{1}{T_{1}^{2}} - \omega_{2_{1}}^{2} + 4 |g_{\alpha_{1}}|^{2} \right) + 4 \frac{\omega_{2_{1}}^{2}}{T_{1}^{2}}}$$

Also,

$$\rho_{23} + \rho_{13} = -\rho_{11} - ---- (3.26)$$

The steady state upper populations,  $\rho_{\rm II}$  and  $\rho_{22}$  decrease markedly on resonance, similar to the behaviour of  $\rho_{33}$  in the 'inverted-V' case.

However, in the 'V' configuration the population does not completely vanish;  $\rho_{11} \neq O$  at resonance. This is because spontaneous emission to the lower level  $|3\rangle$  destroys any coherence between levels, necessary to produce population trapping.

From Eqs. (3.25) and (3.26), we find the steady state equation for the system,

$$Y = X \left( 1 + \frac{16 c \rho_{11}}{x^2} \right)$$
 (3.27)

where X, Y and C are as defined in  $\oint 3.2$ , Eqs. (3.20).

The behaviour of this system, according to Eq. (3.7) is shown in Fig. 3.7: possible bistable behaviour is clearly indicated.



#### CHAPTER 4

#### OPTICAL BISTABILITY IN SEMICONDUCTORS - GENERAL CONSIDERATIONS

As mentioned in Chapter One, two recent experiments have revealed optical bistability in semiconductors: the experiments of Miller et.al. (1979) using InSb, and that of Gibbs et.al. (1979a) involving GaAs.

The suggested mechanism for bistability is quite different for each of these experiments. To aid the discussion of these differences we now briefly review the theory of semiconductors.

### $\phi$ 4.1 Elementary Theory of Semiconductors

According to the band theory of solids, semiconductors consist of a valence band (completely filled with electrons) separated from the empty conduction band by a small energy gap.

If such a solid is optically excited, electrons may move across the energy gap, leaving behind an unoccupied state, known as a hole. Coulomb attraction between electrons and holes leads to the formation of bound electron-hole pairs, called excitons (Knox, 1963).

Excitons are the lowest energy excitations in weakly excited, pure semiconductors. They are electrically neutral and do not contribute to electrical conduction.

If the orbiting radius of electron and hole is of the order of the lattice constant of the material, this tightly bound pair is called a Frenkel exciton. Such excitons are found mainly in molecular solids and are thought to provide a means of energy transfer in these systems.

At the other extreme, the exciton radius extends over many unit cells. These weakly bound pairs are known as Wannier excitons and are the type present in semiconductors. The Wannier exciton has the properties of a Hydrogen-like atom if we neglect exchange effects. The exchange interaction arises because of the identity between electrons in the conduction and valence bands. However, such effects are relatively small in Wannier excitons, in which the electron-hole distance is large.

We will show later that for low exciton concentrations, exciton operators obey boson commutation relations. A great deal of research has been performed assuming excitons are perfect bosons, leading to a theory of Bose condensation in excitonic systems, (Hanamura and Haug, 1977; Keldysh and Kozlov, 1968).

Present day lasers, however, generate sufficient excitation to produce very high exciton concentrations. The boson approximation then breaks down, because of effects associated with the Pauli exclusion principle, as will be discussed later.

We are now in a position to consider in detail the two experiments mentioned at the beginning of the chapter.

The semiconductor used in the experiments of Miller et.al. was InSb, in which the presence of excitons may be ignored (Miller et.al., 1980). The suggested bistability mechanism for this system involved interband excitation; we will discuss this further and develop our own theory of this effect in Chapter 8.

We now direct our attention to the experiments of Gibbs et.al., using GaAs; in which it is suggested that interaction between the light field and excitons comprising the semiconductor gives rise to bistability.

### 

To explain the results of their experiments, Gibbs et.al. suggest that light just below the exciton frequency is absorbed, producing free carriers which alter the exciton absorptivity. The refractive index of the medium thus becomes intensity dependent and provides the necessary mechanism for bistability.

Their assumptions are based on an experimental investigation of the nonlinear spectroscopy of GaAs, (Gibbs et.al., 1979b). They find that the absorption coefficient as a function of intensity is similar to a Bloch-like saturation curve, apart from an unsaturable loss term. Although this suggests that the excitons behave as two-level atoms, Gibbs et.al. conclude only that the free exciton transition saturates like a homogeneously broadened line. Bistable behaviour is explained as follows: absorption of light by excitons saturates because of,

- (a) exciton-exciton collisions;
- (b) screening of Coulombic interaction in electron-hole pairs by free carriers, and
- (c) depletion of electrons and holes necessary to produce new excitons. They also point out that the unsaturable loss term prevents the observation of purely absorptive bistability.

The explanation of the generation of a nonlinear intensity dependent refractive index in GaAs is thus solely based on an experimental determination of excitonic absorptivity. A microscopic theory, essential to the understanding of underlying physical processes was not presented by Gibbs et.al.

We now wish to develop such a theory. To this end we consider the theory of excitons in greater detail.

#### $\phi$ 4.3 Theory of Wannier Excitons

# $\phi$ 4.3(a) Introduction

Conceptually, we view an exciton as an electron-hole pair. The wavefunction for a many particle system consisting of electron and hole states is symmetric to an exchange of excitons. As a symmetric wavefucntion is characteristic of bosons, this suggests excitons may obey Bose statistics.

That excitons exhibit boson-like properties has been discussed extensively (Haken, 1977; Hanamura, 1974a, Keldysh and Kozlov, 1968).

However, at the high exciton densities it is now possible to achieve;  $N \sim 10^{17} - 10^{18} \text{ cm}^{-3}$  (N = exciton density), the average separation between excitons,  $N^{-\frac{1}{3}}$  is of the same order as the exciton's associated Bohr radius,  $\Omega_{0}$ . The fact that excitons consist of fermions now becomes of great importance; as the Pauli exclusion principle prevents electrons (or holes) of different excitons coming close together if they have parallel spins. Thus, when  $N^{-\frac{1}{3}}$  is of the order of  $\Omega_{0}$ , deformation of excitons occurs and we can no longer expect them to behave like bosons.

We formulate this more rigorously as follows: the creation operator for a Wannier exciton can be expressed as a linear combination of electronhole pairs (Hanamura and Haug, 1977),

where  $\Omega_{p_1}$ ,  $d_{p_2}$  are fermion operators for electron (momentum  $p_1$ ) and hole (momentum  $p_2$ );  $\varphi(p)$  is a normalised wavefunction of the ground state of a Hydrogen-like atom;

$$(l(p)) = \frac{8\sqrt{\pi a_0^3}}{(1+p^2 a_0^2)^2}$$
 (4.2)

The expansion (4.1) describes the non-localised nature of the electrons and holes comprising Wannier excitons. For small radius (Frenkel) excitons, we may treat the fermion pair  $d_{p_1}^{\dagger} d_{p_2}^{\dagger}$  as a pseudo-spin operator - leading to a theory very similar to that of a two-level atom (Haken, 1976, Egri, 1979). From Eq. (4.1) we form the commutation relation,

$$[C_{p}, C_{p}^{\dagger}] = 1 - \mathcal{E}_{p,p_{2}} | \varphi_{p} |^{2} (d_{p_{2}}^{\dagger} d_{p_{2}} + a_{p}^{\dagger} a_{p_{1}})$$
(4.3)

Taking matrix elements of Eq. (4.3), we find,

$$\langle n | [C_{p}, C_{p}^{\dagger}] | n \rangle = 1 - O(Na_{o}^{3})$$
 (4.4)

Equation (4.4) indicates that excitons obey boson statistics if  $N\alpha_o^3 \sim 1$ , as found earlier, from physical considerations.

For low density systems then, we may adopt the so-called harmonic approximation in which excitons are treated as non-interacting harmonic oscillators, obeying Bose statistics.

It has been suggested that nonlinear interaction between such bosonlike excitons and other particles comprising semiconductors could lead to laser action. Of the proposed schemes, those most likely to produce laser action are:

1) 
$$(e_x) + (e_x) \rightarrow (h_y)_{photon} + (e_x)$$
 (Benoit a la Guillaume et.al.,  
1969; Haken, 1977)

2) (ex) + electron/hole 
$$\rightarrow$$
 (hv)<sub>photon</sub> + excited electron/hole  
(Benoit à la Guillame et.al., 1973)

3) 
$$(ex) \rightarrow (hv)_{\text{photon}} + (hv)_{\text{phonon}}$$
 (Haken, 1977; Haug, 1968)

where  $(e_X)$  refers to an exciton;  $(h_Y)$  to emitted quanta. Haken et.al.(1975) develop a theory of laser action due to such processes.

A theory of laser action in CdS involving such processes was proposed by Haug (1968). In this substance,

$$N \sim 10^{16} \text{ cm}^{-3}, a_0 = 30 \text{ Å}$$
  
 $\Rightarrow \text{ Na}_0^3 \approx 10^{-4} \ll 1$ 

and the exciton-boson limit is valid.

Returning now to the optical bistability experiments concerning GaAs,

$$N \sim 10^{23}$$
 to  $10^{24}$  m<sup>-3</sup>,  $a_0 = 140$  Å  
 $\Rightarrow N a_0^3 \sim 10^{-1}$  to 1

Clearly the harmonic approximation breaks down in this case and we cannot assume excitons behave as bosons. An alternative approach is thus required.

A quantum theory of excitonic optical bistability, incorporating high density effects, has been developed by Goll and Haken (1980).

# $\oint$ 4.3(b) Excitonic Bistability: Theory of Goll and Haken

Writing the Hamiltonian for the system in terms of fermion operators, Goll and Haken derive Heisenberg equations of motion for the operators:

$$B_{L}^{\dagger} = \hat{\mathcal{E}}_{\boldsymbol{j},\boldsymbol{k}'} \, \mathcal{Q}_{\boldsymbol{j},\boldsymbol{k}'}^{L} \, \mathcal{Q}_{\boldsymbol{j}}^{\dagger} \, \mathcal{Q}_{\boldsymbol{k}'}^{\dagger} \, \mathcal{Q}_{\boldsymbol{k}$$

$$N^{e} = \mathcal{E}_{\mu} \varphi_{\mu}^{L} \varphi_{\mu}^{$$

The  $B^*$ 's and their hermitian conjugates define creation and destruction of excitons at the discrete lattice sites L, and the operators, N, define inversion;  $\varphi_{ll}^{L}$ ' is a wavefunction describing the relative motion of electrons and holes.

The commutation relation,

indicates that the algebra of these operators is not closed, as Eq. (4.6) involves terms that cannot be expressed in terms of  ${\bf B}$  ,  ${\bf B}^+$  or N .

Deterministic equations of motion are obtained by averaging the Heisenberg equations and assuming complete factorisation of expectation values. Including phenomenological damping terms, the following equations are found:

$$\tilde{B}_{L}^{*} = i\gamma \tilde{B}_{L}^{*} + i\gamma_{I} (\tilde{D}_{L} + \varphi(o)) \tilde{B}_{L}^{*} - g\tilde{A}_{L} \tilde{D}_{L} - \delta \tilde{B}_{L}^{*}$$
(4.7a)

$$\tilde{\tilde{D}}_{L} = 2g\left(\tilde{A}_{L}^{\dagger}\tilde{B}_{L}^{\dagger} + \tilde{B}_{L}\tilde{A}_{L}^{\dagger}\right) - \mathcal{Y}_{n}\left(\tilde{D}_{L} + \mathcal{U}(o)\right) \qquad (4.7b)$$

$$\tilde{\tilde{A}}_{L}^{-} = \tilde{\tilde{A}}_{L}^{-} |_{Hight} - G \tilde{\tilde{B}}_{L}^{*}$$
(4.7c)

where  $B_L$ ,  $\widetilde{N}_L$  are C - numbers and,

$$\widetilde{D}_{L} = 2 \widetilde{N}_{L} - \mathcal{Y}(0)$$

 $\gamma$  is the resonance frequency of the excitons;  $\gamma_{I}$  describes the strength of interparticle interactions (e-e, h-h, e-h).

 $\tilde{A}_{L}^{-}$  and  $\tilde{A}_{L}^{+}$  are the negative and positive frequency parts of the vector potential of the light field; and g is the coupling constant for the light field-exciton interaction (G is a similar constant).

The first term in Eq. (4.7c) stems from the Hamiltonian of the free light field in the crystal,  $H_{iight}$ 

 $\lambda$  and  $\lambda_{11}$  are the damping coefficients.

Equations (4.7) bear a strong resemblance to the optical Bloch equations. Bistability is thus expected to follow as in the case of two-level atoms.

Goll and Haken solve Eqs. (4.7) in the steady state and predict bistable behaviour in output intensity, dependent on input intensity.

Such a theory thus provides a simple model of optical bistability in . semiconductors, due to excitonic interaction with the light field. However, it has some limitations: Firstly, damping is included in a phenomenological manner only. Secondly, we cannot formulate the system's Hamiltonian in terms of the exciton operators described by Eqs. (4.5). As a result, we cannot derive the corresponding master equation and Fokker-Planck equation. Hence the statistical properties of the exciton variables cannot be easily calculated.

We thus wish to develop an alternative theory in which the system Hamiltonian is constructed entirely out of exciton operators. To enable the straightforward calculation of statistical averages, we also require such operators to have a well defined, closed algebra.

Once our Hamiltonian is constructed, we can derive the master equation and hence include damping in a systematic fashion.

The approach necessary to formulate such a microscopic theory is discussed in the next chapter.
#### CHAPTER 5

#### BOSON EXPANSIONS OF FERMION OPERATORS

### $\phi$ 5.1 Introduction

In a second quantised formalism, we derive a Hamiltonian for the semiconductor system by defining the field operators as an expansion of fermion operators.

As well as the boson operators describing the light field, such a Hamiltonian would thus consist of electron and hole operators, each obeying separate anticommutation relations.

Instead of deriving statistical averages of the electron and hole variables, we would prefer to obtain information about the excitons formed from the electrons and holes; as in this thesis we are concerned with excitonic optical bistability. We thus wish to express the Hamiltonian in terms of exciton operators and remove all explicit reference to individual electrons and holes. Also, the discussion of the previous chapter showed that a sound treatment of the light-exciton system follows only if the Hamiltonian is constructed from exciton operators which have a well defined algebra.

We have seen that exciton operators, expressed as bilinear combinations of fermion operators, obey boson commutation relations in the low exciton density limit. This suggests that a suitable approach is to treat our fermion system as a boson system. As bosons obey much simpler commutation relations than the anticommutation relations of fermions, this would be a favourable step.

However, in the experiments of Gibbs et.al. (1979), in which bistability was observed, high exciton densities invalidated the low density boson approximation. To develop a suitable model to describe high exciton density systems, we follow in essence a method adopted by Hanamura (1970). This involves the direct transcription from the fermion space to a boson space. Interactions between fermions then become corresponding interactions between bosons, thus all physical effects (e.g. Pauli effects) are consistently included.

Hanamura's work on Wannier excitons was confined mainly to the development of a theory of Bose-condensation (Hanamura, 1974a). He also discussed the optical properties of these excitons, with emphasis on transient effects such as self induced transparency, (Hanamura, 1974b), but did not consider optical bistability. Thus, using a bosonisation transformation to consider optical bistability is a new approach in this field.

The method of boson expansions of many-fermion systems has been used in the development of a quantum theory of several quite different phenomena (Garbaczewski, 1978).

For example, Holstein and Primakoff (1940) introduced a lowtemperature theory of the Heisenberg ferromagnet described in terms of a boson representation of spin waves. Another such bosonised theory of spin waves was presented by Dyson (1956).

More recently, bosonisation of fermion operators has been extensively used in the study of the weak excitation limit of atomic nuclei (Belyaev and Zelevinski, 1962 ; Marumori, 1960; Marumori et.al., 1964; Janssen et.al., 1971; Sorensen, 1967, 1970; Usui, 1960).

The first such transformation was developed by Usui (1960), to formulate a theory of diamagnetism of metals. This transformation unfortunately generated non-hermitian boson Hamiltonians and non-normalisable basis vectors in the boson subspace. However, these defects were removed by Marumori et.al. (1964), who further modified Usui's transformation. In the nuclear models discussed by these authors, the Hamiltonian for the system was written entirely in terms of pairs of electron operators. Thus these theories were concerned with bosonisation of electron-electron pair operators. We refer to such a transformation as bosonisation of generalised bifermion operators.

Our semiconductor system, however, is characterised by electron-hole pairs. To transform our Hamiltonian we thus need to adapt the existing theories of generalised bifermion transformations.

In  $\oint 5.2$  we present a brief summary of the generalised theory of bosonisation of bifermion operators (electron-electron pairs). We extend these ideas to develop a theory of bosonisation of electron-hole pair operators in  $\oint 5.3$ .

# 

The idea that one could replace a fermion space with a corresponding hypothetical boson space was first introduced by Sawada (1957) and Wentzel (1957), in the theory of the high density electron gas.

In the so-called harmonic approximation, Sawada showed that fermionpair operators obeyed approximate boson commutation relations in the high density limit. However, this apparent correspondence leads to an incorrect physical model, as we now show.

In all following discussion, let us designate the boson and fermion spaces by  $V_B$  and  $V_F$  respectively. If the system Hamiltonian is constructed only in terms of pairs of electron operators such as,

the fermion space is characterised by the antisymmetrised basis vectors,

 $|m\rangle = a^{\dagger}_{\alpha_{1}}a^{\dagger}_{\beta_{1}}a^{\dagger}_{\alpha_{2}}a^{\dagger}_{\beta_{2}}\dots a^{\dagger}_{\alpha_{n}}a^{\dagger}_{\beta_{n}}|0\rangle = \prod_{i=1}^{n} a^{\dagger}_{\alpha_{i}}a^{\dagger}_{\beta_{i}}|0\rangle \quad (5.1)$ where  $a^{\dagger}_{\mu}$  is a fermion creation operator for an electron in state  $\mu$ ; and  $|0\rangle$  is the fermion vacuum state. We note that the operator pairs  $a^{\dagger}a^{\dagger}$ , aa and  $a^{\dagger}a$ when applied to the basis state (5.1), will create or destroy an electron pair, or conserve total electron pair number (clearly the state (5.1) could not describe the semiconductor system).

The most general element of a boson space corresponding to this fermion space is,

 $|m\rangle = (b_{\alpha_1\beta_1}^{\dagger})^{N_1} (b_{\alpha_2\beta_2}^{\dagger})^{N_2} \dots (b_{\alpha_n\beta_n}^{\dagger})^{N_n} |0\rangle \qquad (5.2)$ where  $b_{\alpha_i\beta_i}^{\dagger} = boson$  creation operator and  $|0\rangle$  is the boson vacuum state. Let us now restrict discussion to the case  $N_1 = N_2 = \dots = N_n = 1$ 

In the harmonic approximation we replace fermion-pair operators by boson operators; thus the fermion state described by Eq. (5.1) is assumed to correspond to the boson state,

$$|m\rangle = b_{\alpha_1\beta_1} b_{\alpha_2\beta_2} \dots b_{\alpha_n\beta_n} |0\rangle = \prod_{i=1}^{n} b_{\alpha_i\beta_i} |0\rangle ----(5.3)$$

We know, from the theory of Jordon and Wigner (see for example, Schiff (1968), Chapter 14), that the postulate that fermions obey anticommutation relations is sufficient to ensure that a dynamical state can be occupied by only one particle at a time. This is, of course, the traditional expression of the Pauli principle. The anticommutation relations imply the equivalence,

However, no such relationship exists in the corresponding boson state, Eq. (5.3). That is,

 $|m\rangle = \prod_{i=1}^{n} b_{\alpha_i \beta_i}^{\dagger} |0\rangle \quad \text{is linearly independent to the state,}$  $|m\rangle' = (-1)^{p'} P' \prod_{i=1}^{n} b_{\alpha_i \beta_i}^{\dagger} |0\rangle$ 

Thus we cannot assume a strict equivalence between the fermion state, Eq. (5.2) and the boson state, Eq. (5.3). In fact, this simple replacement of a pair of fermion operators with a boson operator only follows if we neglect the effects of the Pauli principle on particles.

We see then, that implicit in both the low density boson approximation (see  $\oint 4.3a$ ) and Sawada's high density harmonic approximation is the assumption of negligible Pauli effects.

These problems are overcome if we define a suitable unitary transformation to map the fermion space to a correctly antisymmetrised boson space.

### $\phi 5.2(a)$ Antisymmetrisation of the Boson Space

We define boson operators, 
$$\vec{b}_{\alpha\beta}$$
 satisfying  
 $\begin{bmatrix} b_{\alpha\beta}, \dot{b}_{\gamma\eta} \end{bmatrix} = \delta_{\alpha\gamma} \delta_{\beta\gamma} - \delta_{\alpha\gamma} \delta_{\beta\gamma}$  (5.5)  
 $\begin{bmatrix} b_{\alpha\beta}, b_{\gamma\gamma} \end{bmatrix} = \begin{bmatrix} \dot{b}_{\alpha\beta}, \dot{b}_{\gamma\gamma} \end{bmatrix} = 0$ 

and introduce a subspace of V g , V g' spanned by the antisymmetrised boson states:

$$Im) = N \mathcal{E}_{p'} (-1)^{p'} P' \dot{b}_{\alpha_1 \beta_1} \dot{b}_{\alpha_2 \beta_2} \dots \dot{b}_{\alpha_n \beta_n} lO)$$
(5.6)

where  $\mathcal{N}$  is a normalisation constant and the permutation operator P results in the state  $|m\rangle$  changing sign when any two indices of the set  $(\alpha_1\beta_1, \ldots, \alpha_n\beta_n)$  are interchanged.

This boson state can also be represented as, (Janssen et.al., 1971),

$$Im) = \mathcal{N} \langle olexp \{ \frac{1}{2} \xi_{\alpha\beta} b_{\alpha\beta} a_{\beta} a_{\beta} a_{\alpha} \} a_{\alpha}^{\dagger} a_{\beta}^{\dagger} \dots a_{\alpha}^{\dagger} a_{\beta n}^{\dagger} | o \rangle | 0 \rangle$$
(5.7)

To find the normalisation factor  $\mathcal{N}$ , we must form (m|m) = 1and thus need to consider the number of distinct permutations of the term  $b_{\alpha_1\beta_1} b_{\alpha_2\beta_2} \dots b_{\alpha_n\beta_n}^+$  with respect to the indices  $(\alpha_1\beta_1, \dots, \alpha_n\beta_n)$ . Firstly, there are (2n)! different arrangements of these indices. However, as

$$b_{\alpha_1\beta_1}b_{\alpha_2\beta_2} = b_{\alpha_2\beta_2}b_{\alpha_1\beta_1}$$

we find that n! of these arrangements are equivalent, so we must divide the total number of arrangements by n!. Also, for each of the n pairs  $(\alpha, \beta_1, \ldots, \alpha_n \beta_n)$  there are two ways of arranging each one. As this is equivalent to having n objects and two containers to put them in (container 1: pairs in order  $\alpha_i \beta_j$ ; container 2: pairs in order  $\beta_j \alpha_i$ ), there are  $2^n$  ways of arranging the pairs amongst each other. As a result, we must divide the total number of arrangements by  $2^n$ . Thus, the number of distinct permutations is,

$$\frac{(2n)!}{2^{n} n!} = \frac{1}{(2n-1)!!}$$

$$\Rightarrow \mathcal{N} = \sqrt{\frac{1}{(2n-1)!!}} \qquad (5.8)$$

where

(2n-1)!! = (2n-1)(2n-3)(2n-5)...

Following Janssen et.al., (1971) we introduce the operator,

$$B_{\alpha\beta}^{\dagger} = b_{\alpha\beta}^{\dagger} - \mathcal{E}_{\gamma\gamma} b_{\alpha\gamma}^{\dagger} b_{\beta\gamma}^{\dagger} b_{\gamma\gamma} \qquad ----(5.9)$$

which generates the required permutations when acting on the ground state. We can then write:

$$|m) = \sqrt{(2n-1)!!} B^{\dagger}_{\alpha_{1}\beta_{1}} \dots B^{\dagger}_{\alpha_{n}\beta_{n}} |0) \qquad ----(5.10)$$

We prove these properties of the operator  $B_{\alpha\beta}$  in  $\oint 5.3(c)$  and the Appendix, in the case of the electron-hole pair transformation.

Thus, we have defined a set of antisymmetrised boson states Im) (Eq. (5.10)) in one-to-one correspondence with the fermion states Im> (Eq. (5.1)). We now consider the explicit form of the operator which will effect such a 'Pauli-principle-conserving' transformation.

# $\oint$ 5.2(b) Usui's Transformation

The first transformation developed to map from a fermion to a boson space was that due to Usui (1960), taking the general form,

$$U_1 = 10 \le 10 \exp \{\frac{1}{2} \mathcal{E}_{ab} \ b^{\dagger}_{ab} \ a_{b} \ a_$$

This operator has the desired property that it maps  $|0\rangle \otimes V_F \rightarrow V'_B \otimes |0\rangle$ but the corresponding basis vectors are not normalised, as one may show from Eq. (5.1), (5.7) and (5.11),

$$U_1(0)|m\rangle = \sqrt{(2n-1)!!} |m\rangle |0\rangle - (5.12a)$$

and

$$(ol(m|U_1 = \sqrt{(2n-1)!!} < ml(ol) - (5.12b))$$

$$\Rightarrow (o| < m|U_1^{\dagger}U_1|m > 10) = (2n-1)!! < o|(m|m)|0 > (5.12c)$$

This defect in the normalisation properties may be rectified by introducing a modified Usui operator (Janssen et.al., 1971),

$$\widetilde{U}_{1} = 10 (0) \left\{ \mathcal{E}_{n=0}^{\infty} \frac{1}{(2n)!} \left[ \mathcal{E}_{\alpha\beta} a^{\dagger}_{\alpha} a^{\dagger}_{\beta} b_{\alpha\beta} \right]^{n} |0\rangle < 0 \right\} \xrightarrow{(5.13)}$$

From Eqs. (5.1), (5.7) and (5.13), we find,

$$\tilde{U}_{1}(m) = \sqrt{(2n-1)!!} (m) = (5.14a)$$

and

$$(0 < m | \tilde{U}_{1} = \sqrt{(2n-1)!!} < 0 | (m)$$
 (5.14b)

Equations (5.12a), (5.12b), (5.14a) and (5.14b) yield,

$$\langle m|\tilde{U}, U, |m'\rangle = \delta m, m'$$
;  $(m|U, \tilde{U}, |m') = \delta m, m' - (5.15)$   
showing that  $\tilde{U}, U$  behaves as unity in the fermion space 10)  $\otimes V_F$   
and  $U, \tilde{U}$ , behaves as unity in the boson subspace  $V'_B \otimes |0\rangle$ .

One may also show that the matrix elements of any operator are unchanged under such a transformation between fermion and boson spaces. Janssen et.al. (1971) derives the relationships associated with this transformation

$$U_{1} a_{\alpha}^{\dagger} a_{\beta}^{\dagger} \widetilde{U}_{1} = B_{\alpha\beta}^{\dagger} \widehat{P},$$

$$U_{1} a_{\beta} a_{\alpha} \widetilde{U}_{1} = b_{\alpha\beta} \widehat{P},$$

$$U_{1} a_{\beta}^{\dagger} a_{\beta} \widetilde{U}_{1} = \widehat{\mathcal{E}}_{\gamma} b_{\alpha\gamma}^{\dagger} b_{\beta\gamma} \widehat{P},$$

$$\widehat{P}$$
is a projection equation.

where  $P_i$  is a projection operator, projecting out the nonantisymmetrised components from each boson state,

$$\widehat{P}_{i} = \pounds_{m} (m) (m) = \pounds_{n=0}^{\infty} \frac{1}{(2n)!} \frac{1}{(2n-1)!!} \pounds_{d_{i}\beta_{i}\dots\beta_{n}}^{\dagger} B_{d_{i}\beta_{i}\dots}^{\dagger} B_{d_{n}\beta_{n}}^{\dagger} 10) (0|B_{\alpha_{n}\beta_{n}\dots}B_{\alpha_{i}\beta_{i}}^{\dagger}}{\beta_{i}\dots\beta_{n}}$$

$$(5.17)$$

The relationships described by Eqs. (5.16) have the advantage that they generate finite expressions for the transformed pair operators.

One drawback of this transformation, however, concerns the nonhermiticity of the resulting operators. That is, if

$$\mathcal{F} = U_1 F \widetilde{U}_1$$

is the boson image of the fermion operator F, then because  $\tilde{U}_1 \neq U_1^{\dagger}$ , if  $F_1 = F_2^{\dagger}$  it is not the case that  $\mathcal{F}_1 = \mathcal{F}_2^{\dagger}$ . This is also apparent from Eqs. (5.16).

Thus any boson Hamiltonian formed using this transformation will not be hermitian.

This problem is not encountered in a modified version of Usui's transformation, developed by Marumori et.al. (1964).

# $\phi 5.2(c)$ Transformation of Marumori et.al.

This transformation is based on the use of the unitary operator,

$$U_{m} = 10 < 01 \leq_{n=0}^{\infty} \frac{1}{(2n)!!} \sqrt{(2n-1)!!} \left[ \leq_{ab} b_{ab} a_{b} a_{a} a_{a} \right]^{n} 10 > (01 - (5.18))$$

It generates the required mapping,

$$U_{M} : |0\rangle \otimes V_{F} \rightarrow V_{B}^{\prime} \otimes |0\rangle$$

and maps to properly normalised states:

$$U_{m}(0)|m\rangle = |m\rangle|0\rangle$$
 (5.19)

yielding, as in the modified Usui transformation

$$\langle m' | U_m U_m | m \rangle = \delta_{m,m'}$$

Calculating the matrix element of any fermion operator  $\mathsf{F}$  we find,

 $\langle mIFIm' \rangle = \langle mIU_{m}^{\dagger}UmFU_{m}^{\dagger}UmIm' \rangle = (mIFIm')$ where  $\mathcal{F} = U_{m}FU_{m}^{\dagger}$ ; indicating that the value of matrix elements is preserved under the transformation  $U_{m}$ .

In contrast to Usui's transformation, we find that hermitian conjugation of operators is valid under  $\ensuremath{\mathsf{U_M}}$  :

$$F_1^{\dagger} = F_2 \Rightarrow F_1^{\dagger} = F_2$$

Janssen et.al.(1971) derived compact formulae for this transformation:

$$U_{M} a_{a}^{\dagger} a_{\beta}^{\dagger} U_{M}^{\dagger} = B_{\alpha\beta}^{\dagger} \sqrt{1 + \hat{N}} \hat{P}_{i}$$

$$U_{M} a_{\beta} a_{\alpha} U_{M}^{\dagger} = \sqrt{1 + \hat{N}} B_{\alpha\beta} \hat{P}_{i}$$

$$U_{M} a_{a}^{\dagger} a_{\beta} U_{M}^{\dagger} = \mathcal{E}_{x} b_{\alpha x}^{\dagger} b_{\beta x} \hat{P}_{i}$$

$$\hat{N} = \mathcal{E}_{\alpha\beta} b_{\alpha\beta}^{\dagger} b_{\alpha\beta}$$

$$\hat{N} = \mathcal{E}_{\alpha\beta} b_{\alpha\beta}^{\dagger} b_{\alpha\beta}$$

where  $P_1$  is the projection operator defined by Eq. (5.17).

The term  $\sqrt{1+\hat{N}}$  occurring in relations (5.20) generates infinite boson expansions. Thus although we avoid the non-hermicity of transformed operators encountered in Usui's approach, we now have to deal with infinite sums of bosons.

In practice we can only use the transformation  $U_M$  in certain limits, when truncation of the infinite expansion is possible.

#### $\phi$ 5.2(d) Other Transformations

Another such transformation was introduced by Belyaev and Zelevinski (1962) and further developed by Sorensen (1967). In this formalism, pairs of fermion operators are replaced by a boson expansion, the expansion coefficients of which are chosen to preserve the commutation relations of the fermion-pair operators. Unfortunately, the wavefunction so formed contains unphysical components which violate the Pauli principle.

A completely different means of deriving a boson representation for a fermion space was presented by Janovici and Schiff (1964): the generatorcoordinate method. As shown by Janssen et.al. (1971) such a method yields the same results as the algebraic method of Marumori et.al.

We now extend the ideas of  $\oint 5.2$  to the special case of the transformation of electron-hole pair operators.

# $\phi 5.3$ Bosonisation of Electron-Hole Pair Operators

In  $\oint 5.2$  we considered the bosonisation of a general fermion system, characterised by bifermion operators.

Such a theory cannot be applied to our semiconductor system, as the fermion state (5.1) has no reference to hole states which are necessary to describe a semiconductor.

In fermion space, the semiconductor Hamiltonian can be written entirely in terms of the pairs of operators,

$$a^{\dagger}a$$
,  $a^{\dagger}d^{\dagger}$ ,  $ad$ ,  $d^{\dagger}d$ 

thus the general fermion state describing the semiconductor system is given by,

$$|m\rangle = a_{a_1}^{\dagger} d_{\beta_1} a_{a_2}^{\dagger} d_{\beta_2} \dots a_{a_n}^{\dagger} d_{\beta_n} |0\rangle$$
 (5.21)

where  $\Delta a_i$  and  $d \beta_i$  are electron and hole operators respectively, for the single particle states  $\alpha_i$ ,  $\beta_i$ .

Hence we wish to develop a transformation that will map electronhole pair operators to boson operators.

Following the work of Marumori, we expect the desired transformation will have the form:

$$U = 10 \times \langle 0 | \mathcal{E}_{n=0}^{\infty} f(n) \left[ \mathcal{E}_{\alpha\beta} \left( b^{\dagger}_{\alpha\beta} d_{\beta} a_{\alpha} \right) \right]^{n} | 0 \rangle \langle 0 |$$

where f(n) is determined from normalisation considerations. We discuss this further in  $\phi$ 5.3(b).

Mappings of interest are:

U ata Ut	,	U d <sup>+</sup> d U <sup>+</sup>
Uad U <sup>+</sup>	,	U at dt Ut

However, mappings of the type

$$Ua^{\dagger}dU^{\dagger}$$
,  $Ua^{\dagger}a^{\dagger}U^{\dagger}$ 

are not considered in our theory, as the application of such pairs of fermion operators to the basis state (5.21), produce states which lie outside the vector space spanning the semiconductor system.

# ∮5.3(a) Boson Subspace

We wish to construct a boson state equivalent to the fermion state (5.21), as

$$(m) = b_{\alpha_1\beta_1} b_{\alpha_2\beta_2} \dots b_{\alpha_n\beta_n} 10) - (5.22)$$

where  $\stackrel{\dagger}{b_{d_i}}_{\beta_i}$  is a boson operator: the  $\alpha_i$  refer to electron states and the  $\beta_i$  refer to hole states.

From anticommutation relations

$$|m\rangle = \prod_i a_{a_i}^{\dagger} d_{\beta_i} |0\rangle = (-1)^{P} \prod_i a_{a_i}^{\dagger} d_{\beta_i}^{\dagger} |0\rangle$$
  
where the operator P generates all possible permutations of electron

indices (hole indices) for fixed hole indices (electron indices).

However, as mentioned in  $\oint 5.2$ , such a correspondence does not exist for the boson state (5.22). In fact, the operator P will produce n!linearly independent states. (There are actually  $(n!)^2$  different permutations of the indices  $(d_1\beta_1, \dots, d_n\beta_n)$  describing all possible states, but only n! of these give rise to distinct boson states).

The required antisymmetrised boson state corresponding to the fermion state (5.21), is,

$$Im) = \sqrt{n!} \quad \hat{\mathcal{E}}_{p} (-1)^{p} P \dot{b}_{a, p} \dots \dot{b}_{an pn} |0) \qquad -(5.23)$$

where the factor  $\frac{1}{\sqrt{n!}}$  is included for normalisation purposes and the summation is over permutations of electron (hole) indices with hole (electron) indices fixed.

# $\oint 5.3(b)$ Transformation of Electron-Hole Pair Operators

We now wish to construct an operator U such that,

$$U(m)(0) = m(0) - (5.24a)$$

and

$$U^{+}(m)|0\rangle = (m)|0\rangle$$
 \_\_\_\_\_(5.24b)

The discussion of section 5.2 suggests we choose an operator of the form:

$$U = 10 \times (01 \sum_{n=0}^{\infty} f(n) \left[ \sum_{a \beta} b_{a \beta} d_{\beta} a_{\alpha} \right]^{n} 10) (01 \qquad (5.25)$$

where f(n) is to be determined.

Applying such an operator to a fermion state, as in Eq. (5.24a), we find

We have included the nth term only of the summation over n, as the fermion state  $|m\rangle$  contains exactly n terms.

(P = permutation of electron indices, for fixed hole indices), as:

$$\langle 0|d\beta n \Delta dn \dots d\beta \Omega d, \Delta_{s}^{\dagger}, d\beta \dots \Delta_{s}^{\dagger}n d\beta n |0\rangle = \delta \beta n \rho n \delta dn \delta n \dots \delta \beta p \delta d, \delta n \dots (5.27b)$$
  
and the summation over  $(d_{1}, \dots d_{n}, \beta_{1}, \dots \beta_{n})$  generates  $n!$  such  
expectation values (for fixed hole indices), with a corresponding sign  
change whenever the order of the fermion operators is inverted.

From Eqs. (5.23), (5.26) and (5.27) we find,

$$U(m)(0) = 10 f(n) \sqrt{n!} n! (m) ----(5.28)$$

As we require the transformation U to have the property described by Eq. (5.24a), we obtain the result,

$$f(n) = \frac{1}{n! \sqrt{n!}}$$

and thus find the necessary transformation,

$$U = 10 \times (01 \ \mathcal{E}_{n=0}^{\infty} \ \frac{1}{n!} \sqrt{n!} \ \left[ \mathcal{E}_{\alpha\beta} \ b_{\alpha\beta} \ d_{\beta} \ \alpha_{\alpha} \right]^{n} 10)(01$$
(5.29)

We also require Eq. (5.24b) to be satisfied: Eqs. (5.23) and (5.29) yield,

The summation over the permutations in Eq. (5.30) generates n!terms. Also, the summation over  $(\alpha_1, \ldots, \alpha_n, \beta_1, \ldots, \beta_n)$  yields another n!terms for each of the permutations generated by P, and the fermion operators produce a sign change to cancel the effect of  $(-1)^P$ . Thus,

$$\sum_{\substack{a_1...a_n\\\beta_1...\beta_n}} a_{a_1}^{\dagger} d_{\beta_1}^{\dagger} \dots a_{a_n}^{\dagger} d_{\beta_n}^{\dagger} |0\rangle \sum_{p} (-1)^{p} P (0| b_{a_n} \beta_n \dots b_{a_1} \beta_1, b_{s_1} b_{s_1} b_{s_n} b_$$

Equations (5.30), (5.31) and (5.1) imply,

$$U^{+}(m)|0\rangle = 10$$
  $a_{31}^{+} d_{p_1}^{+} \cdots a_{3n}^{+} d_{p_n}^{+} |0\rangle = 1m > 10$ 

= Eq. (5.24b) as required.

We have thus derived an operator producing the desired transformation between a fermion space, characterised by electrons and holes, and an antisymmetrised boson space:-

# ∮5.3(c) Antisymmetrised Boson Operators

Following the discussion of  $\oint 5.2$ , we introduce the operator

$$B_{\alpha\beta}^{\dagger} = b_{\alpha\beta}^{\dagger} - \mathcal{E}_{\chi\delta} b_{\alpha\chi} b_{\delta\beta} b_{\delta\chi} \qquad --- (5.33)$$

then we may write the antisymmetrised boson state, Eq. (5.23) as,

$$Im) = \sqrt{n!} \quad \hat{\xi}_{p}(-1)^{p} P \dot{b}_{\alpha,\beta,\dots} \dot{b}_{\alpha,\beta,n}(0) = \sqrt{n!} B^{\dagger}_{\alpha,\beta,\dots} B^{\dagger}_{\alpha,\beta,\dots} |0) \quad -(5.34)$$

In all such terms  $\boldsymbol{\alpha}$  refers to electron states and  $\boldsymbol{\beta}$  to hole states.

Expression (5.34) is proved by induction in the Appendix.

From this definition we find

$$\begin{array}{l} \mathcal{E} \quad B_{\alpha_{1}\beta_{1}}^{\dagger} \dots B_{\alpha_{n}\beta_{n}}^{\dagger} \mid 0 \rangle (0 \mid B_{\alpha_{n}\beta_{n}} \dots B_{\alpha_{n}\beta_{1}} \\
 g_{\dots,\beta_{n}}^{d_{\dots,\beta_{n}}} \\
= \quad \mathcal{E} \quad \mathcal{E}_{p} \left(-1\right)^{p} P \, b_{\alpha_{1}\beta_{1}}^{\dagger} \dots b_{\alpha_{n}\beta_{n}} \mid 0 \rangle (0 \mid B_{\alpha_{n}\beta_{n}} \dots B_{\alpha_{n}\beta_{1}} \\
 g_{\dots,\beta_{n}}^{d_{1}} \dots g_{n} \\
\end{array}$$

Any even permutation of the boson state can be written as,

and any odd permutation as,

$$- \sum_{\substack{\alpha_1,\dots,\alpha_n\\\beta_1,\dots,\beta_n}} b_{\alpha_1} \beta_{\lambda} \ b_{\alpha_2} \beta_{\lambda} \dots \ b_{\alpha_n} \beta_{n-10} (01 \ B_{\alpha_n} \beta_{n-10} \dots B_{\alpha_n} \beta_{n-10})$$

$$= - \sum_{\substack{\alpha_1,\dots,\alpha_n\\\beta_1\dots,\beta_n}} b_{\alpha_1} \beta_{\lambda} \ b_{\alpha_2} \beta_{\lambda} \dots \ b_{\alpha_n} \beta_{n-10} (01 \ B_{\alpha_n} \beta_{n-10} \dots B_{\alpha_n} \beta_{\lambda} \beta_{\lambda} \beta_{\lambda} \beta_{\lambda} \beta_{\lambda} \dots (5.35b)$$

$$= + \sum_{\substack{\alpha_1,\dots,\alpha_n\\\beta_1\dots,\beta_n}} b_{\alpha_1} \beta_{\lambda} \ b_{\alpha_2} \beta_{\lambda} \dots \ b_{\alpha_n} \beta_{n-10} (01 \ B_{\alpha_n} \beta_{n-10} \dots B_{\alpha_n} \beta_{\lambda} \beta_{\lambda} \beta_{\lambda} \beta_{\lambda} \dots \dots (5.35b)$$

as the  $B_{\alpha_i\beta_i}$  are antisymmetrical with respect to the indices  $(\alpha_1, \ldots, \alpha_n)$ with  $(\beta_1, \ldots, \beta_n)$  fixed (or vice versa).

Equations (5.35a) and (5.35b) indicate that the odd and even permutations generate the same terms, and as there are n! such permutations,

$$\sum_{\substack{\alpha_{1},\dots,\alpha_{n}\\\beta_{1},\dots,\beta_{n}}} B_{\alpha_{1}\beta_{1}} \dots B_{\alpha_{n}\beta_{n}} B_{\alpha_{1}\beta_{1}} \dots B_{\alpha_{n}\beta_{n}} \dots B_{\alpha$$

Sim

= 
$$n! (0|b_{yn\delta n} \dots b_{y_1\delta_1}|B_{\alpha_1\beta_1} \dots B_{\alpha_n\beta_n}^+|0)$$

# $\oint 5.3(d)$ Projection Operator

In analogy to Eq. (5.17) we form a projection operator  $\hat{P}$  , which projects out the unphysical components from each boson state:

$$\widehat{P} = \sum_{n} \widehat{P}_{n} = \sum_{m} |m| (m| = \sum_{n=0}^{\infty} \sum_{\substack{\alpha_{1},\dots,\alpha_{n} \\ \beta_{1}\dots,\beta_{n}}} \frac{1}{(n!)^{3}} B_{\alpha_{1}\beta_{1}}^{\dagger} \dots B_{\alpha_{n}\beta_{n}}^{\dagger} |0) (0|$$

$$= \sum_{\substack{\alpha_{1}\dots,\alpha_{n} \\ \beta_{1}\dots,\beta_{n}}} \sum_{\substack{\alpha_{1}\dots,\alpha_{n} \\ \beta_{1}\dots,\beta_{n}}} \frac{1}{(n!)^{2}} B_{\alpha_{1}\beta_{1}}^{\dagger} \dots B_{\alpha_{n}\beta_{n}}^{\dagger} |0) (0| b_{\alpha_{n}\beta_{n}} \dots b_{\alpha_{n}\beta_{n}} \sum_{(5.37a)} (0) (0| b_{\alpha_{n}\beta_{n}} \dots b_{\alpha_{n}\beta_{n}} \sum_{\alpha_{n}\dots,\beta_{n}} (0) (0) (0| b_{\alpha_{n}\beta_{n}} \dots b_{\alpha_{n}\beta_{n}} \sum_{\alpha_{n}\dots,\beta_{n}} (0) (0| b_{\alpha_{n}\beta_{n} \dots b_{\alpha_{n}\beta_{n}} \sum_{\alpha_{n}\dots,\beta_{n}} (0) (0| b_{\alpha_{n}\beta_{n} \dots b_{\alpha_{n}\beta_{n}} \sum_{\alpha_{n}\dots,\beta_{n}} (0) (0| b_{\alpha_{n}\beta_{n}} \dots b_{\alpha_{n}\beta_{n}} \sum_{\alpha_{n}\dots,\beta_{n}} (0) (0| b_{\alpha_{n}\beta_{n} \dots b_{\alpha_{n}\beta_{n}} \sum_{\alpha_{n}\dots,\beta_{n}} (0) (0| b_{\alpha_{n}\beta_{n} \dots b_{\alpha_{n}\beta_{n}} \sum_{\alpha_{n}\dots,\beta_{n}} (0) (0| b_{\alpha_{n}\dots,\beta_{n}\dots,\beta_{n}} \sum_{\alpha_{n}\dots,\beta_{n}} \sum_{\alpha_{n}\dots,\beta_{n}\dots,\beta_{n}} \sum_{\alpha_{n}\dots,\beta_{n}\dots,\beta_{n}} \sum_{\alpha_{n}\dots,\beta_{n}\dots,\beta_{n}\dots,\beta_{n} \dots,\beta_{n} \sum_{\alpha_{n}\dots,\beta_{n}\dots,\beta_{n}\dots,\beta_{n} \dots,\beta_{n} \sum_{\alpha_{n}\dots,\beta_{n}\dots,\beta_{n} \sum_{\alpha_{n}\dots,\beta_{n}\dots,\beta_{n} \dots,\beta_{n} \sum_{\alpha_{n}\dots,\beta_{n}\dots,\beta_{n} \dots,\beta_{n} \sum_{\alpha_{n}\dots,\beta_{n}\dots,\beta_{n} \dots,\beta_{n} \sum_{\alpha_{n}\dots,\beta_{n}\dots,\beta_{n} \dots,\beta_{n} \sum_{\alpha_{n}\dots,\beta_{n}\dots,\beta_{n} \dots,\beta_{n} \sum_{\alpha_{n}\dots,\beta_{n} \dots,\beta_{n} \dots,\beta_{n} \dots,\beta_{n} \sum_{\alpha_{n}\dots,\beta_{n}\dots,\beta_{n} \dots,\beta_{n} \dots,\beta_{n} \dots,\beta_{n} \dots,\beta_{n} \dots,\beta_{n} \dots,\beta_$$

Clearly,

$$\widehat{\rho} = \widehat{\rho}^+ = \widehat{\rho}^2 \qquad ----(5.37b)$$

We now derive some useful relations involving  $\widehat{P}$  ,  $B_{\alpha\beta}$  and  $\widehat{N}$  where

Firstly, consider

$$\widehat{N} \ \widehat{P} = \widehat{\xi}_{n=0}^{\infty} \ \widehat{N} \ \underbrace{\xi}_{\substack{\alpha_{1},\dots,\alpha_{n}\\\beta_{1}\dots,\beta_{n}}}^{1} \frac{1}{(n!)^{2}} \ b_{\alpha_{1}\beta_{1}}^{+} \dots \ b_{\alpha_{n}\beta_{n}}^{+} |0\rangle (0|B_{\alpha_{n}\beta_{n}}\dots B_{\alpha_{1}\beta_{1}}^{-} B_{\alpha_{1}\beta_{1}}^{-} \dots \ b_{\alpha_{n}\beta_{n}}^{+} |0\rangle (0|B_{\alpha_{n}\beta_{n}}\dots B_{\alpha_{1}\beta_{1}}^{-} \dots B_{\alpha_{1}\beta_{1}}^{-} B_{\alpha_{1}\beta_{1}}^{-} \dots \ b_{\alpha_{n}\beta_{n}}^{-} |0\rangle (0|B_{\alpha_{n}\beta_{n}}\dots B_{\alpha_{1}\beta_{1}}^{-} \dots B_{\alpha_{1}\beta_{1}}^{-} \dots \ b_{\alpha_{n}\beta_{n}}^{-} |0\rangle (0|B_{\alpha_{n}\beta_{n}}\dots B_{\alpha_{n}\beta_{n}}^{-} \dots B_{\alpha_{n}\beta_{n}}^{-} \dots B_{\alpha_{n}\beta_{n}}^{-} \dots B_{\alpha_{n}\beta_{n}}^{-} \dots B_{\alpha_{n}\beta_{n}}^{-} \dots \ b_{\alpha_{n}\beta_{n}}^{-} \dots \ b_{\alpha_{n}\beta_{n}}^{-} \dots \ b_{\alpha_{n}\beta_{n}}^{-} \dots B_{\alpha_{$$

and similarly,

$$\hat{P}\hat{N} = \hat{\mathcal{E}}_n \hat{\mathcal{P}}_n n$$
 (5.38b)

Thus, from Eqs. (5.37a), (5.38b),  

$$\widehat{P} \ \widehat{N} \ \overset{+}{\mathsf{bd}} \beta = \begin{pmatrix} \overset{\infty}{n} & \frac{1}{(n!)^2} & \overset{*}{\underset{\beta_1 \dots \beta_n}{\overset{*}{\mathfrak{g}}_{1 \dots \alpha_n}} & \overset{*}{\mathsf{bd}} & \overset{*}{\mathsf{bd}} & \overset{*}{\mathfrak{g}} & \overset{*}{\underset{\beta_1 \dots \beta_n}{\overset{*}{\mathfrak{g}}_{1 \dots \alpha_n}} & \overset{*}{\mathsf{bd}} & \overset{*}{\mathsf{$$

Also, as 
$$[\vec{b}_{\alpha\beta}, \hat{N}] = -\vec{b}_{\alpha\beta}, Eq. (5.39a) \text{ becomes},$$
  
 $\hat{P} \hat{N} \vec{b}_{\alpha\beta} = \hat{P} \vec{b}_{\alpha\beta} (1 + \hat{N}) = B_{\alpha\beta}^{\dagger} \hat{P}$  (5.39b)

Another important relationship follows from,

 $\hat{P} b_{\alpha\beta} \hat{P} = \hat{\mathcal{E}}_{m,m'} [m'] (m' b_{\alpha\beta} [m]) (m = \hat{\mathcal{E}}_{m} [m[\alpha,\beta]) (m) - (5.40)$ where  $[m[\alpha,\beta]) =$  the state [m] with the particular pair  $(\alpha,\beta)$  removed

and 
$$b_{\alpha\beta} \hat{P} = \mathcal{E}_m b_{\alpha\beta} \operatorname{Im}(m) = \mathcal{E}_m \operatorname{Im}[m_{\alpha,\beta}](m) (5.41)$$

$$\widehat{P} b_{\alpha\beta} \widehat{P} = b_{\alpha\beta} \widehat{P} \qquad (5.42)$$

Finally, consider

$$= \sum_{n=0}^{\infty} \frac{n^{2}}{(n!)^{2}} \frac{1}{n!} \sum_{\substack{\alpha'_{1} \dots \alpha'_{n} \\ \beta'_{1} \dots \beta'_{n}}} B_{\alpha'_{1}\beta_{1}}^{\dagger} B_{\alpha'_{1}\beta_{1}}^{\dagger} \dots B_{\alpha'_{n-1}\beta_{n-1}}^{\dagger} |0\rangle(0| B_{\nu\beta_{n}} \dots B_{\nu\beta_{n-1}\beta_{n-1}})$$

$$= \sum_{n=0}^{\infty} \frac{1}{[(n-1)!]^{2}} \frac{1}{n!} \sum_{\substack{\alpha'_{1} \dots \alpha'_{n} \\ \beta'_{1} \dots \beta'_{n}}} B_{\alpha'_{1}\beta_{1}}^{\dagger} \dots B_{\alpha'_{n-1}\beta_{n-1}}^{\dagger} B_{\beta'_{1}\beta_{1}}^{\dagger} \dots B_{\alpha'_{n-1}\beta_{n-1}}^{\dagger} |0\rangle(0| B_{\nu\beta_{n}} B_{\alpha'_{n-1}\beta_{n-1}} \dots B_{\alpha'_{n-$$

$$= \mathcal{E}_{n=0}^{\infty} \frac{n^{2}}{(n!)^{2}} \mathcal{E}_{\substack{\alpha_{1},\dots,\alpha_{n}\\\beta_{1}\dots,\beta_{n}}} \mathbb{B}_{\alpha_{1}\beta_{1}}^{\dagger} \dots \mathbb{B}_{\alpha_{n}\beta_{n}}^{\dagger} 10)(0|b_{\alpha_{n-1}\beta_{n-1}}\dots b_{\alpha_{n}\beta_{1}}(\delta_{\alpha_{n}\mu}\delta_{\beta_{n}\beta_{n}})b_{\nu\beta}$$

$$= \mathcal{E}_{n=0}^{\infty} \frac{n^{2}}{(n!)^{2}} \mathcal{E}_{\substack{\alpha_{1},\dots,\alpha_{n}\\\beta_{1}\dots,\beta_{n}}} \mathbb{B}_{\alpha_{1}\beta_{1}}^{\dagger} \dots \mathbb{B}_{\mu_{n}\beta_{n}}^{\dagger} 10)(0|b_{\nu}\beta_{n}b_{\alpha_{n-1}\beta_{n-1}}\dots b_{\alpha_{n}\beta_{1}}$$

$$= \sum_{n=0}^{1} \frac{\omega}{[(n-1)!]^2} \frac{1}{(n!)} \sum_{\substack{q_1,\dots,q_n \\ \beta_1,\dots,\beta_n}} B_{d_1\beta_1}^{\dagger} \dots B_{d_n-1\beta_{n-1}} B_{d_1\beta_1}^{\dagger} B_{d_1\beta_1}^{\dagger} \dots B_{d_n\beta_1}^{\dagger} B_{d_1\beta_1}^{\dagger} \dots B_{d_n\beta_1}^{\dagger}$$
(5.43b)

(using (5.36a)

Comparing (5.43a) and (5.43b) we find,

$$\widehat{P} \widehat{\mathcal{E}}_{x} \stackrel{\dagger}{b}_{xu} \stackrel{\dagger}{s} \stackrel{}{b}_{xv} = \widehat{\mathcal{E}}_{x} \stackrel{\dagger}{b}_{xu} \stackrel{}{s} \stackrel{}{b}_{vx} \widehat{P}$$
 (5.44)

Frequent use of Eqs. (5.39), (5.42) and (5.44) will be made in the derivation of fermion-pair operator transformations.

# $\oint 5.3(e)$ Properties of U

Having defined the transformation  ${\tt U}$  and projection operator  $\widehat{P}$  , we can now consider the explicit nature of the mapping.

Equations (5.24a) and (5.24b) yield,

$$\langle m'|U^{\dagger}U|m \rangle = (0|\langle m'|U^{\dagger}U|m \rangle|0) = \langle 0|(m|m')|0 \rangle$$
 (5.45a)  
=  $\delta_{m',m}$ 

$$m^{and}(m'|UU^{t}|m) = \langle 0|(m'|U|U^{t}|m)|0 \rangle = (0|\langle m'|m \rangle|0)$$
  
=  $\delta m', m$  (5.45b)

indicating that  $U^{\dagger}U$  behaves as unity in the fermion space, and  $UU^{\dagger}$  as unity in the boson subspace.

The matrix element of an arbitrary operator,  $\mathsf{F}$  between fermion states is then given by,

$$\langle m|F|m' \rangle = \langle m|U^{\dagger}UFU^{\dagger}U|m' \rangle = (m|\mathcal{F}|m')$$

where

$$f = UFU^{\dagger}$$
 (5.46)

is the boson image of F . Thus the matrix element of any operator is unchanged under the action of the operator  $\,U\,$  .

Also, as in the transformation of Marumori et.al., the hermiticity of transformed operators is preserved.

Furthermore, Eq. (5.29) implies  

$$UU^{\dagger} = 10 \times (01 \text{ fm}^{\circ\circ} - \frac{1}{n!} \sqrt{\frac{1}{n!}} \left[ \text{ fap bap dp aa}^{\circ} - \frac{1}{n!} \sqrt{\frac{1}{n!}} \left[ \text{ fap bap dp aa}^{\circ} - \frac{1}{n!} \sqrt{\frac{1}{n!}} \left[ \text{ fap bap dp aa}^{\circ} - \frac{1}{n!} \sqrt{\frac{1}{n!}} \left[ \text{ fap bap dp aa}^{\circ} - \frac{1}{n!} \sqrt{\frac{1}{n!}} \left[ \text{ fap bap dp aa}^{\circ} - \frac{1}{n!} \sqrt{\frac{1}{n!}} \left[ \text{ fap bap dp aa}^{\circ} - \frac{1}{n!} \sqrt{\frac{1}{n!}} \left[ \text{ fap bap dp aa}^{\circ} - \frac{1}{n!} \sqrt{\frac{1}{n!}} \right]^{\circ} \left[ \text{ fap bap dp aa}^{\circ} - \frac{1}{n!} \sqrt{\frac{1}{n!}} \left[ \frac{1}{n!} \sqrt{\frac{1}{n!}} \left[ \frac{1}{n!} \sqrt{\frac{1}{n!}} \sqrt{\frac{1}{$$

$$= 10 \sum_{n=0}^{\infty} \frac{1}{(n!)^3} \sum_{n=0}^{\infty} \langle 0| d\beta_1 a_{d_1} \dots d\beta_n a_{d_n} a_{d_n}^{\dagger} d_{d_n}^{\dagger} \dots \langle 0| \delta_{n} a_{n-1} \rangle$$

$$\dots a_{n-1}^{\dagger} d_{n-1}^{\dagger} a_{n-1} \sum_{n=0}^{\infty} (1-1)^{n-1} ($$

(where  $\Omega$  denotes the summation indices  $\{\alpha'_1, \dots, \alpha'_n, \beta_1, \dots, \beta_n, \delta_1, \dots, \delta_n, \delta_n, \dots, \delta_n\}$ ). The operators  $d\beta_1 a_{d_1} \dots d\beta_n a_{d_n}$  and  $a_{\delta n} d\delta_n \dots a_{\delta n} d\delta_n$  are antisymmetric. with respect to the indices  $(\beta_1 a_1, \dots, \beta_n a_n)$  and  $(\delta_n \delta_n, \dots, \delta_1 \delta_1)$ respectively. The sums in expression (5.47) will thus contain the antisymmetrised components of the states  $b_{\alpha'_1\beta_1} \dots b_{\alpha'n\beta_n} lO$ and  $(O|b_{\delta n} \delta_1 \dots b_{\delta_1\delta_1} \dots only, and we may replace <math>b_{\alpha'_1\beta_1} \dots b_{\alpha'n\beta_n} lO$ ) by the antisymmetrised state  $(\frac{1}{n!}) B_{\alpha'_1\beta_1} \dots B_{\alpha'n\beta_n} lO$ .

$$\Rightarrow UU^{\dagger} = 10 \sum_{n=0}^{\infty} \frac{1}{(n!)^5} \sum_{n=0}^{\infty} \langle 0 | d\beta_1 \, a_{d_1} \dots d\beta_n \, a_{d_n} \, a_{d_n}^{\dagger} \, d_{d_n}^{\dagger} \dots$$

$$\dots a_{x_i}^{\dagger} d_{d_i}^{\dagger} | 0 \rangle B_{\alpha_1\beta_1}^{\dagger} \dots B_{\alpha_n\beta_n}^{\dagger} | 0 \rangle \langle 0 | B_{\alpha_n\beta_n} \dots B_{\alpha_i\beta_i} \langle 0 | (using Eq. (5.27a)).$$

$$= 10 \sum_{n=0}^{\infty} \sum_{\substack{n=0\\ \beta_1 \dots \beta_n}}^{\infty} B_{\alpha_1\beta_1}^{\dagger} \dots B_{\alpha_n\beta_n}^{\dagger} | 0 \rangle \langle 0 | B_{\alpha_n\beta_n} \dots B_{\alpha_i\beta_i} \langle 0 | (using Eq. (5.27a)).$$

In the boson subspace we may replace  $|0\rangle|0\rangle\langle0|(0)|_{by}|0\rangle(0)$  whence,

Equations (5.45b) and (5.47b) indicate that the projection operator acts like the unity operator in the boson subspace.

From Eqs. (5.29) and (5.37) we also find,

$$\widehat{P} U = \sum_{n=0}^{\infty} \frac{1}{(n!)^2} \sum_{\substack{\alpha',\dots,\alpha',\beta \\ \beta',\dots,\beta'n}} B_{\alpha',\beta',\dots} B_{\alpha',\beta'n} \frac{1}{m!} \sum_{\substack{n=0\\ m!}} \frac{1}{m!} \sum_{\substack{\alpha',\dots,\alpha',\beta \\ m!}} B_{\alpha',\beta',n} \frac{1}{m!} \sum_{\substack{\alpha',\dots,\alpha',\beta \\ m!}} B_{\alpha',\beta',n} \sum_{\substack{\alpha',\dots,\alpha',\beta \\ m!}} B_{\alpha',\beta',n} \frac{1}{m!} \sum_{\substack{\alpha',\dots,\alpha',\beta \\ m!}} B_{\alpha',\beta',\dots,\beta'} \sum_{\substack{\alpha',\dots,\alpha',\beta',\dots,\alpha',\beta'}} B_{\alpha',\beta',\dots,\beta'} \sum_{\substack{\alpha',\dots,\alpha',\beta',\dots,\alpha',\beta'}} B_{\alpha',\beta',\dots,\beta'} \sum_{\substack{\alpha',\dots,\alpha',\beta',\dots,\alpha',\beta'}} B_{\alpha',\beta',\dots,\beta'} \sum_{\substack{\alpha',\dots,\alpha',\beta',\dots,\alpha',\beta'}} B_{\alpha',\beta',\dots,\beta'} \sum_{\substack{\alpha',\dots,\alpha',\beta',\dots,\alpha',\beta',\dots,\beta'}} B_{\alpha',\beta',\dots,\beta'} \sum_{\substack{\alpha',\dots,\alpha',\beta',\dots,\alpha',\beta'}} B_{\alpha',\beta',\dots,\beta'} \sum_{\substack{\alpha',\dots,\alpha',\beta',\dots,\alpha',\beta',\dots,\beta'}} B_{\alpha',\beta',\dots,\beta'} \sum_{\substack{\alpha',\dots,\alpha',\beta',\dots,\alpha',\beta'}} B_{\alpha',\beta',\dots,\alpha'} \sum_{\substack{\alpha',\dots,\alpha',\beta',\dots,\alpha',\beta'}} B_{\alpha',\beta',\dots,\alpha'} \sum_{\substack{\alpha',\dots,\alpha',\beta',\dots,\alpha',\beta'}} B_{\alpha',\beta',\dots,\alpha'} B_{\alpha',\beta',\dots,\alpha'$$

= 
$$10 < 01 \leq_{n=0}^{\infty} \sqrt{n!} (n!)^3 \leq_{n=0}^{\infty} B_{\alpha_1\beta_1} \dots B_{\alpha_n\beta_n} 10 (01 b_{\alpha_n\beta_n} \dots b_{\alpha_n\beta_n} b_{\alpha_n\delta_n} \dots b_{\alpha_n\beta_n} b_{\alpha_n\delta_n} \dots b_{\alpha_n\beta_n} (5.48a)$$

We note that,

 $(0 \mid b \, dn \, \beta_n \dots b \, d_1 \, \beta_1 \, \overset{+}{b}_{x_1 \delta_1} \dots \overset{+}{b}_{x_1 \delta_1} \, |0) = \delta \, dn \, x_n \, \delta \, \beta_n \, \delta_n \dots \delta \, d_1 \, x_1 \, \delta \, \beta_1 \, \delta_1$ and as the summation over  $\Omega$  generates n! such terms for fixed  $\{\delta n \delta_n, \dots \delta_1 \delta_1\}$  we find  $\widehat{P} \ U = |0\rangle \langle 0| \, \underbrace{\sum_{n=0}^{\infty} \sqrt{n!}}_{n=0} \frac{1}{(n!)^2} \underbrace{\sum_{\substack{x_1, x_n \\ \delta_1 \dots \delta_n}}}_{\substack{x_1, \delta_1 \dots \delta_n}} B^{\dagger}_{x_1 \delta_1} \dots B^{\dagger}_{x_n \delta_n} \, |0\rangle \, d\delta_1 \, a_{x_1} \dots d\delta_n \, a_{x_n} \, (ol \ \delta_1 \dots \delta_n \, \delta_1 \dots \delta_n \, \delta_n) \, (ol \ \delta_1 \dots \delta_n) \, (ol \ \delta$ 

$$U^{\dagger} \hat{P} = U^{\dagger} \tag{5.48c}$$

As a consequence of Eqs. (5.46), (5.47b), (5.48b) and (5.48c),

$$\mathbf{J} = \hat{\boldsymbol{P}} \mathbf{J} = \mathbf{J} \hat{\boldsymbol{P}} \tag{5.49}$$

implying that the application of an arbitrary transformed (boson) operator to any boson state projects out the non-antisymmetrised states. We have thus successfully extended the ideas of the generalised transformation derived by Marumori et.al. to the case of electron-hole pair operators. Let us now derive the explicit transformations.

# $\oint$ 5.3(f) Derivation of Electron-hole Pair Operator Transformations

Firstly, consider

$$\begin{aligned} Ua_{\mu}^{\dagger}a_{\nu}U^{\dagger} &= 10 \\ <01 \\ & \sum_{n=0}^{\infty} \frac{1}{n!} \sqrt{\frac{1}{n!}} \left[ \sum_{a\beta} b_{a\beta}^{\dagger} d_{\beta} a_{a} \right]^{n} \\ & >10 \\ <01 \\ & \sum_{m=0}^{\infty} \frac{1}{m!} \sqrt{\frac{1}{m!}} \left[ \sum_{x\delta} a_{x}^{\dagger} d_{\delta}^{\dagger} b_{x\delta} \right]^{m} \\ & 10 \\ <01 \\ (5.50) \end{aligned}$$

Expression (5.50) will be non-zero only for those terms in which the summation over  $\{\alpha_1, \dots \alpha_n, \beta_1, \dots \beta_n\}$  generates the same number of electron and hole operators as the summation over  $\{\lambda_1, \dots \lambda_m, \delta_1, \dots \delta_m\}$ .

where  $\boldsymbol{\Lambda}$  again denotes summation over all indices.

As in the derivation of expression (5.47b), we replace the boson state  $b_{\alpha}^{\dagger}, \beta_{1}, \ldots, b_{\alpha}^{\dagger}, \beta_{n} \mid O$  by the antisymmetrised state  $\left(\frac{1}{n!}\right) B_{\alpha}^{\dagger}, \beta_{1}, \ldots, B_{\alpha}^{\dagger}, \beta_{n} \mid O$  (because of the antisymmetrical nature of the fermion operators in the summation).

Consider the summation over  $\Omega$  : By construction, all the subscripts in any given term are different (otherwise the fermion operators generate a zero result). The fermion operators  $a_{\mu}^{+}$  and  $a_{\nu}$  can be moved as follows:

$$\begin{aligned} \hat{\mathcal{E}}_{\mathcal{D}} & \rightarrow \hat{\mathcal{E}}_{\mathcal{D}} \hat{\mathcal{E}}_{r,s} \langle 0| (-a_{\mu}^{\dagger}a_{\alpha_{r}} + \delta_{\mu}a_{r}) [\Pi_{i\neq r} (d_{\beta}ia_{\alpha})] d_{\beta r} d_{\delta_{s}}^{\dagger} \\ & \times [\Pi_{i\neq s} (a_{\delta_{i}}^{\dagger}d_{\delta_{i}}^{\dagger})] (\delta_{r} s_{s} - a_{\delta_{s}}^{\dagger}a_{\gamma}) |0\rangle B_{\alpha_{i}\beta_{i}}^{\dagger} \dots B_{\alpha_{n}\beta_{n}}^{\dagger} |0\rangle \langle 0| B_{\delta_{n}\delta_{n}} \dots B_{\delta_{s}\delta_{s}} \end{aligned}$$

$$\Rightarrow \hat{\mathcal{E}}_{n} n^{2} \delta_{\mu\alpha n} \delta_{\nu\nu n} \langle 0| d_{\beta_{1}} a_{\alpha_{1}} \dots d_{\beta_{n-1}} a_{\alpha_{n-1}} d_{\beta_{n}} d_{\delta_{n}}^{\dagger} a_{\delta_{n-1}} \dots d_{\delta_{n-1}} \dots d_{\delta_{n-1}} \dots d_{\delta_{n-1}} d_{\delta_{n-1}} \dots d$$

The matrix elements in Eq. (5.52) produce Kronecker delta functions as described by Eq. (5.27b). As the sum over  $\Omega$  yields n!(n-i)! such matrix elements for fixed  $\{\alpha_{n-1}, \dots, \alpha_i, \beta_n, \dots, \beta_i\}$  we obtain,

Equ. 5.52 
$$\Rightarrow \sum_{\substack{\alpha_1,\ldots,\alpha_n\\\beta_1,\ldots,\beta_n}} n^2(n-1)! n! B_{\alpha_1\beta_1} \dots B_{\mu\beta_n}^{\dagger}[0)(0| B_{\gamma\delta_1} \dots B_{\gamma_1\delta_1} (5.53))$$

We note that in all further discussion, we include the fermion ground states in the boson states as, 10>  $B_{\alpha_1\beta_1}^{\dagger}$ ...  $B_{\alpha_n\beta_n}^{\dagger}$  10)(01  $B_{\alpha_n\beta_n}$ ...  $B_{\alpha_1\beta_1}^{\dagger}$ ...  $B_{\alpha_n\beta_n}^{\dagger}$  10)10><01(01  $\times B_{\alpha_n\beta_n}$ ...  $B_{\alpha_1\beta_1}^{\dagger}$ ...  $B_{\alpha_n\beta_n}^{\dagger}$  10)(01  $B_{\alpha_n\beta_n}$ ...  $B_{\alpha_1\beta_1}^{\dagger}$ ... (5.54) Thus, substituting (5.53) in (5.51) and using (5.54), we find,

$$Ua_{\mu}a_{\nu}U^{\dagger} = \sum_{n=0}^{\infty} \frac{n(n!)^{2}}{(n!)^{5}} \sum_{\rho} B_{\mu\rho}^{\dagger} \left\{ \sum_{\substack{\alpha_{i}, \dots, \alpha_{n-1} \\ \beta_{i} \dots \beta_{n-1}}} B_{\alpha_{i}\beta_{i}}^{\dagger} \dots B_{\alpha_{n-1}\beta_{n-1}}^{\dagger} 10\right\} (0)$$

(letting  $n \rightarrow n+1$  )

$$= \sum_{\substack{n=1\\\beta \\ n=1}} B_{n=1}^{\dagger} \sum_{\substack{(1+n)^2\\\beta \\ n=1}}^{1} \frac{1}{(n!)^3} \sum_{\substack{\alpha_1,\dots,\alpha_n\\\beta_1\dots,\beta_n}} B_{\alpha_1\beta_1}^{\dagger} \dots B_{\alpha_n\beta_n}^{\dagger} 10) (0|B_{\alpha_n\beta_n}\dots B_{\alpha_n\beta_1}.B_{\alpha_n\beta_n}) B_{\alpha_1\beta_1} \dots B_{\alpha_n\beta_n} B_{\alpha_n\beta_n} \dots B_{\alpha_n\beta_n$$

$$\Rightarrow \bigcup_{\mu} a_{\nu} \bigcup_{\mu} u^{+} = \mathcal{E}_{\rho} B_{\mu\rho}^{+} \mathcal{E}_{n=1}^{\infty} \frac{1}{(1+\widehat{N})^{2}} \frac{1}{(n!)^{3}} \mathcal{E}_{\alpha_{\mu},\alpha_{\mu}} B_{\alpha_{\mu},\beta_{\mu}}^{+} \dots B_{\alpha_{\mu},\beta_{\mu}}^{+} |0\rangle (0|B_{\alpha_{\mu},\beta_{\mu},\beta_{\mu}} - B_{\alpha_{\mu},\beta_{\mu},\beta_{\mu}}^{+} - B_{\alpha_{\mu},\beta_{\mu},\beta_{\mu}}^{+} - B_{\alpha_{\mu},\beta_{\mu},\beta_{\mu}}^{+} - B_{\alpha_{\mu},\beta_{\mu},\beta_{\mu},\beta_{\mu}}^{+} - B_{\alpha_{\mu},\beta_{\mu},\beta_{\mu},\beta_{\mu}}^{+} - B_{\alpha_{\mu},\beta_{\mu},\beta_{\mu},\beta_{\mu}}^{+} - B_{\alpha_{\mu},\beta_{\mu},\beta_{\mu},\beta_{\mu}}^{+} - B_{\alpha_{\mu},\beta_{\mu},\beta_{\mu},\beta_{\mu}}^{+} - B_{\alpha_{\mu},\beta_$$

$$= \hat{\mathcal{E}}_{\rho} \stackrel{+}{\mathcal{B}}_{\rho} \frac{1}{(1+\hat{N})} \stackrel{-}{\hat{P}} \stackrel{+}{b}_{\nu\rho} \stackrel{-}{\hat{P}} (\text{using Eq. (5.42)})$$
$$= \hat{\mathcal{E}}_{\rho} \stackrel{-}{\hat{P}} \stackrel{+}{b}_{\nu\rho} \stackrel{-}{\hat{P}} \stackrel{-}{\rho} \stackrel{-}{$$

$$\Rightarrow Ua_{\mu}^{\dagger}a_{\nu}U^{\dagger} = \hat{\xi}_{\rho} \dot{b}_{\nu\rho} \hat{P} \quad (\text{using Eq. (5.44), (5.37b)}) \quad (5.55)$$
  
where the summation over  $\rho$  is over hole states only, and

Similarly, for the hole pair,

$$U d_{\mu}^{\dagger} d_{\nu} U^{\dagger} = \hat{\xi}_{\mu} d_{\mu} d_{\eta} q^{\dagger} d_{\nu} q^{\dagger} (5.56)$$

where summation over ho' refers to all electron states.

Finally, we consider

$$Ua_{\mu}^{\dagger} d_{\nu}^{\dagger} U^{\dagger} = 10 < 01 \mathcal{E}_{n=0}^{\infty} \frac{1}{n!} \sqrt{n!} \left[ \mathcal{E}_{xs} b_{xs}^{\dagger} d_{p} a_{q} \right]^{n} (0) (01) \times a_{\mu}^{\dagger} d_{\nu}^{\dagger} (0) (01 \mathcal{E}_{m=0}^{\infty} \frac{1}{m!} \sqrt{m!} \left[ \mathcal{E}_{xs} a_{x}^{\dagger} d_{s}^{\dagger} b_{xs} \right]^{m} (0) < 01$$
(5.57)

Non-zero contributions to Eq. (5.57) arise only when the summation over  $\{\alpha'_{1}, ..., \alpha'_{n}, \beta_{1}, ..., \beta_{n}\}$  is raised to a power one greater than the summation over  $\{\lambda_{1}, ..., \lambda_{m}, \delta_{1}, ..., \delta_{m}\}$ ,

$$\Rightarrow |0\rangle \sum_{n=1}^{\infty} \frac{1}{(n!)^2} \frac{1}{\sqrt{n!}} \frac{1}{[(n-1)!]^2} \frac{1}{\sqrt{(n-1)!}} \sum_{n=1}^{\infty} \langle 0| d\beta_1 \alpha_{d_1} \dots d\beta_n \alpha_{d_n} \alpha_{j_n}^{\dagger} d_n^{\dagger} \alpha_{\delta_{n-1}}^{\dagger} d_{\delta_{n-1}}^{\dagger} \dots \alpha_{\delta_n}^{\dagger} d_{\delta_n}^{\dagger} |0\rangle \langle 0| \beta_{\delta_{n-1}\delta_{n-1}} \dots \beta_{\delta_n\delta_n} \langle 0| \qquad (5.58)$$
where we have replaced the boson state  $b_{\alpha_1\beta_1}^{\dagger} \dots b_{\alpha_n\beta_n}^{\dagger} |0\rangle$  by the antisymmetrised state  $(\frac{1}{n!}) \beta_{\alpha_1\beta_1}^{\dagger} \dots \beta_{\alpha_n\beta_n}^{\dagger} |0\rangle$ .

Consider the summation over  ${f \Omega}$  ,

$$\begin{aligned} & \sum_{n} \langle 0| d\beta_{1} a_{di} \dots d\beta_{n} a_{dn} a_{ju}^{\dagger} dv^{\dagger} a_{\delta n-1}^{\dagger} d\delta_{n-1} \dots a_{\lambda 1}^{\dagger} d\delta_{1} |0\rangle B_{\alpha 1 \beta_{1}}^{\dagger} \dots \\ & \dots B_{\alpha n \beta n}^{\dagger} |0\rangle (0| B_{\lambda n-1} \delta_{n-1} \dots B_{\lambda 1} \delta_{1} \\ & = \sum_{\substack{\alpha_{1} \dots \alpha_{n} \\ \beta_{1} \dots \beta_{n}}} (n-1)! (n-1)! n^{2} \langle n_{\alpha 1 \beta_{1}} \dots n_{\alpha n-1} \beta_{n-1} d\beta_{n} a_{\alpha n} a_{ju}^{\dagger} dv^{\dagger} \\ & \qquad (5.59) \\ & \sum_{\substack{\alpha_{1} \dots \alpha_{n} \\ \beta_{1} \dots \beta_{n}}} \chi |n_{\alpha n-1} \beta_{n-1} \dots n_{\alpha 1 \beta_{1}} \rangle B_{\alpha 1 \beta_{1}}^{\dagger} \dots B_{\alpha n \beta_{n}}^{\dagger} |0\rangle (0| B_{\alpha n-1} \beta_{n-1} \dots B_{\alpha 1 \beta_{1}} \\ \end{aligned}$$

The two factors of (n-1)! in Eq. (5.59) arise from the summations over the electron and hole indices  $\{\lambda_{n-1}, \ldots, \lambda_1, \delta_{n-1}, \ldots, \delta_n\}$  for fixed  $\{d, \beta\}$ . The factor  $n^2$  is necessary as we can choose the remaining electron-hole pair (i.e.  $d\beta n \, Odn$ )  $n^2$  ways. Thus,

$$Equ. 5.59 \Rightarrow \begin{pmatrix} c \\ (n!)^2 \\ B_{\alpha'\beta_1} \\ \beta_1 \\ \beta$$

$$U a_{\mu}^{\dagger} d_{\nu}^{\dagger} U^{\dagger} = \sum_{n=1}^{\infty} \frac{1}{\sqrt{n!}} \frac{1}{[(n-1)!]^{2}} \sqrt{\frac{1}{(n-1)!}} B_{\mu\nu}^{\dagger} \sum_{\substack{\alpha_{1},\dots,\alpha_{n-1}\\\beta_{1},\dots,\beta_{n-1}\\\beta_{n-1},\beta_{n-1}}} B_{\alpha_{1}\beta_{1},\dots,\beta_{n-1}}^{\dagger} \dots B_{\alpha_{n}\beta_{n-1}}^{\dagger} \dots B_{\alpha_{n}\beta_{n-1}}^{\dagger} \dots B_{\alpha_{n}\beta_{n-1}}^{\dagger} \dots B_{\alpha_{n}\beta_{n-1}}^{\dagger} \dots B_{\alpha_{n}\beta_{n-1}}^{\dagger} \dots B_{\alpha_{n}\beta_{n-1}}^{\dagger} \dots B_{\alpha_{n-1}\beta_{n-1}}^{\dagger} \dots B_{\alpha_{n-1}\beta_{n-1}}^{$$

$$= B_{\mu\nu}^{T} \mathcal{E}_{n=1}^{\infty} \sqrt{n+1} \quad (n!)^{3} \mathcal{E}_{\beta} B_{\alpha',\beta}^{+} \dots B_{\alpha',\beta_{n}}^{+} lo)(0|B_{\alpha'n} \dots B_{\alpha',\beta_{n}} B_{\alpha',\beta_{n}} \dots B_{\alpha$$

$$U a_{\mu}^{\dagger} d_{\nu}^{\dagger} U^{\dagger} = B_{\mu\nu}^{\dagger} \sqrt{1+\hat{N}} \hat{P}$$
 (5.61)

And, using Eq. (5.39b),

$$U a_{\mu}^{\dagger} d_{\nu}^{\dagger} U^{\dagger} = \hat{P} b_{\mu\nu}^{\dagger} \sqrt{1 + \hat{N}} \qquad ----(5.62)$$

Equations (5.55), (5.56) and (5.62) represent all possible transformations of electron-hole pair operators we may form with the transformation U (defined by Eq. (5.29)).

For completeness we now quote the main results of this section.

Table 5.1: Electron-Hole Transformations

$$\begin{split} U &= 10 \rangle \langle 0| \ \mathcal{E}_{n=0}^{\infty} \ \frac{1}{n!} \ \sqrt{n!} \ \left[ \ \mathcal{E}_{\alpha\beta} \ b_{\alpha\beta}^{\dagger} \ d_{\beta} \ a_{\alpha} \right]^{n} 10 \rangle \langle 0| \\ \widehat{P} &= \mathcal{E}_{n=0}^{\infty} \ \mathcal{E}_{\alpha,\dots,\alpha,n} \ \frac{1}{(n!)^{3}} \ b_{\alpha,\beta,\dots,n}^{\dagger} \ b_{\alpha,\beta,n} \ b_{\alpha,\beta,n} \ 10 \rangle \langle 0| \ B_{\alpha,\beta,\dots,n} \ B_{\alpha,\beta,n} \\ U \ a_{\mu}^{\dagger} a_{\nu} \ U^{\dagger} &= \mathcal{E}_{\rho} \ b_{\mu\rho}^{\dagger} \ b_{\nu\rho} \ \widehat{P} \\ U \ a_{\mu}^{\dagger} a_{\nu} \ U^{\dagger} &= \mathcal{E}_{\rho} \ b_{\rho'\mu} \ b_{\rho'\nu} \ \widehat{P} \\ U \ a_{\mu}^{\dagger} d_{\nu} \ U^{\dagger} &= \mathcal{E}_{\rho'} \ b_{\rho'\mu} \ b_{\rho'\nu} \ \widehat{P} \\ U \ a_{\mu}^{\dagger} d_{\nu}^{\dagger} \ U^{\dagger} &= B_{\mu\nu\nu}^{\dagger} \ \sqrt{1+\widetilde{N}} \ \widehat{P} \ = \widehat{P} \ b_{\mu\nu\nu}^{\dagger} \sqrt{1+\widetilde{N}} \\ \text{where } \ \rho' \ \text{and} \ \rho \ \text{indicate summation over electron and hole states respectively and,} \\ \widehat{N} \ = \ \mathcal{E}_{\alpha\beta} \ b_{\alpha\beta} \ b_{\alpha\beta} \\ and \qquad B_{\alpha\beta}^{\dagger} \ = \ b_{\alpha\beta}^{\dagger} \ - \mathcal{E}_{\nu\nu} \ b_{\alpha\nu} \ b_{\delta\beta} \ b_{\delta\nu} \end{split}$$

Unfortunately the transformed electron-hole operator pair  $O_{\mu} d_{\nu}^{\dagger}$ generates an infinite boson expansion, due to the term  $\sqrt{1 + N}$ . To express such a term we use the general operator expansion derived by Gardiner (Steyn-Ross and Gardiner, 1982),

$$f(\hat{N}) = : \mathcal{E}_{R=0}^{\infty} (\dot{b}_{i} \dot{b}_{i}) : \mathcal{E}_{r=0}^{R} \frac{(-1)^{R-r} f(r)}{r! (R-r)!}$$
(5.63)

:: denotes normal ordered product.

Thus use of the transformation U is limited to certain cases in which the series, Eq. (5.63) converges.

In the next chapter we use this transformation to develop a microscopic theory of excitonic optical bistability in semiconductors.

#### CHAPTER 6

#### THE QUANTUM THEORY OF EXCITONIC OPTICAL BISTABILITY

### $\phi_{6.1}$ Introduction

In this chapter we develop a microscopic theory of the optical bistability arising from the interaction of a coherent light field with excitons comprising a semiconductor.

We start with the Hamiltonian for the system written in terms of electron and hole operators. Following the discussion of Chapter Five, we transform such a Hamiltonian to one characterised by exciton operators. In the exciton Hamiltonian so derived we can identify such processes as exciton-exciton and exciton-field interactions without ambiguity.

We also wish to define exciton operators for our system in order to systematically include damping of excitons. That is, the quantum theory of damping requires us to define a damping Hamiltonian of the form,

 $\begin{array}{l} H_{exciton} = Q_i b_{ex}^{\dagger} + h.c \\ \\ \mbox{[where } Q_i \mbox{ is a reservoir operator and } b_{ex}^{\dagger} \mbox{ an exciton operator]}. \\ \\ \\ \mbox{This is clearly not possible in a fermion system described in terms of } \\ \\ \mbox{electrons and holes.} \end{array}$ 

To achieve the desired exciton Hamiltonian, we use the bosonisation procedure described in Chapter Five to transform pairs of fermion operators to boson operators.

We deal with the infinite boson expansions generated by such a transformation by considering two limiting cases of the system:

i) the low exciton density case ( $\oint 6.3(b)$ ): in which we truncate the boson expansion (as the terms of this series increase in powers of exciton density). We then derive the master equation and Fokker-Planck equation for this system. ii) The high exciton density case ( $\oint 6.6(b)$ ): we perform a canonical transformation on the exciton operators, allowing us to expand about the deterministic solution.

We note that the bistability experiments of Gibbs et.al. correspond to a high exciton density system. Nevertheless, we investigate the possibility of observing bistability in both high and low exciton density systems.

### $\phi$ 6.2 The Model: Fermion Space

We consider the intracavity interaction of a coherent driving field with a semiconductor.

We are interested in the case when the light field excites an electron from the filled valence band to the empty conduction band (thereby creating a hole in the valence band) and so creates an exciton. For simplicity we consider a two-band semiconductor only and neglect electron spin.

Firstly, let us ignore the effects of damping on the system. The discussion of Chapter One,  $\oint 1.3(b)$  indicated that the required Hamiltonian for such a lossless system in a second quantised theory is,

(we discuss damping of the system later in this chapter).

We may write Eq. (6.1a) as, H = Ho,matter + Hmatter-light + Hlight + HI + He.m $+ H_D$  (6.1b)

The first three terms of Eq. (6.1b) stem from expanding the squared bracket in Eq. (6.1a) and noting  $\nabla A = 0$ ; they are:

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1) Ho, matter = 
$$\int \psi^+(\underline{x}) \left\{ \frac{-\nabla^2 \hbar^2}{2m} + \operatorname{Veff}(1\underline{x}I) \right\} \psi(\underline{x}) d^3x$$
 (6.2)

this represents the free Hamiltonian of the semiconductor; (  $Vecc(1\times 1)$  is the potential due to the electrons).

2) The interaction between light and matter,

Hmatter-light = 
$$\int \psi^{+}(\underline{x}) \left(-\frac{e}{m} A(\underline{x}) \cdot \frac{\nabla \hbar}{c}\right) \psi(\underline{x}) d^{3}x \qquad (6.3)$$
  
and

This term describes an interaction between electrons and the light field which is nonlinear in  $\underline{A}$ . For small values of the vector potential  $\underline{A}$  (i.e. small values of its matrix elements) we may neglect this term.

The term  $H_I$  occuring in Eq. (6.1b) describes interaction between particles:

Also, the term He.m. in Eq. (6.1b) describes the free field, which in the single mode approximation becomes,

$$-1_{e.m.} = \omega b b$$
 (6.6a)

where **b** is a boson operator characterising the light field within the cavity.

Finally in Eq. (6.1b),  $H_p$  describes coupling between the external driving field (of frequency  $\omega_L$  and amplitude E) and the cavity mode of interest;

### $\phi$ 6.2(a) Semiconductor Hamiltonian

The Hamiltonian for the semiconductor material alone is given by Eqs. (6.2) and (6.5),

$$H_{s.c.} = \int d^{3}x \ \gamma^{+}(\underline{x}) \ H_{\bullet}(\underline{x}) \ \gamma(\underline{x}) + \frac{1}{2} \iint d^{3}x \ d^{3}y \ \gamma^{+}(\underline{x}) \ \gamma^{+}(\underline{y}) \ \gamma(\underline{x}-\underline{y}) \ \gamma(\underline{y}) \ \gamma(\underline{x})$$
where  $H_{-} = -\frac{\hbar^{2} \nabla^{2}}{2} + V_{ex}(\underline{y})$  is the single partial Hamiltonian

where  $H_0 = -\frac{\kappa - v}{2m} + Veff(IXI)$ , is the single particle Hamiltonian.

As the semiconductor is characterised only by conduction and valence electrons, we expand the field operators as,

where  $\Omega_{R_c}$  and  $\Omega_{R_{v}}$  are fermion annihilation operators for electrons (momentum  $R_c$ ,  $R_v$ ) in the conduction and valence bands respectively.

The arphi's are eigenfunctions of  $\mathsf{H}_{m{o}}$  , satisfying,

$$H_{o} \varphi_{R_{j}} = E_{o,R_{j}} \qquad (j = c, v)$$

Bloch's theorem states that eigenfunctions in an infinite lattice are plane waves modulated by the period of the lattice (see for example Haken, 1976, Chapter 4).

We thus express the eigenfunctions as:

$$\mathcal{U}_{R_j}(\underline{x}) = U_{R_j}(\underline{x}) \exp(i\underline{R}_j,\underline{x})/\sqrt{N}$$
 (6.9)

for an electron with wavevector  $\mathcal{R}$  in band j.  $U\kappa_j(\underline{X})$  is the Bloch function, having the same periodicity as the lattice. We include a factor  $\frac{1}{\sqrt{N}}$ , where N is the number of unit cells in the crystal, so that  $U\kappa_j(\underline{X})$  per lattice cell is normalised.

Expanding the Hamiltonian  $H_{s.c}$  in terms of the field operators as given in Eq. (6.9), we find (see for example Haken, 1976):

where, for example

where, for example  $\langle R_{1c}, R_{2c} | \vee | R_{3c}, R_{4c} \rangle = \iint d^{3} \times d^{3} y \, \varphi^{*}_{R_{1c}}(\underline{x}) \, \varphi^{*}_{R_{2c}}(\underline{y}) \, \vee (|\underline{x} - \underline{y}|)$   $\times \, \varphi_{R_{3c}}(\underline{y}) \, \varphi_{R_{4c}}(\underline{x})$ and  $V(|\underline{x}-\underline{y}|) = \frac{e^2}{|\underline{x}-\underline{y}|}$ 

In the derivation of Eq. (6.10), terms not conserving pair  
excitations, such as 
$$a_{\mathbf{k}c}^{\dagger} a_{\mathbf{k}c}^{\dagger} a_{\mathbf$$

We introduce the concept of a hole via the time reversal operator  ${\sf K}$ (Hanamura and Haug, 1977),

$$a_{Rr} = K^{\dagger} d_{R} K = d_{-R}^{\dagger}$$

- the annihilation of an electron in the valence band having wavevector  ${\sf k}$ is equivalent to the creation of a hole of wavevector  $-\aleph$  .

Equation (6.10) then becomes

$$H_{s.c} = E_{o} + \mathcal{E}_{R} E_{c}(R) a_{R}^{\dagger} a_{R} + \mathcal{E}_{R} E_{v}(R) d_{R}^{\dagger} d_{R}$$

$$+ \frac{1}{2} \sum_{R_{1...R_{4}}} \langle R_{1c}, R_{2c} | V | R_{3c}, R_{4c} \rangle a_{R_{1}}^{\dagger} a_{R_{2}}^{\dagger} a_{R_{3}} a_{R_{4}}$$

$$+ \frac{1}{2} \sum_{R_{...R_{4}}} \langle -R_{1v}, -R_{2v} | V | -R_{3v}, -R_{4v} \rangle d_{R_{1}}^{\dagger} d_{R_{2}}^{\dagger} d_{R_{3}} d_{R_{4}}$$

$$- \mathcal{E}_{R_{1...R_{4}}} (\langle R_{1c}, R_{3v} | V | R_{2v}, R_{4c} \rangle - \langle R_{1c}, R_{3v} | V | R_{4c}, R_{4v} \rangle) \qquad (6.11)$$

$$\times a_{R_{1}}^{\dagger} d_{R_{2}}^{\dagger} d_{R_{3}} a_{R_{4}}$$

where

$$E_{o} = \hat{\xi}_{R} E_{o,v}(R) + \hat{\xi}_{I,R}(\langle R_{v}, I_{v} | V | I_{v}, R_{v} \rangle - \langle R_{v}, I_{v} | V | R_{v}, I_{v} \rangle)$$

$$E_{c}(R) = E_{o,c}(R) + \mathcal{E}_{I,R}(\langle Rc, lv | V | lv, Rc \rangle - \langle Rc, lv | V | Rc, Iv \rangle)$$

$$E_{r}(\mathbf{r}) = -E_{o,v}(\mathbf{r}) - \hat{\mathcal{E}}_{l,\mathbf{r}}(\langle lv, -\mathbf{r} \cdot | V | -\mathbf{r}_{v}, lv \rangle - \langle lv, -\mathbf{r}_{v} | V | lv, -\mathbf{r}_{v} \rangle)$$

 $E_{\circ}$  represents a constant energy term which we will neglect in future calculations; and in the effective mass approximation, (Haken, 1976),

$$E_{r}(R) = \frac{\hbar^2 R^2}{2mh}$$
;  $E_c(R) = E_g + \frac{\hbar^2 R^2}{2me}$ 

where  $m_{\ell}$  and  $m_{h}$  are the respective effective masses for electrons and holes and

$$E_{g} = energy of gap.$$

# Evaluation of Interaction Hamiltonian

We discuss now the approximations employed to evaluate the matrix elements occurring in the interaction terms of Eq. (6.11). Firstly, consider

1) 
$$\frac{1}{2} \sum_{\substack{R_{1},R_{4}}} \langle R_{1}C, R_{2}C | V | R_{3}C, R_{4}C \rangle Q_{R_{1}}^{\dagger} Q_{R_{2}}^{\dagger} Q_{R_{3}} Q_{R_{4}}$$
$$= \frac{1}{2} \sum_{\substack{R_{1},R_{4}}} Q_{R_{1}}^{\dagger} Q_{R_{2}}^{\dagger} Q_{R_{3}} Q_{R_{4}} \iint d^{3}x d^{3}y Q_{R_{1}C}^{\ast} (\underline{x}) Q_{R_{2}C}^{\ast} (\underline{y}) V (|\underline{x} - \underline{y}|) Q_{R_{3}C} (\underline{y})$$
$$\times Q_{R_{4}C} (\underline{x})$$

$$= \frac{1}{2} \sum_{R_1...R_4}^{+} \alpha_{R_1}^{+} \alpha_{R_2}^{+} \alpha_{R_3}^{+} \alpha_{R_4}^{-} I \qquad (6.12)$$

Evaluating the integral I of Eq. (6.12),

$$I = \frac{1}{N^2} \iint d^3 x \, d^3 y \, e^{-i\frac{\kappa}{R} \cdot x} \, U^*_{\kappa,c} (\underline{x}) \, e^{-i\frac{\kappa}{R^2} \cdot y} \, U^*_{\kappa_{2c}} (\underline{y}) \, V(\underline{x} - \underline{y}) \, e^{i\frac{\kappa}{R^3} \cdot y} \, U_{\kappa_{3c}}(\underline{y}) \\ \times e^{i\frac{\kappa}{R^4} \cdot x} \, U_{\kappa_{4c}}(\underline{x}) \\ \text{where we have used the explicit form of the eigenfunctions } \mathcal{Q}_{\kappa,j} \text{ as} \\ \text{defined in Eq. (6.9).}$$

We assume that the exponential factors and the potential  $V(\underline{x},\underline{y})$  are slowly varying functions of X and Y over one lattice cell. The integral I can then be decomposed into summations over the unit cells,

$$I \Rightarrow \frac{1}{N^2} \mathcal{E}_{1,m} \exp \left[ i (\underline{R}_4 - \underline{R}_1) \cdot \underline{I} + i (\underline{R}_3 - \underline{R}_2) \cdot \underline{m} \right] \vee (\underline{I}, \underline{m}) \iint_{V_0} U_{R_1c}(\underline{x}) \\ \times U_{R_2c}^* (\underline{y}) U_{R_3c}(\underline{y}) U_{R_4c}(\underline{x}) d^3 x d^3 y \xrightarrow{(hatticecen)}$$

where the volume of the unit cell is  $v_{\bullet}$  and separate cells have midpoint vectors  $\underline{\textit{l}}$  and  $\underline{m}$  .

We next expand the Bloch functions with respect to  $\mathcal{R}$  and, assuming only small values of  $\mathcal{R}$  are important, retain only lowest order terms. Also, we note the orthogonality of the Bloch functions:

$$\int_{v_0} d^3 x \ U_{R^{\approx 0,c}}^* (\underline{x}) \ U_{R^{\approx 0,c}} (\underline{x}) = 1$$

$$\int_{v_0} d^3 x \ U_{R^{\approx 0,c}}^* (\underline{x}) \ U_{R^{\approx 0,v}} (\underline{x}) = 0$$
(6.13)

(the Bloch functions are normalised to one within a lattice cell).

Using Eq. (6.13), the integral I becomes,  

$$I = \frac{1}{N^2} \underbrace{\&lm} exp[i(\underline{R}_4 - \underline{R}_1).\underline{l} + i(\underline{R}_3 - \underline{R}_2).\underline{m}] V(\underline{l},\underline{m}) \quad (6.14)$$

As  $V(\underline{l},\underline{m})$  and the exponential functions vary slowly (and may be set constant over a lattice cell) we may replace the sum over lattice cells in Eq. (6.14) to an integral over the crystal:

$$\frac{1}{N^2} \mathcal{L}_{\ell} \rightarrow \frac{1}{N^2} \iint d^3 x' d^3 y'$$

(the factor  $\frac{1}{N^2}$  multiplying the summation is replaced by a corresponding factor of  $\frac{1}{V^2}$  outside the integral as there are N lattice cells in the system volume V.)

Thus changing to the centre of mass coordinate,

$$\frac{R}{R} = \frac{x' + y'}{2}$$

and a relative coordinate,  $\underline{r} = \underline{x}' - \underline{y}'$  the integral I becomes

$$I = \sqrt[1]{2} \iint d\underline{R} d\underline{r} \exp [i\underline{R}.(\underline{R}_{4} - \underline{k}_{1} + \underline{k}_{3} - \underline{k}_{2}) + i\underline{\underline{r}}.(\underline{k}_{4} - \underline{k}_{1} + \underline{R}_{2} - \underline{R}_{3})] \\ \times V(\underline{r})$$

[as the Jacobian of the pair  $(\underline{\beta}, \underline{r})$  with respect to  $\underline{X}', \underline{y}'$  is 1].

Integrating over 
$$R$$
 we find,  
 $I = \frac{1}{\sqrt{\delta_{R_4} + \kappa_3}}, \kappa_1 + \kappa_2 \int dr e^{-ir(r_1 - \kappa_4)} V(r)$ 

 $\Rightarrow I = \delta_{\underline{R}_4 + \underline{R}_3, \underline{R}_1 + \underline{R}_2} \quad \mathcal{V}(q) \qquad -----(6.15)$ where  $q = |\underline{R}_1 - \underline{R}_4|$  and  $\mathcal{V}(q) = \frac{1}{\sqrt{V(q)}} \quad V(q)$  is the Fourier transform of  $V(\underline{r})$ .

Thus equations (6.12) and (6.15) yield,  

$$\frac{1}{2} \sum_{\substack{R_{1}...R_{4}}} \langle R_{1c}, R_{2c} | V | R_{3c}, R_{4c} \rangle \Delta_{R_{1}}^{\dagger} \Delta_{R_{2}} \Delta_{R_{3}} \Delta_{R_{4}}$$

$$= \frac{1}{2} \sum_{\substack{R_{1}..R_{2},q}} \bigcup(q) \Delta_{R_{1}+q}^{\dagger} \Delta_{R_{2}-q} \Delta_{R_{2}} \Delta_{R_{1}} \qquad ----(6.16)$$

In a similar manner we evaluate the remaining interaction terms of Eq. (6.11): 2)  $\frac{1}{2} \oint_{\mathbf{R}_{1} \cdot \cdot \mathbf{R}_{4}} \langle -\mathbf{R}_{1} \cdot \mathbf{\sigma}, -\mathbf{R}_{2} \cdot \mathbf{\sigma} | \mathbf{V} | -\mathbf{R}_{3} \cdot \mathbf{\sigma}, -\mathbf{R}_{4} \cdot \mathbf{\sigma} \rangle d_{\mathbf{R}_{1}}^{\dagger} d_{\mathbf{R}_{2}}^{\dagger} d_{\mathbf{R}_{3}} d_{\mathbf{R}_{4}}$  $= \frac{1}{2} \oint_{\mathbf{R}_{1} \cdot \cdot \mathbf{R}_{4}} \mathcal{U}(\mathbf{q}) d_{\mathbf{R}_{1}}^{\dagger} d_{\mathbf{R}_{2}}^{\dagger} d_{\mathbf{R}_{2}}^{\dagger} -\mathbf{q} d_{\mathbf{R}_{1}}^{\dagger} +\mathbf{q} \qquad -----(6.17)$ 

(prime indicates valence band momenta)

and finally, we consider

The second matrix element in Eq. (6.18) vanishes as it generates an average over Bloch functions of the following form:

$$\iint d^3 \times d^3 y \ u_{\bullet,c}^*(\underline{x}) \ u_{\bullet,v}^*(\underline{y}) \ u_{\bullet,c}(\underline{y}) \ u_{\bullet,v}(\underline{x})$$

$$\Rightarrow \int u_{\bullet,c}^*(\underline{x}) u_{\bullet,v}(\underline{x}) d^3 \times \int u_{\bullet,v}^*(\underline{y}) \ u_{\bullet,c}(\underline{y}) \ d^3 y = \mathbf{0} \quad (\text{from } (6.13))$$

Thus expression (6.18) becomes

Hamiltonian terms, we find the Hamiltonian for the semiconductor system alone:

$$H_{s.c.} = \mathcal{E}_{Rc} E_{c}(Rc) a_{Rc}^{\dagger} a_{Rc} + \mathcal{E}_{Rv} E_{v}(Rv) d_{Rv}^{\dagger} d_{Rv}$$

$$+ \frac{1}{2} \mathcal{E}_{q} v(q) \left[ \mathcal{E}_{Rc,Rc} a_{Rc+q}^{\dagger} a_{Rc-q}^{\dagger} a_{Rc} a_{Rc} + \mathcal{E}_{Rv,Rc} d_{Rv}^{\dagger} d_{Rv}^{\dagger} d_{Rv-q}^{\dagger} d_{Rv-q} d_{Rv+q} - 2 \mathcal{E}_{Rc,Rc} a_{Rc+q}^{\dagger} d_{Rv}^{\dagger} d_{Rv-q} a_{Rc} \right] \qquad (6.20)$$

where  $R_c$  and  $K_{\sigma}$  are wavevectors of conduction and valence band states respectively.

The first two terms of Eq. (6.20) represent the kinetic energy of the electrons in the conduction band and the holes in the valence band. The third term describes interactions between particles in the semiconductor: the first term in the square brackets refers to interactions between electrons in the conduction band; the second term to interactions between holes in the valence band; and the third term describes interactions between electrons and holes.

# ∮6.2(b) Semiconductor-Light Field Interaction

The Hamiltonian for this process is given by Eq. (6.3). Substituting the explicit form of the vector potential A (Eq. (1.3),  $\oint$ 1.3(b)(iii)) we find in the rotating wave approximation, (Haken, 1976)

$$H_{INT} = H_{matter-light} = f \sum_{\mu,\nu} (a_{\mu}^{\dagger} d_{\nu}^{\dagger} b g_{\mu\nu} + h.c.) \qquad (6.21a)$$
  
(b = boson operator for cavity mode)

Again we have assumed the single mode approximation (driving field is strongly coupled to one cavity mode only); and

$$g_{\mu\nu} = \sqrt{\frac{2\pi e}{v\omega \hbar m}} \int \varphi_{\mu}^{*}(\underline{x}) \hat{\underline{e}} e^{i\underline{k}\cdot\underline{x}} p (\varphi(\underline{x}) d^{3}x \qquad (6.21b)$$

(cavity mode has frequency  $\omega$  , wavevector  $\underline{k}$  ).

Thus our total lossless system is described by the Hamiltonian,

 $H = H_{s.c.} + H_{INT} + H_{e.m.} + H_{D}$  (6.22) where the terms of Eq. (6.22) are given by Eq. (6.20), (6.21), (6.6a) and (6.6b).

Our aim is to develop a theory of optical bistability in semiconductors arising from excitonic interaction.

However, the Hamiltonian, Eq. (6.22), describes the behaviour of electrons and holes in the semiconductor and does not refer explicitly to the excitons comprising the medium. Thus it is necessary to transform our Hamiltonian to one characterised by excitonic operators.

This is most easily accomplished by performing a bosonisation transformation on the fermion operators and re-expressing the Hamiltonian in terms of exciton operators. Once a suitable definition of exciton operators is achieved, we can include an exciton damping mechanism into our model in a systematic fashion.
## $\phi$ 6.3 Boson Hamiltonian

### $\oint 6.3(a)$ Semiconductor Hamiltonian

Following the theory discussed in Chapter Five, we apply the bosonising transformation on the fermion Hamiltonian  $H_{5,c}$  as follows,

$$H_{s.c.} \xrightarrow{\Rightarrow} U H_{s.c.} U^{\dagger} = \widetilde{H}_{s.c.} \widehat{P} = \left[\widetilde{H}_{o} + \widetilde{H}_{e-e} + \widetilde{H}_{h-h} + \widetilde{H}_{e-h}\right] \widehat{P} - (6.23)$$
  
where  $U$  and  $\widehat{P}$  are the transformation and projection operators  
defined in Chapter Five (see Table 5.1).

The terms of Eq. (6.23) are,

1) 
$$\widetilde{H}_{o}\widehat{P} = \bigcup \left[ \widehat{\mathcal{E}}_{Rc} \operatorname{E}_{c}(Rc) a_{Rc}^{\dagger} a_{Rc} + \widehat{\mathcal{E}}_{Rv} \operatorname{E}_{v}(R) a_{Rv}^{\dagger} d_{Rv} \right] U^{\dagger}$$
  
=  $\widehat{\mathcal{E}}_{Rc,Rv} \left[ \operatorname{E}_{c}(Rc) + \operatorname{E}_{v}(Rv) \right] b_{Rc,Rv}^{\dagger} b_{Rc,Rv} \widehat{P} \qquad (6.24)$ 

where the  $b_{\mathbf{k},\mathbf{k},\mathbf{v}}$  are boson operators.

<sup>2)</sup> 
$$\widetilde{H}_{e-e} \widehat{P} = U \frac{1}{2} \left[ \sum_{\substack{\mathbf{k},\mathbf{k}'_{e} \\ \mathbf{q}}} \nabla(q) a_{\mathbf{k}e+q}^{\dagger} a_{\mathbf{k}'-q}^{\dagger} a_{\mathbf{k}'_{e}} a_{\mathbf{k}'_{e}} \right] U^{\dagger}$$
 (6.25)

Certain ambiguities arise in the transformation described by Eq. (6.25) because the bosonising operator  $\bigcup$  transforms pairs of fermion operators only. As the four fermion operators in Eq. (6.25) can be arranged pair-wise in two distinct ways (with a corresponding sign change) we must consider the transformation of the two possible arrangement of operators:

$$\Rightarrow Equ. 6.25 \Rightarrow U \frac{1}{2} \left[ \begin{array}{c} \mathcal{E} \\ \mathcal{R}_{\epsilon}, \mathcal{R}_{\epsilon} \\ q \end{array} \right] = \left[ \begin{array}{c} a_{\mathbf{R}_{\epsilon}}^{\dagger} + q \\ a_{\mathbf{R}_{\epsilon}^{\dagger} + q \\ a_{\mathbf{R}_{\epsilon}^{$$

$$=\frac{1}{4} \sum_{\substack{\mathbf{R}c,\mathbf{R}c\\\mathbf{q}}} \upsilon(q) \left( U \left\{ -a_{\mathbf{R}c+\mathbf{q}}^{\dagger}a_{\mathbf{R}c}a_{\mathbf{R}c}^{\dagger}-q a_{\mathbf{R}c} + \delta_{\mathbf{R}c-\mathbf{q}},\mathbf{R}c} a_{\mathbf{R}c+\mathbf{q}}^{\dagger}a_{\mathbf{R}c} \right\} U^{\dagger} \right)$$

$$=\frac{1}{4} \sum_{\substack{\mathbf{R}c,\mathbf{R}c\\\mathbf{q}}} \upsilon(q) \left( U \left\{ -a_{\mathbf{R}c+\mathbf{q}}^{\dagger}a_{\mathbf{R}c}a_{\mathbf{R}c}^{\dagger}-q a_{\mathbf{R}c} + \delta_{\mathbf{R}c+\mathbf{q}}a_{\mathbf{R}c} + \delta_{\mathbf{R}c+\mathbf{q}}a_{\mathbf{R}c} + \delta_{\mathbf{R}c+\mathbf{q}}a_{\mathbf{R}c} + \delta_{\mathbf{R}c+\mathbf{q}}a_{\mathbf{R}c}a_{\mathbf{R}c}^{\dagger}-q a_{\mathbf{R}c} \right\} U^{\dagger} \right)$$

$$= \frac{1}{4} \underbrace{\mathcal{E}}_{\substack{\text{Rc,Rc}\\ q}} \underbrace{\upsilon(q)} \left( -U a_{\text{Rc+q}}^{\dagger} a_{\text{Rc}} U^{\dagger} U a_{\text{Rc-q}}^{\dagger} a_{\text{Rc}} U^{\dagger} + U \delta_{\text{Rc-q},\text{Rc}} a_{\text{Rc+q}}^{\dagger} a_{\text{Rc}} U^{\dagger} + U a_{\text{Rc-q}}^{\dagger} a_{\text{Rc}} U^{\dagger} U a_{\text{Rc+q},\text{Rc}}^{\dagger} a_{\text{Rc}} U^{\dagger} \\ -U \delta_{\text{Rc+q},\text{Rc}} a_{\text{Rc}}^{\dagger} u^{\dagger} \right)$$

as  $U^{\dagger}U = 1$  in fermion space (see  $\oint 5.3(e)$ ).

$$\Rightarrow \widetilde{H}_{e-e} \widehat{P} = \frac{1}{4} \underbrace{\sum_{\substack{Re,R',q\\ Rv,R'r}} \overline{U(q)} \left[ \begin{array}{c} b_{R'e-q} R'r & b_{Re+q} Rv & b_{R'e} R'r & b_{ReR'r} \\ & & & & \\ & & & & \\ & & & \\ & & &$$

Similarly,

4) 
$$\widetilde{H}_{e-n} \widehat{P} = - \bigcup_{\substack{\mathbf{k}c, \mathbf{k}' \\ q}} \nabla(q) a_{\mathbf{k}c+q}^{\dagger} d_{\mathbf{k}'} d_{\mathbf{k}'-q} a_{\mathbf{k}c} U^{\dagger}$$
  
=  $-\frac{1}{2} \bigcup_{\substack{\mathbf{k}c, \mathbf{k}' \\ q}} \nabla(q) [a_{\mathbf{k}c+q}^{\dagger} d_{\mathbf{k}'} d_{\mathbf{k}'-q} a_{\mathbf{k}c} + a_{\mathbf{k}c+q}^{\dagger} a_{\mathbf{k}c} d_{\mathbf{k}'} d_{\mathbf{k}'-q}] U^{\dagger} = (6.28)$ 

The first term of expression (6.28) yields

$$=\frac{1}{2} \sum_{\substack{\mathbf{R}c,\mathbf{R}'r\\\mathbf{q}}} \mathcal{V}(\mathbf{q}) \bigcup a_{\mathbf{R}c}^{\dagger} + q d_{\mathbf{R}'r}^{\dagger} \bigcup^{\dagger} \bigcup d_{\mathbf{R}'r-q} a_{\mathbf{R}c} \bigcup^{\dagger}$$

$$=\frac{1}{2} \sum_{\substack{\mathbf{R}c,\mathbf{R}'r\\\mathbf{q}}} \mathcal{V}(\mathbf{q}) B_{\mathbf{R}c}^{\dagger} + q \mathbf{R}'r = \frac{1}{\sqrt{1+\widehat{N}}} \widehat{P} = \frac{1}{\sqrt{1+\widehat{N}}} B_{\mathbf{R}c} \mathbf{R}'r-q \widehat{P} \qquad (\text{see Table 5.1})$$

$$= - \underbrace{\underset{\substack{R_{c}, R_{v}'}}{\widehat{P}}}_{R_{c}+q} \underbrace{\stackrel{R_{v}'}{p}}_{R_{c}+q} \underbrace{\stackrel{R_{v}'}{n}}_{\sqrt{1+\widehat{N}}} \underbrace{\frac{1}{\sqrt{1+\widehat{N}}}}_{R_{c}-R_{v}-q} \underbrace{\stackrel{\widehat{P}}{P}}_{R_{c}-R_{v}-q} \underbrace{\stackrel{\widehat{P}}{P}}_{(6.29)} \underbrace{\stackrel{\widehat{P}}{[\widehat{N} = \underset{\substack{E, E_{v} \\ E_{v} \\ E_{v}}}_{(6.29)}}_{(6.29)}$$
as  $\widehat{P} = 1$  in the boson subspace (see  $\oint 5.3(e)$ ) we ignore the  
presence of  $\widehat{P}$  on the left - hand side of Eq. (6.29).

$$\Rightarrow Equ. 6.29 \Rightarrow -\frac{1}{2} \sum_{\substack{Re,Rv,q\\ Rc,Rv}} v(q) \left[ b_{Rc+q} R_{v+q} b_{RcRv} \right]_{RcRv} \left[ b_{RcRv} b_{RcRv} \right] \widehat{P} - (6.30)$$

The second term of expression (6.28) is,

$$=\frac{1}{2} \sum_{\substack{Rc,R'v\\q}} \mathcal{V}(q) \bigcup a_{Rc+q}^{\dagger} a_{Rc} \bigcup^{\dagger} \bigcup a_{R'v}^{\dagger} d_{R'v-q} \bigcup^{\dagger}$$

$$= -\frac{1}{2} \sum_{\substack{Rc,R'v\\q}} \mathcal{V}(q) \begin{bmatrix} b_{Rc+qRv} & b_{R'cR'v+q} & b_{RcRv} & b_{R'cR'v} + b_{Rc+qRv+q} & b_{RcRv} \end{bmatrix} (6.31)$$

Thus, from Eqs. (6.24), (6.26), (6.27), (6.30) and (6.31) we find the boson Hamiltonian,

where

 $\widetilde{H}_{o} = \underbrace{\mathcal{E}}_{\mathsf{R}_{v},\mathsf{R}_{c}} \left[ \mathsf{E}_{c}(\mathsf{R}_{c}) + \mathsf{E}_{v}(\mathsf{R}_{v}) \right] \overset{\dagger}{\mathsf{b}}_{\mathsf{R}_{c},\mathsf{R}_{v}} \overset{\dagger}{\mathsf{b}}_{\mathsf{R}_{c},\mathsf{R}_{v}} - \underbrace{\mathcal{E}}_{\mathsf{R}_{c},\mathsf{R}_{v}} \upsilon(q) \overset{\dagger}{\mathsf{b}}_{\mathsf{R}_{c}+q} \overset{\dagger}{\mathsf{R}_{v}+q} \overset{\dagger}{\mathsf{b}}_{\mathsf{R}_{c},\mathsf{R}_{v}} \overset{\dagger}{\mathsf{b}}_{\mathsf{R}_{c},\mathsf{R}_{v}} \overset{\dagger}{\mathsf{b}}_{\mathsf{R}_{c}} \overset{\dagger}{\mathsf{R}_{v}} \overset{\dagger}{\mathsf{b}}_{\mathsf{R}_{c},\mathsf{R}_{v}} \overset{\dagger}{\mathsf{b}}_{\mathsf{R}_{c},\mathsf{R}_{v}}} \overset{\dagger}{\mathsf{b}}_{\mathsf{R}_{c},\mathsf{R}_{v}} \overset{\dagger}{\mathsf{b}}_{\mathsf{R},\mathsf{R}_{v},\mathsf{R}_{v}} \overset{\dagger}{\mathsf{b}}_{\mathsf{R},\mathsf{R}_{v}} \overset{\dagger}{\mathsf{b}}_{\mathsf{R$ 

$$\widetilde{H}_{1} = \frac{1}{4} \underbrace{\mathcal{E}}_{\substack{\mathbf{R}v, \mathbf{R}c, \mathbf{q}\\\mathbf{R}v, \mathbf{R}c}, \mathbf{q}} \underbrace{\nabla(\mathbf{q}) \left[ \begin{array}{c} \mathbf{b}_{\mathbf{R}c-\mathbf{q}} & \mathbf{R}v & \mathbf{b}_{\mathbf{R}c+\mathbf{q}} & \mathbf{R}v & \mathbf{b}_{\mathbf{R}c} & \mathbf{R}v + \mathbf{b}_{\mathbf{R}c\mathbf{R}v+\mathbf{q}} & \mathbf{b}_{\mathbf{R}c} & \mathbf{R}v - \mathbf{q} \\ & \mathbf{R}v, \mathbf{R}c & -2 & \mathbf{b}_{\mathbf{R}c+\mathbf{q}} & \mathbf{R}v & \mathbf{b}_{\mathbf{R}c} & \mathbf{R}v + \mathbf{q} \right] & \mathbf{b}_{\mathbf{R}c} & \mathbf{R}v & \mathbf{b}_{\mathbf{R}c} & \mathbf{R}v \\ \widetilde{H}_{2} = -\frac{1}{4} \underbrace{\mathcal{E}}_{\substack{\mathbf{R}v, \mathbf{R}c \\ \mathbf{R}v, \mathbf{R}c}} \underbrace{\nabla(\mathbf{q}) \left[ \begin{array}{c} \mathbf{b}_{\mathbf{R}c+\mathbf{q}} & \mathbf{R}v & \mathbf{b}_{\mathbf{R}c-\mathbf{q}} & \mathbf{R}v \\ \mathbf{R}v, \mathbf{R}c \\ \mathbf{R}v, \mathbf{R}v, \mathbf{R}c \\ \mathbf{R}v, \mathbf{R}v, \mathbf{R}c \\ \mathbf{R}v, \mathbf{R}v, \mathbf{R}c \\ \mathbf{R}v, \mathbf{R}v, \mathbf{R}c \\ \mathbf{R}v, \mathbf{R}v,$$

boson Hamiltonian. Hanamura solved this problem by using a complicated ordering procedure.

We avoid these difficulties by using the transformation U ,

but unfortunately must cope with infinite boson expansions, as discussed in the next section.

#### $\phi$ 6.3(b) Semiconductor-Light Interaction

We wish now to transform the interaction Hamiltonian

$$H_{INT} = \hbar \sum_{\mu,\nu} a_{\mu}^{\dagger} d_{\nu}^{\dagger} b g + h.c$$

However, we must deal with the infinite boson expansion generated by the expression,

$$U a_{\mu}^{+} d_{\nu}^{+} U^{+} = \hat{P} b_{\mu\nu}^{+} (1+\hat{N})^{\nu_{2}} - ----(6.33)$$

Using the generalised operator expansion Eq. (5.63), we find that Eq. (6.33) is a boson series in which the terms increase in powers of  $b_{ij}^{+}b_{ij}$  (exciton density). Thus, for low density systems, higher order terms will become negligible and we may expand Eq. (6.33) to second order only,

$$\Rightarrow \bigcup a_{\mu} d_{\nu}^{\dagger} \bigcup^{\dagger} \approx (\dot{b}_{\mu\nu} - (1 - \sqrt{2}) \dot{b}_{\mu\nu} \mathcal{E}_{ij} \dot{b}_{\mu} \mathcal{E}_{ij} \dot{b}_{\mu} \mathcal{E}_{ij}) \widehat{P} \qquad ---(6.34)$$
Thus, using Eq. (6.34) we find,

$$U H_{INT} U^{\dagger} = \tilde{H}_{INT} \tilde{P} = \hbar \{ g b \mathcal{E}_{ReRV} ( \tilde{b}_{ReRV} - (1 - \sqrt{2}) \mathcal{E}_{R'e,R'v} ( \tilde{b}_{R'e,R'v} - (1 - \sqrt{2}) \mathcal{E}_{R'e,R'v} (\tilde{b}_{R'e,R'v} - \tilde{b}_{R'e,R'v} + h.c. \}$$
(6.35)

We stress that Eq. (6.35) applies only to low density systems.

#### $\phi$ 6.3(c) Low Exciton-Density Hamiltonian

We wish now to transform Eqs. (6.32) and (6.35) to suitably define exciton operators as bound electron-hole pairs. Thus we need to transform the Hamiltonian in such a way as to diagonalise the harmonic term  $\widetilde{H}_{\bullet}$ . This is achieved by using a transformation:

$$b_{RcRv} = \mathcal{E}_{\nu,\kappa} \sqrt{\sqrt{\nu}} \quad \delta_{\kappa,\kappa-\kappa v} \quad f_{\nu} (\alpha \kappa c + \beta \kappa v) C_{\nu,\kappa} \quad ----(6.36)$$

(notation as in Hanamura, 1974a)

(V = system volume)

where  $\int_{\nu} (dR_c + \beta R_{\nu})$  is the Fourier transform of the  $\nu$ -th state of a hydrogen-like atom; d = Mh/(me+mh) and  $\beta = 1 - d$ . Thus,  $C_{\nu,K}$  defines the exciton (boson) destruction operator.

The harmonic term  $\widetilde{H}_{\bullet}$  then assumes the desired form,  $\widetilde{H}_{\bullet}' = \mathcal{E}_{\nu,\kappa} \left[ E_g - \mathcal{E}_{b,n} + \frac{\hbar^2 \kappa^2}{2 M} \right] C_{\nu,\kappa}^+ C_{\nu,\kappa} \equiv \mathcal{E}_{\nu,\kappa} \Omega_{\nu,\kappa} C_{\nu,\kappa}^+ C_{\nu,\kappa}$ where K = centre of mass momentum;  $\mathcal{E}_{b,n}$  = binding energy of the nth excitonic state and M = Me + mh.

Applying the transformation (6.36) to the total Hamiltonian we find

$$\widetilde{H} = \widetilde{H}_{0}' + \widetilde{H}_{ex} + \widetilde{H}_{iNT} + \widetilde{H}_{D}$$
 (6.37)

[As  $\hat{p} = 1$  in the boson subspace, we ignore its explicit presence in further discussion].

where

$$\begin{split} \widetilde{H}'_{o} &= \hat{\mathcal{E}}_{v,\kappa} \ \Omega_{v,\kappa} \ C_{v,\kappa}^{+} C_{v,\kappa} + \omega b^{+} b \\ \widetilde{H}'_{ex} &= \hat{\mathcal{E}}_{M,\kappa',\kappa,q} \\ \gamma',v,\mu' &= \hat{\mathcal{E}}_{\kappav,\kappa'\sigma,\kappa,\kappa'} M_{2} \ C_{\mu',\kappa'+\kappa\sigma-\kappa'}^{+} q \ C_{\mu',\kappa'+\kappa'\sigma-\kappa-q}^{+} C_{v,\kappa} C_{v',\kappa'} \\ \gamma_{v',\mu,\mu',q}^{+} &= \hbar \left\{ g b \left( r_{1} \ C_{\kappa}^{+} - r_{2} \ C_{\kappa}^{+} C_{\kappa}^{+} C_{\kappa} \right) + h.c. \right\} \end{split}$$

$$\widetilde{H}_{D}' = i\hbar (Eexp(-i\omega_{L}t)b^{\dagger} - h.c.)$$

where

$$\begin{split} \mathsf{M}_{1} &= \frac{1}{2^{N^{2}}} \, \mathcal{V}(q_{*}) \left\{ f_{\mu}^{*} \left( \alpha \left( k_{v} + k_{*} + q \right) + \beta k_{v} \right) f_{\mu'}^{*} \left( \alpha \left( k' + k_{v}' - q \right) + \beta k_{v}' \right) \right. \\ &+ f_{\mu}^{*} \left( \alpha \left( k' + k_{v} \right) + \beta \left( k_{v} + q \right) \right) f_{\mu'}^{*} \left( \alpha \left( k + k_{v}' \right) + \beta \left( k_{v}' - q \right) \right) \right. \\ &- 2 f_{\mu}^{*} \left( \alpha \left( k + k_{v} + q \right) + \beta k_{v} \right) f_{\mu'}^{*} \left( \alpha \left( k' + k_{v}' \right) + \beta \left( k_{v}' + q \right) \right) \right\} \\ &\times f_{v} \left( \alpha \left( k + k_{v} \right) + \beta k_{v} \right) f_{\gamma'} \left( \alpha \left( k' + k_{v}' \right) + \beta k_{v}' \right) \right. \\ \\ \mathsf{M}_{2} &= \frac{1}{2^{N^{2}}} \, \mathcal{V}(q) \left\{ f_{\mu}^{*} \left( \alpha \left( k + k_{v} + q \right) + \beta k_{v}' \right) f_{\mu'}^{*} \left( \alpha \left( k' + k_{v}' - q \right) + \beta k_{v} \right) \right. \\ &+ f_{\mu'}^{*} \left( \alpha \left( k + k_{v} \right) + \beta \left( k_{v}' - q \right) \right) f_{\mu'}^{*} \left( \alpha \left( k' + k_{v}' \right) + \beta \left( k_{v}' + q \right) \right) \\ &+ f_{\mu'}^{*} \left( \alpha \left( k + k_{v} \right) + \beta \left( k_{v}' - q \right) \right) f_{\mu'}^{*} \left( \alpha \left( k' + k_{v}' \right) + \beta \left( k_{v}' + q \right) \right) \\ &- 2 f_{\mu'}^{*} \left( \alpha \left( k + k_{v} \right) + \beta \left( k_{v}' - q \right) \right) f_{\mu'}^{*} \left( \alpha \left( k' + k_{v}' \right) + \beta \left( k_{v}' + q \right) \right) \\ &+ f_{\mu'}^{*} \left( \alpha \left( k + k_{v} \right) + \beta \left( k_{v}' - q \right) \right) f_{\mu'}^{*} \left( \alpha \left( k' + k_{v}' \right) + \beta \left( k_{v}' + q \right) \right) \\ &+ f_{\mu'}^{*} \left( \alpha \left( k + k_{v} \right) + \beta \left( k_{v}' - q \right) \right) f_{\mu'}^{*} \left( \alpha \left( k' + k_{v}' \right) + \beta \left( k_{v}' + q \right) \right) \\ &+ f_{\mu'}^{*} \left( \alpha \left( k + k_{v} \right) + \beta \left( k_{v}' - q \right) \right) f_{\mu'}^{*} \left( \alpha \left( k' + k_{v}' \right) + \beta \left( k_{v}' + q \right) \right) \\ &+ f_{\mu'}^{*} \left( \alpha \left( k + k_{v} \right) + \beta \left( k_{v}' - q \right) \right) f_{\mu'}^{*} \left( \alpha \left( k' + k_{v}' \right) + \beta \left( k_{v}' + q \right) \right) \\ &+ f_{\mu'}^{*} \left( \alpha \left( k + k_{v} \right) + \beta \left( k_{v}' - q \right) \right) f_{\mu'}^{*} \left( \alpha \left( k' + k_{v}' \right) + \beta \left( k_{v}' - q \right) \right) \\ &+ f_{\mu'}^{*} \left( \alpha \left( k + k_{v} \right) + \beta \left( k_{v}' - q \right) \right) f_{\mu'}^{*} \left( k' \left( k' + k_{v}' \right) + \beta \left( k_{v}' - q \right) \right) \\ &+ f_{\mu'}^{*} \left( k_{v}' + k_{v}' + \beta \left( k_{v}' - q \right) \right) f_{\mu'}^{*} \left( k_{v}' + k_{v}' + \beta \left( k_{v}' + k_{v}' \right) \right) \\ &+ f_{\mu'}^{*} \left( k_{v}' + k_{v}' + \beta \left( k_{v}' - q \right) \right) f_{\mu'}^{*} \left( k_{v}' + k_{v}' + \beta \left( k_{v}' + k_{v}' \right) \right) \\ &+ f_{v}^{*} \left( k_{v}' + k_{v}' + \beta \left( k_{v}' + k_{v}' + k_{v}' \right) \right)$$

$$-2f_{\mu}^{*}(\alpha(\kappa + \kappa v + q) + \beta \kappa v)f_{\mu'}(\alpha(\kappa' + \kappa v - q) + \beta \kappa v)$$

$$\times f_{\nu}(\alpha(\kappa + \kappa v) + \beta \kappa v)f_{\nu'}(\alpha(\kappa' + \kappa v) + \beta \kappa v)$$

(summation is implied over all indices in  $M_{1}$  and  $M_{2}$  ).

and

$$r_{1} = \sqrt{2} \quad \sum_{kc} f(a kc + \beta (kc - \kappa))$$

$$r_{2} = \frac{1 - \sqrt{2}}{\sqrt{\sqrt{2}}} \quad \sum_{kc} |f(a kc + \beta (kc - \kappa))|^{2} f^{*}(a kc + \beta (kc - \kappa))$$

In the derivation of  $\widetilde{H}'_{iNT}$  in Eq. (6.37) we assumed the cavity mode (governed by the boson operator **b** ) was strongly coupled to a particular exciton mode, of momentum K . Coupling to other exciton modes was assumed to be weak enough to ignore their contribution to the interaction. Also, the summation over  $\gamma$  was dropped, indicating that we only consider one level of the exciton (ground state).

Having successfully formulated the Hamiltonian in terms of the exciton operators  $C_{\kappa}$ ,  $C_{\kappa}^{+}$  we can now introduce damping of excitons into the system.

We firstly note that the terms of  $\widetilde{H}'_{ex}$  describe interactions between all exciton modes. These terms thus give rise to damping which arises from collisions between different excitons.

Additional excitonic damping stems from the coupling of excitons to the crystal lattice. Adopting the usual methods of the quantum theory of damping, we assume the lattice vibrations, i.e. phonon modes, comprise a reservoir in thermal equilibrium which is weakly coupled to the exciton mode we are considering.

The Hamiltonian describing this process is,

A similar exciton-phonon model was considered by Toyozawa (1958, 1959). In his extensive papers, he derives the Hamiltonian:

Hex.-phonon =  $\sum_{i,j,j',l,l'} (b_{i,j-j'} + b_{i,j'-j}) C_{\nu,l}^{\dagger} C_{\nu',l'}$  -----(6.39) where  $b_{ij}^{\dagger}$  is a boson creation operator for the phonon mode; *i* and *j* refer to mode and wavenumber of the phonon.  $\begin{pmatrix} t \\ \nu, \ell \end{pmatrix}$  is the boson creation operator for an exciton;  $\nu$  is the internal quantum number,  $\ell$  is the wavenumber.

Hamiltonians (6.38) and (6.39) will be equivalent if we define the correspondence,

Thus, the work of Toyozawa shows that the reservoir operator of Eq. (6.38) should be viewed as a combination of exciton and phonon operators; and damping of a given exciton occurs via another exciton mode and not solely through the lattice.

Damping of the cavity mode is described by the Hamiltonian,

H cavity damp. = 
$$Q_F b^{\dagger} + h.c.$$
 (6.40)  
The reservoir (described by the operator  $Q_F$  ) could in this

case consist of modes of the radiation field; or the exciton modes we ignored in the derivation of  $\widetilde{H}'_{\rm INT}$ .

## $\phi$ 6.4 Master Equation

In the single mode approximation, the Hamiltonian becomes, (in a frame rotating at the frequency  $\omega_{\textbf{L}}$  )

$$\begin{split} \widetilde{H} &= \hbar \, \delta_1 \, C_k^{\dagger} \, C_k + \hbar \, \delta_2 \, \overline{b} \, b - M_2 \, C_k^{\dagger} \, C_k^{\dagger} \, C_{kCk} + \hbar \, g(r, \overline{b}^{\dagger} \, C_k \\ &- r_2 \, \overline{b}^{\dagger} \, C_k^{\dagger} \, C_{kCk} \, C_k ) + i \hbar \, E \, b + C_k^{\dagger} \, \underbrace{\mathcal{E}}_{k_1 k_2 k_3} \, \chi_1 \, C_{k_1}^{\dagger} \, C_{k_2} \, C_{k_3} \\ &+ \, C_k^{\dagger} \, \underbrace{\mathcal{E}}_{k_1 k_2} \, \chi_2 \, C_{k_1} \, C_{k_2} + C_k^{\dagger} \, C_k \, \underbrace{\mathcal{E}}_{k_1 k_2} \, \chi_3 \, C_{k_1}^{\dagger} \, C_{k_2} + b^{\dagger} \, \underbrace{\mathcal{E}}_{j} \, \chi_4 \, b_{j} \\ &+ \, C_k^{\dagger} \, \underbrace{\mathcal{E}}_{k_1 k_2} \, \chi_5 \, b_{k_1} \, C_{k_2} + h.c. \end{split}$$
where  $\delta_1$  and  $\delta_2$  are detuning terms;  $\delta_1 = \Omega - \omega_L$ ,  $\delta_2 = \omega - \omega_L$ 
 $\chi_1 = M_1 - M_2$ ;  $\chi_2 = M_1 \, (\kappa_1 = \kappa_2 - 2q) - M_2 \, (\kappa_1 = \kappa_2 + 2\kappa'_2 - 2\kappa_2 - 2q); \end{split}$ 

 $\chi_3 = M_1(K_1+q=K_2) - M_2(R_r - R_r + q=0) - M_2(K_2 = K_1 + R_r - R_r + q)$ 

and 
$$\chi_4$$
 = coupling between field mode and reservoir,  
 $\chi_5$  = coupling between exciton mode and exciton-phonon reservoir.

We assume it is possible to consider the exciton ground state only and thus ignore the summation over internal quantum numbers.

The last five terms of Eq. (6.41) describe coupling of cavity and exciton modes to reservoirs. The first three of these damping terms stem from applying the single mode approximation to  $\widetilde{H}'_{ex}$ .

We obtain the master equation for the system following the methods described in Chapter One,  $\oint 1.3(d)$ :

where

$$\frac{\partial \rho}{\partial t} = K_{1,a} (E_{c_{k}}\rho, C_{k}^{\dagger}] + [C_{k}^{\dagger}, \rho C_{k}^{\dagger}]) + K_{1,b} ([C_{k}^{\dagger}\rho, c_{k}]]$$
  

$$\frac{\partial \rho}{\partial t} = K_{1,a} (E_{c_{k}}\rho, C_{k}^{\dagger}) + K_{2,a} (E_{c_{k}}C_{k}\rho, C_{k}^{\dagger}C_{k}^{\dagger}]$$
  

$$+ [C_{k}^{\dagger}, \rho C_{k}]) + K_{2,b} (E_{c_{k}}C_{k}\rho, C_{k}C_{k}]$$
  

$$+ [C_{k}^{\dagger}C_{k}\rho, C_{k}C_{k}]) + K_{3} (E_{k}^{\dagger}C_{k}\rho, C_{k}^{\dagger}C_{k}] + [C_{k}^{\dagger}C_{k}\rho, C_{k}C_{k}]) \quad (6.43)$$

where 
$$K_{1,a} = \pi g(\omega_0) |\chi_1(\omega_0)|^2 \bar{n}_{1,a} (1+\bar{n}_{1,b})(1+\bar{n}_{1,c})$$
  
 $g(\omega_0) = \text{density of states of reservoir modes},$   
 $\omega_0 = \text{exciton resonance frequency},$   
 $\bar{n}_{1,a}; \bar{n}_{1,b}; \bar{n}_{1,c} = \text{thermal population of reservoir mode}$   
and  $K_{1,b} = \pi g(\omega_0) |\chi_1(\omega_0)|^2 \bar{n}_{1,b} \bar{n}_{1,c} ; K_{2,b} = \pi g(\omega_0) |\chi_2|^2 \bar{n}_{2,a} \bar{n}_{2,b};$   
 $K_{2,a} = \pi g(\omega_0) |\chi_1(\omega_0)|^2 (1+\bar{n}_{2,a})(1+\bar{n}_{2,b});$   
 $K_3 = \pi g(\omega_0) |\chi_3|^2 (\bar{n}_{3,a} + \bar{n}_{3,b} + 2\bar{n}_{3,a} \bar{n}_{3,b}).$ 

Thus, in Eq. (6.43), the constants  $K_1$ ,  $K_2$  and  $K_3$  describe the strength of exciton-exciton interactions (collisions). Also,

$$\frac{\partial \rho}{\partial t} = K_5 \left\{ (1 + \bar{n}e_x) ([C_\kappa \rho, C_\kappa^+] + [C_\kappa, \rho C_\kappa^+] + \bar{n}e_\kappa ([C_\kappa^+ \rho, C_\kappa] + [C_\kappa^+, \rho C_\kappa]) \right\}$$
where  $K_5 = \pi g'(\omega_0) |\chi_5|^2$  describes the damping of excitons due to exciton-lattice interactions, (6.44)

and

$$\frac{\partial \rho}{\partial t} \Big|_{\substack{\text{cavily} \\ \text{damp.}}} = K_4 \{ (1+\bar{n}) ( \lfloor b\rho, b^{\dagger} \rfloor + \lfloor b, \rho b^{\dagger} \rfloor ) + \bar{n} ( \lfloor b^{\dagger}\rho, b \rfloor ) \\ + \lfloor b^{\dagger}, \rho b \rfloor ) \} \qquad (6.45)$$

where  $K_4 = \pi g''(\omega_0) |\chi_4|^2$  - describes the radiative damping of the cavity mode.

# $\oint 6.5$ Fokker-Planck and Langevin Equations

We can evaluate the system's quantum fluctuations explicitly by transforming the operator master equation into a Fokker-Planck equation.

Following the discussion of  $\oint 1.3(f)$  we use the generalised P-representation and write:

$$\rho = \int P(\alpha_1, \beta_1, \alpha_2, \beta_2) \frac{|\alpha_1, \alpha_2\rangle < \beta_1^*, \beta_2|}{<\beta_1^*, \beta_2^* |\alpha_1, \alpha_2\rangle} d\alpha_1 d\beta_1 d\alpha_2 d\beta_2$$

where

 where C.C.' means:  $\alpha_1 \rightarrow \beta_1$ ,  $\alpha_2 \rightarrow \beta_2$ ,  $i \rightarrow -i$ and

$$\begin{split} &\chi_{1} = i \, \delta_{1} - 4 \, K_{2,b} + K_{3} + K_{1,a} - K_{1,b} + K_{5} = \chi_{a} + i \, \chi_{b} \\ &g_{1} = g \, r_{1} \qquad ; \quad g_{2} = g \, r_{2} \\ &\chi = 2 \, ( \, K_{2,a} - K_{2,b} - (i/_{h}) \, M_{2}) = \chi_{a} + i \, \chi_{b} \\ &K_{1} = 2 \, [ \, (i/_{h}) \, M_{2} - K_{2,a} - K_{2,b} - K_{3} \, ] \\ &K_{2} = 8 \, K_{2,b} + 2 \, K_{3} \qquad ; \quad \eta = 8 \, K_{2,b} + 2 \, K_{1,b} + 2 \, K_{5} \, \bar{n}_{ex} \\ &\chi_{2} = K_{4} + i \, \delta_{2} = \chi_{c} + i \, \chi_{d} \end{split}$$

In deriving Eq. (6.46) we neglected derivatives higher than second order.

As discussed in  $\oint 1.3(f)$ , in the positive P-representation there exists a Fokker-Planck equation with positive semi-definite drift coefficient, which corresponds to Eq. (6.46).

From such a Fokker-Planck equation we find the Langevin equation,

$$\begin{pmatrix} \dot{\alpha}_{1} \\ \dot{\beta}_{1} \\ \dot{\alpha}_{2} \\ \dot{\beta}_{2} \end{pmatrix} = \begin{pmatrix} -\vartheta_{1}\alpha_{1} - \chi \alpha_{1}\alpha_{1}\beta_{1} - ig_{1}\alpha_{2} + ig_{2} (\beta_{2}\alpha_{1}^{2} + 2\alpha_{2}\alpha_{1}\beta_{1}) \\ -\vartheta_{1}^{*}\beta_{1} - \chi^{*}\beta_{1}\alpha_{1}\beta_{1} + ig_{1}\beta_{2} - ig_{2} (\alpha_{2}\beta_{1}^{2} + 2\beta_{2}\alpha_{1}\beta_{1}) \\ -\vartheta_{2}\alpha_{2} + E - ig_{1}\alpha_{1} + ig_{2}\alpha_{1}\alpha_{1}\beta_{1} \\ -\vartheta_{2}^{*}\beta_{2} + E^{*} + ig_{1}\alpha_{1} - ig_{2}\beta_{1}\alpha_{1}\beta_{1} \\ -\vartheta_{2}^{*}\beta_{2} + E^{*} + ig_{1}\alpha_{1} - ig_{2}\beta_{1}\alpha_{1}\beta_{1} \\ + \begin{pmatrix} \kappa_{1}\alpha_{1}^{2} + 2ig_{2}\alpha_{1}\alpha_{2} & \kappa_{2}\alpha_{1}\beta_{1} + \eta \\ \kappa_{2}\alpha_{1}\beta_{1} + \eta & \kappa_{1}^{*}\beta_{1}^{2} - 2ig_{2}\beta_{1}\beta_{2} & 0 & -ig_{2}\beta_{1}^{2} \\ 0 & -ig_{2}\beta_{1}^{2} & 0 & 0 & 2\kappa_{4}\bar{n} \\ 0 & -ig_{2}\beta_{1}^{2} & 2\kappa_{4}\bar{n} & 0 \end{pmatrix}^{\frac{1}{2}} \begin{pmatrix} \varrho_{1}(t) \\ \varrho_{1}(t) \\ \varrho_{3}(t) \\ \varrho_{4}(t) \end{pmatrix}$$
(6.47)
where the  $\vartheta_{1}(t)$  comprise a Gaussian stochastic process;

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$$\langle z_{i}(t) z_{j}(t') \rangle = \delta_{ij} \delta(t-t')$$

We will discuss Eqs. (6.46) and (6.47) in more detail in Chapter Seven but now turn our attention to the deterministic behaviour of the system; particularly to its bistable nature.

# $\oint$ 6.6 Steady State Behaviour

# $\oint 6.6(a)$ Low Density Case

The low exciton density system is described by the Langevin equations, Eqs. (6.47). As a first approximation we neglect noise and investigate the steady state properties of such a system by considering the deterministic equations:

$$\dot{\alpha}_{1} = -\delta_{1}\dot{\alpha}_{1} - \chi \dot{\alpha}_{1} |\dot{\alpha}_{1}|^{2} - ig_{1}\dot{\alpha}_{2} + ig_{2} (\dot{\alpha}_{2}^{*}\dot{\alpha}_{1}^{2} + 2\dot{\alpha}_{2}|\dot{\alpha}_{1}|^{2})$$
 (6.48a)

$$\dot{\alpha}_{1}^{*} = -\chi_{1}^{*}\dot{\alpha}_{1}^{*} - \chi_{1}^{*}\dot{\alpha}_{1}^{*}|\dot{\alpha}_{1}|^{2} + ig_{1}\dot{\alpha}_{2}^{*} - ig_{2}(\dot{\alpha}_{2}\dot{\alpha}_{1}^{*2} + 2\dot{\alpha}_{2}^{*}|\dot{\alpha}_{1}|^{2})$$
 (6.48b)

$$\dot{\alpha}_2 = -\vartheta_2 \alpha_2 + E - i g_1 \alpha_1 + i g_2 \alpha_1 |\alpha_1|^2$$
 (6.48c)

$$\dot{a}_{2}^{*} = -\delta_{2}^{*}a_{2}^{*} + E^{*} + ig_{1}a_{1}^{*} - ig_{2}a_{1}^{*}|a_{2}|^{2}$$
 (6.48d)

where, in the deterministic limit :  $\beta_1 \rightarrow \alpha_1^*$ ,  $\beta_2 \rightarrow \alpha_2^*$ .

Solving equations in the steady state  $(\dot{\alpha}_1 = \dot{\alpha}_2 = \dot{\alpha}_1^* = \dot{\alpha}_2^* = 0)$ we find,

$$I = n_{1} \left| \frac{\chi_{2}^{*} \left[ (\chi_{b} + \chi_{b}n_{i})(g_{2}n_{i} - g_{i}) + i(\chi_{a} + \chi_{a}n_{i})(3g_{2}n_{i} - g_{i}) \right] + i(g_{2}n_{i} - g_{i})^{2}}{\left| (3g_{2}n_{i} - g_{i})(g_{2}n_{i} - g_{i}) \right|^{2}}$$
(6.49)

and

$$n_{2} = n_{1} \left\{ \frac{\left(\chi_{b} + \chi_{b} n_{1}\right)^{2}}{\left(3g_{2}n_{1} - g_{1}\right)^{2}} + \frac{\left(\chi_{a} + \chi_{a} n_{1}\right)^{2}}{\left(g_{2}n_{1} - g_{1}\right)^{2}} \right\}$$
(6.50)

where  $I = |E|_{s.s.}^2 = steady state input field intensity.$ 

 $n_1 = (|\alpha_1|^2)_{5.5.} = \text{steady state exciton number (intensity)}.$ 

$$n_2 = (|\alpha_2|^2)$$
 s.s. = steady state output field intensity.

Equations (6.49) and (6.50) indicate possible singularities in the system when  $g_2n_1 = g_1$  or  $3g_2n_1 = g_1$ . However, these equations were derived in the low exciton density limit in which we assume,

$$g_2n_1 << g_1$$

As  $g_2 n_i$  approaches  $g_i$  the low density limit becomes invalid and Eqs. (6.49) and (6.50) no longer describe the system.

To determine the possibility of observing bistability in this system, a stability analysis is required. However, the highly nonlinear nature of this four-dimensional system hinders such analytical calculations.

As a first approximation, we consider the very low exciton density limit:

$$g_2 n_1 - g_1 \rightarrow -g_1$$
,  $3g_2 n_1 - g_1 \rightarrow -g_1$ 

Equations (6.49) and (6.50) then become

$$I = \frac{n_{1}}{g_{1}^{2}} \left[ \tilde{p}_{1} n_{1}^{2} + \tilde{p}_{2} n_{1} + \tilde{p}_{3} \right] - ---(6.51a)$$

$$n_{2} = \frac{n_{1}}{g_{1}^{2}} \left[ \left( \chi_{b} + \chi_{b} n_{i} \right)^{2} + \left( \chi_{a} + \chi_{a} n_{i} \right)^{2} \right] \qquad ---(6.51b)$$

where  $\tilde{p}_1 = (\delta_c \chi_b + \delta_d \chi_a)^2 + (\delta_c \chi_a - \chi_b \delta_d)^2$ 

$$\tilde{p}_{2} = 2 \left\{ (\chi_{c}\chi_{b} + \chi_{d}\chi_{a})(\chi_{c}\chi_{b} + \chi_{d}\chi_{a}) + (g_{1}^{2} + \chi_{c}\chi_{a} - \chi_{d}\chi_{b})(\chi_{c}\chi_{a} - \chi_{d}\chi_{b}) \right\}$$

$$\tilde{p}_{3} = (\chi_{c}\chi_{b} + \chi_{d}\chi_{a})^{2} + (g_{1}^{2} + \chi_{c}\chi_{a} - \chi_{d}\chi_{b})^{2}$$
Stability Analysis for Very Low Density Systems

(For a discussion of linearised stability analysis, see Chapter Two,  $\oint 2.1(c)$ ).

In such a low density system  $(i.e.g_2=0)$  we linearise the deterministic Eqs. (6.48) by substituting:

$$a_1 = a_0 + \delta a$$
$$a_2 = \beta_0 + \delta \beta$$

(where  $\alpha_0$ ,  $\beta_0$  are the deterministic means of  $\alpha_1$ ,  $\alpha_2$  respectively;  $\delta \alpha$ ,  $\delta \beta$  are small deviations from these means); and retaining only constant terms and terms linear in  $\delta \alpha$ ,  $\delta \beta$  in Eqs. (6.48). In this way we find the linearised equations:

$$\begin{split} \delta \dot{\alpha} &= (-\delta_1 - 2\chi_{1d_0})^2 \delta \alpha - \chi_{\alpha^2} \delta \alpha^* - ig_1 \delta \beta \\ \delta \dot{\alpha}^* &= (-\delta_1^* - 2\chi_{1d_0})^2 \delta \alpha^* - \chi^* \alpha_0^{*2} \delta \alpha + ig_1 \delta \beta \\ \delta \dot{\beta}^* &= -\delta_2 \delta \beta - ig_1 \delta \alpha \\ \delta \dot{\beta}^* &= -\delta_2^* \delta \beta^* + ig_1 \delta \alpha^* \end{split}$$
(6.52)

Equations (6.52) yield the dispersion equation:

$$M_{0}\lambda^{4} + m_{1}\lambda^{3} + m_{2}\lambda^{2} + m_{3}\lambda + m_{4} = 0$$
  
where

$$\begin{split} m_{0} &= 1 \quad ; \quad m_{1} = \operatorname{Re}(\aleph_{2}) - \operatorname{Re}(-\aleph_{1} - 2 \,\chi_{1d_{0}1^{2}}) \\ m_{2} &= 2g_{1}^{2} - |\chi|^{2}(|d_{0}|^{2})^{2} + |\aleph_{2}|^{2} + |\aleph_{1} + 2 \,\chi_{1d_{0}1^{2}}|^{2} - \operatorname{Re}(-\aleph_{1} - 2 \,\chi_{1d_{0}1^{2}})\operatorname{Re}(\aleph_{2}) \\ m_{3} &= g_{1}^{2} \,m_{1} - |\chi|^{2}(|d_{0}1^{2})^{2}\operatorname{Re}(\aleph_{2}) - |\aleph_{2}|^{2}\operatorname{Re}(-\aleph_{1} - 2 \,\chi_{1d_{0}1^{2}}) + \operatorname{Re}(\aleph_{2})|\aleph_{1} + 2 \,\chi_{1d_{0}1^{2}}|^{2} \\ m_{4} &= \frac{\partial I}{\partial n_{1}} \quad ; \qquad \text{obtained from Eq. (6.51a).} \end{split}$$

## Stability Conditions:

The Hurwitz stability criteria for this system are

- (1)  $m_0 > 0$
- (2)  $m_i > 0$
- (3)  $m_1 m_2 m_3 > 0$

(4) 
$$m_3(m_1m_2-m_3) - m_1^2 \frac{\partial I}{\partial n_1} > 0$$

and (5)  $\frac{\partial I}{\partial n_1} \left[ m_3 (m_1 m_2 - m_3) - m_1^2 \frac{\partial I}{\partial n_1} \right] > 0$ 

The system exists in a stable state only when conditions (1) to (5) are satisfied. Given that the parameters can be chosen such that all these conditions are met, conditions (4) and (5) imply the system is only stable when,

is unstable when

and is critically unstable when  $\frac{\partial I}{\partial n_1} = 0$ 

The steady state behaviour corresponding to Eq. (6.51) is illustrated in Figs. 6.a and 6.b.



According to the stability analysis, the top and bottom branches of Fig. 6.a (branches (i) and (iii) will be stable to small fluctuations (i.e.  $\partial I/\partial n_i > O$  in these regions). Branch (ii) in Fig. 6.a will be unstable and the turning points ( P and Q ) correspond to bistable transition points. Bistability thus occurs as indicated in Fig. 6.b.

We note that in the limit of large exciton damping (in which the excitons may be adiabatically eliminated), the parameter  $\delta_1$  is very much larger than all other system parameters. In this case:

 $m_{o} = 1 , m_{i} \sim + \operatorname{Re}(\vartheta_{i}) , m_{2} \sim \left[\operatorname{Re}(\vartheta_{i})\right]^{2}$   $m_{3} \sim \left[\operatorname{Re}(\vartheta_{1})\right]^{2} \cdot \operatorname{Re}(\vartheta_{2}) , m_{4} \equiv \frac{\partial I}{\partial m_{i}} \sim \frac{I}{g_{i}^{2}} \left(\vartheta_{d}^{2} + \vartheta_{c}^{2}\right) \left[\operatorname{Re}(\vartheta_{i})\right]^{2}$ The Hurwitz criteria for stability then become (large  $\vartheta_{1}$  limit): (1)  $m_{o} = 1 > O$ (2)  $m_{i} \sim + \operatorname{Re}(\vartheta_{i}) > O$ 

(3) 
$$m_1 m_2 - m_3 \sim [Re(s_1)]^3 > 0$$

(4) 
$$m_3(m_1m_2 - m_3) - m_1^2 \frac{\partial I}{\partial n_1} \sim \left[ \operatorname{Re}(\mathfrak{X}_1) \right]^3 > C$$

and

(5) 
$$\frac{\partial I}{\partial n_1} \left\{ m_3(m_1m_2 - m_3) - m_1^2 \frac{\partial I}{\partial n_1} \right\} \sim \left[ Re(\aleph_1) \right]^7 > 0$$

Hence, conditions (1) to (5) are always satisfied in the adiabatic limit (  $\delta_1$  large).

We may thus conclude that bistability as indicated in Fig. 6.b will occur in the low density system in the adiabatic limit of large  $\aleph_1$ .

The conditions for bistability are determined by requiring the equation  $\partial I/\partial n$ ,=0 to have real positive solutions. This implies the bistability conditions,

 $2\tilde{p}_2 < 0$ ,  $4\tilde{p}_2^2 - 12\tilde{p}_1\tilde{p}_3 > 0$  \_\_\_\_\_(6.53)

The inequalities (6.53) will obviously be satisfied for a large range of parameters.

We note that the bistability depends solely on the exciton-exciton interaction.

Let us consider the absorptive limit ,  $\delta d = \delta_b = 0$  . In this case,  $\tilde{p}_2 = 2(g_1^2 + \delta_c \delta_a) \delta_c \chi_a$ , which is negative only if  $\delta_a$  is negative, as all other parameters are positive. This requires either very large values of the thermal occupation numbers of the reservoir modes, or strong exciton-exciton interactions. Thus, purely absorptive bistability is possible only when exciton-exciton collisions become dominant.

If we now include a non-zero value of  $g_2$  into the system, we do not expect the bistable behaviour to change significantly - as long as  $g_2 << 1$  and  $n_1$  remains relatively small.

Figs. 6.1 and 6.2 show plots of  $n_1$  vs.  $I_{and}$   $n_2$  vs.  $I_{for small}$   $g_2$ . (The values of  $n_1$  at the points A, B, C, and D in Fig. 6.1 correspond to the values of  $n_1$  at the points A, B, C and D in Fig. 6.2).

Dotted arrows indicate expected bistability. That is, from the stability analysis for the case  $g_2 = 0$ , we find that the middle branch of Fig. 6.1 will be unstable, as  $\partial I / \partial n_1 < 0$ ; and the top and bottom branches will be stable as  $\partial I / \partial n_1 > 0$ . Bistable transitions occur at the points A and C on Fig. 6.1 as  $\partial I / \partial n_1 = 0$  at these points.

As output intensity depends parametrically on I through  $n_1$ , we expect a corresponding bistability will be observed in output intensity  $(n_2)$ , dependent on input intensity I. This is indicated in Fig. 6.2.

We see than that bistability is possible in a low exciton density system. However in experiments involving GaAs (in which bistability was observed) exciton densities are too high to justify the neglect of higher order terms in the expansion, Eq. (6.34). We need to consider the high density case separately.



Fig. 6.1: Variation of Exciton Number with Input Intensity in the Low Density Limit ( $\delta_a = \delta_c = \delta_d = g_1 = 1$ .;  $\delta_b = -10$ .;  $g_2 = .01$ ;  $\chi_a = 1$ .;  $\chi_b = 5$ .).



Fig. 6.2: Variation of Output Intensity with Input Intensity in the Low Density Limit (Data as in Fig. 6.1).

# $\phi$ 6.6(b) High Density Case

Problems arise when we wish to describe the high exciton density system, as the infinite boson expansion occurring in  $\tilde{H}'_{\rm INT}$  (see Eq. (6.33)) now becomes divergent. We overcome this difficulty by performing a transformation on the exciton operators which generates a finite expression for  $\tilde{H}'_{\rm INT}$ .

Consider the explicit form of the interaction Hamiltonian: (i.e. from Eqs. (6.21a) and (6.33)),

$$\widetilde{H}_{INT}' = \pi g b \left[ \epsilon_{1} c_{\kappa}^{\dagger} - \epsilon_{2} c_{\kappa}^{\dagger} c_{\kappa}^{\dagger} c_{\kappa} \right] \left[ 1 + \epsilon_{3} c_{\kappa}^{\dagger} c_{\kappa} \right]^{-1/2} + h.c. \quad -(6.54)$$

where

$$\mathcal{E}_{1} = \sqrt{\nabla} \mathcal{E}_{Rc} f (aRc + \beta (Rc - K))$$

$$\mathcal{E}_{2} = \sqrt{\nabla \nabla} \left\{ \mathcal{E}_{Rc} \left| f \left( \alpha R_{c} + \beta \left( R_{c} - K \right) \right) \right|^{2} f^{*} \left( \alpha \left( R_{c} - K \right) + \beta \left( 2K - R_{c} \right) \right) \right\}$$

$$\mathcal{E}_{3} = \frac{1}{\sqrt{2}} \left\{ \mathcal{E}_{Rc} \left| f \left( d Rc + \beta (Rc - K) \right) \right|^{2} \right\}$$

We now set,

$$C_{\kappa} = A + \lambda$$
,  $C_{\kappa}^{\dagger} = A^{\dagger} + \lambda^{*}$  -----(6.55)

where A is a boson operator such that  $\langle A \rangle = O$  and  $\lambda$  is a constant where

 $\langle c \rangle = \lambda$ 

The large exciton density limit thus corresponds to large  $\lambda$  and we may expand perturbatively about the deterministic solution.

Substituting Eqs. (6.55) into Eq. (6.54), we expand the denominator to first order and retain terms of order  $\lambda^2$  and  $\lambda$  only:

$$\Rightarrow Equ. 6.54 \Rightarrow \widetilde{H}_{INT} = -\frac{\kappa}{2} g b \mathcal{E}_2 (\mathcal{E}_3 | \lambda |^2)^{-1/2} \lambda^* \{ (3A^+ + 2\lambda^*) \lambda (6.56) + A \lambda^* \} + h.c.$$

The Hamiltonian described by Eq. (6.56) yields the following contributions to the deterministic equations:

$$\langle b \rangle = i \chi \langle (|\lambda|^2)^{-\frac{1}{2}} \lambda \{ (3A + 2\lambda) \lambda^* + A^* \lambda \} \rangle \qquad (6.57)$$

$$\langle C_{\kappa} \rangle = i \chi \langle 3 b (|\lambda|^2)^{-1/2} |\lambda|^2 + b^{\dagger} (|\lambda|^2)^{-1/2} \lambda^2 \rangle - ----(6.58)$$

where  $\langle \rangle$  denotes average value, and

$$\chi = \frac{9 \varepsilon_2 \varepsilon_3}{2}$$

Using the definitions  $\langle A \rangle = 0$ ,  $\langle c \rangle = \lambda = \alpha_1$ ,  $\langle b \rangle = \alpha_2$ , we combine Eqs. (6.57) and (6.58) with the deterministic equations obtained from  $\widetilde{H}'_0 + \widetilde{H}'_{ex} + \widetilde{H}'_0 + \widetilde{H}_{damping}$  (i.e. Eqs. (6.48) with  $g_1 = g_2 = 0$ ) to find the high exciton density equations,

$$\dot{\alpha}_{1} = -\aleph_{1}\alpha_{1} - \aleph_{1}\alpha_{1}|\alpha_{1}|^{2} + i \aleph(|\alpha_{1}|^{2})^{-l_{2}}(3\alpha_{2}|\alpha_{1}|^{2} + \alpha_{2}^{*}\alpha_{1}^{2}) - (6.59)$$

$$\dot{\alpha}_{2} = -\aleph_{2}\alpha_{2} + E + 2i \aleph\alpha_{1}(|\alpha_{1}|^{2})^{-l_{2}} - (6.60)$$

and complex conjugate equations.

Equations (6.59) and (6.60) can be solved in the steady state to yield,

$$I = \frac{(l_{1} + n_{1} l_{2})^{2} + (l_{3} + l_{4} n_{1})^{2}}{4 \chi^{2}} \qquad (6.61)$$

$$n_{2} = \frac{1}{4 \chi^{2}} \left\{ \frac{1}{4} (\chi_{b} + \chi_{b} n_{1})^{2} + (\chi_{a} + \chi_{a} n_{1})^{2} \right\} \qquad (6.62)$$

$$l_1 = \frac{\aleph_c \aleph_b}{2} + \aleph_d \aleph_a \qquad ; \quad l_2 = \frac{\aleph_c \aleph_b}{2} + \aleph_d \aleph_a$$

$$l_3 = \chi_c \chi_a - \frac{\chi_d \chi_b}{2}$$
;  $l_4 = \chi_c \chi_a - \frac{\chi_d \chi_b}{2} + 4 \chi^2$ 

Equations (6.61) and (6.62) show that the steady state now displays a quadratic nonlinearity.

When graphing the behaviour of the high exciton density system, we note that for low values of  $n_1$ , Eqs. (6.61) and (6.62) become invalid. Thus, in this region we must use the theory developed in  $\oint 6.5(a)$  for the low density system.

Fig. 6.3 shows the variation of exciton number  $(n_i)$  with input intensity (I). The graph is a combination of two curves: the upper part describes the high density behaviour (Eq. (6.61)) and the lower part corresponds to the low density case (Eq. (6.49)).

In these two distinct regions, the respective theories accurately predict the system's behaviour. However, in the intermediate region (indicated by broken lines in Fig. 6.3) we are unsure of the applicability of either theory. We expect the actual behaviour of the system to follow Fig. 6.3, except in the intermediate region; where we assume a smooth transition between high and low density behaviour will occur.

It is assumed that the point G in Fig. 6.3 corresponds to a point of critical instability and that a bistable transition will occur as indicated.

As we are unsure of the precise behaviour of the system in the region around the point E in Fig. 6.3, the stability analysis necessary to determine the actual nature of the hysteresis was not performed. Such investigations are proceeding.

Nevertheless, we do expect a transition to occur in the neighbourhood of the point E, and that bistability will arise as indicated in Fig. 6.3.

Fig. 6.4 shows the corresponding variation of output intensity with input intensity. (The values of  $\gamma_1$  at the points E, F, G and H in Fig. 6.3 correspond to the values of  $\gamma_1$  at the points E, F, G and H in Fig. 6.4). The upper and lower curves were obtained from the high and low density theories respectively. Again, broken lines indicate regions in which neither theory is valid.

As the input intensity is increases, exciton number  $n_1$  varies as in Fig. 6.3, until at  $I_2$  a transition occurs, indicated by  $\overrightarrow{\mathsf{EF}}$ . Because  $n_2$  depends parametrically on I, via  $n_1$  we thus expect





Fig. 6.4: Output Intensity Vs. Input Intensity Including Effects of High Exciton Density. (Data as for Fig. 6.3).

a similar unstable behaviour in output intensity at  $I_2$ . Such a transition is indicated by the arrow  $\vec{EF}$  in Fig. 6.4. Similarly, the transition  $\vec{GH}$  at  $I_1$  occurs in Fig. 6.4

As in the low density case, the possibility of observing bistability requires  $\partial I/\partial n_{,} = 0$  to have real, positive solutions. This results in the condition,

$$-(l_1 l_2 + l_3 l_4) > 0$$

In contrast to the low density case, a non-zero  $\chi$  is no longer necessary for bistability. However, absorptive bistability still requires,

i.e. dominant exciton-exciton interaction.

Thus, bistability and hysteresis in output intensity dependent on input intensity is also displayed in the high exciton density system.

We note that this behaviour follows the experimental curves of Gibbs et.al. (1979), showing output intensity versus input intensity for GaAs.

## ∮6.7 Optical Bistability in Semiconductors

We have thus presented a theory of light-semiconductor interaction which includes effects such as exciton-exciton interactions and radiative damping. As such it represents an improvement on the phemonenological theory of Goll and Haken (1980).

After developing a boson Hamiltonian, we were able to model the system in the two limiting cases of low and high exciton densities.

Steady state analysis showed the system exhibits bistability in both exciton number and output intensity dependent on input intensity.

At present though, excitonic bistability of output intensity dependent on input intensity has been observed in GaAs only in the high density limit. Our work, however, shows that bistability can occur in materials having lower exciton density: In this limit, dispersive and absorptive bistability were shown to depend critically on exciton-exciton interactions. Thus, such interactions provide the nonlinearity necessary to produce an intensity dependent refractive index. We also see that these excitonexciton interactions generate the cooperative effects necessary for bistability. (An example of such a low exciton density system is CdS, in which nonlinear optical effects have already be observed (Benoit à la Guillame et.al., 1969; Haug, 1968).

At high exciton densities, in the dispersive limit, bistability was shown to occur without the presence of exciton-exciton interactions. The nonlinearity producing bistability in this case arose from the interaction between the light field and the medium. From the expression for  $\widetilde{H}'_{1NT}$  (Eq. (6.56)) we see that this involves the interaction of a photon with two or three excitons. This agrees in essence with the suggested bistability mechanism proposed by Gibbs et.al. (1979): light absorbed just below the exciton resonance frequency produces carriers, changing the absorptivity and polarisability of the medium, thus leading to an intensity dependent refractive index.

We also predicted purely absorptive bistability at high exciton densities, which was seen to require strong exciton-exciton interactions. So far, only dispersive bistability has been experimentally observed.

Finally, we note that Eqs. (6.48) with  $g_2 = O$  are identical to the equations of motion for the driven anharmonic oscillator. ( $\alpha_1 \leftrightarrow$  amplitude of oscillator,  $\alpha_2 \leftrightarrow$  field mode).

Such a system is known to exhibit bistability, depending critically on the anharmonicity, i.e.  $\chi$  in this case (Steyn-Ross, 1979).

Thus we see that weakly driven excitonic systems can display bistability if the parameter  $\chi$  is sufficiently large. As  $\chi$  depends on excitonexciton collisions (clearly a temperature associated effect) such a bistability is a consequence of exciton-exciton interactions.

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#### CHAPTER 7

# QUANTUM FLUCTUATIONS IN LOW EXCITON-DENSITY SYSTEMS

#### $\oint$ 7.1 Introduction

We now discuss quantum fluctuations in the low exciton density semiconductor system by considering the Fokker-Planck equation, Eq. (6.46).

From this equation we wish to derive the system's photon statistics. As potential conditions are not satisfied in this highly nonlinear system, the statistical properties are most easily calculated via linearised fluctuation theory (Chaturvedi et.al., 1977). However, for systems of dimensionality higher than two, these calculations become very complicated. A reduction of the excitonic system's dimensionality is thus required to enable the straightforward calculation of statistical properties.

Fortunately, in most of the systems we model, actual experimental conditions will be such that we do not have to explicitly consider certain variables.

For example, in intracavity experiments we can construct the cavity so that the cavity mode has a much longer lifetime than the excitons. We are then able to adiabatically eliminate the excitons.

Alternatively, in experiments using very fast pulsed lasers ( $\circ$  picosecond pulses) the excitons can have a longer lifetime than the cavity mode which may then be adiabatically eliminated.

This chapter concerns how these two types of variable eliminations can be performed in a low exciton density system, with a view to calculating photon statistics.

As mentioned in  $\oint 1.3(g)$ , adiabatic elimination of variables is well defined within a deterministic theory, but problems arise when we introduce noise into the system. Thus, we firstly discuss how such an elimination Can be performed within the framework of a stochastic theory.

# 67.2 General Adiabatic Elimination Methods

Let us consider a general nonlinear two-dimensional system, described by the coupled stochastic equations,

$$\dot{a}_1 = f_1(a_1, a_2) + f_{a_1}(t)$$
 (7.1)  
 $\dot{a}_2 = f_2(a_1, a_2) + f_{a_2}(t)$  (7.2)

where  $f_1$  and  $f_2$  are arbitrary functions and  $f_{\alpha_1}(t)$ ,  $f_{\alpha_2}(t)$  are stochastic fluctuating terms where,

$$\langle f_{di}(t) \rangle = 0$$
  $i = 1, 2$ 

and

$$\langle f_{a_i}(t) f_{a_j}(t') \rangle = D_{ij} \delta(t-t')$$

where  $D_{ij}$  is a diffusion matrix element of the Fokker-Planck equation corresponding to Eqs. (7.1) and (7.2).

We assume  $lpha_1$  is damped on a much faster time scale than  $lpha_2$  , thus enabling the elimination of  $lpha_1$  .

As a first approximation we may follow Haken (1978) and neglect all noise terms associated with the variable  $\alpha_1$  (i.e. set  $\int_{\alpha_1} (t) = O$ in Eq. (7.1)). We then substitute the stationary mean of  $\alpha_1$  into Eq. (7.2), as in the deterministic case discussed in  $\oint 1.3(g)$ . The reduced system is thus described by the single stochastic differential equation,

$$\dot{\alpha}_2 = f_2((\alpha_1)_{ss}, \alpha_2) + f_{\alpha_2}(t)$$
 (7.3)

where  $(\alpha_1)_{55}$  was obtained from solving Eq. (7.1) with  $\dot{\alpha}_1 = 0$  and  $\hat{\alpha}_1(t) = 0$ .

A major drawback of this method is that the only noise in the reduced system now stems from the variable  $\alpha_2$ . We expect that this neglect of noise in  $\alpha_1$  will be justified when the noise in mode 2 is large in comparison. When this is not the case however, major errors in the calculation of important physical properties will occur.

The need to include the effects of noise from eliminated variables was recognised by Haken (1970) and Louisell (1973) in their treatment of adiabatic elimination of atomic variables in the laser.

Following their approach, elimination of  $\alpha_1$  proceeds by setting  $\dot{\alpha}_1 = O$  in Eq. (7.1) thus yielding,

$$(d_1)_{55} = f_3(d_2, f_{d_1}(t))$$
 (7.4)

The steady state mean of  $d_1$  now contains explicit reference to noise terms. The reduced system is then described by the equation

$$\dot{\alpha}_2 = f_2 [f_3(\alpha_2, f_{\alpha_1}(t)), \alpha_2] + f_{\alpha_2}(t) -----(7.5)$$

At a first glance, such an approach seems promising, as Eq. (7.5) contains noise terms arising from both variables. However, in a nonlinear system the function  $f_2$  occurring in Eq. (7.5) could well contain terms of the form,

$$[f_{\alpha_1}(t)]^2$$
 \_\_\_\_\_(7.6)

Such a product of fluctuating terms is infinite and does not describe any real physical situation.

The procedure we have been describing actually has no rigorous basis, and as we have seen, involves a suspect treatment of system noise.

That is, although setting  $\alpha_1 = 0$  in Eq. (7.1) properly describes the stationary regime, this procedure assumes the noise term  $\int \alpha_1(t)$  remains constant.

However, we assume that the correct adiabatic noise limit will be found by performing a systematic expansion of the noise terms. Such methods, directly applicable to stochastic differential equations do not exist.

Thus, in this chapter we adopt what is believed to be the first systematic method of adiabatic elimination of variables from the Fokker-Planck equation - that proposed by Gardiner (1982). This procedure, based on a projection operator technique, follows the methods of Wilemski (1976) and Papanicolaou (1977): A projection operator in terms of the stationary distribution function  $(P_s)$  of the variable to be eliminated is constructed. Such an operator projects vectors into a subspace in which all vectors are expressed as a product of the stationary distribution  $P_s$ , and an arbitrary function of the remaining variables. This new subspace thus represents a reduced system, describing only the variables not eliminated.

Hence the dimensionality of the system is reduced in this new subspace, providing the necessary simplification we require to derive the system's statistical properties.

#### \$7.3 Projection Operator Technique: General Procedure

Before considering the explicit case of the semiconductor system, we will review the basic steps of the projection operator method, as presented by Gardiner (1982).

The procedure assumes it is always possible to scale the system variables such that the Fokker-Planck equation can be written as,

$$\frac{\partial P(\underline{\alpha})}{\partial t} = (\chi^2 L_1 + L_2 + L_3) P(\underline{\alpha}) \qquad -----(7.7)$$

where  $\delta$  is a dimensionless parameter, and  $\delta \rightarrow \infty$  describes the adiabatic limit. Physically, we identify  $\delta$  with the damping coefficient of the variable to be eliminated, as adiabatic elimination requires the damping coefficient to be large in comparison to other system parameters.

We restrict this discussion to the simplified case of a two-variable system in which  $\underline{\alpha} = (\alpha_1, \alpha_2)$  and  $\alpha_1$  represents the variable we wish to eliminate.

In Eq. (7.7), the operator  $L_1$  concerns the variable to be eliminated,  $\alpha_1$ ,

$$L_{1} = \frac{\partial f(\alpha_{1}, \alpha_{2})}{\partial \alpha_{1}} + \frac{1}{2} \frac{\partial^{2} D(\alpha)}{\partial \alpha_{1}^{2}} - \dots - (7.8)$$

and  $L_2$  and  $L_3$  are operators involving  $\alpha_1$  and  $\alpha_2$ , to be discussed shortly.

We now wish to derive a Fokker-Planck equation involving the variable  $\alpha_2$  only, and thus describing the distribution function,

This can be achieved by a method based on the following simple argument: In the large  $\delta$  limit,  $L_1$  dominates in the Fokker-Planck equation, thus a solution of the equation will be a solution of

$$\frac{\partial P}{\partial t} = \delta^2 L, P \qquad (7.10)$$

multiplied by some function of  $\alpha_2$  (yet to be determined).

As & is very large, the solution of Eq. (7.10) rapidly approaches its steady state and we thus only need consider the stationary solution, described by,

$$L, P = O$$
 (7.11)

More rigorously, we define a projection operator,  $m{arphi}$  as,

$$(\beta f)(\alpha_1, \alpha_2) = P_s(\alpha_1) \int d\alpha_1 f(\alpha_1, \alpha_2) - \dots (7.12)$$

where f is an arbitrary function and  $P_s(A_i)$  satisfies Eq. (7.11).

With regard to the vector space containing all functions of  $(a_1, a_2)$ , projects any vector to a subspace consisting of vectors of the form,

$$U(a_1,a_2) = P_s(a_1) \widehat{U}(a_2)$$

where  $\widehat{\mathcal{U}}(a_2)$  is an arbitrary function of  $a_2$ .

Thus by applying P to the Fokker-Planck equation we project out any explicit reference to  $\alpha_1$  and obtain an equation in terms of  $\alpha_2$ only.

To this end we define,

and we are interested in obtaining an equation for  $\, {f V} \,$  alone.

The operators  $L_2$  and  $L_3$  of Eq. (7.7) describe, in the main, the behaviour of the variable  $\alpha_2$ .  $L_3$  forms the basis of the new, reduced Fokker-Planck equation and must involve only  $\alpha_2$ . All remaining terms of the Fokker-Planck equation are contained in  $L_2$ .

In practice,  $L_1$  and  $L_2$  are chosen so that  $L_3$  can be written in terms of  $\alpha_2$  only and such that  $L_2$  satisfies,

$$\mathcal{P} \mathsf{L}_2 \mathcal{P} = \mathsf{O} \tag{7.14}$$

a condition essential to the elimination procedure, as we will see later.

As an illustrative example, let us consider the following system (Gardiner, 1982),

$$\frac{\partial P}{\partial t} = \frac{\partial}{\partial \alpha_1} \frac{\delta^2 \left[ f(\alpha_2) - A(\alpha_1) \right]}{\partial \alpha_2} - \frac{\partial}{\partial \alpha_2} \left[ b(\alpha_2) \alpha_1 + a(\alpha_2) \right]}{\left[ + \delta^2 \frac{\partial^2}{\partial \alpha_1^2} B(\alpha_1) + \frac{1}{2} \frac{\partial^2}{\partial \alpha_2^2} \left[ c(\alpha_2) \right]^2 \right]}$$
(7.15)

where f, A, b and a are in general nonlinear functions. Equation (7.15) can be cast in the form of Eq. (7.7) by setting,

The remainder of Eq. (7.15) forms the operator  $L_3$ ; however we need to remove reference to the variable  $\alpha_1$  occurring in the second term. This is achieved by defining the statistical average

$$m(a_2) = \langle a_1 \rangle_{a_2} = \int P_5(a_1) a_1 da_1 - ----(7.17)$$

and forming the operator  $L_2$  ,

$$L_2 = -\frac{\partial}{\partial \alpha_2} b(\alpha_2) \left[ \alpha_1 - m(\alpha_2) \right] \qquad -----(7.18)$$

Thus,

$$L_{3} = -\frac{\partial}{\partial d_{2}} \left[ a(\alpha_{2}) + b(\alpha_{2})m(\alpha_{2}) \right] + \frac{1}{2} \frac{\partial^{2}}{\partial \alpha_{2}^{2}} \left[ c(\alpha_{2}) \right]^{2} \qquad ----(7.19)$$

The operators  $L_1$ ,  $L_2$ ,  $L_3$  described by Eqs. (7.16), (7.18) and (7.19) combine to produce the Fokker-Planck equation, Eq. (7.15), which can now be written in the form of Eq. (7.7).

We also note,

$$PL_{2}Pf = -P_{s}(\alpha_{1})\int d\alpha_{1}'\frac{\partial}{\partial \alpha_{2}}b(\alpha_{2})[\alpha_{1} - m(\alpha_{2})]P_{s}(\alpha_{1}')\int d\alpha_{1}''f(\alpha_{1}'',\alpha_{2})$$

$$= -P_{s}(\alpha_{1})\frac{\partial}{\partial \alpha_{2}}b(\alpha_{2})\int d\alpha_{1}'(\alpha_{1} - m(\alpha_{2}))P_{s}(\alpha_{1}')\int d\alpha_{1}''f(\alpha_{1}'',\alpha_{2})$$

$$= 0$$

$$\left(1 - 1\left(1 - m(\alpha_{2})\right)P_{s}(\alpha_{1}')\right) = 0$$

as 
$$\int dd'_{1}(d_{1} - m(d_{2})) P_{s}(d'_{1}) = 0$$
 from Eq. (7.17).  
Thus,  $PL_{2}P = 0$ 

$$\begin{aligned} \mathcal{P}: Equ. 7.7 \Rightarrow \mathcal{P} \frac{\partial P}{\partial t} &= \mathcal{P}(\mathcal{Y}^2 L_1 + L_2 + L_3) \\ &= \mathcal{P}(\mathcal{Y}^2 L_1 + L_2 + L_3)[\mathcal{P} P + (1 - \mathcal{P})P] \end{aligned}$$

and using  $PL_1 = L_1P = 0$  and  $PL_2P = 0$  we find,

$$\frac{\partial}{\partial t}PP = \frac{\partial v}{\partial t} = PL_2w + PL_3(v+w)$$
(7.20)

where  $\mathcal V$  and  $\mathcal W$  are defined by Eqs. (7.13a) and (7.13b).

Secondly, we derive an equation for 
$$\mathcal{W}$$
 from Eq. (7.7)  
 $(I-\mathcal{P}) \stackrel{\partial}{\partial t} \mathcal{P} = (I-\mathcal{P})(\mathcal{V}^{2}L_{1}+L_{2}+L_{3})\mathcal{P} = (I-\mathcal{P})(\mathcal{V}^{2}L_{1}+L_{2}+L_{3})[\mathcal{P}\mathcal{P}]$   
 $+(I-\mathcal{P})\mathcal{P}]$ 

(Using (7.11) and (7.14))

$$\Rightarrow \frac{\partial w}{\partial t} = \chi^2 L_1 w + (1 - \beta) L_2 w + L_2 v + (1 - \beta) L_3 (v + w) - (7.21)$$

The elimination procedure, in which all reference to the dynamical behaviour of w is removed, follows by solving Eqs. (7.20) and (7.21) in the large  $\delta$  limit.

The coupled equations (7.20) and (7.21) can be solved using a Laplace transform method, in which we define,

and

Assuming W(0) = 0, Eqs. (7.20) and (7.21) become, on application of Eqs. (7.22), (7.23),

$$5 \tilde{v}(s) = \mathcal{V}(L_{2} + L_{3})\tilde{w}(s) + \mathcal{V}L_{3}\tilde{v}(s) + v(o) -----(7.24)$$

$$5 \tilde{w}(s) = [\lambda^{2}L_{1} + (1 - \beta)(L_{2} + L_{3})]\tilde{w}(s) + [L_{2} + (1 - \beta)L_{3}]\tilde{v}(s)$$

Solving Eqs. (7.24), the following expression, to second order in & is obtained - in the large & limit

$$5\tilde{v}(s) \approx \left[ \mathcal{P}L_3 - \sqrt[3]{-2}\mathcal{P}(L_2 + L_3)L_1^{-1}(L_2 + (1 - \mathcal{P})L_3) \right]\tilde{v}(s) + v(0) \qquad ---(7.25)$$

If L<sub>3</sub> has no  $\delta$  dependence, the term  $\ell L_3$  dominates in Eq. (7.25). The Fokker-Planck equation then becomes, in the adiabatic limit,

$$\frac{\partial \hat{P}(d_2)}{\partial t} = -\frac{\partial}{\partial d_2} \left[ a(d_2) + b(d_2) m(d_2) \right] P + \frac{1}{2} \frac{\partial^2}{\partial d_2^2} \left[ c(d_2) \right]^2 P - \frac{1}{2} \frac{\partial^2}{\partial d_2^2} \left[ c(d_2) \right]^2 P - \frac{1}{2} \frac{\partial^2}{\partial d_2^2} \left[ c(d_2) \right]^2 P$$

where we have set  $\mathcal{U}(t) = P_{s}(\alpha_{1}) \hat{P}(\alpha_{2})$ .

We see then that to lowest order, the elimination procedure is equivalent to the first approximation of Haken (1978), discussed in  $\oint 7.2$ : in which all noise in  $\alpha_1$  is ignored and  $\alpha_1$  replaced by its stationary value. This follows as in this case the noise of variable  $\alpha_1$ is comparatively small and we may assume the deterministic result:  $m(\alpha_2) \approx (\alpha_1)_{55}$ , where  $(\alpha_1)_{55}$  is the deterministic mean. Equation (7.26) will be applicable only if  $L_3$  has terms of higher order than  $\chi^{-2}$  in its drift and diffusion coefficients.

However, it is often the case that the terms of  $L_3$  have only a small noise contribution. In this instance we must include all other terms,  $O(\delta^{-1})$ , in Eq. (7.25).

As shown by Gardiner (1982), Eq. (7.25) then becomes,

$$5\tilde{v}(s) \approx [PL_3 - \delta^{-2}PL_2L_1^{-1}\{L_2 + [L_3, P]\}]\tilde{v}(s) + v(0)$$
 (7.27)

We discuss the evaluation of these additional terms with reference to the explicit case of the semiconductor system.

In the remainder of this chapter we consider the two limiting cases:

i) Adiabatic elimination of excitons and

ii) Adiabatic elimination of cavity mode,

and derive the statistical properties of the system for case i).

#### $\phi$ 7.4 Adiabatic Elimination of Excitons in the Semiconductor System

This regime assumes the excitons are damped on a much faster time scale than the cavity mode.

The linewidth of excitons in semiconductors is typically 0.1% of the exciton energy. This is approximately 0.1 eV, corresponding to a lifetime of  $10^{-12}$ sec. As a typical cavity lifetime is about  $10^{-7}$ sec, our assumption is justified.

## ∮7.4(a) Linearised Theory

As we will be using a linearised fluctuation theory to calculate the photon statistics, we first linearise the system. To this end we define, Exciton Mode:  $\alpha_1 = \alpha_0 + \alpha$ ,  $\beta_1 = \alpha_0^* + \alpha^+$ Field Mode:  $\alpha_2 = \beta_0 + \beta$ ,  $\beta_2 = \beta_0^* + \beta^+$  (7.28) where  $\alpha_{o}$  and  $\beta_{o}$  are the deterministic means of  $\alpha_{1}$  and  $\alpha_{2}$ , and  $\alpha_{3}$  and  $\beta_{4}$  and  $\beta_{4}$  and  $\beta_{4}$  are small deviations from these means.

We note that the variables  $\alpha$ ,  $\alpha^+$  (and similarly  $\beta$ ,  $\beta^+$ ) are not necessarily complex conjugates; as the stochastic variables  $\alpha'_1$  and  $\beta_1$ are not complex conjugates (Drummond and Gardiner, 1980). Thus, in all further discussion, pairs of variables such as  $(\alpha, \alpha^+)$  are assumed to vary independently over separate complex planes, and are only complex conjugate in the deterministic mean.

Linearisation of the system follows by substituting Eqs. (7.28) into the Fokker-Planck equation (Eq. (6.46)); and retaining only terms linear in  $\alpha$  and  $\beta$  in the drift matrix, and only constant terms in the diffusion matrix (Chaturvedi et.al., 1977). This yields the linearised Fokker-Planck equation:

$$\frac{\partial P}{\partial t} = \begin{cases} \frac{\partial}{\partial \alpha} \left[ \chi_1 \alpha + \chi \left( 2\alpha 1\alpha 0 \right)^2 + \alpha^4 \alpha_0^2 \right) + ig_1 \beta - ig_2 \beta^4 \alpha_0^2 \\ -2ig_2 \beta_0^* \alpha \alpha_0 - 2ig_2 \beta 1\alpha_0 1^2 - 2ig_2 \beta_0 \left(\alpha^4 \alpha_0 + \alpha_0^* \alpha\right) \right] \end{cases}$$

$$+\frac{1}{2}\frac{\partial^{2}\left[K_{1}\alpha_{0}^{2}+2ig_{2}\alpha_{0}\beta_{0}\right]}{\partial\alpha^{2}}+\frac{1}{2}\frac{\partial^{2}\left[K_{2}\alpha_{0}\alpha_{1}^{2}+\eta\right]}{\partial\alpha^{2}}+\frac{\partial^{2}\left[ig_{2}\alpha_{0}^{2}\right]}{\partial\alpha^{2}\beta}}{\partial\alpha^{2}\beta}$$
$$+\frac{\partial}{\partial\beta}\left[\delta_{2}\beta+ig_{1}\alpha-ig_{2}\left(2\alpha_{1}\alpha_{0}\alpha_{1}^{2}+\alpha^{2}\alpha_{0}^{2}\right)\right]+\frac{1}{2}\frac{\partial^{2}}{\partial\beta^{2}\beta^{2}}K_{4}\bar{n}+c.c.\right]P$$
(7.29)

where 'C.C.' means:  $\alpha \rightarrow \alpha^+$ ,  $\beta \rightarrow \beta^+$ ,  $\alpha \rightarrow \alpha^*$ ,  $\beta \rightarrow \beta^*$ ,  $i \rightarrow -i$ and the constants  $\delta_1$ ,  $\delta_2$ ,  $\chi$ ,  $g_1$ ,  $g_2$ ,  $\kappa_1$ ,  $\kappa_2$ ,  $\eta$  and  $\kappa_4$  were defined in Chapter Six after Eq. (6.46).

#### 

In order to implement the elimination procedure described in  $\oint 7.3$ , we must write Eq. (7.29) in the form of Eq. (7.7).

This requires a suitable scaling of variables and parameters. As the final form of the Fokker-Planck equation (i.e. after elimination) depends crucially on the choice of scaling, such scaling must be justifiable on physical grounds.

The most obvious scaling concerns the linear damping coefficient  $\chi_1 \sim K_5$ . In the adiabatic limit we are considering, the excitons have a relatively short lifetime and thus  $K_5$  is large in comparison to other parameters. We then expect,

where  $\delta \rightarrow \infty$  corresponds to the adiabatic limit (see  $\oint 7.3$ ).

For the remaining variables, however, the choice of scaling is not as obvious.

A reasonable assumption is that the form of the system's deterministic equations (Eqs. 6.48), should be preserved after scaling. That is, we expect the system's steady state behaviour to remain unchanged after the elimination procedure.

With this in mind, the correct  $\delta^2$  dependence of the operator  $L_1$  follows only if we set  $\chi \sim O(\delta^2)$ . However, the noise terms of  $L_1$  then have a  $\delta^4$  dependence, as  $K_2 \sim \chi^2$ . We are thus not able to follow this approach.

Another method is to evaluate the various parameters and by determining their relative magnitudes, define a suitable scaling.

Hanamura (1974a) investigated the parameters and found the following functional dependence:

$$\chi = \chi \left(\frac{\alpha^3}{\nu}\right) , g_1 = g_1 \left(\sqrt{\frac{\nu}{\alpha^3}}\right) , g_2 = g_2 \left(\sqrt{\frac{\alpha^3}{\nu}}\right)$$

where  $\Omega_{\circ}$  is the Bohr radius and V is the volume. In general, we may choose V >>  $\Omega_{\circ}^{3}$  and thus expect the ratio  $V / \Omega_{\circ}^{3}$  to be very large.
This presents a possible criterion for the scaling of these variables; if we assume

$$\sqrt{\frac{V}{a_0^3}} \sim O(\delta)$$
 (7.31a)

thus,

$$\chi \equiv \chi_1 \chi^{-2}$$
,  $g_1 \equiv c \chi$ ,  $g_2 \equiv d \chi^{-1}$  (7.31b)

where  $\chi_{\iota}$ , c and d are all constants.

The scaling  $\chi \sim O(\gamma^{-2})$  implies, (see the definitions following Eq. (6.46)):

$$K_{2} \sim O(\delta^{-4}) \equiv K_{2}' \delta^{-4}$$
 (7.32a)

and

$$K_{1} \sim O(\delta^{-2}) \equiv K_{1}^{\prime} \delta^{-2}$$
 (7.32b)

where  $K_1'$  and  $K_2'$  are constants.

Also,

$$\delta_{1} = \delta'_{1} + \delta_{1}$$
 (7.33a)

where

Similarly, from the definitions following Eq. (6.46),

$$\eta = \delta'_{I} \eta_{I} + 2 \delta_{II} \bar{n}_{ex}$$
 (7.34a)

(  $\eta_I$  is a constant)

$$\Rightarrow \eta = \eta_{I} \delta^{-4} + 2 \kappa' \bar{n}_{ex} \delta^{2} \qquad (7.34b)$$

We note that there is no dimensional relationship between  $\sqrt{\frac{V}{a_0^3}}$ , and  $\sqrt{\frac{V}{a_0^3}}$  defined in Eq. (7.30). However we expect that K5 and  $\sqrt{\frac{V}{a_0^3}}$  will both become very large.

The scaling defined in Eq. (7.31) will always hold in this semiconductor system, and is not a feature of the adiabatic limit alone. In contrast, the scaling  $K_5 \sim O(\delta^2)$  holds only in the adiabatic limit of large exciton damping.

We stress that the scaling defined by Eqs. (7.31) represents only one possible choice. Clearly there exists a whole range of different scalings. However, our particular choice will produce the desired form of the Fokker-Planck equation, Eq. (7.7) (required to carry out the elimination procedure); and also correctly describes the relative magnitudes of the variables  $\chi$ ,  $g_1$  and  $g_2$ . We believe this to be sufficient justification for the scaling regime defined by Eqs. (7.31).

We redefine the system variables as,

 $d_1 \leftrightarrow d + d_0 \delta$ 

(7.35)

$$d_2 \longleftrightarrow \delta\beta + \beta_c$$

Defining the variables as such results in the cavity mode becoming large, and the exciton variable approaching its deterministic mean as  $\delta$  becomes large. This is clearly the expected behaviour in the adiabatic limit.

All remaining system parameters are not scaled in any way; and thus the scaling of the system in this adiabatic limit is defined by Eqs. (7.31), (7.32), (7.33), (7.34), and (7.35).

Substituting these equations into Eq. (7.29), we find the scaled Fokker-Planck equation,

$$\frac{\partial P}{\partial t} = \left\{ \begin{array}{l} \chi^{2} \frac{\partial}{\partial \alpha} \left[ \kappa' \alpha' + ic\beta - id\beta' \alpha_{o}^{2} - 2id\beta |\alpha_{o}|^{2} \right] + \chi^{2} \frac{\partial^{2}}{\partial \alpha} 2\kappa' \bar{n}ex \\ + \frac{\partial}{\partial \alpha} \left[ \chi_{1} \left( 2\alpha |\alpha_{o}|^{2} + \alpha^{+} \alpha_{o}^{2} \right) - 2id\beta_{o}^{*} \alpha' \alpha_{o} - 2id\beta_{o} \left( \alpha^{+} \alpha_{o} + \alpha_{o}^{*} \alpha' \right) + \chi_{I} \alpha' \right] \\ + \frac{1}{2} \frac{\partial^{2}}{\partial \alpha^{2}} \left[ \kappa_{1}' \alpha_{o}^{2} + 2id\alpha_{o} \beta_{o} \right] + \frac{1}{2} \frac{\partial^{2}}{\partial \alpha^{+} \partial \alpha} \left[ \frac{\kappa_{2}' |\alpha_{o}|^{2}}{\chi^{2}} + \frac{\eta_{I}}{\chi^{4}} \right] + \frac{\partial^{2}}{\partial \alpha \beta} id\alpha_{o}^{2} \\ + \frac{\partial}{\partial \beta} \left[ \chi_{2} \beta + ic\alpha - id(2\alpha |\alpha_{o}|^{2} + \alpha^{+} \alpha_{o}^{2}) \right] + \frac{1}{\chi^{2}} \frac{\partial^{2}}{2} \frac{2\kappa_{4} \bar{n}}{\beta \beta \beta \beta^{+}} + c.c. \right\} P \quad (7.36)$$

We wish to cast Eq. (7.36) into the form of Eq. (7.7). The operator L will be formed from the first two terms of Eq. (7.36). Much of the calculation in the elimination procedure requires knowledge of the explicit form of  $P_s(\underline{\alpha})$  satisfying,

$$L_1 P_s(\underline{\alpha}) = 0$$

To simplify L, and thus more easily obtain  $\rho_s(\underline{\alpha})$  we transform  $\alpha$  to a variable describing an Ornstein-Uhlenbeck process. Such a process represents the simplest of all stochastic systems and is described by the Fokker-Planck equation,

$$\frac{\partial f(\underline{a})}{\partial t} = \left\{ \begin{array}{c} \frac{\partial}{\partial a} & a + \frac{\partial}{\partial a \partial a^{+}} \end{array} \right\}^{D_{1}} + c.c. \left\{ \begin{array}{c} P(\underline{a}) & \underline{\phantom{a}} \end{array} \right\}$$
(7.37)

It has the stationary distribution function,

where  $\mathcal{N}$  is a normalisation factor,  $\mathcal{N} = (\pi D_{i}^{V_{2}})^{-1}$ 

The distribution, Eq. (7.38) predicts the expectation values,

$$\langle \alpha \rangle_s = \int P_s(\underline{\alpha}) d d^2 d = 0$$
 (7.39)

and

$$\langle \alpha^+ \alpha \rangle_s = \int P_s(\underline{\alpha}) \alpha^+ \alpha d^2 \alpha = D_1$$
 (7.40)

 $\oint$ 7.4(c) Transformation to An Ornstein-Uhlenbeck Process

We transform  $\alpha$  as follows,

$$X = \alpha + \frac{1}{k'} \left[ ic\beta - id\beta^{+}\alpha^{2} - 2id\beta |\alpha_{0}|^{2} \right] = \alpha + F_{0} \qquad (7.41a)$$

and

$$X^{+} = a^{+} + F_{o}^{+}$$
 (7.41b)

[where X ,  $\chi^+$  are not necessarily complex conjugates].

Further defining

$$y = \beta$$
,  $y^+ = \beta^+$  (7.42)

[again, y,  $y^{\dagger}$  are not complex conjugate, except in the mean], we obtain the transformations,

$$\frac{\partial}{\partial \alpha} = \frac{\partial}{\partial x} ; \qquad \frac{\partial}{\partial \alpha^{+}} = \frac{\partial}{\partial x^{+}} \qquad ----(7.43a)$$

$$\frac{\partial}{\partial \beta} = F_{1} \frac{\partial}{\partial x} + F_{2} \frac{\partial}{\partial x^{+}} + \frac{\partial}{\partial y} \qquad ----(7.43b)$$

$$\frac{\partial}{\partial \beta^{+}} = F_{2}^{+} \frac{\partial}{\partial x} + F_{1}^{+} \frac{\partial}{\partial x^{+}} + \frac{\partial}{\partial y^{+}}$$

where

and

$$F_2 = \frac{1}{K'} [ida^{+2}]$$
 (7.44c)

The Jacobian of the transformation defined by Eqs. (7.41) is unity. Thus the probability distribution is still properly normalised after this transformation. We note that if the change of variables was such that the Jacobian was not equal to unity, the probability distribution function would have to be suitably transformed in order to yield the correct Fokker-Planck equation.

Substituting Eqs. (7.41), (7.42), (7.43) in Eq. (7.36) yields,  

$$\frac{\partial P}{\partial t} = \begin{cases} \frac{\sqrt{2}}{\partial x} \frac{\partial}{\partial x} K' X + \frac{\sqrt{2}}{2} \frac{\partial^{2}}{\partial x \partial x^{+}} 2K' \bar{n} ex \\ + \frac{\partial}{\partial x} \left[ (x - F_{o}) (2 \chi_{1} | d_{o} |^{2} - 2i d \beta_{o} d_{o} - 2i d \beta_{o} d_{o}^{*}) + (x^{+} - F_{o}^{+}) (\chi_{1} d_{o}^{2} - 2i d \beta_{o} d_{o}) \right] \\ + \frac{1}{2} \frac{\partial^{2}}{\partial x^{2}} \left[ K'_{1} d_{o}^{2} + 2i d d_{o} \beta_{o} \right] + \frac{\partial}{\partial \chi} \left( \frac{\partial}{\partial y} + F_{1} \frac{\partial}{\partial x} + F_{2} \frac{\partial}{\partial x^{+}} \right) i d d_{o}^{2} \\ + \frac{1}{\sqrt{4}} \left( \frac{\partial}{\partial \chi} \chi_{I} (x - F_{o}) + \frac{1}{2} \frac{\partial^{2}}{\partial x \partial x^{+}} \eta_{I} \right) + \frac{1}{2\sqrt{2}} \frac{\partial^{2}}{\partial x \partial x^{+}} K'_{2} | d_{o} |^{2} \\ + \left[ \frac{\partial}{\partial y} + F_{1} \frac{\partial}{\partial \chi} + F_{2} \frac{\partial}{\partial \chi^{+}} \right] \left[ \chi_{2} y + (x - F_{o}) i (c - 2d | d_{o} |^{2}) \\ - i d d_{o}^{2} (x^{+} - F_{o}^{+}) \right] \\ + \frac{1}{2\sqrt{2}} \left( \frac{\partial}{\partial y} + F_{1} \frac{\partial}{\partial x} + F_{2} \frac{\partial}{\partial \chi^{+}} \right) \left( \frac{\partial}{\partial y} + F_{1} \frac{\partial}{\partial \chi^{+}} + F_{2} \frac{\partial}{\partial \chi} \right) 2K_{+} \bar{n} \\ + c \cdot c \cdot \int P$$

$$(7.45)$$

The variable X now describes an Ornstein-Uhlenbeck process and we may write Eqs. (7.45) as,

where

$$L_{1} = \frac{\partial}{\partial x} K' X + \frac{1}{2} \frac{\partial^{2}}{\partial x \partial x^{+}} 2 K' \bar{n} ex + c.c. \qquad ----(7.47)$$

$$\begin{split} L_{2} &= \frac{\partial}{\partial x} \begin{bmatrix} (x - F_{o}) \left( 2 \chi_{1} | d_{o}|^{2} - 2id \beta_{o}^{*} d_{o} - 2id \beta_{o} d_{o}^{*} + iF_{1} \left( c - 2d | d_{o}|^{2} \right) + id d_{o}^{*2} F_{2}^{*} \right) \\ &+ (x^{*} - F_{o}^{*}) \left( \chi_{1} d_{o}^{2} - 2id \beta_{o} d_{o} - id d_{o}^{2} F_{1} - iF_{2}^{*} \left( c - 2d | d_{o}|^{2} \right) \right) \\ &+ \chi_{2} y F_{1} + \chi_{2} y + F_{2}^{*} \end{bmatrix} \\ &+ \frac{1}{2} \frac{\partial}{\partial x^{2}} \begin{bmatrix} K_{1}' d_{o}^{2} + 2i d d_{o} \beta_{o} \end{bmatrix} + \frac{\partial}{\partial x} \left( \frac{\partial}{\partial y} + F_{1} \frac{\partial}{\partial x} + F_{2} \frac{\partial}{\partial x^{*}} \right) i d d_{o}^{2} \\ &+ \frac{\partial}{\partial y} \begin{bmatrix} i x \left( c - 2d | d_{o}|^{2} \right) - i x^{*} d d_{o}^{2} \end{bmatrix} \\ &+ \frac{1}{\chi^{2}} \left( \frac{1}{2} \left\{ \left| \frac{\partial}{\partial y} + F_{1} \frac{\partial}{\partial x} + F_{2} \frac{\partial}{\partial x^{*}} \right|^{2} - \frac{\partial^{2}}{\partial y \partial y^{*}} \right\} 2 K_{4} \tilde{n} + \frac{1}{2} \frac{\partial^{2}}{\partial x \partial x^{*}} K_{2}' | d_{o}|^{2} \right) \\ &+ \frac{1}{\chi^{4}} \left[ \frac{\partial}{\partial x} \chi_{1} \left( x - F_{o} \right) + \frac{1}{2} \frac{\partial^{2}}{\partial x \partial x^{*}} \eta_{1} \right] + c.c. \\ \end{array}$$

and  

$$L_{3} = \frac{\partial}{\partial y} \left[ \lambda_{2} y - i F_{o} \left( c - 2d | d_{o}|^{2} \right) + i d a_{o}^{2} F_{o}^{+} \right] + \frac{1}{2\lambda^{2}} \frac{\partial^{2}}{\partial y \partial y^{+}} 2\kappa_{4} \bar{n} + c.c. \quad (7.49)$$
From Eqs. (7.37) and (7.47) we see,  

$$L_{1} P_{s}(\underline{x}) = O$$

$$\Rightarrow P_{s}(\underline{x}) = \left[ \pi \kappa' \bar{n}_{ex} \right]^{-1} exp \left( -\frac{1 \times 1^{2}}{1 \times 1^{2}} \right) \quad ----(7.50)$$

where

and

We thus define the projection operator,

$$P_{\mathcal{F}}(x,y) = P_{s}(\underline{x}) \int d^{2}x \, \mathcal{F}(x,y) \qquad -----(7.52)$$

for an arbitrary function z(x,y) .

We now show that  $\mathcal{P}L_2 \mathcal{P} = O$  : The operator  $L_2$  (Eq. (7.48)) contains terms only of the form,

i) 
$$\frac{\partial^{n+m}}{\partial x^n \partial x^{+m}} (0, (x, y))$$
 where  $n = 0, 1, 2$   
 $m = 0, 1, 2$ 

and

$$\frac{\partial}{\partial y} \begin{array}{c} \varphi_2(x) \\ \frac{\partial}{\partial y} \end{array} , \begin{array}{c} \frac{\partial^2}{\partial y \partial y^+} \\ \frac{\partial^2}{\partial y \partial y^+} \end{array}$$

Forming  $\beta L_2 \beta$  for terms described by i), we find,

$$\mathscr{P}_{L_{2}}\mathscr{P}_{s}(x,y) = \mathcal{P}_{s}(\underline{x}') \int d^{2}x \frac{\partial^{n+m}}{\partial x^{n} \partial x^{+m}} \mathscr{Q}_{i}(x,y) \mathcal{P}_{s}(\underline{x}) \int d^{2}x'' \mathscr{Z}(x'',y) \qquad (7.53)$$

and as  $F_{\varsigma}(\underline{X})$  and all its derivatives are zero at  $+\infty$  and  $-\infty$  to ensure a bounded distribution function,

$$\int_{-\infty}^{\infty} d^2 x \frac{\partial^{n+m}}{\partial x^n \partial x^{+m}} (\mathcal{Q}_i(x,y) \mathcal{P}_s(\underline{x}) = 0$$
(7.54)

thus terms such as Eq. (7.53) vanish.

Also, for terms described by ii),

$$\begin{split} & \beta L_2 \, \beta_3 (x,y) = P_s \left( \underline{x}' \right) \frac{\partial}{\partial y} \int d^2 x \, \varphi_2(x) \, P_s(\underline{x}) \int d^2 x'' \, z(x'',y) \quad (7.55) \\ & \text{As} \quad \int d^2 x \, \varphi_2(x) \, P_s(\underline{x}) = \varphi_2(\langle x \rangle_s) = O \quad , \quad \text{Eq. (7.55) vanishes.} \\ & \text{Thus, Eqs. (7.53) and (7.55) indicate the desired result:} \end{split}$$

 $\mathcal{P}L_{2}\mathcal{P}=0$ 

Having suitably defined the operators  $L_1$ ,  $L_2$  and  $L_3$  we can follow the procedure defined in  $\oint 7.3$  to adiabatically eliminate the variable X.

Equation (7.49) shows that the operator  $L_3$  has noise terms of order  $\chi^{-2}$  only. Thus, as indicated in  $\phi$ 7.3, as well as the terms of  $L_3$ , the required Fokker-Planck equation must include all terms of order  $\chi^{-2}$  arising in the elimination process.

Hence we need to evaluate the additional terms,

$$\mathcal{P}L_2 L_1 L_2 \widetilde{\mathcal{V}}(S)$$
 (7.56)

and

$$\mathcal{P}L_{2}L_{1}^{-1}[L_{3},\mathcal{P}]\widetilde{\mathcal{V}}(5)$$
 -----(7.57)

Equations (7.49) and (7.52) clearly show,

$$[L_3, 8] = 0$$

and we need only consider the term Eq. (7.56).

We note that expression (7.56) is already of order  $\delta^{-2}$  in the elimination expansion (Eq. (7.27)), thus we may neglect all terms of order  $\delta^{-2}$  occurring in  $L_2$  when evaluating Eq. (7.56).

For convenience we rewrite  $L_2$  as,

$$L_{2} = L_{2}^{x_{1}} + L_{2}^{x_{2}} + L_{2}^{xy_{1}} + L_{2}^{xy_{2}} + L_{2}^{y_{1}} + L_{2}^{y_{2}} - \dots - (7.58)$$

where

$$L_{2}^{X_{1}} = \frac{\partial}{\partial X} \left[ \tilde{b}_{0} + \tilde{b}_{1} X + \tilde{b}_{2} X^{\dagger} \right] + \frac{1}{2} \frac{\partial^{2}}{\partial X^{2}} \tilde{b}_{3} + \frac{\partial^{2}}{\partial X \partial X^{\dagger}} \tilde{b}_{4} - \dots (7.59a)$$

$$L_{2}^{X_{2}} = \left( L_{2}^{X_{1}} \right)^{\dagger} - \dots (7.59b)$$

$$L_{2}^{y_{1}} = \frac{\partial}{\partial y} \left[ i \times M_{o} - i \times^{+} d \alpha_{o}^{2} \right] ; \quad L_{2}^{y_{2}} = \left( L_{2}^{y_{1}} \right)^{+} \quad (7.61)$$

and

$$\begin{split} \widetilde{b}_{0} &= -F_{0}\left(2\chi_{1}|a_{0}|^{2} - 2id\beta_{0}^{*}a_{0} - 2id\beta_{0}a_{0}^{*}\right) - iF_{0}F_{1}\left(C - 2d|a_{0}|^{2}\right) - iF_{0}F_{1}^{*}da_{0}^{2} \\ &- F^{+}\left(\chi_{1}a_{0}^{2} - 2id\beta_{0}a_{0}\right) - ida_{0}^{2}F_{1}F_{0}^{*} - iF_{0}^{*}F_{2}\left(C - 2d|a_{0}|^{2}\right) + \delta_{2}\left(yF_{1} + y^{+}F_{2}^{+}\right)^{*} \\ \widetilde{b}_{1} &= 2\chi_{1}|a_{0}|^{2} - 2id\beta_{0}^{*}a_{0} - 2id\beta_{0}a_{0}^{*} + iF_{1}\left(C - 2d|a_{0}|^{2}\right) + ida_{0}^{*2}F_{2}^{+} \\ \widetilde{b}_{2} &= \chi_{1}a_{0}^{2} - 2id\beta_{0}a_{0} - ida_{0}^{2}F_{1} - iF_{2}\left(C - 2d|a_{0}|^{2}\right) \\ \widetilde{b}_{3} &= K_{1}a_{0}^{2} + 2id\beta_{0}a_{0} \quad ; \quad \widetilde{b}_{4} = iF_{2}da_{0}^{2} \\ M_{0} &= -iF_{1}K' \end{split}$$

and  $F_0$ ,  $F_1$ ,  $F_2$  are defined in Eqs. (7.44), and c, d,  $\chi_1$  and  $K'_1$  are defined in Eqs. (7.31), (7.32). Defining

$$\widetilde{v}(s) = P_s(\underline{x}) \widetilde{P}(\underline{y})$$

we may write the explicit form of expression (7.56),

$$\mathscr{P}_{L_{2}}L_{1}^{-1}L_{2}\widetilde{v}(5) = P_{S}(\underline{x}')\int d^{2}x \left[L_{2}^{y_{1}}+L_{2}^{y_{2}}\right]L_{1}^{-1}L_{2} P_{S}(\underline{x})\widetilde{P}(\underline{y}) \qquad (7.62)$$

We have dropped the terms  $L_2^{x_1}$ ,  $L_2^{x_2}$ ,  $L_2^{xy_1}$  and  $L_2^{xy_2}$  from the operator  $L_2$  occurring on the left hand side of  $L_1^{-1}$  in Eq. (7.62). These terms contain derivatives with respect to X and thus yield vanishing integrals, as indicated by Eq. (7.54).

To evaluate expression (7.62), we use the general relationship (Gardiner, 1982),

$$\int d^2 x f_1(\underline{x}) L_1^{-1} f_2(\underline{x}) P_S(\underline{x}) = -\int_0^{\infty} dt \langle f_1(\underline{x},t) f_2(\underline{x},0) \rangle_s \quad (7.63)$$
  
In this case,  $f_1$  contains only linear terms in  $\times$ , i.e.

$$f_1 = L_2^{y_1} (L_2^{y_2}) = i M_0 X - i d_{x_0}^2 X^{\dagger}$$

Thus, as X describes an Ornstein-Uhlenbeck process, the only non-vanishing correlations arising from Eq. (7.63) will be,

$$D = \int_{0}^{\infty} \langle x^{\dagger}(t) \chi(0) \rangle_{s} dt \qquad ----(7.64a)$$

From Eq. (7.51) and the quantum regression theorem (Louisell, 1973), we find,

We also note that the derivatives with respect to X occurring in  $L_2$  act directly on  $P_s(\underline{x})$  as,

$$\frac{\partial}{\partial x} P_{s}(\underline{x}) = - \frac{x^{+}}{\overline{n}ex} P_{s}(\underline{x}) \qquad ----(7.65a)$$

and

$$\frac{\partial}{\partial x^{+}} P_{s}(\underline{x}) = - \underline{x} P_{s}(\underline{x}) - (7.65b)$$

Using Eqs. (7.65), Eq. (7.62) becomes,

where  $\lambda$  is the expression occurring within the curly brackets in Eq. (7.66b).

The reduced Fokker-Planck equation for the variable y in the adiabatic limit now follows from Eq. (7.27),

$$S \tilde{v}(s) = \mathscr{P}[L_3 - \mathscr{Y}^2 L_2 L_1^{-1} L_2] \tilde{v}(s) + v(o)$$
 \_\_\_\_\_(7.68)  
Inverting Eq. (7.68),

$$\Rightarrow \frac{d}{dt} \tau(t) = \mathscr{B} \left[ L_3 - \mathscr{V}^2 L_2 L_1^{-1} L_2 \right] \tau(t)$$

Defining  $v(t) = P_s(\underline{x}) \hat{p}(\underline{y})$  we find (using Eq. (7.67))

 $\frac{d}{dt} P_{s}(x) \hat{P}(y) = P_{s}(\underline{x}) \int d^{2}x' L_{3} P_{s}(x') \hat{P}(y) + \delta^{-2} P_{s}(\underline{x}) \mathcal{L} \hat{P}(y) \qquad (7.69)$ As L<sub>3</sub> depends only on Y, and  $\int d^{2}x' P_{s}(x') = 1$ , Eq. (7.69) becomes,

Thus, from Eqs. (7.49) and (7.66) we find the reduced Fokker-Planck equation:

$$\frac{\partial P}{\partial t} = \left\{ \frac{\partial}{\partial y} \left[ y \left( \aleph_{2} + \frac{1}{k'} \left[ c^{2} - 4 c d |\alpha_{0}|^{2} + 3 d^{2} (|\alpha_{0}|^{2})^{2} \right] + \aleph^{-2} q_{1} \right) + \aleph^{-2} q_{2} y^{+} \right] + \frac{1}{\gamma^{2}} \left[ \frac{1}{2} \frac{\partial^{2}}{\partial y^{2}} (2 \kappa_{4} \bar{n} + 2 \frac{M_{0} d \alpha_{0}^{2}}{K'} (2 \bar{n} e_{x} + 1)) + \frac{1}{2 \kappa'} \frac{\partial^{2}}{\partial y \partial y^{+}} + d^{2} (|\alpha_{0}|^{2})^{2} (1 + \bar{n} e_{x}) \right] \right\} + c.c. \right\} P \qquad (7.70)$$

where

$$q_{1} = \frac{L}{K'} (da_{0}^{2} e_{2}^{+} - M_{0}e_{1})$$

$$q_{2} = \frac{L}{K'} (da_{0}^{2} e_{1}^{+} - M_{0}e_{2})$$

and

$$e_{1} = F_{1} \left[ -(2\chi_{1}|d_{0}|^{2} - 2id\beta_{0}d_{0} - 2id\beta_{0}d_{0}) - iF_{1}(c - 2d|d_{0}|^{2}) - iF_{1}^{+}dd_{0}^{2} + \chi_{2} \right] + F_{2} \left[ -(\chi_{1}|d_{0}|^{2} - 2id\beta_{0}d_{0}) - idd_{0}^{2}F_{1} - iF_{2}(c - 2d|d_{0}|^{2}) \right]$$

$$\begin{aligned} e_2 &= F_2^+ \left[ -(2\chi_1 | d_0|^2 - 2id\beta^* d_0 - 2id\beta_0 d_0^*) - iF_1(c - 2d|d_0|^2) + \delta_2 \\ &- iF_2^+ d_0^2 \right] + F_1^+ \left[ -(\chi_1 d_0^2 - 2id\beta_0 d_0) - idd^2 F_1 - iF_2(c - 2d|d_0|^2) \right] \\ \text{and 'c.c.' means} \quad y \to y^+, \ i \to -i \\ &(\text{etr.}) \end{aligned}$$

Equation (7.70) completely describes the behaviour of the cavity mode, including statistical fluctuations, in the limit of heavily damped excitons.

We note that the drift coefficient contains terms of order  $\delta^{-2}$ which did not arise from the operator  $L_3$ . Such terms, derived in part from diffusion terms, would not be generated by the approximate elimination methods described in  $\oint 7.2$ . That is, the drift terms of the corresponding reduced Fokker-Planck equation derived by these methods stem solely from the drift coefficient of the original Fokker-Planck equation. This is an indication of the shortcomings of these approximate methods. The diffusion coefficients of Eq. (7.70) are all of order  $\chi^{-2}$ . This is as expected, as L<sub>3</sub> (the leading term in the elimination expansion, Eq. (7.27)) only had noise terms of order  $\chi^{-2}$ .

Finally in this section we note that the form of Eq. (7.70) depends critically on our initial choice of scaling. For example, this scaling leads to the effects of exciton-exciton collisions (described by the parameter  $\chi$  ) being evident only in terms of order  $\chi^{-2}$  in the drift coefficient.

As we derive statistical properties of the system from this equation, we must be quite sure that the scaling is justified on physical grounds.

#### $\phi$ 7.5 Photon Statistics for Cavity Modes

Having derived a linearised two-dimensional Fokker-Planck equation we can now use the theory developed by Chaturvedi et.al. (1977) to derive expectation values and spectra for the system.

From this theory, we find that a two-dimensional system:  $\Delta = (\alpha_1, \alpha_2)$  obeying the linearised Langevin equations,

(where A, D are matrices,  $\xi(t)$  is a vector of stochastic terms) has correlation matrix

$$\underline{\underline{C}} = \begin{pmatrix} \langle \alpha_1^2 \rangle & \langle \alpha_1 \alpha_2 \rangle \\ \langle \alpha_2 \alpha_1 \rangle & \langle \alpha_2^2 \rangle \end{pmatrix}$$
(7.72)

where

$$\underline{\underline{C}} = \underline{\underline{P}} \cdot \det(\underline{\underline{A}}) + (\underline{\underline{A}} - I \cdot Tr(\underline{\underline{A}})) \underline{\underline{P}} (\underline{\underline{A}} - I \cdot Tr(\underline{\underline{A}}))^{\prime}$$
(7.73)  
2 Tr(\underline{\underline{A}}) det(\underline{\underline{A}})

and spectrum,

$$\sum_{i=1}^{n} (\Delta_{i} + i\omega)^{-1} = (\Delta_{i}^{T} - i\omega)^{-1} \qquad (7.74)$$

We apply these results to our system by forming the linearised Langevin equation corresponding to Eq. (7.70),

$$\frac{d}{dt} \begin{pmatrix} y \\ y^{+} \end{pmatrix} = - \begin{pmatrix} a_{1} & a_{2} \\ a_{2}^{+} & a_{1}^{+} \end{pmatrix} \begin{pmatrix} y \\ y^{+} \end{pmatrix} + \begin{pmatrix} d_{1} & d_{2} \\ d_{2} & d_{1}^{+} \end{pmatrix}^{\frac{1}{2}} \begin{pmatrix} \mathcal{E}_{1}(t) \\ \mathcal{E}_{2}(t) \end{pmatrix}$$
(7.75)

where  $\xi_{1}(t)$ ,  $\xi_{1}(t)$  are Gaussian stochastic terms, and

$$a_{1} = -\aleph_{2} + \frac{1}{\kappa} \left[ c^{2} - 4 c d |a_{0}|^{2} + 3 d^{2} (|a_{0}|^{2})^{2} \right] + \aleph^{-2} q_{1}$$

$$a_{2} = \aleph^{-2} q_{2}$$
(7.76)

$$d_{1} = \frac{1}{\lambda^{2}} \left[ 2\kappa_{4} \bar{n} + \frac{2M \cdot dd^{2}}{\kappa'} (2\bar{n}e_{x} + 1) \right]$$

$$d_{2} = \frac{1}{\lambda^{2}\kappa'} \left[ 2M^{2}_{0} \bar{n}e_{x} + d^{2}(1d_{0}1^{2})^{2}(1+\bar{n}e_{x}) \right]$$

From Eqs. (7.73), (7.74) and (7.75) we then find,

$$\begin{split} & \subseteq = \frac{1}{(a_{1}+a_{1}^{+})(1a_{1}1^{2}-1a_{2}1^{2})} \begin{pmatrix} a_{1}^{+2}d_{1} + a_{2}^{2}d_{1}^{+} - 2a_{1}^{+}a_{2}d_{2} & -a_{1}^{+}a_{2}^{+}d_{1} - a_{1}a_{2}d_{1}^{+} \\ + d_{1}(1a_{1}1^{2}-1a_{2}1^{2}) & + 2d_{2}1a_{1}1^{2} \\ -a_{1}^{+}a_{2}^{+}d_{1} - a_{1}a_{2}d_{1}^{+} & a_{2}^{+2}d_{1} + a_{1}^{+}a_{1}^{+}-2a_{1}a_{2}^{+}d_{2} \\ + 2d_{2}1a_{1}1^{2} & + d_{1}^{+}(1a_{1}1^{2}-1a_{2}1^{2}) \\ & (7.77) \end{split}$$

$$\begin{split} & \subseteq (\omega) = (\Delta_{1}\Delta_{2})^{-1} \begin{pmatrix} (a_{1}^{+}-i\omega)[d_{1}(a_{1}^{+}+i\omega)-d_{2}a_{2}] & -a_{2}^{+}[d_{1}(a_{1}^{+}+i\omega)-d_{2}a_{2}] \\ -a_{2}[d_{2}(a_{1}^{+}+i\omega)-d_{1}^{+}a_{2}] & +(a_{1}-i\omega)[d_{2}(a_{1}^{+}+i\omega)] \\ -a_{2}[d_{2}(a_{1}^{+}+i\omega)-d_{1}^{+}a_{2}] & +(a_{1}-i\omega)[d_{2}(a_{1}^{+}+i\omega)] \\ -a_{2}[-d_{2}a_{2}^{+}+d_{1}^{+}(a_{1}+i\omega)] & -a_{2}^{+}[-d_{1}a_{2}^{+}+d_{2}(a_{1}+i\omega)] \\ -a_{2}[-d_{2}a_{2}^{+}+d_{1}^{+}(a_{1}+i\omega)] & +(a_{1}-i\omega)[-d_{2}a_{2}^{+} \\ +d_{1}^{+}(a_{1}+i\omega)] \end{pmatrix} \\ & (7.78) \end{split}$$

## $\oint 7.5(a)$ Moment Calculations

From Eqs. (7.76), we see

 $d_1, d_2 \sim O(\delta^{-2})$ 

$$\mathcal{Q}_{1} \sim \mathcal{O}(\mathcal{Y}^{\circ}, \mathcal{Y}^{-2})$$

 $\alpha_{2} \sim O(\gamma^{-2})$ 

and

As each term of  $\underline{\subseteq}$  and  $\underline{\underline{\subseteq}}(\omega)$  contain either a factor of  $d_1$ or  $d_2$ , we obtain the highest order contributions to these matrices by neglecting terms of order  $\overset{}{\delta}^{-2}$  occurring in  $a_1$  and completely neglecting  $a_2$ .

Thus to highest order in X , we find the total emitted intensity (to first order in the linearised asymptotic expansion),

$$I_y = |\beta_0|^2 + \langle y^+ y \rangle = |\beta_0|^2 + C_{12}$$

$$\Rightarrow Iy = n_2 - \frac{2[\bar{n}e_x(c^2 - 4cdn_i + 5d^2n_i^2) + d^2n_i^2]}{\chi^2 K'[\chi_2 + \frac{1}{k_i}(c^2 - 4cdn_i + 3d^2n_i^2)]}$$
(7.79)

where  $n_2 = |\beta_0|^2$  = steady state emitted intensity in deterministic limit,

and

 $n_1 = |\alpha_0|^2$  = deterministic exciton intensity.

From Eq. (7.79) we note that to this order, the emitted intensity depends only on the strength of the exciton-light coupling (i.e. the parameters C, d). There is no dependence on exciton-exciton collisions assumed small in this limit.

We may also derive the second order correlation function,

$$g^{1}(0) = 1 + \frac{2}{n_{2}} \left[ \langle y^{+}y \rangle + \operatorname{Re}\left(\beta_{0}^{*} \langle y^{2} \rangle\right) \right]$$
  
= 1 + \frac{2}{n\_{2}} \left[ C\_{12} + \operatorname{Re}\left(\beta\_{0}^{\*} C\_{11}\right) \right] -----(7.80)

The evaluation of expression (7.80) requires knowledge of the deterministic mean of the field mode amplitude,  $\beta_0$ . As  $C_{11}$  and  $C_{12}$  are both positive, the possibility of observing photon antibunching depends on the form of  $\beta_0$ .

## $\oint$ 7.5(b) Fluctuation Spectrum

Again, we retain only terms of highest order in  $\lambda$  in  $\leq (\omega)$ and find the spectrum of the transmitted light,

$$[S(\omega)]_{21} = \frac{d_2}{|Q_1|^2 + \omega^2}$$
(7.81)

### $\phi$ 7.6 Adiabatic Elimination of Cavity Mode

## ∮7.6(a) Scaling

We now consider the limit in which the excitons have a much longer lifetime than the cavity mode.

The obvious choice of scaling in this case is,

$$\delta_2 \sim O(\delta^2) \Rightarrow K_4 \sim O(\delta^2)$$
 (large cavity damping)

Thus let,

$$\delta_2 \equiv K_c \delta^2$$
 and  $K_4 \equiv K_c \delta^2$  -----(7.82)

In  $\oint 7.4$  (b) we defined the scaling of the semiconductor parameters  $\chi$ ,  $g_1$  and  $g_2$ . Such a scaling is determined solely from our theoretical model and is independent of the various adiabatic limits we adopt.

Thus we again assume this scaling and set,

$$\chi = \chi_1 \chi^{-2}$$
,  $K_1 = K_1' \chi^{-2}$ ,  $K_2 = K_2' \chi^{-4}$  -----(7.83)  
 $g_1 = c \chi$ ,  $g_2 = d \chi^{-1}$ 

(where  $\chi_1$ ,  $\kappa'_1$ ,  $\kappa'_2$ , C and d are constants).

In this case the exciton damping coefficient is no longer scaled, i.e.

hence from Eqs. (7.33a) and (7.34a) we find

 $K_5 \sim O(\delta^{\circ})$ 

$$\chi_1 = \chi^{-4} \chi_1 + K_5$$
 \_\_\_\_\_(7.84)  
 $\eta = \eta_1 \chi^{-4} + 2K_5 \bar{n}_{ex}$ 

We also assume,

$$E \sim O(\lambda^2) \Rightarrow E \equiv E, \lambda^2$$
 (7.85)

The large driving field scaling described by Eq. (7.85) is necessary to produce the  $\chi^2$  dependence of the operator  $L_1$ .

In this limit, we do not scale the variable  $\alpha_2$  (cavity mode) but define

$$d_1 \rightarrow \delta d_1$$
,  $\beta_1 \rightarrow \delta \beta_1$  -----(7.86)

Thus, in the adiabatic limit  $X \to \infty$  , the exciton amplitude becomes very large.

We note that the scaling defined in Eqs. (7.85) and (7.86) is consistent with large exciton amplitude in the adiabatic limit.

Substituting Eqs. (7.82), (7.83), (7.84), (7.85) and (7.86) into the semiconductor Fokker-Planck equation (Eq. (6.46)) we find the scaled equation:

$$\frac{\partial P}{\partial t} = \begin{cases} \chi^2 \frac{\partial}{\partial a_2} [K_c a_2 - E_1 + i c a_1 - i d a_1 a_1 \beta_1] + \frac{\chi^2}{2} \frac{\partial^2}{\partial a_2} 2K_c \bar{n} \\ \frac{\partial P}{\partial a_2} \end{bmatrix}$$

$$+ \frac{\partial}{\partial \alpha_{1}} \left[ \alpha_{1} \left( \chi_{1} \chi^{-4} + K_{5} \right) + \alpha_{1} \alpha_{1} \beta_{1} \chi_{1} + i c \alpha_{2} - i d \left( \beta_{2} \alpha_{1}^{2} + 2 \alpha_{2} \beta_{1} \alpha_{1} \right) \right] \\ + \frac{1}{2} \frac{\partial^{2}}{\partial d_{1}^{2}} \left[ \chi^{-2} K_{1}^{\prime} \alpha_{1}^{2} + 2 i d \chi^{-2} \alpha_{1} \alpha_{2} \right] + \frac{1}{2} \frac{\partial^{2}}{\partial \alpha_{1} \partial \beta_{1}} \left[ \chi^{-4} K_{2}^{\prime} \alpha_{1} \beta_{1} + 2 \frac{K_{5} \overline{n} e_{x}}{\chi^{2}} \right] \\ + \frac{\partial^{2}}{\partial \alpha_{1} \partial \alpha_{2}} \left[ i d_{2} \alpha_{1}^{2} \right] + c.c. \right\} P$$
(7.87)

As in  $\oint 7.4$ , we wish to write Eq. (7.87) in the form of Eq. (7.7). The operator  $L_1$  of such an equation will consist of the first two terms (and their complex conjugates) of Eq. (7.87). We see now that the choice  $E \sim O(\delta^2)$  gives rise to the desired  $\delta^2$  dependence of  $L_1$ . Following the method of  $\oint 7.4$ , we ease computations by transforming  $d_2$  to a variable describing an Ornstein-Uhlenbeck process.

# $\oint$ 7.6(b) Transformation to Ornstein-Uhlenbeck Process ( $d_2$ )

This is achieved via the definitions:

and

$$X = \alpha_1$$
;  $X^+ = \beta_1$  (7.89)

Again, the pairs of variables  $(x, x^+)$  and  $(y, y^+)$  are complex conjugate only in the steady state.

We thus find the transformations:

$$\frac{\partial}{\partial d_2} = \frac{\partial}{\partial y} , \quad \frac{\partial}{\partial \beta_2} = \frac{\partial}{\partial y^+}$$

$$\frac{\partial}{\partial \beta_2} = G_1 \frac{\partial}{\partial y} + G_2 \frac{\partial}{\partial y^+} + \frac{\partial}{\partial x}$$

$$\frac{\partial}{\partial \beta_1} = G_1^+ \frac{\partial}{\partial y^+} + G_2^+ \frac{\partial}{\partial y} + \frac{\partial}{\partial x^+}$$
(7.90)
(7.90)
(7.91)

where

$$G_{0} = \frac{1}{\kappa_{c}} \left[ ic X - E_{1} - id X^{2} X^{+} \right] - \frac{1}{\kappa_{c}} \left[ ic - 2id X^$$

and

$$G_2 = \frac{1}{\kappa_c} [i dx^{+2}]$$
 (7.92c)

Substituting Eqs. (7.90), (7.91) and (7.92) into Eq. (7.87) we find the required Fokker-Planck equation,

$$\frac{\partial P}{\partial t} = (\lambda^2 L_1 + L_2 + L_3)P - \dots (7.93)$$

where

$$L_{1} = \frac{\partial}{\partial y} \kappa_{c} y + \frac{1}{2} \frac{\partial^{2}}{\partial y \partial y^{+}} 2 \kappa_{c} \bar{n} + c.c. \qquad (7.94)$$

$$L_{2} = \frac{\partial}{\partial x} [icy - idx^{2}y^{+} - 2idyxx^{+}] \\ + \left( \begin{array}{c} G_{1} \frac{\partial}{\partial y} + G_{2} \frac{\partial}{\partial y^{+}} \\ & -idE(y^{+} - G_{\bullet}^{+})x^{2} + 2(y - G_{\bullet})xx^{+}] \end{array} \right) \\ + \frac{\partial}{\partial y} \left[ \begin{array}{c} G_{1} \frac{\partial}{\partial y} + G_{2} \frac{\partial}{\partial y^{+}} + \frac{\partial}{\partial x} \end{array} \right] idx^{2} + termsO(8^{-2}) + c.c.$$

(7.95)

$$L_{3} = \frac{\partial}{\partial x} \left[ x \left( \delta_{I} \delta^{-4} + K_{5} \right) + X^{2} \chi^{+} k_{1} + ic \left( -g_{0} \right) + i d \chi^{2} g_{0}^{+} + 2i d g_{0} \chi \chi^{+} \right] \right] \\ + \frac{1}{2} \frac{\partial^{2}}{\partial \chi^{2}} \left[ \delta^{-2} K_{1}' \chi^{2} - \delta^{-2} 2i d \chi g_{0} \right] \\ + \frac{1}{2} \frac{\partial^{2}}{\partial \chi \partial \chi^{+}} \left[ \delta^{-4} K_{2}' \chi \chi^{+} + \eta_{I} \delta^{-6} + \delta^{-2} K_{5} 2 \bar{n} e \chi \right] + c.c. \qquad (7.96)$$

Clearly the operator  $L_{\star}$  describes an Ornstein-Uhlenbeck process, thus,

where

and

$$|u|^2 = \bar{n} = \langle y^+ y \rangle_s$$
 \_\_\_\_\_(7.98)

and

We thus define the projection operator

$$P_1 z(x, y) = P_s(y) \int d^2 y z(x, y)$$
 (7.100)

for an arbitrary function ~ ~ .

As the operator  $L_2$  (including terms of order  $\delta^{-2}$  ) consists only of terms of the form,

it follows that  $\mathcal{P}_1 L_2 \mathcal{P}_1 = O$  (See the discussion of  $\oint 7.4(c)$ , especially Eqs. (7.53), (7.54), and (7.55)).

The highest order noise terms occurring in L<sub>3</sub> are  $O(\gamma^{-2})$ . Thus we must again include all terms of order  $\gamma^{-2}$  arising in the elimination process. As  $[L_3, \beta_1] = 0$ , we need only evaluate,

We need not consider the explicit form of the terms of order  $\chi^{-2}$  occurring in  $L_2$  as these only appear in terms of order  $\chi^{-4}$  in the elimination expansion.

To simplify further calculations, we write  $L_2$  as,

$$L_{2} = L_{2}^{x_{1}} + L_{2}^{x_{2}} + L_{2}^{y_{1}} + L_{2}^{y_{2}} + L_{2}^{xy_{1}} + L_{2}^{xy_{2}} - -----(7.102)$$

where

$$L_{2}^{x_{1}} = \frac{\partial}{\partial x} \left[ m_{1}y + m_{2}y^{+} \right] ; \quad L_{2}^{x_{2}} = \left( L_{2}^{x_{1}} \right)^{+} \qquad (7.103)$$

$$L_{2}^{y_{1}} = \frac{\partial}{\partial y} \left( m_{3} + m_{4}y + m_{5}y^{+} \right) + \frac{\partial^{2}}{\partial y^{2}} m_{6} + \frac{\partial^{2}}{\partial y^{2}} m_{7} ; \quad L_{2}^{y_{2}} = \left( L_{2}^{y_{1}} \right)^{+} \qquad (7.104)$$

and

$$L_{2}^{xy_{1}} = \frac{\partial^{2}}{\partial y \partial x} m_{g}$$
;  $L_{2}^{xy_{2}} = (L_{2}^{xy_{1}})^{+}$  (7.105)

and where

$$m_{1} = ic - 2idxx^{+}, m_{2} = idx^{2},$$

$$m_{3} = G_{1}(xk_{5} + x^{2}x^{+}\chi_{1} - icG_{0} + idx^{2}G_{0}^{+} + 2idG_{0}xx^{+})$$

$$+ G_{2}^{+}(x^{+}k_{5} + x^{+2}x\chi_{1} + icG_{0}^{+} - idx^{+2}G_{0} - 2idG_{0}^{+}xx^{+}),$$

$$m_{4} = G_{1}(ic - 2idxx^{+}) + G_{2}^{+}(idx^{+2}), m_{6} = iG_{1}dx^{2},$$

$$m_{5} = -G_{1}idx^{2} + G_{2}^{+}(-ic + 2idxx^{+}), m_{7} = iG_{2}dx^{2},$$

$$m_{8} = idx^{2}$$
(7.106)

The integrals occurring in Eq. (7.107) give rise to time integrals of correlation functions:

As y describes an Ornstein-Uhlenbeck process, the only non-zero correlation functions are,

$$\langle y^{+}(t) y(0) \rangle_{s} = \bar{n} e^{-\kappa_{t} t}$$
 (7.109)

Thus using Eqs. (7.108) and (7.109) in Eq. (7.107) we find,  $P_1 L_2 L_1^{-1} L_2 \widetilde{v}(s) = -P_s(\underline{y}) \frac{\overline{n}}{K_c} \left\{ \frac{\partial}{\partial x} (2idx^+ m_2 + 2idx m_1 + 2idx m_1^+ + m_1 m_3 + m_2 m_3^+ + 2idx^+ m_8) + \frac{\partial^2}{\partial x^2} (2m_1 m_2 + m_1 m_8) + \frac{\partial^2}{\partial x^2} (2m_1 m_1 + m_1 + m_1 m_1 + m_1$ 

Following the discussion of  $\oint 7.4(c)$ , we find the reduced Fokker-Planck equation:

$$\frac{\partial}{\partial t} \widehat{P}(\underline{x}) = [L_3 + \underline{y}^{-2} \underline{f}'] \widehat{P}(\underline{x}) \qquad ----(7.112)$$

where  $L_3$  is given by Eq. (7.96) and  $\chi'$  is defined by Eqs. (7.110) and (7.111).

Retaining only terms of order  $\delta^{-2}$  and higher in  $L_3$ , Eq. (7.112) becomes,

$$\frac{\partial \hat{P}(x)}{\partial t} = \begin{cases} \frac{\partial}{\partial x} \left( [x K_5 + x^2 x^4 \hat{x}_1 - ic G_0 - id x^2 G_0^4 + 2i d G_0 x x^4] \right) \\ + 2i d G_0 x x^4] \\ + 8^{-2} \frac{\bar{n}}{K_c} [2i d x^4 m_2 + 2i d x m_1 + 2i d x m_1^4 + m_1 m_3 + m_2 m_3^4 + 2i d x^4 m_3] \\ + 2i d x^4 m_3] \end{cases} \\ + \frac{1}{2 N^2} \left( \frac{\partial^2}{\partial x^2} [K_1' x^2 - 2i d x G_0 + 4 m_1 m_2 + 2 m_1 m_3] \\ + \frac{\partial^2}{\partial x \partial x^4} [2 K_5 \bar{n} e x + 2 m_1 m_1^4 + 2 m_2 m_3^4 + m_2^3] + c.c. \right) \hat{P}(x)$$
(7.113)

(where 'C.C.' means,  $x \rightarrow x^+$ ,  $i \rightarrow -i$ )

Equation (7.113) represents the Fokker-Planck equation for the exciton mode X , in the adiabatic limit of heavily damped cavity modes.

We note again the presence of terms of order  $\delta^{-2}$  in the drift matrix - not generated by the approximate methods of  $\oint 7.2$ .

If required, one may now apply the linearised fluctuation theory of  $\oint 7.5$  to this system and obtain statistical information.

### $\oint 7.7$ Discussion

We have seen how the projection operator method can be employed to successfully reduce the dimensionality of a Fokker-Planck equation of a nonlinear system. This represents the most accurate and systematic method known to adiabatically eliminate variables in stochastic systems. However, the correct choice of scaling, crucial to the procedure, is not always obvious. This scaling determines the final form of the reduced Fokker-Planck equation and thus the predicted statistical properties of the system, so we must be able to satisfactorally justify our choice. In general we base this choice on physical considerations (e.g. the magnitude of damping rates) and experimental evidence. We also scale variables and parameters to preserve the system's deterministic behaviour.

Once the scaling is defined, the most straightforward approach is to transform the variable to be eliminated to one describing an Ornstein-Uhlenbeck process. This is the best understood of all stochastic systems, having a known stationary distribution, and thus simplifies calculations greatly.

Essential to the definition of a Ornstein-Uhlenbeck process is the thermal noise defining the diffusion coefficient. We note that a common practice in other approximate elimination methods is to assume thermal noise is negligible, and then proceed with the elimination of variables. This is clearly a major error as the more accurate projection operator method depends critically on the inclusion of thermal noise.

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#### CHAPTER 8

### INTERBAND EXCITATION IN SEMICONDUCTORS

### ∮8.1 Introduction

As well as in the experiments involving GaAs, optical bistability has also been observed in InSb by Miller et.al., (1979). The large dielectric constant of this material inhibits the Coulomb effects necessary for exciton formation. The excitons are thus screened and we cannot use the theory developed in Chapter Six to explain the experimentally found bistability.

Weaire et.al., (1979) estimated the refractive index of InSb and found it corresponded to a third order susceptibility  $(\chi^{(3)})$  much larger than that expected due to valence electron effects.

To explain the existence of such a high  $\chi^{(3)}$ , Miller et.al. (1980) proposed a model which included two associated effects: saturation and power broadening.

It was assumed that the incident field excited electrons from valence to conduction bands. However, the excitation in these experiments occurred at energies less than the gap; and as impurity effects are saturated in InSb, transitions from impurity levels could not account for this. The mechanism for interband absorption is thus unclear.

Nevertheless, Miller et.al. (1980) do present a model of interband excitation in which each vertical transition is viewed as a two-level oscillator. Radiative transitions couple only pairs of states having the same k-value in different bands. Using standard theory (Yariv, 1975) they find the susceptibility of a collection of N two-level systems (with only the lower states initially occupied) and sum over the appropriate band states to find the refractive index. This so-called 'direct saturation' model adequately explains the system's nonlinearities. There do though, exist problems in the inclusion of relaxation terms. The only damping included in the model is interband radiative recombination, in which the electron and hole forming the two-level system recombine with the subsequent emission of a photon. However, an electron forming the upper state of a given two-level system may be scattered to another state in the conduction band, thereby destroying the electron-hole pair. Thus the effects of this intraband interaction must be included into the theory.

In this chapter we present a very simple model to explain the intracavity interaction between a light field and a semiconductor such as InSb. We assume the electrons and holes (in respective conduction and valence bands) can be modelled as N two-level systems and form the interactive medium.

Our theory relies on several simplifying assumptions, explained in later sections of this chapter.

By deriving a master equation for such a system, we include interand intra-band damping in a consistent fashion. Bistability, similar to that of atomic systems is found in the steady state.

#### 68.2 The Model

We consider the intracavity interaction of coherent light with a two-band semiconductor. We assume the incident field couples strongly to only one cavity mode (frequency  $\omega$  ), which in turn couples to the medium. The Hamiltonian for such a lossless system was derived in Chapter Six,  $\oint 6.2$ ,

 $H = H_{e.m.} + H_{b} + H_{b} + H_{INT}$  ------(8.1)

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where

$$H_{e.m.} = \omega b^{\dagger} b$$
 (8.2)

(b is a boson operator associated with a cavity mode of frequency  $\omega$ ). H<sub>D</sub> is the Hamiltonian term describing the coupling of the driving field (amplitude E and frequency  $\omega_{L}$  ) with the cavity mode:

$$H_{\mathfrak{o}} = i\hbar (Eexp(-i\omega_{\mathfrak{c}}t)b^{\dagger} - h.c.) \qquad ------(8.3)$$

 $H_{s.c.}$  is the Hamiltonian for the semiconductor system alone:

$$H_{s.c.} = \mathcal{E}_{\kappa_{c}} E_{c}(\kappa_{c}) a_{\kappa_{c}}^{\dagger} a_{\kappa_{c}} + \mathcal{E}_{\kappa_{v}} E_{v}(\kappa_{v}) d_{\kappa_{v}}^{\dagger} d_{\kappa_{v}}$$

$$+ \mathcal{E}_{q} \upsilon(q) \left\{ \mathcal{E}_{\kappa_{c},\kappa_{c}} a_{\kappa_{c}+q}^{\dagger} a_{\kappa_{c}-q}^{\dagger} a_{\kappa_{c}} a_{\kappa_{c}} + \mathcal{E}_{\kappa_{v},\kappa_{v}} d_{\kappa_{v}}^{\dagger} d_{\kappa_{v}}^{\dagger} d_{\kappa_{v}}^{\dagger} d_{\kappa_{v}} d_{\kappa_{v}}^{\dagger} d_{\kappa_{v}} d_$$

where  $\alpha_{R_c}, d_{R_v}$  are fermion operators for electrons and holes respectively;  $k_c$  and  $R_v$  are wave-vectors for conduction and valence band states;

$$E_{r}(R_{r}) = \frac{\hbar^{2}R_{r}^{2}}{2m_{h}}$$
;  $E_{c}(R_{c}) = E_{g} + \frac{\hbar^{2}R_{c}^{2}}{2m_{e}}$ 

where  $m_{e}$ ,  $m_{h}$  are the respective electron and hole effective masses;

 $\mathcal{V}(q)$  is the Fourier transform of  $V(\underline{r}) = \frac{e^2}{|\underline{r}|}$ , the Coulomb interaction potential.

 $H_{\text{INT}}$  describes the interaction between the cavity mode and the semiconductor,

$$H_{INT} = \hbar \mathcal{E}(ba_{\mu}^{\dagger} d_{\nu}^{\dagger} g + h.c.)$$
 (8.5)

where g is the light-matter coupling constant (see Eq. (6.21b)).

As well as the terms of the Hamiltonian, Eq. (8.1), we must also include the effects of damping into our model - these are discussed later.

In forming the system Hamiltonian we have applied the single mode approximation to the modes of the cavity. We now assume the single driven cavity mode excites only one electron across the gap, that is it couples strongly to only one two-level oscillator.

We thus confine our attention to one electron state and one hole state only and assume all other electronic states form a reservoir for the system.

The Hamiltonian, Eq. (8.1) then becomes,

$$H = H_{Res.} + H_s + H_{ve}$$
 (8.6)

where  $H_{S}$  is the Hamiltonian describing the system involving the cavity, electron and hole states of interest:

$$H_{s} = \hbar \omega b^{\dagger} b + \hbar \omega e a^{\dagger}_{\kappa_{\circ}} a_{\kappa_{\circ}} + \hbar \omega_{h} d^{\dagger}_{\kappa'} d_{\kappa'} d_{\kappa$$

The first three terms of Eq. (8.7) describe the free part of the Hamiltonian. The fourth term describes the light-matter interaction; and the final term stems from eq. (8.4) and describes an electron-hole interaction.

Hges is the electronic reservoir Hamiltonian:

$$H_{Res.} = \left\{ H_{s.c.} \left( \mathcal{R}_c \neq \mathcal{R}_o, \mathcal{R}_u \neq \mathcal{R}'_o \right) \right\}_{Free \text{ part}} (8.8)$$

Finally,  $H_{ve}$  describes the interaction between the electron and hole states of interest  $(R_{v} = k_{o}'; k_{c} = k_{o})$  and electronic reservoir modes:

$$H_{ve} = \frac{1}{2} \left\{ a_{R_{o}}^{+} \hat{\Sigma} e_{i} a_{R_{i}}^{+} a_{R_{i}}^{+} a_{R_{i}}^{+} a_{R_{o}} \hat{\Sigma} e_{2} a_{R_{i}}^{+} a_{R_{i}}^{+} a_{R_{i}}^{+} \hat{\Sigma} e_{2} a_{R_{i}}^{+} a_{R_{i}}^{+} a_{R_{i}}^{+} \hat{\Sigma} e_{2} a_{R_{i}}^{+} a_{R_{i}}^{+} a_{R_{i}}^{+} d_{R_{i}}^{+} d_{R_{i}}^$$

Each of the terms of Eq. (8.9) represents a form of electronic damping. The first three refer to scattering of the electron into the reservoir states of the conduction and valence bands; and the next three terms describe scattering of the hole into reservoir states. The final term of Eq. (8.9) describes the destruction of the electron-hole pair (twolevel oscillator) through scattering into conduction and valence band states.

All the terms of Eq. (8.9) describe a type of intraband relaxation which, as indicated in  $\oint 8.1$ , should be included into our model.

We note that the first six terms of Eq. (8.9) refer to the intraband scattering of isolated electrons and holes; whereas the final term refers explicitly to the decay of an electron-hole pair via intraband relaxation. As we are concerned only with the behaviour of the electron-hole pair forming the two-level oscillator, we ignore the first six terms of Eq. (8.9), assuming these processes have a negligible effect on the twolevel system. This is a major simplifying assumption.

Thus, Hve describing intraband damping is,

 $H_{ve} = a_{R_o}^{\dagger} d_{R_o}^{\dagger} \pounds e_7 a_{R_i} d_{R_2}^{\dagger} + h.c. = a_{R_o}^{\dagger} d_{R_o}^{\dagger} Q_{e_i} + h.c. \quad (8.10)$ where  $Q_{e_i}$  is a combination of electron and hole reservoir operators.

The discussion of  $\oint 8.1$  showed that as well as intraband damping we must also consider radiative damping of the two-level system. This has the Hamiltonian

 $H_{\text{elec.damp.}} = a_{\text{ko}}^{\dagger} d_{\text{ko}}^{\dagger} Q_{e_2} + \text{h.c.} \qquad (8.11)$ 

where  $Q_{e_2}$  represents an operator associated with a radiation field forming a reservoir. Equation (8.11) describes a process in which the electron-hole pair recombines to emit a photon. Finally we must include damping of the cavity mode: the Hamiltonian assumes the usual form,

 $H_{cavity} = b^{\dagger}Q_{F} + h.c. \qquad (8.12)$  damp.

where  $Q_F$  is a reservoir operator (mode of radiation field).

Thus the Hamiltonian for our system is,

$$H = H_R + H_S + H_{damping} \qquad -----(8.13)$$

where  $H_R$  is the Hamiltonian describing the reservoir formed from conduction and valence band states and modes of the radiation field.

Hs is the system Hamiltonian, given by Eq. (8.7) and Hdamping refers to system damping,

Hamping = Hve + Helec. + H cavity (8.14)where the terms of Eq. (8.14) are given by Eqs. (8.10), (8.11) and (8.12).

Equation (8.13) describes a system in which a driven cavity mode excites a single electron from the conduction band to the valence band: This excitation is viewed as a two-level oscillator.

As such, the system is described in terms of the operators  $a, d, a^{\dagger}$ and  $d^{\dagger}$ , and is thus four-dimensional. We can simplify the Hamiltonian by recalling the discussion of  $\oint 1.3(b)$  (ii); in which the formal equivalence between a two-level system and a spin- $\frac{1}{2}$  system was shown.

That is, we assume the correspondence,

$$a^{+}d^{+} \leftrightarrow \sigma^{+}$$
$$a d \leftrightarrow \sigma^{-}$$
$$\frac{1}{2}(a^{+}a - d^{+}d) \leftrightarrow \sigma^{3}$$

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where  $\sigma^+$ ,  $\sigma^-$  and  $\sigma^3$  are the spin-flip operators defined in  $\oint 1.3(b)$  (ii).

If we set our zero of energy as follows



(where  $\hbar \omega_{\bullet}$  is the energy separation between electron and hole). Then the Hamiltonian, Eq. (8.13), becomes,

$$H = H_{R} + \pi \omega_{0} \sigma^{3} + \pi \omega_{0} b^{\dagger} b + \pi \sigma_{1} \sigma^{\dagger} \sigma^{-} + \pi (g \sigma^{\dagger} b + h.c.)$$

$$+ i \pi (E b^{\dagger} e^{-i \omega_{L} t} - h.c.) + (Q_{e_{1}} \sigma^{\dagger} + Q_{e_{2}} \sigma^{+} + h.c.)$$

$$+ (Q_{F} b^{\dagger} + h.c.) - (8.15)$$

Eq. (8.15) thus describes a system of a driven two-level system in which we have included the effects of radiative (interband) and intraband damping of the electron-hole pair. Also present is the term

 $\upsilon_{\iota} \ \sigma^{+} \sigma^{-}$  , describing a type of dipole-dipole interaction.

Apart from this term, Eq. (8.15) is identical to the Hamiltonian describing a driven two-level atom (Drummond and Walls, 1980b). As bistability is found in such a system, we expect similar behaviour in the semiconductor system.

#### $\oint 8.2(a)$ Master Equation

Transforming to a frame rotating at the laser frequency  $\omega_{\iota}$  , we find the master equation,

where the last two terms of Eq. (8.16) were derived using the quantum theory of damping, discussed in Chapter One,  $\oint 1.3(d)$ ;

$$\frac{\partial \rho}{\partial t}\Big|_{\substack{\text{Elec.} \\ \text{damp.}}} = \mathcal{E}_{i=1}^{2} \forall e_{i} \left[ (1 + \bar{n}e_{i})([\sigma^{-}\rho,\sigma^{+}] + [\sigma^{-},\rho\sigma^{+}]) \right]$$

+  $\overline{n}e_i([\sigma^+\rho,\sigma^-]+[\sigma^+,\rho\sigma^-])]$  -----(8.17)

where  $(\delta e_1, \overline{n} e_1)$  and  $(\delta e_2, \overline{n} e_1)$  are the damping coefficients and reservoir occupation numbers for intraband and interband damping respectively.

$$\frac{\partial \rho}{\partial t} = \frac{1}{2b\rho b^{\dagger}} - \frac{b^{\dagger}b}{\rho b^{\dagger}} - \frac{b^{\dagger}b}{\rho} + 2\bar{n} [[b,\rho],b] \right\} - (8.18)$$

where  $\delta_1$  is the cavity damping coefficient and  $\bar{n}$  the thermal occupation number of the reservoir of radiation modes.

Also,  $\delta_1 = \omega_{\bullet} - \omega_{\perp}$  is the relative detuning of the laser with the two-level system; and  $\delta_2 = \omega - \omega_{\perp}$  is the cavity detuning.

In order to produce a more realistic model of a semiconductor we consider the effect of many such two-level oscillators; and consider the interaction of a coherent driving field with N two-level systems.

The master equation, Eq. (8.16), then becomes,

$$\frac{\partial \rho}{\partial t} = -i \hat{\mathcal{E}}_{v} \delta_{v} \left[ \sigma_{v}^{2}, \rho \right] - i \delta_{2} \left[ \dot{b}^{\dagger} b, \rho \right] - i \hat{\mathcal{E}}_{v\mu} \nabla_{v\mu} \left[ \sigma_{v}^{\dagger} \sigma_{v}^{\dagger}, \rho \right]$$

$$- i \hat{\mathcal{E}}_{v} \left[ g_{v} \sigma_{v}^{\dagger} b + g_{v}^{*} \sigma_{v}^{\dagger} b^{\dagger}, \rho \right] + \left[ Eb^{\dagger} - E^{*}b, \rho \right]$$

$$+ \frac{\partial \rho}{\partial t} \left| \frac{Elec.}{damp.} + \frac{\partial \rho}{\partial t} \right| \frac{cavity}{damp.}$$

$$(8.19)$$

where the summation over  $arphi,\, \mu$  extends over the N oscillators.

In the deterministic limit, we neglect quantum fluctuations and assume a complete decorrelation of system variables. We then find the average values of system operators from Eq. (8.19): <u>Cavity</u>:

$$\dot{\beta} = -(\vartheta_1 + i\delta_2)\beta + E - i\hat{\xi}_{\gamma} g_{\gamma}^* \langle \sigma_{\gamma}^- \rangle \qquad (8.20)$$
where  $\beta = \langle b \rangle$  and we have set  $\bar{n} = 0$ .

Setting  $\bar{n}_{e}$ , =  $\bar{n}_{e_1} = 0$  in Eqs. (8.17) we find the oscillator equations:

#### Oscillators:

$$\langle \sigma_{\nu}^{*} \rangle = \left[ i \left( \upsilon_{\nu} - \delta_{\nu} \right) - \delta_{E\nu} \right] \langle \sigma_{\nu}^{*} \rangle + 2i \underbrace{\xi_{\mu}}_{\nu \neq \mu} \upsilon_{\mu\nu} \langle \sigma_{\nu}^{*} \rangle \langle \sigma_{\nu}^{*} \rangle$$

$$- 2ig_{\nu}^{*} \langle \sigma_{\nu}^{*} \rangle \beta^{*} (8.21)$$

$$\langle \dot{\sigma}_{v}^{-} \rangle = \left[ -i(\upsilon_{v} - \delta_{v}) - \vartheta_{Ev} \right] \langle \sigma_{v}^{-} \rangle - 2i \mathcal{E}_{\mu} \quad \upsilon_{\nu\mu} \langle \sigma_{v}^{-} \rangle \\ + 2ig_{v} \langle \sigma_{v}^{-} \rangle \beta$$

$$(8.22)$$

$$\langle \sigma_{\gamma}^{*} \rangle = ig_{\gamma}^{*} \langle \sigma_{\gamma}^{-} \rangle \beta^{*} - ig_{\gamma} \langle \sigma_{\gamma}^{+} \rangle \beta$$
$$- \aleph_{E_{\gamma}} [2 \langle \sigma_{\gamma}^{*} \rangle + 1] \qquad (8.23)$$

where summation over  $\forall$  is implied in Eqs. (8.21), (8.22) and (8.23) and  $\forall \epsilon_{\gamma} = (\forall e_{\gamma} + \forall e_{2})_{\gamma}$ 

### $\oint 8.3$ Steady State Analysis

To investigate bistability in the system, we wish to solve Eqs. (8.20), (8.21), (8.22), and (8.23) in the steady state. The summations over Vand  $\mu$  in these equations complicate such calculations.

We now adopt a further simplifying assumption: We assume the light field interacts equally strongly with each of the N oscillators (i.e.  $g_{\gamma} = g = \text{constant}$ ); and that the light-oscillator does not affect the behaviour of the neighbouring oscillators. We may thus replace the summation in Eq. (8.20) by the factor N,

i.e. 
$$\hat{\xi}_{\gamma} g_{\gamma}^* \langle \sigma_{\gamma}^- \rangle \rightarrow Ng^* \langle \sigma_{\gamma}^- \rangle$$

and hence find the equation of motion for the cavity mode:

$$Eq\mu. 8.20 \Rightarrow \dot{\beta} = -(\aleph_1 + i\delta_2)\beta + E - iNg^* \langle \sigma_{\nu}^- \rangle \qquad (8.24)$$

The oscillator equations of motion are complicated by the term,

To simplify this term we assume the steady state values of each oscillator are approximately equal:

$$\Rightarrow$$
 In steady state,  $\langle \sigma_{\mu}^{+} \rangle \approx \langle \sigma_{\nu}^{+} \rangle$ 

The summation in Eq. (8.21) then becomes,

$$\mathcal{E}_{\mu} \mathcal{T}_{\nu\mu} \langle \sigma_{\nu}^{*} \rangle \langle \sigma_{\nu}^{*} \rangle \xrightarrow{s.s.} \langle \sigma_{\nu}^{*} \rangle \langle \sigma_{\nu}^{*} \rangle \mathcal{E}_{\mu} \mathcal{T}_{\nu\mu}$$

Assuming it is possible to perform the summation over the  $u_{\nu}$  , we thus find

Substituting Eq. (8.26) into Eqs. (8.21), (8.22), (8.23) we can obtain stationary solutions for this system by setting  $\langle \dot{\sigma}^+ \rangle = \langle \dot{\sigma}^- \rangle = \langle \dot{\sigma}^+ \rangle = 0$ 

However, this procedure generates a cubic equation describing the steady state value of  $\langle \sigma^{\gamma} \rangle$ . As this is difficult to solve analytically, we employ a further simplifying assumption and use a perturbative method of solution, treating  $\tilde{v}$  as an expansion parameter, i.e. we let,

$$\langle \sigma_{\gamma}^{3} \rangle_{ss} = \langle \sigma_{\sigma}^{3} \rangle + \tilde{\nabla} \langle \sigma_{\gamma}^{3} \rangle + \tilde{\nabla}^{2} \langle \sigma_{2}^{3} \rangle + \cdots - (8.27)$$

By adopting the expansion (8.27), we assume the interaction between different oscillators (described by Eq. (8.26)) produces only a small perturbation to the light-matter interaction.

Zeroth Order:

$$\langle \sigma_{o,\gamma}^{+} \rangle_{ss} = - \frac{2ig^{*}\beta^{*}(\eta^{2} + \delta_{E})}{(i\eta - \delta_{E})\omega}$$
 (8.28)

First Order:

$$\langle \sigma_{i,\nu}^{+} \rangle_{s.s.} = \frac{2ig^{*}\beta^{*}(-i\eta - \delta_{E})(|6|g|^{2}|\beta|^{2}\eta - \partial^{2})}{\partial^{2}(\partial - 2v_{i}\eta + 2iv_{i}\delta_{E})}$$
 (8.29)

where  $\eta = v_1 - \delta_1$ ,  $\vartheta = 4 i g i^2 i \beta j^2 + 2(\eta^2 + \delta_E^2)$ 

### $\oint 8.3(a)$ Equation of State

We obtain the equation of state for the system by substituting Eqs. (8.28), (8.29) in Eq. (8.24) and setting  $\dot{\beta} = 0$ :

Zeroth Order:

$$E = \beta \left( \chi_{1} + i \delta_{2} - \frac{2 N I g I^{2} (i \eta - \chi_{E})}{4 I g I^{2} I \beta I^{2} + 2 (\eta^{2} + \chi_{E}^{2})} \right) - (8.30)$$

First Order:

$$E = \beta \left( \vartheta_1 + i \vartheta_2 + 2N I g I^2 (i \eta - \vartheta_{\bar{e}}) (I \theta I g I^2 | \beta I^2 \upsilon_1 \eta - \mathcal{D}^2) \right) \qquad (8.31)$$

Equations (8.30) and (8.31) illustrate the steady state behaviour of the semiconductor system.

The first order equation (8.31) describes the system in which we have included the effects of interactions between the N two-level oscillators.

Equation (8.30), the zeroth order approximation, greatly resembles the equation of state of a system of N driven two-level atoms (Drummond, 1979). We now compare Eq. (8.30) with the two-level atom case.

Introducing the definitions:

$$\delta_{I} = \delta_{I} + i \delta_{2} = \delta_{I} (I + i \delta_{A}) \qquad (8.32a)$$
  
$$\delta_{II} = i \eta - \delta_{E} = \delta_{E} (i \delta_{B} - I) \qquad (8.32b)$$

And the scaling (c.f. Eqs. (3.20), Chapter Three),

$$C = \frac{N \log^2}{2 \sqrt[3]{3} \sqrt[3]{5}}$$
(8.33)

$$X = \beta \sqrt{\frac{2 |q|^2}{\chi_{E}^2}}$$
;  $Y = \frac{E}{\chi_{I}} \sqrt{\frac{2 |q|^2}{\chi_{E}^2}}$  (8.34)

Eq. (8.30) becomes,

$$Y = X \left[ 1 + i\delta_A + \frac{2C(1 - i\delta_B)}{1 + \delta_B^2 + 1\times 1^2} \right]$$
(8.35)

Equation (8.35) is identical to the state equation for a travelling wave interacting with two-level atoms (Drummond, 1979).

We may thus use the results of the atomic system to determine the conditions for bistability in the semiconductor system.

#### 

#### i) Absorptive Bistability

We note firstly that the semiconductor and atomic systems are not identical; because of the presence of  $\, {\cal V}_I \,$  in the 'detuning' term  $\eta \,$  .

Thus,  $\delta_1 = \delta_2 = 0$  in the semiconductor system does not correspond to the absorptive regime, as  $\delta_B \neq 0$ .

However, when  $\delta_1 = v_1$  we have the case  $\gamma = 0$  and Eq. (8.35) becomes,

$$Y = X \left[ 1 + \frac{2C}{1 + |X|^2} \right]$$
(8.36)

which is identical to the absorptive bistability state equation derived by Bonifacio and Lugiato (1976).

Equation (8.36) predicts bistability in the system when:

$$C > 4$$
 \_\_\_\_\_(8.37)

#### ii) Cavity on Resonance

This case arises when  $\delta_A = O$ ,  $\delta_B \neq O$ . Following Drummond (1979), we find bistability occurs when,

$$C \gg 1$$
 and  $C^2 > \frac{27\delta\bar{B}}{4}$  (8.38)

This situation can occur even if the oscillators are on resonance, as,

$$\delta_1 = 0 \Rightarrow \delta_B = \delta_E \tau_i \neq 0$$

#### iii) Cavity Detuned

We consider now the case  $\delta_A \neq 0$  and  $\delta_B = 0$ . This occurs only if  $v_1 = \delta_1$ .

Then, for c >> 1 bistability occurs if (Drummond, 1979),

$$C^2 > \frac{27 \delta_A^2}{4}$$
(8.39)

#### iv) Dispersive Bistability

Finally, if  $\delta_A \neq 0$ ,  $\delta_B \neq O$  we have two situations (Drummond, 1979):

(a) If  $\delta_A \delta_B < O$  then if C,  $|\delta_A|$  and  $|\delta_B|$  are all very much greater than unity, bistability occurs when

(b) If  $\delta_A \delta_B > O$  then bistability occurs if,

$$C > \frac{1}{2} |\delta_A \delta_B|$$
 (8.41)

Hence, (subject to all simplifying assumptions) the bistability of the semiconductor system is completely described in terms of the behaviour of the atomic system. The major difference between the two systems is the presence of  $V_i$  in the term  $\eta$ . The term  $U_i$  originates from a type of dipole-dipole interaction ( $\sigma * \sigma^-$ ) which does not occur in the atomic system. The effect is to shift the resonant frequency of the oscillators.

#### 

We now consider Eq. (8.31), describing the behaviour of a system in which oscillator-oscillator interactions have been included.

Using the definitions, Eqs. (8.32), (8.33) and (8.34), Eq. (8.31) becomes,

$$Y = X \begin{bmatrix} 1 + i\delta_{A} - \frac{4c(1-i\delta_{B})[8 v_{1}\eta x^{2} - 4\delta_{E}^{2}(1+|x|^{2}+\delta_{B}^{2})^{2}]}{4(1+|x|^{2}+\delta_{B}^{2})^{2}[2\delta_{E}^{2}(1+|x|^{2}+\delta_{B}^{2}) - 2v_{1}\eta - 2iv_{1}\delta_{E}]} \end{bmatrix}_{(8,42)}$$

To examine the change in the system's behaviour due to the first order perturbation, we graphically compare Eq. (8.42) with the zeroth order equation of state, Eq. (8.35), for specific values of system parameters.

This is illustrated by Fig. 8.1 - a plot of X versus Y: parameters have been chosen such that we are within the bistable region.

The solid curve in Fig. 8.1 describes Eq. (8.35) and the dotted curve corresponds to Eq. (8.42) for  $U_1 = 10$ . Hence, even when  $U_1$ is the same order as the other system parameters, the effect of the first order perturbation is minimial.

We thus conclude that the zeroth order perturbation is sufficient to describe the semi-conductor system: the effects of oscillatoroscillator interactions can be ignored.


Fig. 8.1. Variation of Output Field (X) with Input Field (Y) for Oscillator-Light System (C = 50.,  $\delta_1 = 10.$ ,  $\delta_A = 0$ ,  $\delta_B = 5.$ ; DOTTED CURVE,  $v_1 = 10.$ )

## 

We have thus presented a fully quantum mechanical theory of interband excitation in semiconductors.

The fermion system of electrons and holes was viewed as a collection of N electron-hole pairs, each behaving as a two-level oscillator (the upper level of which corresponded to an electron state and the lower level corresponded to a hole state).

The applicability of our model depends on the validity of the many simplifying assumptions incorporated into the theory.

That is, the system Hamiltonian we used was derived from first principles in Chapter Six. However, in order to obtain results from this many body Hamiltonian, certain approximations were made.

Firstly, the only type of intraband relaxation included in the model was that in which the electron and hole comprising the oscillator were simultaneously scattered into fermion reservoirs in their respective bands. Intraband scattering in which only the electron (upper level of the oscillator) is scattered into a reservoir while the hole (lower level) remains in the same state was considered unlikely. We thus neglected Hamiltonian terms describing such single electron or hole scattering.

Secondly, we assumed the interaction between the light field and each of the N oscillators was identical; and each oscillator-light interaction was completely independent. These conditions will be satisfied for suitable field strengths and if the oscillators are sufficiently widely spaced to ensure negligible interference between oscillators.

Thirdly, the stationary value of a given oscillator variable (e.g. upper state population) was assumed equal for each oscillator. 199

Finally, we assumed the interaction between oscillators (described by the coupling coefficient  $\mathcal{V}_{\nu\mu}$ ) was small in comparison to the lightoscillator interaction. We then used  $\widetilde{\mathcal{V}} = \underbrace{\mathcal{E}}_{\mathcal{V}_{\nu}\mu}$  as an expansion parameter in a perturbative method of solution.

Our model thus presents only a very simple picture of the lightsemiconductor system.

Nevertheless, effects such as inter-band and intra-band relaxation of electron-hole pairs were systematically included into our theory. These effects were manifested in the equation of state via the parameter

 $\delta_E$ ; thus both intra- and inter-band scattering gives rise to the same macroscopic damping effects.

Also, analysis showed that zeroth order perturbation terms were sufficient to describe the behaviour of the system. To this order, the system was shown to exhibit bistability similar to that of a two-level atomic system (subject to the validity of all the simplifying assumptions we adopted).

Finally, we note that such a spin-½ system can be used to describe a system of Frenkel excitons (Haken, 1977). As Frenkel excitons characterise molecular solids, optical bistability may occur in such solids if all parameters are such as to satisfy the appropriate conditions.

## APPENDIX - PROOF OF EQUATION (5.34)

We prove this expression by induction: Firstly, we show it is valid  
for 
$$n = 2, 3$$
.  
 $n = 2$ : Equ. 5.34  $\Rightarrow$   $(m) = \sqrt{2!} B_{d_1\beta_1}^+ B_{d_2\beta_2}^+ |0\rangle$   
Equ. 5.33  $\Rightarrow \sqrt{\frac{1}{2!}} B_{d_1\beta_1}^+ B_{d_2\beta_2}^+ |0\rangle = \sqrt{\frac{1}{2!}} (b_{\alpha_1\beta_1} - \mathcal{E}_{\gamma_5} b_{\alpha_1\beta_1} b_{\delta\gamma_1} b_{\delta\gamma_1}) (b_{d_2\beta_2}^- - \mathcal{E}_{\gamma_5'} b_{\alpha_1\beta_1} b_{\delta\gamma_1} b_{\delta\gamma_1}) (b_{d_2\beta_2}^- - \mathcal{E}_{\gamma_5'} b_{\alpha_1\beta_1} b_{\delta\gamma_1} b_{\delta\gamma_1} b_{\delta\gamma_2}) (b_{d_2\beta_2}^- - \mathcal{E}_{\gamma_5'} b_{\alpha_1\beta_1} b_{\delta\gamma_1} b_{\delta\gamma_2} b_{\delta\gamma_2}) |0\rangle$   
 $= \sqrt{\frac{1}{2!}} (b_{\alpha_1\beta_1} b_{\alpha_2\beta_2} - \mathcal{E}_{\gamma_5} b_{\alpha_1\beta_2} b_{\alpha_2\beta_1}) |0\rangle$  (1)  
From the definition of the two-boson state (Eq. (5.23));  
 $|m\rangle = \sqrt{\frac{1}{2!}} \mathcal{E}_p(-1)^p P b_{\alpha_1\beta_1} b_{\alpha_2\beta_2} - b_{\alpha_1\beta_2} b_{\alpha_2\beta_1} |0\rangle$  (2)  
where  $P$  means a permutation of the  $\alpha'_5$  ( $\beta'_5$ ) with fixed  $\beta(\alpha')$ .

where P means a permutation of the  $\alpha$ 's ( $\beta$ 's) with fixed  $\beta$  ( $\alpha$ ) As (1) = (2), we find Eq. (5.34) holds for n = 2.

Similarly, for 
$$n = 3$$
 Eq. (5.34)  $\Rightarrow$ 

$$|m) = \sqrt{3!} B_{\alpha_{1}\beta_{1}} B_{\alpha_{2}\beta_{2}} B_{\alpha_{3}\beta_{3}} |0)$$

$$= \sqrt{\frac{1}{3!}} (b_{\alpha_{1}\beta_{1}} - \mathcal{E}_{x_{1}\delta_{1}} b_{\alpha_{1}\delta_{1}} b_{\delta_{1}\beta_{1}} b_{\delta_{1}\delta_{1}} b_{\delta_{1}\delta_{2}}) (b_{\alpha_{2}\beta_{2}} - \mathcal{E}_{x_{2}\delta_{2}} b_{\alpha_{2}\lambda_{2}} b_{\delta_{2}\beta_{2}} b_{\delta_{2}\delta_{2}} b_{\delta_{2}\delta_{2}} b_{\delta_{2}\delta_{2}} b_{\delta_{2}\delta_{2}} b_{\delta_{2}\delta_{2}} b_{\delta_{3}\delta_{3}} b_{\delta$$

$$= \sqrt{3!} (b_{\alpha_1\beta_1} b_{\alpha_2\beta_2} b_{\alpha_3\beta_3} - b_{\alpha_1\beta_1} b_{\alpha_3\beta_1} b_{\alpha_3\beta_3} - b_{\alpha_1\beta_3} b_{\alpha_3\beta_1} b_{\alpha_3\beta_2} \\ - b_{\alpha_1\beta_1} b_{\alpha_2\beta_3} b_{\alpha_3\beta_3} + b_{\alpha_1\beta_1} b_{\alpha_3\beta_1} b_{\alpha_3\beta_3} + b_{\alpha_1\beta_3} b_{\alpha_3\beta_1} b_{\alpha_3\beta_2}) |0)$$

which is equivalent to the definition of the three-boson state, obtained from Eq. (5.23)

Finally, we must show that if Eq. (5.24) holds for  $\,n\,$  , then it holds for  $\,n\,$  + i  $\,$  : we assume,

$$B_{\alpha_1\beta_1}^{\dagger}$$
 ...  $B_{\alpha_n\beta_n}^{\dagger} 10) = \mathcal{E}_{\rho} (-1)^{\rho} P B_{\alpha_1\beta_1}^{\dagger} \dots B_{\alpha_n\beta_n}^{\dagger} 10)$  (3)

then, 
$$B_{di\beta_1}^{\dagger}$$
, ...,  $B_{dn\beta_n}^{\dagger}$ ,  $B_{dn+1\beta_{n+1}}^{\dagger}$ ,  $B_{dn+1\beta_{n+1}}^{\dagger}$ ,  $E_p(-1)^p P b_{di\beta_1}^{\dagger}$ ,  $b_{dn\beta_n}$ ,  $B_p(-1)^p P b_{di\beta_1}^{\dagger}$ ,  $b_{dn\beta_n}^{\dagger}$ ,  $b_{dn\beta_n}^{\dagger$ 

$$= \dot{b}_{dn+1}\beta_{n+1} \hat{\varepsilon}_{p} (-1)^{p} P \dot{b}_{d1}\beta_{1} \dots \dot{b}_{dn\beta_{n}} lo) - \hat{\varepsilon}_{p} (-1)^{p} P \hat{\varepsilon}_{p=1}^{-1} \dot{b}_{dn+1}\beta_{p} \dot{b}_{dp}\beta_{n+1} \prod_{\substack{i=1\\i\neq p}}^{n} \dot{b}_{di}\beta_{i} lo)$$
(4)

The terms of Eq. (4) produce all the required permutations, and we may write,

Equ. (4) 
$$\Rightarrow \mathcal{E}_{p}(-1)^{p'} p' b_{\alpha n+1} \beta_{n+1} b_{\alpha} \beta_{1} \dots b_{\alpha n} \beta_{n} 10)$$

where P' means a permutation of the set (d1,..,dn+1) with ( $\beta_1,..,\beta_{n+1}$ ) fixed, or vice versa.

Equations (3) and (4) indicate

$$B_{\alpha_{1}\beta_{1}}^{+} \dots B_{\alpha_{n}\beta_{n}}^{+} |0\rangle = \mathcal{E}_{p}(-1)^{p} P b_{\alpha_{1}\beta_{1}}^{+} \dots b_{\alpha_{n}\beta_{n}}^{+} |0\rangle$$

$$\Rightarrow B_{\alpha_{1}\beta_{1}}^{+} \dots B_{\alpha_{n+1}\beta_{n+1}}^{+} |0\rangle = \mathcal{E}_{p}(-1)^{p} P b_{\alpha_{1}\beta_{1}}^{+} \dots b_{\alpha_{n+1}\beta_{n+1}}^{+} |0\rangle$$

which is the required result.

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