ASPECTS AND APPLICATIONS OF TWO-TIME AVERAGES IN OPEN QUANTUM MARKOFFIAN SYSTEMS

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Very often a theoretical physicist is confronted with systems of exceedingly high complexity about which he requires only a limited amount of information. The familiar problems of statistical mechanics come immediately to mind. The master equation method has been developed in both formal and practical spheres to an extent which provides us now with a most elegant and powerful approach to such problems.

ABSTRACT

The use of a quantum-mechanical operator master equation for the description of an open Markoffian system is discussed in two particular aspects. In both the evaluation of operator averages at two times is a central feature. We deal first with the concept of detailed balance, so familiar from classical statistical mechanics, and hope to bring the various usages of this term under the common perspective provided by the formal generalisation of classical results.

The application of detailed balancing principles to quantum systems obeying Markoffian master equations is discussed and the formal generalisation of the classical principle of detailed balance introduced. A view of detailed balance as the macroscopic consequence of microreversibility is taken. Here the close association between two-time averages and detailed balance requires that the various properties of the former be developed as a basis to our discussions. Both operator and phase-space formulations for evaluating averages are therefore discussed, and from them the respective operator and phase-space expressions for quantum detailed balance constructed. From here the classical limit with $\hbar \to 0$ is shown to correspond to the introduction of detailed balance to open quantum Markoffian systems via a literal interpretation of "quantum-mechanical Fokker-Plank processes". The relationship of full quantum detailed balance to the detailed balancing of transitions within the energy representation, such as is encountered in the Pauli master equation, is established through introducing the concept of diagonal master equations. For these

utilisation of the energy representation allows a direct identification with the Pauli situation. Finally, a generalised master equation accounting for a variety of reservoir couplings is derived, and studied in relation to both the operator detailed balance and the diagonal master equations for thermal equilibrium. It is shown that the presence of certain features in the reservoir interactions generally ensure the failure of full detailed balance, and simultaneously provides deviations from the diagonal form. These observations are to be associated with an inadequacy in the approximations of the master equation method.

A second section of this work tackles the theoretical description of a particular physical system within the framework of the master equation formalism. The concept of the open Markoffian system is applied in relation to resonance fluorescence and the properties of two-time averages used in the determination of field correlation functions. Very intense illumination, which gives rise to the dynamical Stark effect, is studied with particular interest.

Within the context of a quantum-mechanical master equation for incident field plus atomic scattering centre we develop a full description of the resonance fluorescence phenomenon. From a stationary autocorrelation function fluorescence spectra corresponding to arbitrary incident field strengths may be evaluated for the conditions pertaining under atomic saturation. Spectra are also available for the nonstationary field associated with the transient region of atomic dynamics. Both semiclassical and one-photon approximations are illuminated in a simple fashion. Particular interest is taken in deriving the second-order correlation function for the scattered light. This is readily available from our formalism and exhibits the interesting feature of photon-antibunching at weak incident intensities. The

region of weak scattering is familiar in its description by the Lorentzian electron oscillator model and this purely quantum feature provides a possible check for Q.E.D. versus recent improved semiclassical theories. The second-order correlation function is also discussed in relation to its potential for the experimental study of fluorescence spectra, particularly those in the region of the dynamical Stark effect. Possibilities for the extraction of spectral detail presently unattainable are clearly evident.

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PART I

AN INTRODUCTION TO THE

OPEN MARKOFFIAN SYSTEM

No system in the physical universe is truly isolated. The contention of Newton's first law of motion is far from obvious within common experience where dissipation is the rule. When applicable Markoffian behaviour greatly simplifies the inclusion of interaction with the surroundings.

CHAPTER I

THE MASTER EQUATION APPROACH TO THE OPEN MARKOFFIAN SYSTEM

1.1. Introduction

Every student of physics learns very early in his career Newton's three laws governing the mechanics of classical particles. It is not generally until much later, however, that the extent of Newton's insight is appreciated. In particular, Newton's first law is so often taken for granted and may even appear self-evident. It is nonetheless with considerable surprise and fascination that we first encounter a device such as the linear air track and a demonstration of frictionless dynamics. The movement of a glider back and forth without apparent loss of speed seems decidedly unnatural, and so it should, since where in our everyday experience do we meet with such behaviour? Thus it is a notion very fundamental to our experience that all physical systems are open in that they interact with their environment. Of course, the loss of energy by dissipation is not the only exchange which may be mediated by such an interaction. Energy may be pumped into a system, as for example in the laser, or, on the other hand, matter may be transported between a system and its surroundings. The latter is the case with open chemically reacting systems. We are concerned throughout this thesis with aspects of open systems and more specifically those requiring a quantum-mechanical description.

Inasmuch as the surrounding sources and sinks of energy, and/or matter, should be included for an exact description of open systems, it becomes a question of perspective as to whether we consider a very complicated closed system or an essentially simple open system modified slightly by its surroundings. It is convenient to specify a composite closed system $S \oplus R$ where S is an open system and R is some reservoir, the two being coupled in a manner dependent on the physical circumstances. The open system S appears then as a subsystem of the closed system $S \oplus R$. With the mathematical realisation of this specification the conception of an open system need not then be restricted to circumstances in which a physical division is apparent between S and R. The separation becomes one between dynamical variables; macroscopic variables defining S and the physical description with which we have ultimate interest, and microscopic variables defining $S \oplus R$. In this, therefore, hydrodynamic phenomena are included. With these a description is sought in terms of a few macroscopic variables such as temperature and pressure while underlying the changes in these gross features are processes at the molecular level.

In general, any interaction is a two-way affair and such is the case with the open system. While it is influenced by its surroundings there must also be some effect in the reverse direction on the surroundings themselves. Thus, in its subsequent interaction with a slightly modified environment an open system is meeting with the consequences of its earlier dynamical evolution. Generally, therefore, the history of an open system is reflected back on itself through its reservoir. If this history influences significantly the subsequent evolution we refer to a system with memory, if not the system has no memory and exhibits Markoffian behaviour. The requirement for the blanking of memory and the origin of Markoffian behaviour is essentially one of time scales. Dynamical variables constituting the description of S must vary on a time scale much larger than that which typifies the time taken for the reservoir to recover from any perturbations imposed upon it. This for a thermal reservoir is merely the time scale on which relaxation to equilibrium takes place. Our own concern will be solely with the Markoffian behaviour in open quantum systems.

It is in meeting the requirement of moving from the exact description of the closed system $S \oplus R$ to usable equations for S alone that the master equation approach has developed. Introduction of the master equation concept into quantum statistical mechanics was made by Pauli (1928) whose objective was to describe the relaxation to equilibrium of a large closed system. Central to his derivation was a repeated random phase assumption so that each state is identified by its probability for occupation rather than a complex probability amplitude. The Pauli master equation is, therefore, an equation for only part of the full density operator, namely, that giving the diagonal matrix elements. It is in this feature that contact with the requirements for the open system may be made. Here the part of the full density operator for $S \oplus R$ required is that obtained by tracing over reservoir variables. Accomplishing this task, from the Schrödinger equation describing $S \oplus R$ an operator equation in variables of S alone is obtained (Carmichael, 1972; Carmichael and Walls, 1973). A unified perspective on the master equation concept is provided by the projector formalism (Haake, 1973; Agarwal, 1973a) which leads to the so-called generalised master equation (Zwanzig, 1964). Here a 'relevant part' of the full density operator is projected out of the microscopic Liouville equation by a projection operator. This may itself remain arbitrary. The appropriate specification of the projector then allows the treatment of a wide variety of problems. In particular it forms the basis for a rigorous derivation of both Pauli's master equation (Zwanzig, 1961) and the operator master equations describing open quantum systems (Agarwal, 1973a, Haake, 1973).

The first two chapters of this thesis are devoted to general features of the master equation approach to open quantum Markoffian systems. Our purpose is to introduce the concepts and mathematical structure underlying the developments of later sections. We begin in section 1.2 with a brief derivation by the projector technique of the formal master equation for an open quantum Markoffian system. We then develop a specific operator form for general interaction with thermal reservoirs in section 1.2. This master equation serves as a general basis from which the full operator master equation for any system interacting with thermal reservoirs may be written down directly. It is used further on as the foundation for discussions concerning the effect of internal couplings and non-energy conserving reservoir interactions on detailed balance. Chapter II is concerned with the evaluation of two-time operator averages within the context of an operator master equation. In section 2.1 we outline the extraction from exact microscopic expressions of a form for these averages using the master equation for S alone. Section 2.2 then deals with the single Boson system and the Phase-Space evaluation of two-time averages. More specific features arising for normal and anti-normal orderings are discussed in section 2.3.

1.2 Formal Derivation of the Master Equation

The generalised master equations were originally derived independently by Van Hove (1957), Nakajima (1958), Zwanzig (1960), Prigagine and Resibois (1961) and Montroll (1962). These quantum-mechanical master equations were derived to serve as kinetic equations for the diagonal elements of the density operator for a large system. The equivalence of the various independent approaches has been discussed by Zwanzig (1964). With the projector method we may derive in a similar fashion a dynamical equation for the reduced density operator $\rho(t)$ of an open system. If $\chi(t)$ is the full density operator for $S \oplus R$ then

$$\rho(t) = tr_R\, \chi(t) \tag{1.2.1}$$

Our treatment here follows essentially that of Haake (1973) and Agarwal (1973a).

The microscopic Liouville equation reads

$$\frac{d\chi(t)}{dt} = (-i/\hbar)[H, \chi(t)] = -iL\chi(t) \tag{1.2.2}$$

where the Hamiltonian H and Liouville operator L may be expanded with

$$\begin{aligned} H &= H_S + H_R + H_{SR} \\ L &= L_S + L_R + L_{SR} \end{aligned} \tag{1.2.3}$$

H_S and H_R describe free dynamics for S and R respectively and H_{SR} is a coupling term. We may define a general projector P with

$$P^2 = P \tag{1.2.4}$$

and thus

$$(1-P)^2 = (1-P) \quad , \quad P(1-P) = 0 \tag{1.2.5}$$

then writing

$$\chi(t) = P\chi(t) + (1-P)\chi(t) \tag{1.2.6}$$

from (1.2.2) equations for $P\chi(t)$ and $(1 - P)\chi(t)$ follow.

$$\frac{d(P\chi(t))}{dt} = -iPL(P\chi(t)) - iPL(1-P)\chi(t) \tag{1.2.7}$$

and

$$\frac{d(1-P)\chi(t)}{dt} = -i(1-P)LP\chi(t) - i(1-P)L(1-P)\chi(t) \tag{1.2.8}$$

Integrating the second of these directly we find

$$\begin{aligned}(1-P)\chi(t) = {} & \exp[-i(1-P)Lt](1-P)\chi(0) \\ & -i\int_0^t d\tau (1-P)L\exp[-i(1-P)L\tau](P\chi(t))\end{aligned} \tag{1.2.9}$$

Substituting into (1.2.7) this gives an equation for the projected part of $\chi(t)$ which may be written in the form

$$\frac{d(P\chi(t))}{dt} = -iPL(P\chi(t)) + \int_0^t d\tau\, K(\tau)(P\chi(t-\tau)) + I(t) \qquad (1.2.10)$$

where the Kernel $K(\tau)$ and inhomogeneity $I(t)$ are defined by

$$K(\tau) = PL\exp[-i(1-P)L\tau](1-P)L$$
$$I(t) = -iPL\exp[-i(1-P)Lt](1-P)\chi(0) \qquad (1.2.11)$$

This equation is coupled to the irrelevant part of the density operator through the inhomogeneity $I(t)$.

The specific application of this projection operator formalism to an open system S, subsystem of $S \oplus R$, now arises with the definition of P and introduction of approximations appropriate to the Hamiltonian (1.2.3). Since we will be concerned later on specifically with reservoirs in thermal equilibrium we define the projector here by

$$P = R_{TH}\,tr_R \qquad (1.2.12)$$

where R_{TH} describes the thermal equilibrium state of the reservoir. More generally R_{TH} may be replaced by a reference state of the reservoir chosen in view of the application at hand (Haake, 1973). Using first the commutivity of operators in H_S and H_R and secondly the cyclic property of the trace we may then write

$$PL_S = L_SP$$
$$PL_R = L_RP = 0 \qquad (1.2.13)$$

which gives also

$$(1-P)(L_S+L_R) = (L_S+L_R)(1-P) \qquad (1.2.14)$$

Thus, considering the integrand $K(\tau)(P\chi(t-\tau))$ in (1.2.10) we have

$$K(\tau)(P\chi(t-\tau)) = PL\exp[-i(1-P)L\tau](1-P)L(P\chi(t-\tau)) \qquad (1.2.15)$$

using (1.2.5) and (1.2.14) this gives

$$K(\tau)(PX(t-\tau)) = PL_{SR}\exp[-i(1-P)L\tau](1-P)L_{SR}(PX(t-\tau)) \tag{1.2.16}$$

We therefore may write the Kernel for (1.2.10) as

$$K(\tau) = PL_{SR}\exp[-i(1-P)L\tau](1-P)L_{SR} \tag{1.2.17}$$

Setting

$$\mathcal{L}(\tau) = \exp[-i(1-P)L\tau]\mathcal{L}(0) \tag{1.2.18}$$

with $\mathcal{L}(0) = (1 - P)\, L_{SR}$

it then follows that

$$\begin{aligned}\frac{d\mathcal{L}(\tau)}{d\tau} &= -i(1-P)L\mathcal{L}(\tau)\\ &= -i(L_S+L_R)\mathcal{L}(\tau) - i(1-P)L_{SR}\mathcal{L}(\tau)\end{aligned} \tag{1.2.19}$$

where we have used again (1.2.5) and (1.2.14). With

$$\hat{\mathcal{L}}(\tau) = e^{i(L_S+L_R)\tau}\mathcal{L}(\tau) \tag{1.2.20}$$

we then find

$$\frac{d\hat{\mathcal{L}}(\tau)}{d\tau} = -i(1-P)\hat{L}_{SR}(\tau)\hat{\mathcal{L}}(\tau) \tag{1.2.21}$$

where

$$\hat{L}_{SR}(\tau) = e^{i(L_S+L_R)\tau}L_{SR}\,e^{-i(L_S+L_R)\tau} \tag{1.2.22}$$

Integrating this equation and iterating repeatedly it follows that

$$\hat{\mathcal{L}}(\tau) = T\exp\left\{-i\int_0^\tau (1-P)\hat{L}_{SR}(\tau)(1-P)\right\}(1-P)L_{SR} \tag{1.2.23}$$

where T is the Dyson time ordering operator. We then write from (1.2.17), (1.2.18) and (1.2.20)

$$K(\tau) = PL_{SR}\,e^{-i(L_S+L_R)\tau}\,U(\tau)(1-P)L_{SR} \tag{1.2.24}$$

with

$$U(\tau) = T\exp\left\{-i\int_0^\tau (1-P)\hat{L}_{SR}(\tau)(1-P)\right\}L_{SR} \tag{1.2.25}$$

It is convenient that we transform to the interaction picture with density operator $\hat{\chi}(t)$ defined by

$$\hat{\chi}(t) = e^{i(L_s+L_R)t}\chi(t) \ . \tag{1.2.26}$$

We find from (1.2.10)

$$\frac{d(P\hat{\chi}(t))}{dt} = -iP\hat{L}_{SR}(P\hat{\chi}(t)) + e^{i(L_s+L_R)t}\int_0^t K(\tau)\,e^{-i(L_s+L_R)(t-\tau)}(P\hat{\chi}(t-\tau)) + e^{i(L_s+L_R)t}I(t) \tag{1.2.27}$$

where $K(\tau)$ is now given by (1.2.24). Considering then the term $e^{i(L_S + L_R)t}K(\tau)\,e^{-i(L_S + L_R)(t - \tau)}$ we may write

$$e^{i(L_s+L_R)t}K(\tau)\,e^{-i(L_s+L_R)(t-\tau)} = P\hat{L}_{SR}(t)\,e^{i(L_s+L_R)(t-\tau)}U(\tau)\,e^{-i(L_s+L_R)(t-\tau)}(1-P)\hat{L}_{SR}(t-\tau) \tag{1.2.28}$$

It is easily shown that

$$e^{i(L_s+L_R)(t-\tau)}U(\tau)\,e^{-i(L_s+L_R)(t-\tau)} = \mathcal{U}(t,\tau) = T\exp\left\{-i\int_{t-\tau}^{t} dt'\,(1-P)\hat{L}_{SR}(t')(1-P)\right\} \tag{1.2.29}$$

and thus, substituting (1.2.29) into (1.2.27) we find

$$\frac{d(P\hat{\chi}(t))}{dt} = -iP\hat{L}_{SR}(P\hat{\chi}(t)) + \int_0^t d\tau\,P\hat{L}_{SR}(t)\,\mathcal{U}(t,\tau)(1-P)\hat{L}_{SR}(t-\tau)(P\hat{\chi}(t-\tau)) + e^{i(L_s+L_R)t}I(t) \tag{1.2.30}$$

We now introduce the various approximations appropriate to the uses with which we are concerned in this thesis. They are as follows:

(i) The system and reservoir are initially uncorrelated so that

$$\chi(0) = \rho(0)R_{TH} \tag{1.2.31}$$

Clearly then

$$(1-P)\chi(0) = 0 \tag{1.2.32}$$

and the inhomogeneity I(t) vanishes.*

(ii) The interaction Hamiltonian has no matrix elements diagonal in the reservoir variables so that

$$P L_{SR}(P\dot{X}(t)) = 0 \tag{1.2.33}$$

(iii) The interaction is weak and therefore we may take the first Born approximation by setting $\mathcal{U}(t,\tau)$ to unity. For a general discussion of the Born approximation see Haake (1969).

(iv) The behaviour is Markoffian and hence $P\hat{\chi}(t-\tau)$ may be replaced by $P\hat{\chi}(t)$.

With these approximations (1.2.30) then becomes

$$\frac{d\hat{\rho}(t)}{dt} = \int_0^t tr_R(\hat{L}_{SR}(t)\hat{L}_{SR}(t-\tau)R_{TH})\ \hat{\rho}(t) \tag{1.2.34}$$

Transforming back from the interaction picture the reduced density operator $\rho(t)$ for the open system S then obeys the equation

$$\frac{d\rho(t)}{dt} = \mathcal{L}\,\rho(t) \tag{1.2.35}$$

where $\mathcal{L}$ is a generalised Liouville operator to be determined

$$\mathcal{L} = -i\mathcal{L}_R + \mathcal{L}_I \tag{1.2.36}$$

Here we have made a division into reversible and irreversible components with respect to time inversion (Prigogine *et al.*, 1973), $\mathcal{L}_R$ and $\mathcal{L}_I$

$$\mathcal{L}_R = L_S \tag{1.2.37}$$

and

$$\mathcal{L}_I = e^{-iL_S t}\int_0^t d\tau\, tr_R(\hat{L}_{SR}(t)\hat{L}_{SR}(t-\tau)R_{TH})\, e^{iL_S t} \tag{1.2.38}$$

---

*Although assumption (i) is commonly made it may be avoided for Markoffian systems where the decay of reservoir correlations on a time scale much shorter than that on which $\rho(t)$ evolves quickly destroys correlations in the initial state (Haake, 1971, 1973).

1.3 A Generalised Master Equation for Thermal Reservoirs

We derive now in this section a specific form for the operator $\mathcal{L}$ of equation (1.2.35) allowing for the general reservoir interaction given in the Hamiltonian

$$\begin{aligned} H &= H_S + H_R + H_{SR} \\ H_R &= \sum_\lambda H_R^\lambda \\ H_{SR} &= \sum_\lambda H_{SR}^\lambda \end{aligned} \tag{1.3.1}$$

Here the reservoir R consists of a series of independent subsystems. The λth subsystem is taken in thermal equilibrium at a temperature T_λ and thus

$$R_{TH} = \prod_\lambda R_{TH}^\lambda = \prod_\lambda e^{-\frac{H_R^\lambda}{kT_\lambda}} \Big/ tr_{R_\lambda}\left(e^{-\frac{H_R^\lambda}{kT_\lambda}}\right) \tag{1.3.2}$$

The master equation derived will be used further on in discussions relating to the quantum detailed balance.

We assume a boson reservoir with

$$H_R = \sum_j \hbar\omega_j r_j^\dagger r_j \tag{1.3.3}$$

and write the interaction H_{SR}^λ in the form

$$H_{SR} = S_\lambda \Gamma_\lambda^\dagger + S_\lambda^\dagger \Gamma_\lambda \tag{1.3.4}$$

where S_λ is an unspecified system operator and Γ_λ is given by

$$\Gamma_\lambda = \sum_j \hbar \kappa_j^\lambda r_j^\lambda \tag{1.3.5}$$

r_j^λ is a boson annihilation operator corresponding to the boson mode with frequency ω_j^λ and κ_j^λ is a coupling constant. We work in the representation formed by the energy eigenstates $|E_n>$ of S. These states being defined by

$$H_S|E_n\rangle = E_n|E_n\rangle \tag{1.3.6}$$

Expanding the operator S_λ in this representation we have

$$S_\lambda = \sum_{n,m} |E_n\rangle\langle E_m| S^\lambda_{n,m} \tag{1.3.7}$$

Writing

$$\omega_{n,m} = |E_n - E_m|/\hbar \tag{1.3.8}$$

we then divide S_λ into positive and negative frequency parts $S_\lambda^{(+)}$ and $S_\lambda^{(-)}$ according to the scheme

$$S_\lambda = S_\lambda^{(+)} + S_\lambda^{(-)} \tag{1.3.9}$$

where

$$\begin{aligned} S_\lambda^{(+)} &= \sum_\mu S_\lambda^{(+)}(\omega_\mu) \\ S_\lambda^{(+)}(\omega_\mu) &= \sum_{\substack{n,m \\ E_n < E_m}} |E_n\rangle\langle E_m| S^\lambda_{n,m}\, \delta_{\omega_{n,m},\omega_\mu} \end{aligned} \tag{1.3.10}$$

and

$$\begin{aligned} S_\lambda^{(-)} &= \sum_\mu S_\lambda^{(-)}(\omega_\nu) \\ S_\lambda^{(-)}(\omega_\nu) &= \sum_{\substack{n,m \\ E_n > E_m}} |E_n\rangle\langle E_m| S^\lambda_{n,m}\, \delta_{\omega_{n,m},\omega_\nu} \end{aligned} \tag{1.3.11}$$

In this fashion we separate positive and negative frequency parts of the interaction operator S_λ.

Corresponding to Hamiltonian (1.3.1) we may write the Liouvillian

$$\begin{aligned} L &= L_s + L_R + L_{SR} \\ L_R &= \sum_\lambda L_R^\lambda \\ L_{SR} &= \sum_\lambda L_{SR}^\lambda \end{aligned} \tag{1.3.12}$$

and then the operator $\mathcal{L}$ from (1.2.36) may be written

$$\begin{aligned} \mathcal{L} = -iL_s + e^{-iL_st} \sum_\lambda \int_0^t tr_{R_\lambda}\left(\hat{L}^\lambda_{SR}(t)\hat{L}_{SR}(t-\tau)R^\lambda_{TH}\right) e^{iL_st} \\ -(1/\hbar^2) \sum_{\substack{\lambda\lambda' \\ \lambda\neq\lambda'}} \int_0^t tr_{R_\lambda}\left(\hat{L}^\lambda_{SR}(t)\left(tr_{R_{\lambda'}}\hat{L}^{\lambda'}_{SR}(t-\tau)R^{\lambda'}_{TH}\right)R^\lambda_{TH}\right) e^{iL_st} \end{aligned} \tag{1.3.13}$$

Under assumption (ii) of the previous section that the interaction has no matrix elements diagonal in the energy representation the second of these sums of terms vanishes and we have the form

$$\mathcal{L}\rho = -iL_s + e^{-iL_st}\sum_\lambda \int_0^t tr_{R_\lambda}\left(\hat{L}^\lambda_{SR}(t)\hat{L}^\lambda_{SR}(t-\tau)R^\lambda_{TH}\right)e^{iL_st}\rho \qquad (1.3.14)$$

Now

$$L^\lambda_{SR} = \frac{1}{\hbar}[H^\lambda_{SR}, \;\;] \qquad (1.3.15)$$

and hence from (1.2.22) and the identity

$$e^{iLt}O = e^{\frac{iH}{\hbar}t}\,O\,e^{-\frac{iH}{\hbar}t} \qquad (1.3.16)$$

O being an arbitrary operator

$$\hat{L}^\lambda_{SR}(t) = (1/\hbar)[\hat{H}^\lambda_{SR}(t), \;\;] \qquad (1.3.17)$$

where

$$\hat{H}^\lambda_{SR}(t) = e^{\frac{i(H_s+H_R)t}{\hbar}}\,H^\lambda_{SR}\,e^{\frac{-i(H_s+H_R)t}{\hbar}} \qquad (1.3.18)$$

using the specification (1.3.3) and (1.3.4) for H^λ_{SR} then

$$\hat{H}^\lambda_{SR}(t) = S_\lambda(t)\Gamma^\dagger_\lambda(t) + S^\dagger_\lambda(t)\Gamma_\lambda(t) \qquad (1.3.19)$$

with

$$\Gamma_\lambda(t) = e^{\frac{iH_Rt}{\hbar}}H^\lambda_{SR}e^{\frac{-iH_Rt}{\hbar}} = \sum_j \hbar K^\lambda_j r^\lambda_j e^{i\omega_jt} \qquad (1.3.20)$$

and

$$S_\lambda(t) = e^{\frac{iH_st}{\hbar}}S_\lambda e^{\frac{-iH_st}{\hbar}} = \sum_\mu S^{(+)}_\lambda(\omega_\mu)e^{-i\omega_\mu t} + S^{(-)}_\lambda(\omega_\mu)e^{i\omega_\mu t} \qquad (1.3.21)$$

Substituting from (1.3.15), (1.3.17), (1.3.18) and (1.3.19) into (1.3.14) we may then carry out the trace and do the integrals in a standard manner (see for example Carmichael, 1972). This leads to a master equation of the form (1.2.35) where the irreversible term is given by

$$\mathcal{L}_I\rho = \sum_\lambda \sum_\nu (\gamma_\lambda(\omega_\nu)/2)\Big\{[S_\lambda^{(+)}(\omega_\nu)\rho, S_\lambda^\dagger] + [S_\lambda, \rho S_\lambda^{(+)\dagger}(\omega_\nu)]$$
$$+ \bar{n}_\lambda(\omega_\nu)\big([S_\lambda^{(+)}(\omega_\nu)\rho, S_\lambda^\dagger] + [S_\lambda, \rho S_\lambda^{(+)\dagger}(\omega_\nu)]$$
$$+ [S_\lambda^{(+)\dagger}(\omega_\nu)\rho, S_\lambda] + [S_\lambda^\dagger, \rho S_\lambda^{(+)}(\omega_\nu)]\big)\Big\} \tag{1.3.22}$$

Here the damping constants $\gamma_\lambda(\omega_\nu)$ are given in terms of a strength function $g_\lambda(\omega_\nu)$ and coupling constants

$$\gamma_\lambda(\omega_\nu) = 2\pi g_\lambda(\omega_\nu)|K_\lambda(\omega_\nu)|^2 \tag{1.3.23}$$

and

$$\bar{n}_\lambda(\omega_\nu) = [\exp(\hbar\omega_\nu/kT_\lambda) - 1]^{-1} \tag{1.3.24}$$

CHAPTER II

TWO-TIME AVERAGES IN THE MASTER EQUATION FORMALISM

2.1 Operator Averages at Two Times

With the derivation of (1.2.35) for the reduced density operator all single time averages $<S^{(\alpha)}(t)>$, where $S^{(\alpha)}$ is any system operator, may be obtained in terms of system variables alone. We have

$$\begin{aligned}\langle S^{(\alpha)}(t)\rangle &= tr_{S\oplus R}\,\chi(t)\,S^{(\alpha)}(0) = tr_S[tr_R\,\chi(t)]\,S^{(\alpha)}(0)\\ &= tr_S\,\rho(t)\,S^{(\alpha)}(0)\end{aligned} \tag{2.1.1}$$

Now the two-time average $<S^{(\alpha)}(t+\tau)\,S^{(\beta)}(t)>$, $\tau \geq 0$, when considered in relation to the complete system $S \oplus R$ with full density operator $\chi(t)$ may be written $<S^{(\alpha)}(t+\tau)\,S^{(\beta)}(t)>_{\chi(t)}$ and is given exactly by the formal expression

$$\langle S^{(\alpha)}(t+\tau)\,S^{(\beta)}(t)\rangle_{\chi(t)} = tr_{S\oplus R}\,\chi(0)\,S^{(\alpha)}(t+\tau)\,S^{(\beta)}(t) \tag{2.1.2}$$

The time-dependent operators are given in the Heisenberg picture by

$$\begin{aligned}S^{(\alpha)}(t+\tau) &= U(t+\tau)\,S^{(\alpha)}(0)\\ S^{(\beta)}(t) &= U(t)\,S^{(\beta)}(0)\end{aligned} \tag{2.1.3}$$

and $U(t)$ is the time-development operator with, for arbitrary operator O,

$$U(t)\,O = e^{iLt}\,O = e^{\frac{iHt}{\hbar}}\,O\,e^{\frac{-iHt}{\hbar}} \tag{2.1.4}$$

It is then desirable to eliminate the trace over the reservoir from (2.1.2) to obtain, as in (2.1.1), an expression in terms of the dynamics of S alone. We therefore envisage evaluation of the two-time average from $\rho(t)$ and the generalised Liouville operator $\mathcal{L}$ governing its dynamics. We will specify separate from (2.1.2) a macroscopic average $\langle s^{(\alpha)}(t+\tau)\, s^{(\beta)}(t)\rangle_{\rho(t)}$ and derive its explicit form in what follows.

Beginning with (2.1.2), (2.1.3) and (2.1.4) we have

$$\langle S^{(\alpha)}(t+\tau)S^{(\beta)}(t)\rangle_{\chi(t)} = tr_{S\otimes R}\,\chi(0)\, e^{\frac{iH}{\hbar}(t+\tau)}\, S^{(\alpha)}(0)\, e^{\frac{-iH}{\hbar}(t+\tau)} \cdot e^{\frac{iH}{\hbar}t}\, S^{(\beta)}(0)\, e^{\frac{-iH}{\hbar}t} \tag{2.1.5}$$

and, using the cyclic property of the trace this may be written

$$\langle S^{(\alpha)}(t+\tau)S^{(\beta)}(t)\rangle_{\chi(t)} = tr_{S\otimes R}\,\chi(t)\, e^{\frac{iH}{\hbar}\tau}\, S^{(\alpha)}(0)\, e^{\frac{-iH}{\hbar}\tau}\, S^{(\beta)}(0) \tag{2.1.6}$$

where with (1.3.16) the formal solution to (1.2.3) is

$$\chi(t) = e^{\frac{-iH}{\hbar}t}\,\chi(0)\, e^{\frac{iH}{\hbar}t} \tag{2.1.7}$$

A further use of the cyclic property of the trace then yields

$$\langle S^{(\alpha)}(t+\tau)S^{(\beta)}(t)\rangle_{\chi(t)} = tr_S\left[tr_R\, e^{\frac{-iH}{\hbar}\tau}\, S^{(\beta)}(0)\,\chi(t)\, e^{\frac{iH}{\hbar}\tau}\right] S^{(\alpha)}(0) \tag{2.1.8}$$

Defining the operator $\mathcal{S}^{(\beta)}(\tau)$ by

$$\mathcal{S}^{(\beta)}(\tau) = e^{\frac{-iH}{\hbar}\tau}\, S^{(\beta)}(0)\,\chi(t)\, e^{\frac{iH}{\hbar}\tau} \tag{2.1.9}$$

this is clearly just the formal solution to the equation

$$\frac{d\mathcal{S}^{(\beta)}(\tau)}{d\tau} = (-i/\hbar)[H, \mathcal{S}^{(\beta)}(\tau)] = -iL\,\mathcal{S}^{(\beta)}(\tau) \tag{2.1.10}$$

This equation is equivalent to (1.2.2) and may be treated in a similar fashion via the projection technique to arrive at the form

$$\frac{d\hat{\mathcal{S}}^{(\beta)}(\tau)}{d\tau} = \mathcal{L}\,\hat{\mathcal{S}}^{(\beta)}(\tau) \tag{2.1.11}$$

analogous to (1.2.35), with

$$\hat{\mathcal{S}}^{(\beta)}(\tau) \simeq tr_R\,\mathcal{S}^{(\beta)}(\tau) = tr_R\, e^{\frac{-iH}{\hbar}\tau}\, S^{(\beta)}(0)\,\chi(t)\, e^{\frac{iH}{\hbar}\tau} \tag{2.1.12}$$

The approximation appearing here reflects in particular the Born and Markoffian approximations (iii) and (iv) of section 1.2. In place of approximation (i) of the same section we now require

$$\chi(t) \approx \rho(t) R_{TH} \tag{2.1.13}$$

as, for example, is taken in the treatment of two-time averages by Lax (1963). However, the footnote to approximation (i) is again here relevant. The Markoffian assumption is sufficient to ensure the decay of correlations in $\chi(t)$ on a time scale much shorter than that with which we are concerned in following the dynamics of S (Haake, 1971, 197

Solving (2.1.11) formally with $\hat{\mathcal{S}}^{(\beta)}(0)$ given by

$$\hat{\mathcal{S}}^{(\beta)}(0) = tr_R \mathcal{S}^{(\beta)}(0) = tr_R S^{(\beta)}(0)\,\chi(t) = S^{(\beta)}(0)\,\rho(t) \tag{2.1.14}$$

we may then write

$$\hat{\mathcal{S}}^{(\beta)}(\tau) = e^{\mathcal{L}\tau} S^{(\beta)}(0)\,\rho(t) \tag{2.1.15}$$

and combining this with (2.1.8) and (2.1.12) find for $\langle S^{(\alpha)}(t+\tau)S^{(\beta)}$

$$\langle S^{(\alpha)}(t+\tau)\,S^{(\beta)}(t)\rangle_{\rho(t)} = tr_S\left[e^{\mathcal{L}\tau} S^{(\beta)}(0)\,\rho(t)\right] S^{(\alpha)}(0) \tag{2.1.16}$$

Clearly

$$\langle S^{(\alpha)}(t+\tau)\,S^{(\beta)}(t)\rangle_{\rho(t)} \approx \langle S^{(\alpha)}(t+\tau)\,S^{(\beta)}(t)\rangle_{\chi(t)} \tag{2.1.17}$$

where the approximation arises in (2.1.12). We have here essentially the quantum-regression theorem of Lax (1963, 1967, 1968) and a relationship to Lax's theorem is drawn explicitly in appendix A.

To conclude we note that for the average with alternative time ordering $\langle S^{(\alpha)}(t)\ S^{(\beta)}(t+\tau)\rangle$, $\tau \geq 0$, an analogous method yields the result

$$\langle S^{(\alpha)}(t)\,S^{(\beta)}(t+\tau)\rangle_{\rho(t)} = tr_S\left[e^{\mathcal{L}\tau} \rho(t)\,S^{(\alpha)}(0)\right] S^{(\beta)}(0) \tag{2.1.18}$$

with

$$\langle S^{(\alpha)}(t) S^{(\beta)}(t+\tau) \rangle_{\rho(t)} \approx \langle S^{(\alpha)}(t) S^{(\beta)}(t+\tau) \rangle_{\chi(t)} \tag{2.1.19}$$

2.2 Phase-Space Forms for Single Boson Systems

During the last decade, the single boson system, corresponding to a single mode of the electromagnetic field, has attracted considerable attention. This, of course, reflects the widespread interest in the single mode laser (for a review see Haken, 1970). Arising initially in the coherent-state representation of Glauber (Glauber, 1963), a formulation of quantum dynamics for this single boson system has developed allowing all single-time operator averages to be obtained via integration in phase space. Here cumberson operator traces may be avoided using the so-called Glauber-Sudarshan P-representation for the density operator (Glauber, 1963; Sudarshan, 1966). Generalisations of the phase-space formulation have been discussed by a number of authors (Sudarshan, 1966; Cahill and Glauber, 1969; Agarwal and Wolf, 1970). In particular, an approach has been worked out in detail by Agarwal and Wolf (1970). A correspondence between operators $\hat{S}(a,a^{\dagger})$ of the boson system and c-number functions $S^{(\Omega)}(\alpha,\alpha^{*})$ is set up in the form of a linear mapping $S^{(\Omega)}(\alpha,\alpha^{*}) \to \hat{S}(a,a^{\dagger})$ accomplished by the mapping operator $\hat{\Omega}$;

$$\hat{\Omega}\, S^{(\Omega)}(\alpha,\alpha^{*}) = \hat{S}(a,a^{\dagger}) \tag{2.2.1}$$

Requiring the operator $\hat{S}(a,a^{\dagger})$ to admit expression as a Fourier integral,

$$\hat{S}(a,a^{\dagger}) = \int d^{2}\beta\, G(\beta,\beta^{*}) \exp(\beta a^{\dagger} - \beta^{*} a) \tag{2.2.2}$$

with

$$G(\beta,\beta^{*}) = 1/\pi\, tr_{s}\, \hat{S}(a,a^{\dagger}) \exp(\beta^{*} a - \beta a^{\dagger}) \tag{2.2.3}$$

the mapping operator is defined explicitly by relating this expression to the Fourier integral for $S^{(\Omega)}(\alpha,\alpha^*)$. We require

$$G(\beta,\beta^*) = \Omega(\beta,\beta^*)\, F^{(\Omega)}(\beta,\beta^*) \tag{2.2.4}$$

where

$$S^{(\Omega)}(\alpha,\alpha^*) = \int d^2\beta\, F^{(\Omega)}(\beta,\beta^*) \exp(\beta\alpha^* - \beta^*\alpha) \tag{2.2.5}$$

and

$$F^{(\Omega)}(\beta,\beta^*) = (1/\pi^2) \int d^2\alpha\, S^{(\Omega)}(\alpha,\alpha^*) \exp(\beta^*\alpha - \beta\alpha^*) \tag{2.2.6}$$

Here $\Omega(\beta,\beta^*)$ is a function in complex phase-space acting as a filter function. A broad class of mappings may be specified by imposing appropriate restrictions on this filter function. It is taken throughout this present work that $\Omega(\beta,\beta^*)$ be a complex function of the general form

$$\Omega(\beta,\beta^*) = \exp(\mu\beta^2 + \nu\beta^{*2} + \lambda\beta\beta^*) \tag{2.2.7}$$

with μ, ν, and λ as real constants.

The appeal of the phase-space calculus arises in the fact that single-time averages may be evaluated now by integration in phase space. As is shown in appendix B, generally, for any two operators $\hat{S}_1(a,a^\dagger)$ and $\hat{S}_2(a,a^\dagger)$, we have the result

$$\mathrm{tr}_s\, \hat{S}_1(a,a^\dagger)\, \hat{S}_2(a,a^\dagger) = (1/\pi) \int d^2\alpha\, S_1^{(\Omega)}(\alpha,\alpha^*)\, S_2^{(\bar{\Omega})}(\alpha,\alpha^*) \tag{2.2.8}$$

In the specific instance where $P^{(\Omega)}(\alpha,\alpha^*,t)$ is the Ω-equivalent for $\rho(a,a^\dagger,t)$,

$$\hat{\Omega}\, P^{(\Omega)}(\alpha,\alpha^*,t) = \rho(a,a^\dagger,t) \tag{2.2.9}$$

then for any operator $\hat{S}(a,a^\dagger)$

$$\begin{aligned} \langle \hat{S}(a,a^\dagger)\rangle &= \mathrm{tr}_s\, S(a,a^\dagger)\, \rho(a,a^\dagger,t) \\ &= (1/\pi) \int d^2\alpha\, P^{(\Omega)}(\alpha,\alpha^*,t)\, S^{(\bar{\Omega})}(\alpha,\alpha^*) \end{aligned} \tag{2.2.10}$$

where the inverse mapping $\bar{\Omega}$ has

$$\bar{\Omega}(\beta,\beta^*) = \Omega(\beta,\beta^*)^{-1} \qquad (2.2.11)$$

In the case where $\hat{S}(a,a^\dagger)$ is a normally ordered operator and $\bar{\Omega}$ is taken to correspond to normal ordering, $S^{(\bar{\Omega})}(\alpha,\alpha^*)$ is very simply constructed by the substitution $a \to \alpha$, $a^\dagger \to \alpha^*$. In this instance $F^{(\Omega)}(\alpha,\alpha^*,t)$ is the Glauber-Sudarshan P-function (Glauber, 1963; Sudarshan, 1966). Furthermore, for time ordered, normally ordered products of operators, equally simple procedures produce a large number of multitime averages (Lax, 1968).

We consider here the results (2.1.16) and (2.1.18) with a view to arriving at a phase-space expression of these results for the general Ω-mapping defined according to (2.2.7). To avoid confusion in the use of Greek letters we will drop the superscripts (α) and (β) of the previous section and write

$$\langle \hat{S}_1(t+\tau)\hat{S}_2(t)\rangle_{\rho(t)} = tr_S[e^{\mathcal{L}\tau}\hat{S}_2(0)\rho(t)]\hat{S}_1(0) \qquad (2.2.12)$$

and

$$\langle \hat{S}_1(t)\hat{S}_2(t+\tau)\rangle_{\rho(t)} = tr_S[e^{\mathcal{L}\tau}\rho(t)\hat{S}_1(0)]\hat{S}_2(0) \qquad (2.2.13)$$

Now, as in (2.1.15), we have

$$\hat{\mathcal{S}}_2(a,a^\dagger,\tau) = e^{\mathcal{L}\tau} S_2(a,a^\dagger,0)\rho(a,a^\dagger,t) \qquad (2.2.14)$$

and thus, we may write immediately from (2.2.9)

$$\langle \hat{S}_1(t+\tau)\hat{S}_2(t)\rangle_{\rho(t)} = (1/\pi)\int d^2\beta\, S_1^{(\Omega)}(\beta,\beta^*)\,\mathcal{S}^{(\bar{\Omega})}(\beta,\beta^*,\tau) \qquad (2.2.15)$$

where

$$\hat{\Omega}\,\mathcal{S}_2^{(\bar{\Omega})}(\beta,\beta^*,\tau) = \hat{\mathcal{S}}_2(a,a^\dagger,\tau) \qquad (2.2.16)$$

We then define for the Liouville operator $\mathcal{L}(a,a^\dagger)$ an $\bar{\Omega}$-equivalent such that

$$\hat{\bar{\Omega}}\left[\mathcal{L}^{(\bar{\Omega})}(\beta,\beta^*)S^{(\bar{\Omega})}(\beta,\beta^*)\right] = \mathcal{L}(a,a^\dagger)\,\hat{S}(a,a^\dagger) \tag{2.2.17}$$

Thus, from (2.2.16) and (2.2.14)

$$\begin{aligned}\hat{\bar{\Omega}}\,\mathcal{S}_2^{(\bar{\Omega})}(\beta,\beta^*,\tau) &= e^{\mathcal{L}\tau}\,\hat{\mathcal{S}}_2(a,a^\dagger,0)\\ &= \hat{\bar{\Omega}}\exp[\mathcal{L}^{(\bar{\Omega})}(\beta,\beta^*)\tau]\,\mathcal{S}_2^{(\bar{\Omega})}(\beta,\beta^*,0)\end{aligned}$$

whence

$$\mathcal{S}_2^{(\bar{\Omega})}(\beta,\beta^*,\tau) = \exp[\mathcal{L}^{(\bar{\Omega})}(\beta,\beta^*)\tau]\,\mathcal{S}_2^{(\bar{\Omega})}(\beta,\beta^*,0) \tag{2.2.18}$$

To proceed, we may write

$$\mathcal{S}_2^{(\bar{\Omega})}(\beta,\beta^*,0) = \int d^2\alpha\;\delta^2(\alpha-\beta)\,\mathcal{S}_2^{(\bar{\Omega})}(\alpha,\alpha^*,0) \tag{2.2.19}$$

and with $\Delta^{(\Omega)}(\beta - a,\ \beta^* - a^\dagger)$ defined as the Ω-equivalent of $\delta^2(\beta-\alpha)$:

$$\hat{\Omega}\,\delta^2(\beta-\alpha) = \Delta^{(\Omega)}(\beta-a,\ \beta^*-a^\dagger) \tag{2.2.20}$$

equation (2.2.11) indicates equation (2.2.19) to be the phase-space form of the result

$$\mathcal{S}_2^{(\bar{\Omega})}(\beta,\beta^*,0) = \pi\,\mathrm{tr}_S\,\Delta^{(\Omega)}(\beta-a,\ \beta^*-a^\dagger)\,\hat{S}_2(a,a^\dagger)\,\rho(a,a^\dagger,t) \tag{2.2.21}$$

From the cyclic property of the trace, this yields

$$\mathcal{S}_2^{(\bar{\Omega})}(\beta,\beta^*,0) = \pi\,\mathrm{tr}_S\,\rho(a,a^\dagger,t)\,\Delta^{(\Omega)}(\beta-a,\ \beta^*-a^\dagger)\,\hat{S}_2(a,a^\dagger) \tag{2.2.22}$$

We must now invoke a general result established by Agarwal and Wolf (1970) and outlined in appendix B: for any two operators $\hat{S}_1(a,a^\dagger)$ and $\hat{S}_2(a,a^\dagger)$, where the product $\hat{S}_1(a,a^\dagger)\ \hat{S}_2(a,a^\dagger)$ is designated $\hat{S}_{12}(a,a^\dagger)$, we have

$$\hat{S}_{12}^{(\Omega)}(a,a^\dagger) = S_1^{(\Omega)}(\alpha,\alpha^*)\exp[\overleftrightarrow{\Lambda}^{(\Omega)}(\alpha,\alpha^*)]\,S_2^{(\Omega)}(\alpha,\alpha^*) \tag{2.2.23}$$

with

$$\overleftrightarrow{\Lambda}^{(\Omega)}(\alpha,\alpha^*) = -2\nu\frac{\overleftarrow{\partial}}{\partial\alpha}\frac{\overrightarrow{\partial}}{\partial\alpha} - 2\mu\frac{\overleftarrow{\partial}}{\partial\alpha^*}\frac{\overrightarrow{\partial}}{\partial\alpha^*} + (\lambda+1/2)\frac{\overleftarrow{\partial}}{\partial\alpha}\frac{\overrightarrow{\partial}}{\partial\alpha^*} + (\lambda-1/2)\frac{\overleftarrow{\partial}}{\partial\alpha^*}\frac{\overrightarrow{\partial}}{\partial\alpha} \tag{2.2.24}$$

Here the arrows ← and → indicate differentiation of the functions to the left and right of the differential operator respectively.

Using now the fundamental result (2.2.11), together with (2.2.23), we may write (2.2.22) as

$$\mathcal{S}_2^{(\bar{\Omega})}(\beta,\beta^*,0) = \int d^2\alpha\, P^{(\Omega)}(\alpha,\alpha^*,t)\exp[\overleftrightarrow{\Lambda}^{(\Omega)}(\alpha,\alpha^*)]\,\delta^2(\alpha-\beta,\alpha^*-\beta^*) \cdot S_2^{(\bar{\Omega})}(\alpha,\alpha^*,0) \tag{2.2.25}$$

We substitute this into equation (2.2.18) and arrive at the form

$$\mathcal{S}_2^{(\bar{\Omega})}(\beta,\beta^*,\tau) = \int d^2\alpha\, P^{(\Omega)}(\alpha,\alpha^*,t)\exp[\overleftrightarrow{\Lambda}^{(\Omega)}(\alpha,\alpha^*)]\,\mathcal{P}^{(\bar{\Omega})}(\beta,\beta^*,\tau|\alpha,\alpha^*,0) \cdot S_2^{(\bar{\Omega})}(\alpha,\alpha^*,0) \tag{2.2.26}$$

having introduced in $\mathcal{P}^{(\bar{\Omega})}(\beta,\beta^*,\tau|\alpha,\alpha^*,0)$ the Green's function solution to the master equation (1.2.35) when cast into phase-space form by the mapping operator $\hat{\bar{\Omega}}$. By definition

$$\mathcal{P}^{(\bar{\Omega})}(\beta,\beta^*,\tau|\alpha,\alpha^*,0) = \exp[\mathcal{L}^{(\bar{\Omega})}(\beta,\beta^*)\tau]\,\delta^2(\alpha-\beta,\alpha^*-\beta^*) \tag{2.2.27}$$

where the corresponding phase-space form of the master equation is

$$\frac{dP^{(\bar{\Omega})}}{dt}(\alpha,\alpha^*,t) = \mathcal{L}^{(\bar{\Omega})}(\alpha,\alpha^*)\,P^{(\bar{\Omega})}(\alpha,\alpha^*,t) \tag{2.2.28}$$

To achieve our objective it remains only to substitute (2.2.26) into (2.2.15); we arrive at the result

$$\langle S_1(t+\tau)S_2(t)\rangle_{\rho(t)} = \int d^2\alpha\int d^2\beta\, \overleftarrow{\mathcal{W}}^{(\Omega)}(\beta,\beta^*,t+\tau;\alpha,\alpha^*,t) \cdot S_1^{(\Omega)}(\beta,\beta^*)\,S_2^{(\bar{\Omega})}(\alpha,\alpha^*) \tag{2.2.29}$$

where $\overleftarrow{\mathcal{W}}^{(\Omega)}(\beta,\beta^*; t+\tau; \alpha,\alpha^*; 0)$ is defined by

$$\overleftarrow{\mathcal{W}}^{(\Omega)}(\beta,\beta^*,t+\tau;\alpha,\alpha^*,t) = P^{(\Omega)}(\alpha,\alpha^*,t)\exp[\overleftrightarrow{\Lambda}^{(\bar{\Omega})}(\alpha,\alpha^*)]\,\mathcal{P}^{(\bar{\Omega})}(\beta,\beta^*,\tau|\alpha,\alpha^*,0) \tag{2.2.30}$$

An analogous argument yields from (2.2.14) the result

$$\langle \hat{S}_1(a,a^\dagger,t)\hat{S}_2(a,a^\dagger,t+\tau)\rangle_{\rho(t)} = \int d^2\alpha \int d^2\beta\; \overrightarrow{\mathcal{W}}^{(\Omega)}(\beta,\beta^*,t+\tau;\alpha,\alpha^*,t)\, S_2^{(\Omega)}(\beta,\beta^*)\, S_1^{(\bar{\Omega})}(\alpha,\alpha^*) \tag{2.2.31}$$

where

$$\overrightarrow{\mathcal{W}}^{(\Omega)}(\beta,\beta^*,t+\tau;\alpha,\alpha^*,t) = \mathcal{P}^{(\bar{\Omega})}(\beta,\beta^*,t+\tau|\alpha,\alpha^*,0)\exp[\overleftrightarrow{\Lambda}^{(\bar{\Omega})}(\alpha,\alpha^*)]\,P^{(\Omega)}(\alpha,\alpha^*,t) \tag{2.2.32}$$

It is worthwhile at this stage to notice the similarity in form of equations (2.2.29) and (2.2.32) to the familiar double integration over phase-space encountered in classical statistical mechanics (see, for example, De Groot and Mazur, 1972). For a system described by a classical Fokker-Plank equation a joint probability density is constructed for the evaluation of two-time correlations in relevant stochastic variables. In the present formalism $\overleftarrow{\mathcal{W}}^{(\Omega)}(\beta,\beta^*; t+\tau; \alpha,\alpha^*; t)$ and $\overrightarrow{\mathcal{W}}^{(\Omega)}(\beta,\beta^*; t+\tau; \alpha,\alpha^*; t)$ act as pseudo probability densities relevant to the 'quantum Fokker-Plank equation' originating in the Ω-mapping of the operator master equation (1.2.35). The presence of the operator $\exp[\overleftrightarrow{\Lambda}^{(\Omega)}(\alpha,\alpha^*)]$ gives one indication of the deviation of the quantum problem from a classical stochastic model. A distinction between $\overleftarrow{\mathcal{W}}^{(\Omega)}(\beta,\beta^*; t+\tau; \alpha,\alpha^*; t)$ and $\overrightarrow{\mathcal{W}}^{(\Omega)}(\beta,\beta^*; t+\tau; \alpha,\alpha^*; t)$ must be drawn, which of course arises in the non-commutivity of operators $\hat{S}_1(a,a^\dagger)$ and $\hat{S}_2(a,a^\dagger)$. Further discussion of these features is left until section 4.2.

2.3 Normal and Antinormal Ordering

In the previous section equations (2.2.29) to (2.2.32) have been derived as the expression of two-time correlation functions for the open quantum Markoffian system. In these results the role of a pseudo joint probability density is played by the functions $\overleftarrow{\mathfrak{W}}^{(\Omega)}(\beta,\beta^*,t+\tau;\ \alpha,\alpha^*,t)$ and $\overrightarrow{\mathfrak{W}}^{(\Omega)}(\beta,\beta^*,t+\tau;\ \alpha,\alpha^*,t)$. We introduce now a further function, $W^{(\Omega)}(\beta,\beta^*,t+\tau;\ \alpha,\alpha^*,t)$, also with a claim to the status of a pseudo joint probability function. The definition is here given by

$$W^{(\Omega)}(\beta,\beta^*,t+\tau;\alpha,\alpha^*,t)=P^{(\Omega)}(\alpha,\alpha^*,t)\,\mathcal{P}^{(\Omega)}(\beta,\beta^*,\tau|\alpha,\alpha^*,0) \qquad (2.3.1)$$

Such an expression arises naturally from a literal interpretation of the underlying 'quantum Fokker-Plank equation' within the familiar structure of classical stochastic theory. Extrapolating this formal analogy one might make the error of postulating the expression of the average $\langle\hat{S}_1(t+\tau)\,\hat{S}_2(t)\rangle_{\rho(t)}$ in the form

$$\int d^2\alpha\int d^2\beta\, W^{(\Omega)}(\beta,\beta^*,t+\tau;\alpha,\alpha^*,t)\,S_1^{(\Omega)}(\beta,\beta^*)\,S_2^{(\Omega)}(\alpha,\alpha^*).$$

It is not the purpose of the present section to make any comparison of this statement with the correct results of the previous section; this topic is reserved for section 4.2. However, to make complete our investigation of the two-time averages, and for future reference, we will demonstrate here how in fact the factored form of $W^{(\Omega)}(\beta,\beta^*,t+\tau;\ \alpha,\alpha^*,t)$ may also be used for the construction of two-time correlations.

The most usual mappings encountered in relation to the single boson system are those accompanying the normal and antinormal operator orderings. Since we must now consider the specific form of the operator $\exp\left[\overleftrightarrow{\Lambda}^{(\Omega)}(\alpha,\alpha^*)\right]$, we restrict ourselves here to these two examples. In this instance the filter functions $\Omega(\beta,\beta^*)$ are familiar (Cahill and Glauber, 1969; Agarwal and Wolf, 1970) and are furthermore inverse to one another. For normal ordering $\Omega^{(N)}(\beta,\beta^*) = \beta\beta^*$, while for antinormal ordering $\Omega^{(A)}(\beta,\beta^*) = -\beta^*\beta$. Then $\bar{\Omega}^{(N)}(\beta,\beta^*) = \Omega^{(A)}(\beta,\beta^*)$, and of course $\bar{\Omega}^{(A)}(\beta,\beta^*) = \Omega^{(N)}(\beta,\beta^*)$. Making the association between (2.2.7) and (2.2.24) we may clearly write

$$\overleftrightarrow{\Lambda}^{(N)}(\alpha,\alpha^*) = \frac{\overleftarrow{\partial}}{\partial\alpha}\frac{\overrightarrow{\partial}}{\partial\alpha^*} \tag{2.3.2}$$

and

$$\overleftrightarrow{\Lambda}^{(A)}(\alpha,\alpha^*) = -\frac{\overleftarrow{\partial}}{\partial\alpha^*}\frac{\overrightarrow{\partial}}{\partial\alpha} \tag{2.3.3}$$

If we then begin with equations (2.2.29) to (2.2.32) and introduce these differential operators explicitly, we may write for normal ordering

$$\langle\hat{S}_1(t+\tau)\hat{S}_2(t)\rangle_{\rho(t)} = \int d^2\alpha\int d^2\beta\,[P^{(N)}(\alpha,\alpha^*,t)\exp(\frac{\overleftarrow{\partial}}{\partial\alpha}\frac{\overrightarrow{\partial}}{\partial\alpha^*})\,\mathcal{P}^{(A)}(\beta,\beta^*,\tau|\alpha,\alpha^*,0)]\,S_1^{(N)}(\beta,\beta^*)\,S_2^{(A)}(\alpha,\alpha^*) \tag{2.3.4}$$

$$\langle\hat{S}_1(t)\hat{S}_2(t+\tau)\rangle_{\rho(t)} = \int d^2\alpha\int d^2\beta\,[\mathcal{P}^{(A)}(\beta,\beta^*,\tau|\alpha,\alpha^*,0)\exp(\frac{\overleftarrow{\partial}}{\partial\alpha}\frac{\overrightarrow{\partial}}{\partial\alpha^*})\,P^{(N)}(\alpha,\alpha^*,t)]\,S_1^{(A)}(\alpha,\alpha^*)\,S_2^{(N)}(\beta,\beta^*) \tag{2.3.5}$$

and for antinormal ordering

$$\langle\hat{S}_1(t+\tau)\hat{S}_2(t)\rangle_{\rho(t)} = \int d^2\alpha\int d^2\beta\,[P^{(A)}(\alpha,\alpha^*,t)\exp(-\frac{\overleftarrow{\partial}}{\partial\alpha^*}\frac{\overrightarrow{\partial}}{\partial\alpha})\,\mathcal{P}^{(N)}(\beta,\beta^*,\tau|\alpha,\alpha^*,0)\,S_1^{(A)}(\beta,\beta^*)\,S_2^{(N)}(\alpha,\alpha^*) \tag{2.3.6}$$

$$\langle\hat{S}_1(t)\hat{S}_2(t+\tau)\rangle_{\rho(t)} = \int d^2\alpha\int d^2\beta\,[\mathcal{P}^{(N)}(\beta,\beta^*,\tau|\alpha,\alpha^*,0)\exp(-\frac{\overleftarrow{\partial}}{\partial\alpha^*}\frac{\overrightarrow{\partial}}{\partial\alpha})\,P^{(A)}(\alpha,\alpha^*,t)]\,S_1^{(N)}(\alpha,\alpha^*)\,S_2^{(A)}(\beta,\beta^*) \tag{2.3.7}$$

The road to our objective, as taken from here, is basically one of algebraic manipulation. Thus we state now two operator identities to be used in this respect. If n, p, q, and r are integer variables, and $\phi_1(\alpha,\alpha^*)$ and $\phi_2(\alpha,\alpha^*)$ are arbitrary functions, then first, it is shown in appendix C that

$$\sum_{n=0}^{\infty} (-1)^n/n! \cdot \frac{\partial^n}{\partial \alpha^{*n}} \left(\alpha^{*q} \frac{\partial^n}{\partial \alpha^n}\right) = \left(\alpha^* - \frac{\partial}{\partial \alpha}\right)^q \exp\left(-\frac{\partial^2}{\partial \alpha \partial \alpha^*}\right) \quad (2.3.8)$$

and further, that

$$\left(\alpha^* - \frac{\partial}{\partial \alpha}\right)^p \phi_1(\alpha,\alpha^*)\phi_2(\alpha,\alpha^*) = \sum_{r=0}^{p} [(-1)^r p!/r!(p-r)!] \left[\frac{\partial^r}{\partial \alpha^r}\phi_1(\alpha,\alpha^*)\right]\left[\left(\alpha^* - \frac{\partial}{\partial \alpha}\right)^{p-r} \phi_2(\alpha,\alpha^*)\right] \quad (2.3.9)$$

Where we have dispensed with arrows and the conventional direction of differentiation is to be understood. At the outset we also define $S^{(N)}_{1,2}(\alpha^*,\alpha)$ and $S^{(A)}_{1,2}(\alpha,\alpha^*)$ in power series expansions, with

$$S^{(N)}_{1,2}(\alpha^*,\alpha) = \sum_{pq} {}^{q}_{p}S^{(N)}_{1,2}\, \alpha^{*p} \alpha^q \quad (2.3.10)$$

$$S^{(A)}_{1,2}(\alpha,\alpha^*) = \sum_{pq} {}^{q}_{p}S^{(A)}_{1,2}\, \alpha^{p} \alpha^{*q} \quad (2.3.11)$$

and then operator functions:

$$\hat{S}^{(N)}_{1,2}\left(\alpha^*, \alpha + \frac{\partial}{\partial \alpha^*}\right) = \sum_{pq} {}^{q}_{p}S^{(N)}_{1,2}\, \alpha^{*p}\left(\alpha + \frac{\partial}{\partial \alpha^*}\right)^q \quad (2.3.12)$$

$$\hat{S}^{(N)}_{1,2}\left(\alpha, \alpha^* + \frac{\partial}{\partial \alpha}\right) = \sum_{pq} {}^{q}_{p}S^{(N)}_{1,2}\, \alpha^{q}\left(\alpha^* + \frac{\partial}{\partial \alpha}\right)^p \quad (2.3.13)$$

$$\hat{S}^{(A)}_{1,2}\left(\alpha, \alpha^* - \frac{\partial}{\partial \alpha}\right) = \sum_{pq} {}^{q}_{p}S^{(A)}_{1,2}\, \alpha^{p}\left(\alpha^* - \frac{\partial}{\partial \alpha}\right)^q \quad (2.3.14)$$

$$\hat{S}^{(A)}_{1,2}\left(\alpha^*, \alpha - \frac{\partial}{\partial \alpha^*}\right) = \sum_{pq} {}^{q}_{p}S^{(A)}_{1,2}\, \alpha^{*q}\left(\alpha - \frac{\partial}{\partial \alpha^*}\right)^p \quad (2.3.15)$$

and

$$\hat{S}^{(N)}_{1,2}\left(\alpha^* - \frac{\partial}{\partial \alpha}, \alpha\right) = \sum_{pq} {}^{q}_{p}S^{(N)}_{1,2}\left(\alpha^* - \frac{\partial}{\partial \alpha}\right)^p \alpha^q \quad (2.3.16)$$

$$\hat{S}^{(N)}_{1,2}\left(\alpha - \frac{\partial}{\partial \alpha^*}, \alpha^*\right) = \sum_{pq} {}^{q}_{p}S^{(N)}_{1,2}\left(\alpha - \frac{\partial}{\partial \alpha^*}\right)^q \alpha^{*p} \quad (2.3.17)$$

$$\hat{S}^{(A)}_{1,2}\left(\alpha + \frac{\partial}{\partial \alpha^*}, \alpha^*\right) = \sum_{pq} {}^{q}_{p}S^{(A)}_{1,2}\left(\alpha + \frac{\partial}{\partial \alpha^*}\right)^p \alpha^{*q} \quad (2.3.18)$$

$$\hat{S}^{(A)}_{1,2}\left(\alpha^* + \frac{\partial}{\partial \alpha}, \alpha\right) = \sum_{pq} {}^{q}_{p}S^{(A)}_{1,2}\left(\alpha^* + \frac{\partial}{\partial \alpha}\right)^q \alpha^{p} \quad (2.3.19)$$

Now the four expressions listed as (2.3.4) to (2.3.7) may be treated in an essentially similar manner so as to restate their results in terms of the factored form of the pseudo probability $W^{(\Omega)}(\beta,\beta^*;\ t+\tau;\ \alpha,\alpha^*;\ t)$. We will follow this development explicitly for (2.3.4). Expanding the exponential as a power series we are considering the relation

$$\langle \hat{S}_1(t+\tau)\hat{S}_2(t)\rangle_{\rho(t)} = \int d^2\alpha \int d^2\beta \sum_{n=0}^{\infty} (1/n!)\left[\frac{\partial^n}{\partial\alpha^n} P(\alpha,\alpha^*,t)\right] \left[\frac{\partial^n}{\partial\alpha^{*n}} \overset{(A)}{\mathcal{P}}(\beta,\beta^*,\tau|\alpha,\alpha^*,0)\right] \overset{(N)}{S_1}(\beta,\beta^*)\, \overset{(A)}{S_2}(\alpha,\alpha^*) \quad (2.3.20)$$

If we integrate by parts with respect to α^* n times, assuming the integrated parts vanish, we find

$$\langle \hat{S}_1(t+\tau)\hat{S}_2(t)\rangle_{\rho(t)} = \int d^2\alpha \int d^2\beta \sum_{n=0}^{\infty} (-1)^n/n!\ \cdot \frac{\partial^n}{\partial\alpha^{*n}}\left[\overset{(A)}{S_2}(\alpha,\alpha^*) \frac{\partial^n}{\partial\alpha^n} \overset{(N)}{P}(\alpha,\alpha^*,t)\right] \overset{(A)}{\mathcal{P}}(\beta,\beta^*,\tau|\alpha,\alpha^*,0)\, \overset{(N)}{S_1}(\beta,\beta^*) \quad (2.3.21)$$

With (2.3.11) taken together with (2.3.8) it then follows that

$$\sum_{n=0}^{\infty} (-1)^n/n! \cdot \frac{\partial^n}{\partial\alpha^{*n}}\left[\overset{(A)}{S_2}(\alpha,\alpha^*) \frac{\partial^n}{\partial\alpha^n} \overset{(N)}{P}(\alpha,\alpha^*,t)\right] = \sum_{pq} \overset{(A)}{S}{}^{q}_{2\,p}\, \alpha^p \left(\alpha^* - \frac{\partial}{\partial\alpha}\right)^q \exp\left(-\frac{\partial^2}{\partial\alpha\partial\alpha^*}\right) \overset{(N)}{P}(\alpha,\alpha^*,t) \quad (2.3.22)$$

and thus, acknowledging (2.3.14), we have

$$\sum_{n=0}^{\infty} (-1)^n/n!\ \frac{\partial^n}{\partial\alpha^{*n}}\left[\overset{(A)}{S_2}(\alpha,\alpha^*) \frac{\partial^n}{\partial\alpha^n} \overset{(N)}{P}(\alpha,\alpha^*,t)\right] = \overset{\wedge(A)}{S_2}\left(\alpha,\alpha^* - \frac{\partial}{\partial\alpha}\right) \exp\left(-\frac{\partial^2}{\partial\alpha\partial\alpha^*}\right) \overset{(N)}{P}(\alpha,\alpha^*,t) \quad (2.3.23)$$

Finally, we may quote from the papers of Agarwal and Wolf (1970) the result

$$\exp\left(-\frac{\partial^2}{\partial\alpha\partial\alpha^*}\right) \overset{(N)}{P}(\alpha,\alpha^*,t) = \overset{(A)}{P}(\alpha,\alpha^*,t) \quad (2.3.24)$$

Therefore, from (2.3.21) and (2.3.23) we may extract the expression

$$\langle \hat{S}_1(t+\tau)\hat{S}_2(t)\rangle_{\rho(t)} = \int d^2\alpha \int d^2\beta \, [\hat{S}_2^{(A)}(\alpha,\alpha^* - \frac{\partial}{\partial\alpha}) P^{(A)}(\alpha,\alpha^*,t)] \\ \mathcal{P}^{(A)}(\beta,\beta^*,\tau|\alpha,\alpha^*,0)\, S_1^{(N)}(\beta,\beta^*) \qquad (2.3.25)$$

Accompanying this there are three similar results arising in (2.3.5) to (2.3.7), namely

$$\langle \hat{S}_1(t)\hat{S}_2(t+\tau)\rangle_{\rho(t)} = \int d^2\alpha \int d^2\beta \, [\hat{S}_1^{(A)}(\alpha^*,\alpha - \frac{\partial}{\partial\alpha^*}) P^{(A)}(\alpha,\alpha^*,t) \\ \mathcal{P}^{(A)}(\beta,\beta^*,\tau|\alpha,\alpha^*,0)\, S_2^{(N)}(\beta,\beta^*) \qquad (2.3.26)$$

and

$$\langle \hat{S}_1(t+\tau)\hat{S}_2(t)\rangle_{\rho(t)} = \int d^2\alpha \int d^2\beta \, [\hat{S}_2^{(N)}(\alpha^*,\alpha + \frac{\partial}{\partial\alpha^*}) P^{(N)}(\alpha,\alpha^*,t)] \\ \mathcal{P}^{(N)}(\beta,\beta^*,\tau|\alpha,\alpha^*,0)\, S_1^{(A)}(\beta,\beta^*) \qquad (2.3.27)$$

$$\langle \hat{S}_1(t)\hat{S}_2(t+\tau)\rangle_{\rho(t)} = \int d^2\alpha \int d^2\beta \, [\hat{S}_1^{(N)}(\alpha,\alpha^* + \frac{\partial}{\partial\alpha}) P^{(N)}(\alpha,\alpha^*,t)] \\ \mathcal{P}^{(N)}(\beta,\beta^*,\tau|\alpha,\alpha^*,0)\, S_2^{(A)}(\beta,\beta^*) \qquad (2.3.28)$$

In these four integrals we have achieved expression in terms of the factored forms of $W^{(A)}(\beta,\beta^*,\tau;\ \alpha,\alpha^*\ 0)$ and $W^{(N)}(\beta,\beta^*,\tau;\ \alpha,\alpha^*\ 0)$.

We wish to investigate one further development before leaving this section. Equations (2.3.25) to (2.3.28) utilise explicitly the operators (2.3.12) to (2.3.15). With little extra effort we may re-express these results in forms which draw on the operator definitions (2.3.16) to (2.3.19). As a result, phase-space integrals are reached which use normal order equivalents for both operators $\hat{S}_1(a,a^\dagger)$ and $\hat{S}_2(a,a^\dagger)$ together with antinormal equivalents for the pseudo joint probability, and of course, also visa versa.

We begin with the consideration of the relationship between the normal order equivalent $S^{(N)}(\alpha^*,\alpha)$ and the antinormal order equivalent

$S^{(A)}(\alpha,\alpha^*)$ for any operator $\hat{S}(a,a^\dagger)$. As a generalisation of (2.3.24), we have in fact

$$\overset{(A)}{S}(\alpha,\alpha^*) = \exp\left(-\frac{\partial^2}{\partial\alpha\partial\alpha^*}\right)\overset{(N)}{S}(\alpha^*,\alpha) \tag{2.3.29}$$

With a view to expressing $S^{(A)}(\alpha,\alpha^*)$ explicitly in terms of the coefficients ${}^{q}_{p}S^{(N)}$ of (2.3.10) we then take a function $\Phi(\alpha,\alpha^*)$, where

$$\Phi(\alpha,\alpha^*) = \exp\left(-\frac{\partial^2}{\partial\alpha\partial\alpha^*}\right)\overset{(N)}{S}(\alpha^*,\alpha)\,\phi(\alpha,\alpha^*) \tag{2.3.30}$$

$\phi(\alpha,\alpha^*)$ being arbitrary. With reference to (2.3.10) we then find

$$\Phi(\alpha,\alpha^*) = \sum_{pq} {}^{q}_{p}\overset{(N)}{S} \sum_{n=0}^{\infty} (-1)^n/n!\left[\frac{\partial^n}{\partial\alpha^{*n}}\left(\alpha^{*p}\frac{\partial^n}{\partial\alpha^n}\right)\right]\alpha^q\,\phi(\alpha,\alpha^*) \tag{2.3.31}$$

and invoking (2.3.8) this yields

$$\Phi(\alpha,\alpha^*) = \sum_{pq} {}^{q}_{p}\overset{(N)}{S}\left(\alpha^*-\frac{\partial}{\partial\alpha}\right)^p \sum_{n=0}^{\infty}(-1)^n/n!\left[\frac{\partial^n}{\partial\alpha^n}\left(\alpha^q\frac{\partial^n}{\partial\alpha^{*n}}\right)\right]\phi(\alpha,\alpha^*) \tag{2.3.32}$$

which, again with (2.3.8), yields

$$\Phi(\alpha,\alpha^*) = \sum_{pq} {}^{q}_{p}\overset{(N)}{S}\left(\alpha^*-\frac{\partial}{\partial\alpha}\right)^p\left(\alpha-\frac{\partial}{\partial\alpha^*}\right)^q\exp\left(-\frac{\partial^2}{\partial\alpha\partial\alpha^*}\right)\phi(\alpha,\alpha^*) \tag{2.3.33}$$

Taking specifically $\phi(\alpha,\alpha^*) = 1$, in this result, we have our expression for $S^{(A)}(\alpha,\alpha^*)$ in terms of the coefficients ${}^{q}_{p}S^{(N)}$ belonging to $S^{(N)}(\alpha^*,\alpha)$:

$$\begin{aligned}\overset{(A)}{S}(\alpha,\alpha^*) &= \sum_{pq}\sum_{r=0}^{p} {}^{q}_{p}\overset{(N)}{S}(-1)^r/r!(p-r)!\left(\frac{\partial^r}{\partial\alpha^r}\alpha^q\right)\alpha^{*p-r} \\ &= \Bigg(\sum_{\substack{pq\\ p\le q}}\sum_{r=0}^{p} + \sum_{\substack{pq\\ p>q}}\sum_{r=0}^{q}\Bigg){}^{q}_{p}\overset{(N)}{S}\left[(-1)^r p!\,q!/r!(p-r)!(q-r)!\right] \\ &\qquad\cdot\alpha^{q-r}\alpha^{*\,p-r}\end{aligned} \tag{2.3.34}$$

Use of the expansion (2.3.34) is now made by writing

$$\overset{\wedge(A)}{S}\left(\alpha,\alpha^*-\frac{\partial}{\partial\alpha}\right) = \Bigg(\sum_{\substack{pq\\ p\le q}}\sum_{r=0}^{p} + \sum_{\substack{pq\\ p>q}}\sum_{r=0}^{q}\Bigg){}^{q}_{p}\overset{(N)}{S}\left[(-1)^r p!\,q!/r!(p-r)!(q-r)!\right]\alpha^{q-r}\left(\alpha^*-\frac{\partial}{\partial\alpha}\right)^{p-r} \tag{2.3.35}$$

which may further be expressed in the form of (2.3.34):

$$\overset{\wedge(A)}{S}\left(\alpha,\alpha^*-\frac{\partial}{\partial\alpha}\right) = \sum_{p,q=0}^{\infty} {}^{q}_{p}\overset{(N)}{S}\sum_{r=0}^{p}\left[(-1)^r p!/(p-r)!\right]\left(\frac{\partial^r}{\partial\alpha^r}\alpha^q\right)\left(\alpha^*-\frac{\partial}{\partial\alpha}\right)^{p-r} \tag{2.3.36}$$

Hence, having (2.3.9) it is established that

$$\hat{S}^{(A)}(\alpha,\alpha^*-\frac{\partial}{\partial\alpha}) = \hat{S}^{(N)}(\alpha^*-\frac{\partial}{\partial\alpha},\alpha) \qquad (2.3.37)$$

In a like manner it may also be shown that

$$\hat{S}^{(A)}(\alpha^*,\alpha-\frac{\partial}{\partial\alpha^*}) = \hat{S}^{(N)}(\alpha-\frac{\partial}{\partial\alpha^*},\alpha^*) \qquad (2.3.38)$$

$$\hat{S}^{(N)}(\alpha^*,\alpha+\frac{\partial}{\partial\alpha^*}) = \hat{S}^{(A)}(\alpha+\frac{\partial}{\partial\alpha^*},\alpha^*) \qquad (2.3.39)$$

$$\hat{S}^{(N)}(\alpha,\alpha^*+\frac{\partial}{\partial\alpha}) = \hat{S}^{(A)}(\alpha^*+\frac{\partial}{\partial\alpha},\alpha) \qquad (2.3.40)$$

By the introduction of these results to (2.3.25) to (2.3.28) we then acquire, in addition to the same, the following phase-space integrals for two-time averages:

$$\langle\hat{S}_1(t+\tau)\hat{S}_2(t)\rangle_{\rho(t)} = \int d^2\alpha\int d^2\beta\,[\hat{S}_2^{(N)}(\alpha^*-\frac{\partial}{\partial\alpha},\alpha)P^{(A)}(\alpha,\alpha^*,t)]\,\mathcal{P}^{(A)}(\beta,\beta^*,\tau|\alpha,\alpha^*,0)\,S_1^{(N)}(\beta,\beta^*) \qquad (2.3.41)$$

$$\langle\hat{S}_1(t)\hat{S}_2(t+\tau)\rangle_{\rho(t)} = \int d^2\alpha\int d^2\beta\,[\hat{S}_1^{(N)}(\alpha-\frac{\partial}{\partial\alpha^*},\alpha^*)P^{(A)}(\alpha,\alpha^*,t)]\,\mathcal{P}^{(A)}(\beta,\beta^*,\tau|\alpha,\alpha^*,0)\,S_2^{(N)}(\beta,\beta^*) \qquad (2.3.42)$$

$$\langle\hat{S}_1(t+\tau)\hat{S}_2(t)\rangle_{\rho(t)} = \int d^2\alpha\int d^2\beta\,[\hat{S}_2^{(A)}(\alpha+\frac{\partial}{\partial\alpha^*},\alpha^*)P^{(N)}(\alpha,\alpha^*,t)]\,\mathcal{P}^{(N)}(\beta,\beta^*,\tau|\alpha,\alpha^*,0)\,S_1^{(A)}(\beta,\beta^*) \qquad (2.3.43)$$

$$\langle\hat{S}_1(t)\hat{S}_2(t+\tau)\rangle_{\rho(t)} = \int d^2\alpha\int d^2\beta\,[\hat{S}_1^{(A)}(\alpha^*+\frac{\partial}{\partial\alpha},\alpha)P^{(N)}(\alpha,\alpha^*,t)]\,\mathcal{P}^{(N)}(\beta,\beta^*,\tau|\alpha,\alpha^*,0)\,S_2^{(A)}(\beta,\beta^*) \qquad (2.3.44)$$

These final four expressions correspond exactly to the form of the quantum regression theorem given by Louisell in his Varenna Summer School notes (Louisell, 1969).

PART II

DETAILED BALANCE IN THE OPEN QUANTUM MARKOFFIAN SYSTEM

A system subject to statistical description may be maintained in equilibrium in the presence of an underlying dynamical structure by a nett balance between transitions into and out of each physical state. Considerable simplification of perspective is possible when transitions between any pair of states admit to a "balancing in detail".

CHAPTER III

INTRODUCTION

3.1 An Introduction to Detailed Balance

The probings of men into the nature of physical systems have their results classified within the various theoretical formalisms of the physical sciences. Those falling within the scope of statistical mechanics represent one of the boldest ventures of theoretical physicists; by their very nature the problems encountered in this field are ones of extreme complexity. It is, nevertheless, a tribute to the beauty of our physical world that within such complex situations there are often relationships of exceeding simplicity pointing the way to an understanding at a very fundamental level. One such is the principle of detailed balance, a principle conceived in response to the concept of detailed balancing of transitions introduced by Fowler (1924) in the early 1920s. It is in reference to this principle that the following five chapters are presented.

To set detailed balance within its context it is necessary for us to first appreciate that any statistical equilibrium, while a stationary situation in the macroscopic sense, is, at a more fundamental level, a circumstance of intense dynamical activity. We are well familiar with the thermodynamic equilibrium accompanying the maximisation of entropy. Then in this instance, a gas maintained in equilibrium at a temperature T presents, on close examination, a vast array of molecular motions and collisions; these in fact working to maintain equilibrium. It is in this environment of microscopic motions that we discover the origins of detailed balance.

Let us grasp first the concept of a balance in detail as against balance in general. It is clear that any dynamical behaviour persisting throughout equilibrium must admit to an overall balance which maintains the structure of the equilibrium probabilities. Thus, let us envisage, for example, the circumstance illustrated in Fig.(3.1.1). Here a body exists in thermodynamic equilibrium at a temperature T with the walls of its container and the intervening radiation field. Granting the equilibrium conditions, the situation at a more fundamental level is nonetheless one of flux as radiation is being continually emitted and absorbed. A balance is then implied and is simple expressed by

$$\text{Radiated Power} = \text{Absorbed Power} \tag{3.1.1}$$

without this relationship the equilibrium temperature T may not be maintained. In reference to this condition we see that it is not at all restrictive with respect to details of the manner in which balance is achieved. Perhaps, on the one had, radiation is emitted in preference at one frequency and balanced by preferential absorption at some different frequency. Or, on the other, may it be that balance is attained for each selected frequency, polarisation, and direction of momentum, thus achieving overall balancing through the balance of individual parts. It is in fact the second proposition which holds true, of which one may easily be convinced by arguing with the aid of an imaginary filter encompassing the enclosed body.* In recognising this we are moving from the gross balance of (3.1.1) to a balance in detail.

The principle of detailed balance was used throughout the infancy

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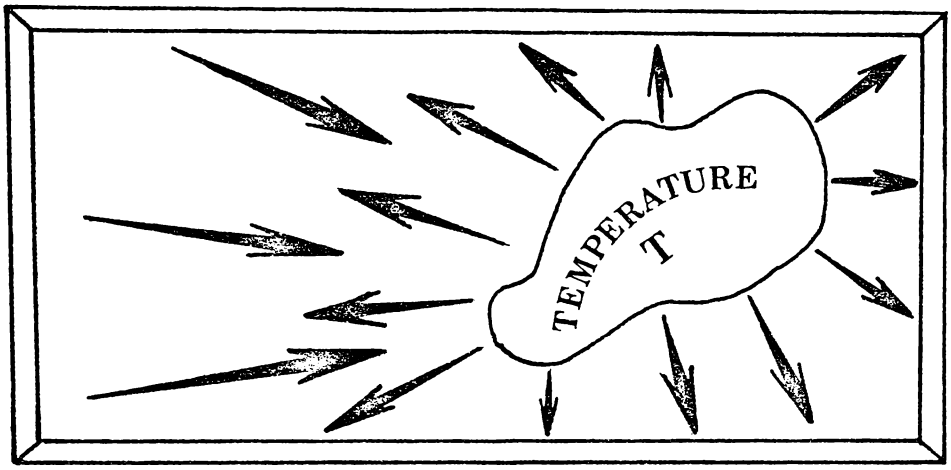
*This illustration is taken from Reif (1965).

(a)



(b)

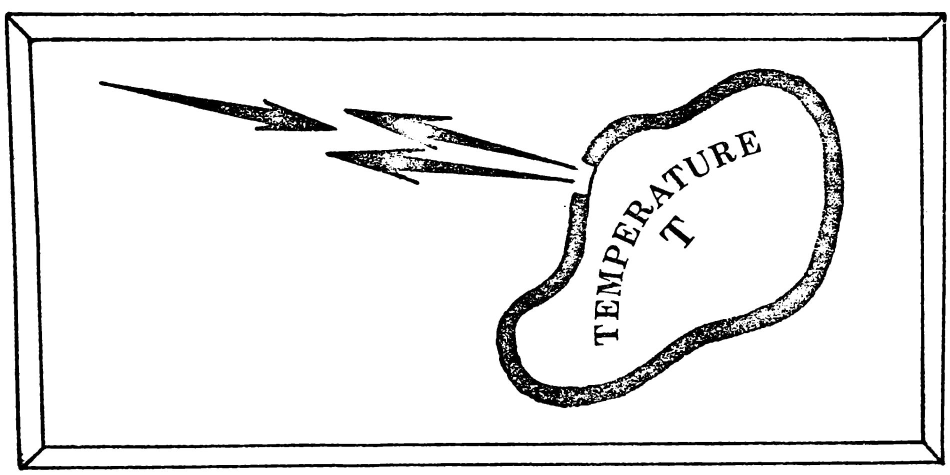


Fig (3·1·1) Balance of absorbed and radiated power at equilibrium: (a) overall balance, (b) balance in detail.

of statistical mechanics by various authors facing the problems of atoms, electrons, and photons in equilibrium (Kramers, 1923; Pauli, 1923; Milne, 1924; Fowler, 1924; Dirac, 1924). Its explicit expression differs somewhat depending on the context in which it is conceived, although throughout these applications the underlying concept of "detailed balancing" (Fowler, 1924) is fundamental. (As an example, its occurrence within the context of Boltzmann's theory for gases is accounted in Tolman's classic text (Tolman, 1967).) For ourselves we choose an introduction in relation to the much studied Pauli master equation. The circumstances in which this applies are familiar, we take an isolated system suffering a small perturbation or Hamiltonian interaction term so that transitions between the various states of the free Hamiltonian are possible. The master equation may then be taken from one of the many standard texts (see, for example, Kittel, 1958; Reif, 1965; Dennery, 1972). We write

$$\frac{dp_n}{dt} = \sum_m \gamma_{m,n} p_m - \gamma_{n,m} p_n \tag{3.1.2}$$

Here p_n is the probability for the acquisition of the nth state and $\gamma_{n,m}$ is a time proportional probability for transition between the nth and mth states respectively. The temporal evolution here is clearly discernible. The first term represents transitions from all states m to the nth state, while the second term represents a loss from this nth state through transitions to all states m. Now clearly, for steady-state or equilibrium conditions, (3.1.2) calls for an overall balance of transitions into and out of each nth state. Thus there is the requirement

$$\sum_m \gamma_{m,n} p_m^{ss} = \sum_n \gamma_{n,m} p_n^{ss} \qquad \forall n \tag{3.1.3}$$

or, for every state n

No. of transitions per unit time from all states m to state n	=	No. of transitions per unit time from state n to all states m	(3.1.4)

Then within the highly complex dynamical environment embodying the various transitions between states detailed balance arises to impose a profound simplicity. It proposes a "detailed balancing" of transitions, a balance in frequency of each transition with its inverse. This is the simplest way of achieving the conditions of (3.1.3) and may be taken as a ground base definition for detailed balance with expression in the form

$$\gamma_{m,n}\, p_m^{ss} = \gamma_{n,m}\, p_n^{ss} \qquad \forall\, n,m \tag{3.1.5}$$

or, for all pairs of states n and m

No. of transitions per unit time from state m to state n	=	No. of transitions per unit time from state n to state m	(3.1.6)

3.2 Microreversibility as a Basis for Detailed Balance

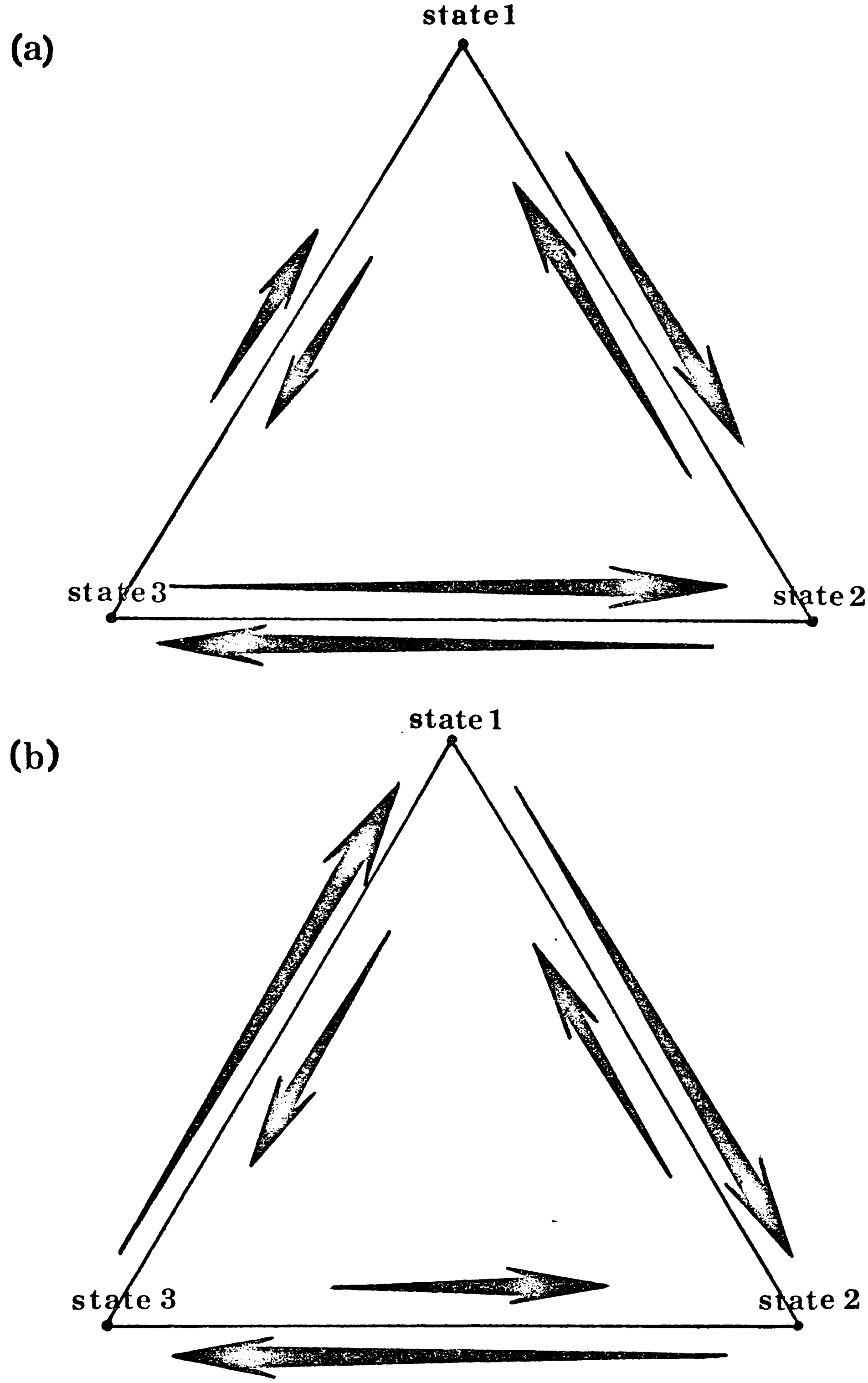
Detailed balance has been seen in the preceding section to be sufficient for equilibrium requirements, and moreover, to attribute a profound simplicity to the microdynamics underlying equilibrium conditions. However, nothing accounted so far will justify the assertion that (3.1.5) constitutes a necessary condition at equilibrium. Indeed it may not be proved from thermodynamic considerations alone that equilibrium is actually maintained by this exact balancing of each transition with its inverse. It was Fowler (1924) who, in the words of Dirac (1924), showed that "there may be cycles of inseparable processes, each of which does not balance individually, although the whole cycle forms a 'unit mechanism' which balances itself in any assembly in statistical equilibrium". This concept of cyclical, or orbital, balancing mechanisms is illustrated for a simple three state system in Fig.(3.2.1). It has been recognised as the usual requirement for nonequilibrium situations that stationarity be preserved by this cyclic balance (Klein, 1955; Tomita and Tomita, 1974) apart from a few instances where a detailed balance is assured by symmetry considerations (Graham, 1973). We must now meet the difficulty of ruling out the possibility of cyclic balancing in the equilibrium situation.

It was recognised in the early literature that there was fortunately in fact a simple resolution here. From Dirac (1924) we find, in answer to the raising of this problem in Fowler's paper of 1924:

> "It seems possible, however, to suppose that all atomic processes are reversible, or, more exactly, that if after any encounter all the velocities are reversed, then the whole process would just repeat itself backwards, the systems finally leaving the scene of action being the same as the original systems in the first process and having the reverse velocities. With this assumption, to which there are no known exceptions, each kind of encounter must be just as likely to occur as its converse in which every velocity has changed sign, the whole process taking place backwards, since there is now perfect symmetry between past and future time."

Thus it is in the reversibility of atomic motions that at a most fundamental level a foundation is found for detailed balance.

It was Tolman (1924, 1925) who, in his discussion of molecular collision processes in a gas at equilibrium, formulated this reversibility of microscopic processes into his "principle of microscopic reversibility". Here he established equal frequencies for the occurrence of any molecular collision Fig.(3.2.2a) and its reverse Fig.(3.2.2b) at equilibrium. From this, for molecules possessing spherical symmetry an equivalent relationship may be drawn for any collision and its inverse Fig.(3.2.2c). The term, principle of microscopic reversibility, or, principle of microreversibility, has come down to us today with expression in various forms (Messiah, 1970; De Groot and Mazur, 1962; Reif, 1965; Graham and Haken, 1971). For



arrow lengths are proportional to transition rates at equilibrium

Fig(3·2·1) Equilibrium dynamics for a three state system: (a) with detailed balance, (b) with a cyclic balance.

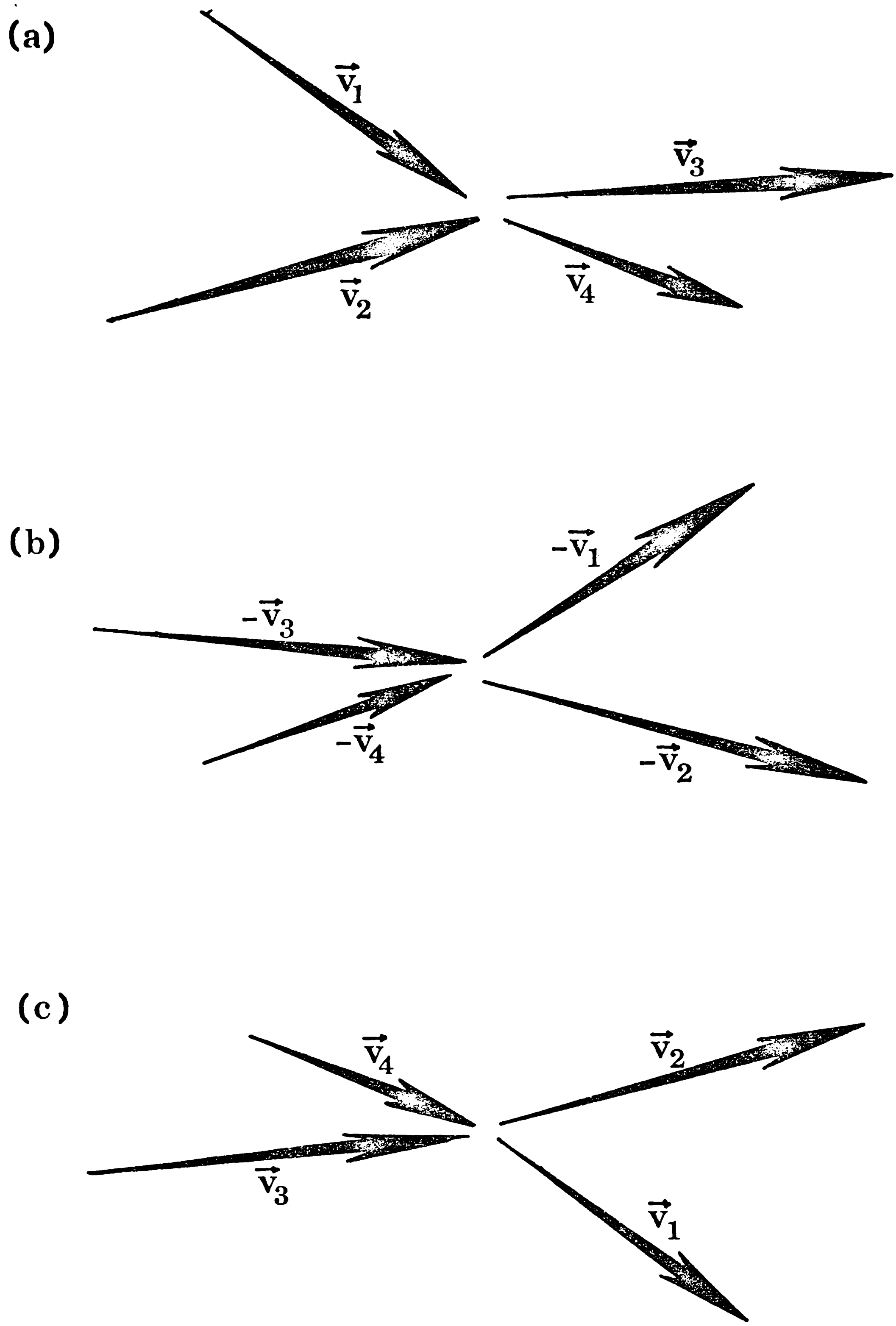


Fig (3·2·2) (a) Molecular collision, (b) Reverse collision, (c) Inverse collision.

ourselves we may take as fundamental to it the invariance under time reversal of those equations basic to the microscopic dynamics. Thus, for example, for any trajectory Fig.(3.2.3a), a solution to some Heisenberg equations of motion, the time reversed trajectory Fig.(3.2.3b) is equally well a solution to these same Heisenberg equations taken with a negative time coordinate.

With reference to the Pauli master equation (3.1.2), to be consistent with its origin within microreversibility, the detailed balance (3.1.5) should perhaps be presented in the form

$$\gamma_{m,n}\, p_m^{ss} = \gamma_{\tilde{n},\tilde{m}}\, p_{\tilde{n}}^{ss} \tag{3.2.1}$$

where ~ denotes the time reversed state. However, in this context the distinction between reverse and inverse processes becomes immaterial due to formulation in terms of transition probabilities and the early adoption of statistical assumptions (in particular that of random phases) in the derivation of the Pauli equation (Tolman, 1967). Then inasmuch as the transition probabilities are proportional to the matrix elements of some interaction Hamiltonian H_I

$$\gamma_{n,m} \propto |\langle n|H_I|m\rangle|^2 \tag{3.2.2}$$

the probability for any transition is assured to be equal to that of its inverse by the Hermitian quality of this Hamiltonian. Coupled with the conservation of energy which requires all states m accessible from n to be of the same energy, and hence, at equilibrium, to occur with equal probability, (3.1.5) is then trivially ensured for equilibrium by the two separate relationships

$$\left.\begin{array}{l}\gamma_{n,m} = \gamma_{m,n} \\ p_n^{ss} = p_m^{ss}\end{array}\right. \qquad \forall_{n,m} \tag{3.2.3}$$

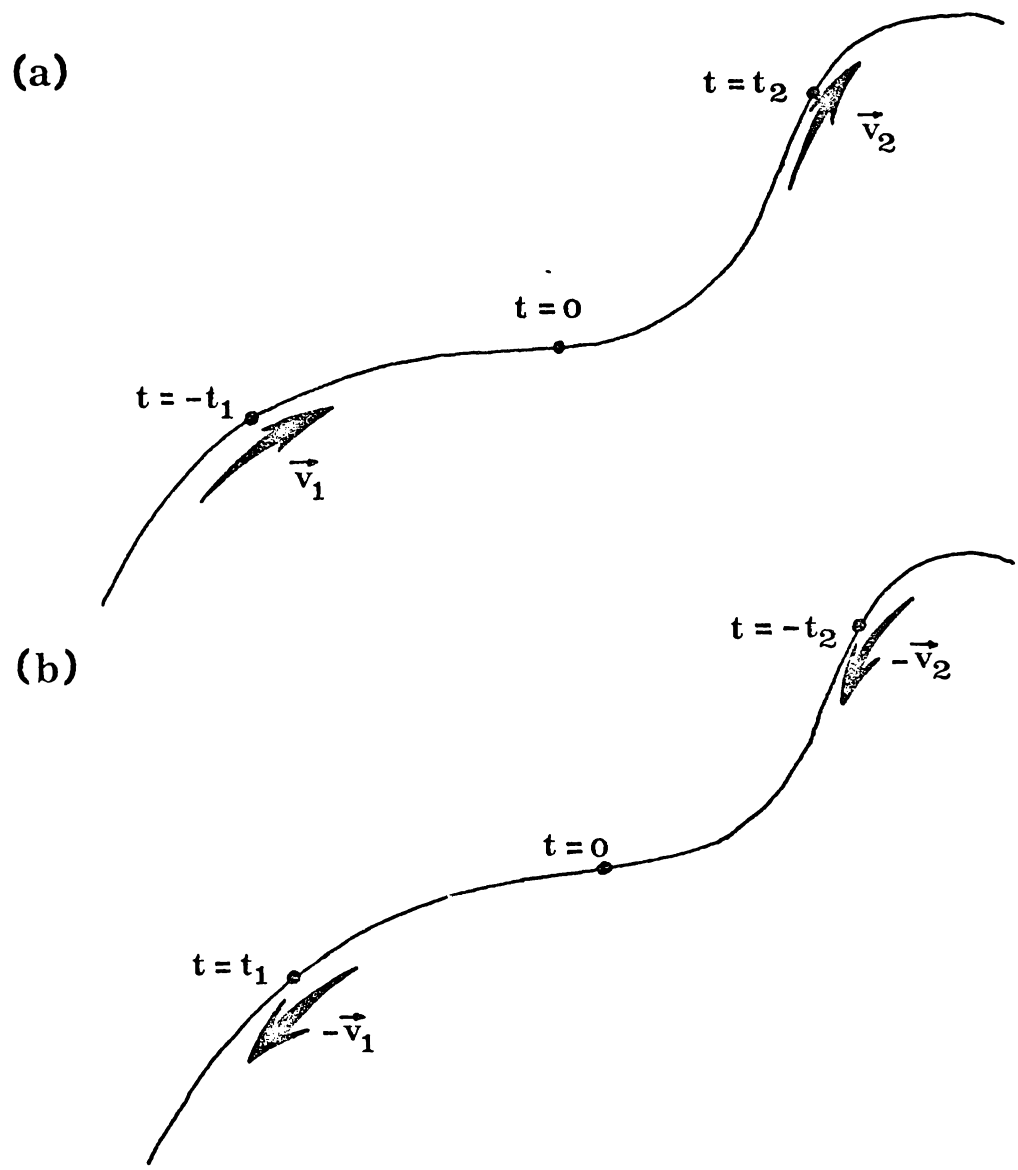


Fig (3·2·3) Time reversal of a classical trajectory: (a) single particle trajectory, (b) time reversed trajectory.

3.3 The Question of Open Quantum Systems

Apart from taking its place in standard texts on statistical mechanics detailed balance has attracted little attention in its own right since its conception in the papers of Fowler and his contemporaries. Recently, however, there has been a resurgence of interest following the proposition by Graham and Haken (1971) that this condition underlies certain unexpected features of nonequilibrium steady states. The origin of their suggestion is to be found in the steady state of the single mode laser (Haken, 1970), responsible for so many innovations in theoretical physics. This steady state admits analysis in terms of an analogue of the equilibrium thermodynamic potential (Graham, 1973) and the Landau theory of phase transitions (Grossmann and Richter, 1971), a behaviour completely unexpected for a nonequilibrium steady state. In the wake of Graham and Haken's observations, and contemporary with considerable present interest in the far from equilibrium steady state in general (Graham, 1973, 1975; Haken, 1975), various authors have investigated in new detail the consequences of detailed balance for situations bearing description by a classical Fokker-Plank equation (Risken, 1972; Agarwal, 1973b; Wöhrstein and Haken, 1973; Tomita and Tomita, 1974). It is within this environment that there arose a paper by Agarwal (1973c) bringing our attention to the whole question of formulating detailed balance for a purely quantum-mechanical system. What follows in the next four chapters of this thesis is the product of our response to this question.

The Pauli equation (3.1.2) is certainly a quantum-mechanical equation, it appears as a foundation-stone in quantum statistical mechanics. It is, however, built on various assumptions and approximations, perfectly admissible in themselves, but which lead to an equation in time-proportional transitions, and hence, fundamentally to the equation of a classical Markoff process. It is just this nature of the Pauli equation which admits so readily the detailed balancing concept of Fowler. But here there is a fundamental oversight, as is so aptly indicated by Kittel (1958): "The picture of a system jumping from one definite eigenstate to another is not sound quantum mechanics". Our contention is not at all with this Pauli equation, indeed this is itself quite adequately vindicated for appropriate circumstances in Zwanzig's statistical mechanics of irreversibility (1961). What we, in the following four chapters, are concerned with however is the fundamental formulation of detailed balance for an open quantum Markoffian system falling into the general description of the operator master equation (1.2.35). Having here a full operator equation access is open to much more information than is ever available from the Pauli equation alone, and in relation to this, microreversibility may hold consequences unforeseen in Pauli detailed balance. Furthermore, without the concept of a steady state maintained by detailed balancing of transition, how indeed are we going to transfer the ideas of the familiar detailed balance itself into the present environment? We approach this question in the following section.

3.4 Formulation in terms of Two-Time Averages

Before broaching the problem outlined in the previous section let us restate the master equation (3.1.2), adopting here an expression in matrix notation. We write

$$\frac{d\,p(n)}{dt} = (\gamma)^{t}\, p(n) \tag{3.4.1}$$

where p(n) is taken to be a column matrix

$$p(n) = \begin{pmatrix} p_1 \\ \vdots \\ p_n \\ \vdots \end{pmatrix} \tag{3.4.2}$$

and (γ) is the matrix

$$(\gamma)_{n,m} = \gamma_{n,m} \qquad m \neq n$$
$$(\gamma)_{n,n} = -\sum_{k} \gamma_{n,k} \tag{3.4.3}$$

We may then take directly as the formal solution

$$p(n,t) = (\exp[(\gamma)^{t} t])\, p(n,o) \tag{3.4.4}$$

where p(n,o) is some initial distribution of states.

Now, with the view of a system of discrete states intermediated by time-proportional transitions, we may take (3.4.1) as the equation of a discrete classical Markoff process. In this instance we may define a joint probability distribution $W(n, t + \tau; m,t)$ as yielding probabilities for the successive occupation of states m at time t and n at time $t + \tau$. Restricting then our attention to the stationary conditions of equilibrium, with joint distribution $W_{ss}(n,\tau; m,o)$, any two-time average, or correlation function, $\langle s^{(\alpha)}(\tau)\, s^{(\beta)}(o)\rangle_{ss}$ finds expression in the form

$$\langle S^{(\alpha)}(\tau)\, S^{(\beta)}(o)\rangle_{ss} = \sum_{n,m} S^{(\alpha)}(n)\, S^{(\beta)}(m)\, W_{ss}(n,\tau; m,o) \tag{3.4.5}$$

where $s^{(\alpha)}$ and $s^{(\beta)}$ are any two relevant observables.

Now our objective is to cast the detailed balance of (3.1.5) into a form which does not draw directly for its interpretation on the concept of detailed balancing of states. To this end we first adopt the explicit expression $W_{ss}(n,\tau; m,o)$ offered by (3.4.4) when taken together with the equilibrium solution $p^{ss}(n)$ to (3.4.1). If δ_n is a column vector of all zeros other than a unit value in the nth row, we have

$$\begin{aligned} W_{ss}(n,\tau; m,o) &= (\exp[(\gamma)^{t}\tau]\,\delta_m)_n\, p_m^{ss} \\ &= (\exp[(\gamma)^{t}\tau])_{n,m}\, p_m^{ss} \\ &= (\exp[(\gamma)\tau])_{m,n}\, p_m^{ss} \end{aligned} \tag{3.4.6}$$

Then, as it is readily seen that the detailed balance statement (3.1.5) leads by induction to the more general relationship

$$(\gamma^k)_{m,n}\, p_m^{ss} = (\gamma^k)_{n,m}\, p_n^{ss} \tag{3.4.7}$$

for an aribtrary integer k, it trivially follows that equivalent to this detailed balance is the result

$$W_{ss}(n,\tau;m,0) = W_{ss}(m,\tau;n,0) \tag{3.4.8}$$

Of course, this statement in joint probabilities is but one further expression of the verbal statement (3.1.6). Fulfilment of our objective is now accomplished inasmuch as it clearly follows from (3.4.5) that (3.4.8), and hence the statement of detailed balancing for (3.4.1), is equivalent to the requirement

$$\langle S^{(\alpha)}(\tau)\, S^{(\beta)}(0)\rangle_{ss} = \langle S^{(\beta)}(\tau)\, S^{(\alpha)}(0)\rangle_{ss} \tag{3.4.9}$$

to be adopted for arbitrary $S^{(\alpha)}$ and $S^{(\beta)}$. We have in this escaped from the conceptual strictures of detailed balancing of states to a purely formal statement, which, with the aid of the results of Chapter II, may obviously be formulated within the full scope of our master equation (1.2.35). This therefore forms the obvious basis for the transfer of detailed balance into the fully quantum-mechanical environment.

3.5 Outline of Following Chapters

The following two chapters are concerned with the formulation of detailed balance within the context of the master equation for the open quantum Markoffian system (1.2.35). The first of these constitutes work already published in Physics Letters (Carmichael and Walls, 1975a) and Zeitschrift für physik (Carmichael and Walls, 1975b), while in Chapter V we present results as yet unpublished.

We begin with the derivation of detailed balance from microdynamics and the principle of microreversibility in section 4.1. Then in section 4.2, we cast this detailed balance into a phase-space form. After this initial formulation within the phase-space calculus the accruing detailed balance conditions are subjected, in section 4.3, to a reduction through the formal classical limit to a form bearing direct relationship to the detailed balance of the classical Fokker-Plank equations. In this we conclude the position of the detailed balance obtained by literal interpretation of the quantum-mechanical Fokker-Plank equations to be the classical limit of the fully quantum-mechanical form.

We move, in section 4.4, from this phase-space treatment for the open quantum Markoffian system to its description within the energy representation. By casting the fully quantum-mechanical detailed balance conditions into a form in terms of the matrix elements of the energy representation we may move, in section 4.5, to an identification with the Pauli master equation of the previous sections. Here we isolate a class of master equations, designated diagonal master equations, as being equivalent to the Pauli situation. In these circumstances the full detailed balance conditions reduce, in a limited sense, to the simple conditions of (3.1.5) and make direct contact with the classical concept of detailed balancing of transitions. There is nonetheless a more stringent requirement in the full conditions insomuch as we now have access to the correlation of nondiagonal operators. Various simple applications and illustrations of the results of these four sections 4.2, 4.3, 4.4 and 4.5 are given in section 4.6.

We conclude Part II of this thesis in Chapter V with an examination of the generalised master equation of section 1.3 within the context of full quantum detailed balance. After establishing the general solution to this equation at equilibrium in section 5.1, the operator detailed balance conditions are tested in section 5.2. We find that terms oscillatory in the energy representation may arise from either internal couplings in the open system or nonenergy conserving terms in the system-reservoir interaction, and that these give rise to the failure of full quantum detailed balance. In section 5.3 it is shown that master equations having such terms are just those which fail to fall into the classification of diagonal master equations.

CHAPTER IV

DETAILED BALANCE IN THE OPEN QUANTUM MARKOFFIAN SYSTEM

4.1 Detailed Balance from Microreversibility

We are considering here an open Markoffian system S whose coupling to the reservoir R defines a composite closed system $S \oplus R$. The dynamics of this closed system form the microscopic foundation for the behaviour of S variables alone.

In section 1.2 we designated the density operator for $S \oplus R$ by $\chi(t)$ and that for S by $\rho(t)$ with

$$\rho(t) = tr_R\, \chi(t) \tag{4.1.1}$$

The complete Hamiltonian H divides into three parts

$$H = H_S + H_R + H_{SR} \tag{4.1.2}$$

and correspondingly the Liouvillian L is given by

$$L = (1/\hbar)[H,\ ] = L_S + L_R + L_{SR} \tag{4.1.3}$$

H_S and H_R respectively govern the free dynamics of S and R, and H_{SR} constitutes an interaction term. We assume that the Hamiltonian ensures microreversibility requiring

$$KHK^{\dagger} = \tilde{H} = H \tag{4.1.4}$$

where K is the quantum-mechanical time reversal operator (Messiah, 1970). Now $\chi(t)$ evolves in time under the unitary operator

$$U(t,t_0) = \exp[-iL(t-t_0)] \tag{4.1.5}$$

while, as demonstrated in section 1.2, the evolution of $\rho(t)$ is determined by the nonunitary operator

$$\mathcal{U}(t,t_0) = \exp[-i\mathcal{L}(t-t_0)] \tag{4.1.6}$$

with, from (1.2.36), (1.2.37) and (1.2.38)

$$\mathcal{L} = L_s + e^{-iL_st}\int_0^t tr_R(\hat{L}_{SR}(t)\hat{L}_{SR}(t-\tau)R_{TH})\, e^{iL_st} \tag{4.1.7}$$

Then, for arbitrary operators $S^{(\alpha)}$ and $S^{(\beta)}$ of S, the correlation $\langle S^{(\alpha)}(t_0+\tau)\ S^{(\beta)}(t_0)\rangle_{\rho(to)}$, evaluated in section 2.1 in terms of $\rho(t_0)$, closely approximates its corresponding microscopic form $\langle S^{(\alpha)}(t_0+\tau)\ S^{(\beta)}(t_0)\rangle_{\chi(to)}$.

The nonunitary contribution to $\mathcal{L}$ introduces to S a dynamical irreversibility (George *et al.*, 1972; Prigogine *et al.*, 1973; Prigogine, 1973). As a consequence the system exhibits a long time evolution to a stationary state ρ_{ss} with

$$\mathcal{L}\rho_{ss} = 0 \tag{4.1.8}$$

Generally, this macroscopic steady state cannot presuppose microscopic stationarity, that is, it cannot presuppose the existence of a χ_{ss} such that

$$L\rho_{ss} = 0 \tag{4.1.9}$$

As for the classical circumstance (De Groot and Mazur, 1962), the quantum detailed balance is necessitated by microreversibility only in the equilibrium situation where microscopic stationarity does develop. Restricting ourselves to this situation we write

$$\begin{aligned}\langle S^{(\alpha)}(\tau)\, S^{(\beta)}(0)\rangle_{\chi_{ss}} &= tr_{S\oplus R}\,\chi_{ss}[U(\tau,0)\, S^{(\alpha)}(0)]\, S^{(\beta)}(0)\\ &= tr_{S\oplus R}\,\chi_{ss}\exp[(iH/\hbar)\tau]\, S^{(\alpha)}(0)\exp[-(iH/\hbar)\tau]\, S^{(\beta)}(0)\end{aligned} \tag{4.1.10}$$

Now, using the antilinear property of K (Messiah, 1970), it follows that for an arbitrary operator 0

$$\mathrm{tr}\, O = \mathrm{tr}\, \tilde{O}^{\dagger} \tag{4.1.11}$$

Applying this together with microreversibility in the form (4.1.4), and also noting the microscopic stationarity (4.1.8), we have

$$\begin{aligned}
&\langle \overset{(\alpha)}{S}(\tau)\, \overset{(\beta)}{S}(0)\rangle_{\chi_{ss}} \\
&= \mathrm{tr}_{S\oplus R}\, \tilde{S}^{(\beta)\dagger}(0) \exp[-(iH/\hbar)\tau]\, \tilde{S}^{(\alpha)\dagger}(0) \exp[(iH/\hbar)\tau]\, \tilde{\chi}_{ss} \\
&= \mathrm{tr}_{S\oplus R}\, \tilde{\chi}_{ss} \exp[(iH/\hbar)\tau]\, \tilde{S}^{(\beta)\dagger}(0) \exp[-(iH/\hbar)\tau]\, \tilde{S}^{(\alpha)\dagger}(0) \\
&= \mathrm{tr}_{S\oplus R}\, \tilde{\chi}_{ss} [U(\tau,0)\, \tilde{S}^{(\beta)\dagger}(0)]\, \tilde{S}^{(\alpha)\dagger}(0)
\end{aligned} \tag{4.1.12}$$

thus, subject to the condition

$$\tilde{\chi}_{ss} = \chi_{ss} \tag{4.1.13}$$

we have established

$$\langle \overset{(\alpha)}{S}(\tau)\, \overset{(\beta)}{S}(0)\rangle_{\chi_{ss}} = \langle \tilde{S}^{(\beta)\dagger}(\tau)\, \tilde{S}^{(\alpha)\dagger}(0)\rangle_{\chi_{ss}} \tag{4.1.14}$$

Conditional then only on the validity of the Markoffian and weak coupling approximations leading to (4.1.6), in equilibrium microreversibility ensures the relationship

$$\langle \overset{(\alpha)}{S}(\tau)\, \overset{(\beta)}{S}(0)\rangle_{\rho_{ss}} = \langle \tilde{S}^{(\beta)\dagger}(\tau)\, \tilde{S}^{(\alpha)\dagger}(0)\rangle_{\rho_{ss}} \tag{4.1.15}$$

which we may adopt as the full quantum-mechanical form of detailed balance sought in the previous section. This is just the definition proposed by Agarwal (1973b). The absence of the Hermitian conjugation in his definition merely reflects alternative definition of the time reversal operator.

We should recognise that the procedure followed here in (4.1.10) to (4.1.14) runs parallel to the corresponding classical development. If the usual classical discussion in terms of a joint probability density (De Groot and Mazur, 1962) is substituted by one in terms of two-time averages, use of the Liouville formalism allows both classical and quantum-mechanical treatments to be couched in the same language. We would also emphasise the difference between the statements of (4.1.14) and (4.1.15). The Markoffian and weak coupling approximations may indeed lead to a loss of detailed balance (as defined by (4.1.15)), even in equilibrium. This possibility will be revealed in the following chapter.

4.2 Detailed Balance in the Phase-Space Calculus

We have outlined the essentials of the phase-space calculus of Agarwal and Wolf (1970) in section 2.2 and appendix B. When introduced to the open Markoffian system $\rho(t)$ is replaced by the quasiprobability distribution function $P^{(\Omega)}(\alpha, \alpha^*; t)$:

$$\hat{\Omega}\, P^{(\Omega)}(\alpha,\alpha^*,t) = (1/\pi)\,\rho(a,a^\dagger,t) \tag{4.2.1}$$

where the factor $(1/\pi)$ ensures normalisation to unity. The operator master equation

$$\frac{d\rho}{dt} = \mathcal{L}\rho \tag{4.2.2}$$

becomes a differential equation, generally of the Fokker-Plank type, for $P^{(\Omega)}(\alpha, \alpha^*; t)$:

$$\frac{d P^{(\Omega)}(\alpha,\alpha^*,t)}{dt} = \mathcal{L}^{(\Omega)}(\alpha,\alpha^*)\, P^{(\Omega)}(\alpha,\alpha^*,t) \tag{4.2.3}$$

where

$$\hat{\Omega}\, \mathcal{L}^{(\Omega)}(\alpha,\alpha^*)\, S^{(\Omega)}(\alpha,\alpha^*) = \mathcal{L}\, \hat{S}(a,a^\dagger) \tag{4.2.4}$$

Let us consider the quantum detailed balance in the form

$$\langle \hat{S}_1(\tau)\,\hat{S}_2(0)\rangle_{\rho_{ss}} = \langle \tilde{\hat{S}}_2^{\dagger}(\tau)\,\tilde{\hat{S}}_1^{\dagger}(0)\rangle_{\rho_{ss}} \tag{4.2.5}$$

Time correlation functions are to be evaluated by double integration in phase space and we have from (2.2.27), (2.2.29) and (2.2.30)

$$\langle \hat{S}_1(\tau)\,\hat{S}_2(0)\rangle_{\rho_{ss}} = \int d^2\alpha \int d^2\alpha_0\, S_1^{(\Omega)}(\alpha,\alpha^*)\, S_2^{(\tilde{\Omega})}(\alpha_0,\alpha_0^*)\, \overleftarrow{W}_{ss}^{(\Omega)}(\alpha,\alpha^*,\tau;\alpha_0,\alpha_0^*,0) \tag{4.2.6}$$

with the stationary quasiprobability density

$$\overleftarrow{W}_{ss}^{(\Omega)}(\alpha,\alpha^*,\tau;\alpha_0,\alpha_0^*,0) = P_{ss}^{(\Omega)}(\alpha_0,\alpha_0^*)\exp[\overleftrightarrow{\Lambda}(\alpha_0,\alpha_0^*)]\, \exp[\mathcal{L}^{(\tilde{\Omega})}(\alpha,\alpha^*)\tau]\,\delta^{(2)}(\alpha-\alpha_0) \tag{4.2.7}$$

Considering then the Ω-equivalent $\tilde{S}^{(\Omega)}(\alpha,\alpha^*)$ of $\tilde{\hat{S}}(a,a^{\dagger})^{\dagger}$ we are lead to define the $\tilde{\Omega}$-mapping with $\tilde{\Omega}(\beta,\beta^*) = \Omega(-\beta^*,-\beta)$. It then follows from (2.2.4) to (2.2.6) that

$$\tilde{S}^{(\Omega)}(\alpha,\alpha^*) = S^{(\tilde{\Omega})}(\alpha^*,\alpha) \tag{4.2.8}$$

and hence, using the Ω-mapping, we may write

$$\begin{aligned}\langle \tilde{\hat{S}}_2^{\dagger}(\tau)\,\tilde{\hat{S}}_1^{\dagger}(0)\rangle_{\rho_{ss}} &= \int d^2\alpha \int d^2\alpha_0\, \tilde{S}_2^{(\tilde{\Omega})}(\alpha,\alpha^*)\, \tilde{S}_1^{(\tilde{\Omega})}(\alpha_0,\alpha_0^*)\, \overleftarrow{W}_{ss}^{(\tilde{\Omega})}(\alpha,\alpha^*,\tau;\alpha_0,\alpha_0^*,0)\\ &= \int d^2\alpha \int d^2\alpha_0\, S_2^{(\tilde{\Omega})}(\alpha^*,\alpha)\, S_1^{(\Omega)}(\alpha_0^*,\alpha_0)\, \overleftarrow{W}_{ss}^{(\tilde{\Omega})}(\alpha,\alpha^*,\tau;\alpha_0,\alpha_0^*,0)\end{aligned}$$

Making the change of variables $\alpha \to \alpha_0^*$, $\alpha_0 \to \alpha^*$

$$\langle \tilde{\hat{S}}_2^{\dagger}(\tau)\,\tilde{\hat{S}}_1^{\dagger}(0)\rangle_{\rho_{ss}} = \int d^2\alpha \int d^2\alpha_0\, S_1^{(\Omega)}(\alpha,\alpha^*)\, S_2^{(\tilde{\Omega})}(\alpha_0,\alpha_0^*)\, \overleftarrow{W}_{ss}^{(\tilde{\Omega})}(\alpha_0^*,\alpha_0,\tau;\alpha^*,\alpha,0) \tag{4.2.9}$$

Since $S_1^{(\Omega)}(\alpha,\alpha^*)$ and $S^{(\bar{\Omega})}(\alpha_0,\alpha_0^*)$ are arbitrary a concise statement of detailed balance follows from (4.2.5), (4.2.6) and (4.2.9):

$$\overleftarrow{W}^{(\Omega)}_{ss}(\alpha,\alpha^*,\tau;\alpha_0,\alpha_0^*,0) = \overleftarrow{W}^{(\tilde{\bar{\Omega}})}_{ss}(\alpha_0^*,\alpha_0,\tau;\alpha^*,\alpha,0) \tag{4.2.10}$$

A similar formulation of detailed balance in phase-space form has recently been derived by Agarwal (1975), and, in a somewhat different context, detailed balance in essentially this form arises in the quantum statistical mechanics of nonequilibrium thermodynamics by Vlieger *et al.* (1961).

Let us now define operators $\tilde{\mathcal{L}}$ and $\bar{\mathcal{L}}$ in Hilbert space by

$$\begin{aligned} \widetilde{\mathcal{L}\hat{S}} &= \tilde{\mathcal{L}}\,\tilde{\hat{S}} \\ tr_s\,\hat{S}_1\mathcal{L}\hat{S}_2 &= tr_s\,\hat{S}_2\bar{\mathcal{L}}\hat{S}_1 \end{aligned} \tag{4.2.11}$$

Specifying the associated differential operators in phase-space by $\tilde{\mathcal{L}}^{(\Omega)}(\alpha,\alpha^*)$ and $\bar{\mathcal{L}}^{(\Omega)}(\alpha,\alpha^*)$ it is readily shown from (4.2.4), (4.2.8) and (4.2.11) that

$$\tilde{\mathcal{L}}^{(\Omega)}(\alpha,\alpha^*) = \mathcal{L}^{(\tilde{\Omega})}(\alpha^*,\alpha) \tag{4.2.12}$$

and

$$\bar{\mathcal{L}}^{(\Omega)}(\alpha,\alpha^*) = \mathcal{L}^{(\bar{\Omega})^{+}}(\alpha,\alpha^*) \tag{4.2.13}$$

where + denotes the adjoint. We may now derive necessary and sufficient conditions for (4.2.10). Expanding the quasiprobability densities according to (4.2.7) this reads

$$\begin{aligned} &P_{ss}^{(\Omega)}(\alpha_0,\alpha_0^*)\exp[\overleftrightarrow{\Lambda}^{(\Omega)}(\alpha_0,\alpha_0^*)]\exp[\mathcal{L}^{(\bar{\Omega})}(\alpha,\alpha^*)\tau]\,\delta^{(2)}(\alpha-\alpha_0) \\ &= P_{ss}^{(\tilde{\bar{\Omega}})}(\alpha^*,\alpha)\exp[\overleftrightarrow{\Lambda}^{(\tilde{\bar{\Omega}})}(\alpha^*,\alpha)]\exp[\mathcal{L}^{(\tilde{\Omega})}(\alpha_0^*,\alpha_0)\tau]\,\delta^{(2)}(\alpha-\alpha_0) \end{aligned} \tag{4.2.14}$$

and from (4.2.8), (4.2.12) and the properties of $\overleftrightarrow{\Lambda}^{(\Omega)}(\alpha,\alpha^*)$

become

$$P_{ss}^{(\Omega)}(\alpha_0,\alpha_0^*)\exp[\overleftrightarrow{\Lambda}^{(\Omega)}(\alpha_0,\alpha_0^*)]\exp[\mathcal{L}^{(\bar{\Omega})}(\alpha,\alpha^*)\tau]\delta^{(2)}(\alpha-\alpha_0)$$

$$= \exp[\tilde{\mathcal{L}}^{(\Omega)}(\alpha_0,\alpha_0^*)\tau]\delta^{(2)}(\alpha-\alpha_0)\exp[\overleftrightarrow{\Lambda}^{(\bar{\Omega})}(\alpha,\alpha^*)]\tilde{P}_{ss}^{(\bar{\Omega})}(\alpha,\alpha^*) \quad (4.2.15)$$

To convert this to an operator condition we then multiply both sides by the arbitrary function $S^{(\Omega)}(\alpha,\alpha^*)$ and integrate with respect to α. For the left hand side we may write

$$P_{ss}^{(\Omega)}(\alpha_0,\alpha_0^*)\exp[\overleftrightarrow{\Lambda}^{(\Omega)}(\alpha_0,\alpha_0^*)]\int d^2\alpha\, S^{(\Omega)}(\alpha,\alpha^*)$$
$$\exp[\mathcal{L}^{(\bar{\Omega})}(\alpha,\alpha^*)\tau]\delta^{(2)}(\alpha-\alpha_0)$$

$$= P_{ss}^{(\Omega)}(\alpha_0,\alpha_0^*)\exp[\overleftrightarrow{\Lambda}^{(\Omega)}(\alpha_0,\alpha_0^*)]\int d^2\alpha\,\delta^{(2)}(\alpha-\alpha_0)$$
$$\exp[\mathcal{L}^{(\bar{\Omega})+}(\alpha,\alpha^*)\tau]S^{(\Omega)}(\alpha,\alpha^*)$$

$$= P_{ss}^{(\Omega)}(\alpha_0,\alpha_0^*)\exp[\overleftrightarrow{\Lambda}^{(\Omega)}(\alpha_0,\alpha_0^*)]\exp[\mathcal{L}^{(\bar{\Omega})+}(\alpha_0,\alpha_0^*)\tau]S^{(\Omega)}(\alpha_0,\alpha_0^*)$$

$$= P_{ss}^{(\Omega)}(\alpha_0,\alpha_0^*)\exp[\overleftrightarrow{\Lambda}^{(\Omega)}(\alpha_0,\alpha_0^*)]\exp[\bar{\mathcal{L}}^{(\Omega)}(\alpha_0,\alpha_0^*)\tau]S^{(\Omega)}(\alpha_0,\alpha_0^*) \quad (4.2.16)$$

where we have used (4.2.13). Then for arbitrary operators $\hat{S}_1(a,a^\dagger)$ and $\hat{S}_2(a,a^\dagger)$ with Ω-equivalents $S_1^{(\Omega)}(\alpha,\alpha^*)$ and $S_2^{(\Omega)}(\alpha,\alpha^*)$ there is established in appendix B the general result

$$S_{12}^{(\Omega)}(\alpha,\alpha^*) = S_1^{(\Omega)}(\alpha,\alpha^*)\exp[\overleftrightarrow{\Lambda}^{(\Omega)}(\alpha,\alpha^*)]S_2^{(\Omega)}(\alpha,\alpha^*) \quad (4.2.17)$$

where $S_{12}^{(\Omega)}(\alpha,\alpha^*)$ is the Ω-equivalent of the product $\hat{S}_1(a,a^\dagger)\ \hat{S}_2(a,a^\dagger)$. Thus, from the right hand side, using (B.34) we may write

$$\exp[\tilde{\mathcal{L}}^{(\Omega)}(\alpha_0,\alpha_0^*)\tau]\int d^2\alpha\, S^{(\Omega)}(\alpha,\alpha^*)[\delta^{(2)}(\alpha-\alpha_0)\exp[\overleftrightarrow{\Lambda}^{(\bar{\Omega})}(\alpha,\alpha^*)]$$
$$\tilde{P}_{ss}^{(\bar{\Omega})}(\alpha,\alpha^*)]$$

$$= \exp[\tilde{\mathcal{L}}^{(\Omega)}(\alpha_0,\alpha_0^*)\tau]\,\pi\,\mathrm{tr}_s\, S(a,a^\dagger)\Delta^{(\bar{\Omega})}(\alpha_0-a,\alpha_0^*-a^\dagger)\,\tilde{\rho}_{ss}$$

$$= \exp[\tilde{\mathcal{L}}^{(\Omega)}(\alpha_0,\alpha_0^*)\tau]\,\pi\,\mathrm{tr}_S\,\tilde{\rho}_{ss}\,S(a,a^\dagger)\,\Delta^{(\bar{\Omega})}(\alpha_0-a,\alpha_0^*-a^\dagger)$$

$$= \exp[\tilde{\mathcal{L}}^{(\Omega)}(\alpha_0,\alpha_0^*)\tau]\int d^2\alpha\;\delta^{(2)}(\alpha-\alpha_0)\,\tilde{P}_{ss}^{(\Omega)}(\alpha,\alpha^*)$$
$$\cdot\exp[\overleftrightarrow{\Lambda}^{(\Omega)}(\alpha,\alpha^*)]\,S^{(\Omega)}(\alpha,\alpha^*)$$

$$= \exp[\tilde{\mathcal{L}}^{(\Omega)}(\alpha_0,\alpha_0^*)\tau]\,\tilde{P}_{ss}^{(\Omega)}(\alpha_0,\alpha_0^*)\,\exp[\overleftrightarrow{\Lambda}^{(\Omega)}(\alpha_0,\alpha_0^*)]\,S^{(\Omega)}(\alpha_0,\alpha_0^*) \tag{4.2.18}$$

where we have introduced $\Delta^{(\bar{\Omega})}(\alpha_0 - a,\ \alpha_0^* - a^\dagger)$ which, as in appendix B, is simply the $\bar{\Omega}$-equivalent for $\delta^{(2)}(\alpha - \alpha_0)$. From (4.2.16) and (4.2.18) it follows that (4.2.10) is equivalent to the requirement

$$P_{ss}^{(\Omega)}(\alpha,\alpha^*)\,\exp[\overleftrightarrow{\Lambda}^{(\Omega)}(\alpha,\alpha^*)]\,\bar{\mathcal{L}}^{(\Omega)}(\alpha,\alpha^*)^n$$
$$= \tilde{\mathcal{L}}^{(\Omega)}(\alpha,\alpha^*)^n\,\tilde{P}_{ss}^{(\Omega)}(\alpha,\alpha^*)\,\exp[\overleftrightarrow{\Lambda}^{(\Omega)}(\alpha,\alpha^*)] \tag{4.2.19}$$

where n may take values from 0 to ∞ and $\overleftrightarrow{\Lambda}^{(\Omega)}(\alpha,\alpha^*)$ acts on everything to the right. It is then readily shown by induction that necessary and sufficient conditions are given by

$$P_{ss}^{(\Omega)}(\alpha,\alpha^*) = \tilde{P}_{ss}^{(\Omega)}(\alpha,\alpha^*) \tag{4.2.20}$$

$$P_{ss}^{(\Omega)}(\alpha,\alpha^*)\,\exp[\overleftrightarrow{\Lambda}^{(\Omega)}(\alpha,\alpha^*)]\,\bar{\mathcal{L}}^{(\Omega)}(\alpha,\alpha^*)$$
$$= \tilde{\mathcal{L}}^{(\Omega)}(\alpha,\alpha^*)\,\tilde{P}_{ss}^{(\Omega)}(\alpha,\alpha^*)\,\exp[\overleftrightarrow{\Lambda}^{(\Omega)}(\alpha,\alpha^*)] \tag{4.2.21}$$

Agarwal has derived operator conditions ensuring the detailed balance of (4.2.5) (Agarwal, 1973c) and the conditions obtained here are just what we would expect from applying $\hat{\Omega}$ to these operator conditions of Agarwal.

4.3 The Classical Limit of Detailed Balance in the Phase-Space form

For a classical Fokker-Plank process with probability density $P(\{x\},t)$ obeying

$$\frac{dP(\{x\},t)}{dt} = \mathcal{L}(\{x\})\, P(\{x\},t) \tag{4.3.1}$$

the detailed balance requirement is as in (3.4.8) and reads

$$W_{ss}(\{x\},\tau;\{x_0\},0) = W_{ss}(\{\tilde{x}_0\},\tau;\{\tilde{x}\},0) \tag{4.3.2}$$

with the stationary joint probability density $W_{ss}(\{x\},\tau;\ \{x_0\},0)$ given by

$$\begin{aligned} &W_{ss}(\{x\},\tau;\{x_0\},0) \\ &\qquad = P_{ss}(\{x_0\})\exp[\mathcal{L}(\{x\})\tau]\,\delta(\{x\}-\{x_0\}) \end{aligned} \tag{4.3.3}$$

Literal interpretation of the quantum-mechanical Fokker-Plank equation (4.2.3) adopts this statement directly:

$$W_{ss}^{(\Omega)}(\alpha,\alpha^*,\tau;\alpha_0,\alpha_0^*,0) = W_{ss}^{(\Omega)}(\alpha_0^*,\alpha_0,\tau;\alpha^*,\alpha,0) \tag{4.3.4}$$

with

$$\begin{aligned} &W_{ss}^{(\Omega)}(\alpha,\alpha^*,\tau;\alpha_0,\alpha_0^*,0) \\ &\qquad = P_{ss}^{(\Omega)}(\alpha_0,\alpha_0^*)\exp[\mathcal{L}^{(\Omega)}(\alpha,\alpha^*)\tau]\,\delta^{(2)}(\alpha-\alpha_0) \end{aligned} \tag{4.3.5}$$

Contrasted with (4.2.10) this is generally inadequate for a fully quantum-mechanical system.

Pursuing this comparison we invoke the results (4.2.12) and (4.2.13), and express (4.2.20) and (4.2.21) in the form

$$P_{ss}^{(\Omega)}(\alpha,\alpha^*) = P_{ss}^{(\tilde{\Omega})}(\alpha^*,\alpha) \tag{4.3.6}$$

$$P_{ss}^{(\Omega)}(\alpha,\alpha^*)\exp[\overleftrightarrow{\Lambda}^{(\Omega)}(\alpha,\alpha^*)]\mathcal{L}^{(\bar{\Omega})^+}(\alpha,\alpha^*)$$
$$=\mathcal{L}^{(\tilde{\Omega})}(\alpha^*,\alpha)\,P_{ss}^{(\Omega)}(\alpha,\alpha^*)\exp[\overleftrightarrow{\Lambda}^{(\Omega)}(\alpha,\alpha^*)] \quad (4.3.7)$$

This form is suitable for comparison with the classical detailed balance conditions of Graham and Haken (1971) and Risken (1972). In the language of (4.3.4) these read

$$P_{ss}^{(\Omega)}(\alpha,\alpha^*) = P_{ss}^{(\Omega)}(\alpha^*,\alpha) \quad (4.3.8)$$

$$P_{ss}^{(\Omega)}(\alpha,\alpha^*)\,\mathcal{L}^{(\Omega)^+}(\alpha,\alpha^*) = \mathcal{L}^{(\Omega)}(\alpha^*,\alpha)\,P_{ss}^{(\Omega)}(\alpha,\alpha^*) \quad (4.3.9)$$

Clearly two considerations maintain a distinction between these alternative sets of conditions: the deviation of the operator $\exp[\overleftrightarrow{\Lambda}^{(\Omega)}(\alpha,\alpha^*)]$ from unity and the nonequivalence of the Ω, $\bar{\Omega}$ and $\tilde{\Omega}$ mappings. Both distinctions vanish however in the classical limit $\hbar\to 0$. We may demonstrate this by introducing the coordinate and momentum operators $\hat{q}$ and $\hat{p}$, where

$$\begin{aligned} a &= (2\hbar\omega)^{-1/2}(\omega\hat{q}+i\hat{p}) \\ a^\dagger &= (2\hbar\omega)^{-1/2}(\omega\hat{q}-i\hat{p}) \end{aligned} \quad (4.3.10)$$

With real variables q, p and u, v defined by

$$\begin{aligned} q &= (\hbar/2\omega)^{1/2}(\alpha+\alpha^*) \\ p &= -i(\hbar\omega/2)^{1/2}(\alpha-\alpha^*) \end{aligned} \quad (4.3.11)$$

and

$$\begin{aligned} u &= (2\hbar\omega)^{-1/2}(\beta+\beta^*) \\ v &= i\omega(2\hbar\omega)^{-1/2}(\beta-\beta^*) \end{aligned} \quad (4.3.12)$$

the fundamental equations (2.2.2), (2.2.3) and (2.2.5), (2.2.6) may be written in the form

$$
\begin{aligned}
\hat{\mathcal{S}}(\hat{q},\hat{p}) &= \int du\,dv\,\mathcal{G}(u,v)\exp[-i(v\hat{q}+u\hat{p})] \\
\mathcal{G}(u,v) &= (\hbar/2\pi)\,\mathrm{tr}_S\,\hat{\mathcal{S}}(\hat{q},\hat{p})\exp[i(v\hat{q}+u\hat{p})]
\end{aligned}
\tag{4.3.13}
$$

and

$$
\begin{aligned}
\mathcal{S}^{(\Omega)}(q,p) &= \int du\,dv\,\mathcal{F}^{(\Omega)}(u,v)\exp[-i(vq+up)] \\
\mathcal{F}^{(\Omega)}(u,v) &= (1/2\pi)^2\int dq\,dp\,\mathcal{S}^{(\Omega)}(q,p)\exp[i(vq+up)]
\end{aligned}
\tag{4.3.14}
$$

where

$$\mathcal{G}(u,v) = \Pi(u,v)\,\mathcal{F}^{(\Omega)}(u,v) \tag{4.3.15}$$

Here

$$
\begin{aligned}
\mathcal{S}^{(\Omega)}(q,p) &= S^{(\Omega)}(\alpha,\alpha^*) \\
\mathcal{F}^{(\Omega)}(u,v) &= F^{(\Omega)}(\beta,\beta^*) \\
\hat{\mathcal{S}}(\hat{q},\hat{p}) &= \hat{S}(a,a^\dagger) \\
\mathcal{G}(u,v) &= G(\beta,\beta^*)
\end{aligned}
\tag{4.3.16}
$$

Within this presentation, for (2.2.7) and (2.2.24), we have

$$\Omega(\beta,\beta^*) = \Pi(u,v) = \exp(\mu' u^2 + \nu' v^2 - i\lambda' uv) \tag{4.3.17}$$

and

$$
\begin{aligned}
\overleftrightarrow{\Lambda}^{(\Omega)}(\alpha,\alpha^*) &= \overleftrightarrow{\Xi}^{(\Omega)}(q,p) \\
&= -2\nu'\frac{\overleftarrow{\partial}}{\partial q}\frac{\overrightarrow{\partial}}{\partial q} - 2\mu'\frac{\overleftarrow{\partial}}{\partial p}\frac{\overrightarrow{\partial}}{\partial p} - i(\lambda'+\hbar/2)\frac{\overleftarrow{\partial}}{\partial p}\frac{\overrightarrow{\partial}}{\partial q} \\
&\quad - i(\lambda'-\hbar/2)\frac{\overleftarrow{\partial}}{\partial q}\frac{\overrightarrow{\partial}}{\partial p}
\end{aligned}
\tag{4.3.18}
$$

with

$$\mu' = (\hbar\omega/2)\,[\lambda + (\mu + \nu)]$$
$$\nu' = (\hbar/2\omega)\,[\lambda - (\mu + \nu)] \qquad (4.3.19)$$
$$\lambda' = \hbar(\mu - \nu)$$

Clearly, for the classical limit

$$\lim_{\hbar \to 0} \Omega(\beta, \beta^*) = 1$$
$$\lim_{\hbar \to 0} \exp[\overleftrightarrow{\Lambda}^{(\Omega)}(\alpha, \alpha^*)] = 1 \qquad (4.3.20)$$

Under such conditions (4.3.6) and (4.3.7) reduce to (4.3.8) and (4.3.9). Thus, the detailed balance (4.3.4) obtained from a literal interpretation of the quantum-mechanical Fokker-Plank process is formally the classical limit of the full quantum-mechanical detailed balance.

4.4 Detailed Balance in the Energy Representation

We turn now from the phase-space calculus of the previous two sections to the expression of the detailed balance requirement of (4.1.15) within the energy representation. The energy representation is specified by the complete set of states $|\varepsilon,\{\lambda\}\rangle$, where

$$H_s|\varepsilon,\{\lambda\}\rangle = \varepsilon|\varepsilon,\{\lambda\}\rangle \qquad (4.4.1)$$

and $\{\lambda\}$ is a set of quantum numbers accounting for an energy degeneracy. For notational simplicity we will distinguish these states by a single index writing $|n\rangle$ for $|\varepsilon\{\lambda\}\rangle$. Taking matrix elements the operator master equation (4.2.2) then reads

$$\frac{d\rho_{n,n'}}{dt} = \sum_{m,m'} \gamma^{n,n'}_{m,m'}\, \rho_{m,m'} \qquad (4.4.2)$$

where

$$\gamma^{n,n'}_{m,m'} = \langle n|(\mathcal{L}|m\rangle\langle m'|)|n'\rangle \tag{4.4.3}$$

Now, when taken together with the result (2.1.6) for evaluation of two-time averages, the full quantum detailed balance reads

$$tr_s[\exp(\mathcal{L}\tau)S^{(\beta)}\rho^{ss}]S^{(\alpha)} = tr_s[\exp(\mathcal{L}\tau)\tilde{S}^{(\alpha)\dagger}\rho^{ss}]\tilde{S}^{(\beta)\dagger} \tag{4.4.4}$$

which, with the help of the antiunitary property of the time reversal operator (Messiah, 1970) may be converted to the form

$$tr_s[\exp(\mathcal{L}\tau)S^{(\beta)}\rho^{ss}]S^{(\alpha)} = tr_s[\exp(\tilde{\mathcal{L}}\tau)S^{(\alpha)}]S^{(\beta)} \tag{4.4.5}$$

where $\tilde{\mathcal{L}}$ is as defined in (4.2.11). Thus, writing for the operators $S^{(\alpha)}$ and $S^{(\beta)}$ the matrix element forms

$$\begin{aligned} S^{(\alpha)} &= \sum_{n,n'} S^{(\alpha)}_{n,n'}|n\rangle\langle n'| \\ S^{(\beta)} &= \sum_{m,m'} S^{(\beta)}_{m,m'}|m\rangle\langle m'| \end{aligned} \tag{4.4.6}$$

we may write for full detailed balance within the energy representation

$$\begin{aligned} &\sum_{n,n'}\sum_{m,m'} S^{(\alpha)}_{n,n'}S^{(\beta)}_{m,m'}\langle n'|[\exp(\mathcal{L}\tau)|m\rangle\langle m'|\rho^{ss}]|n\rangle \\ &= \sum_{n,n'}\sum_{m,m'} S^{(\alpha)}_{n,n'}S^{(\beta)}_{m,m'}\langle m'|[\exp(\tilde{\mathcal{L}}\tau)\tilde{\rho}^{ss}|n\rangle\langle n'|]|m\rangle \end{aligned} \tag{4.4.7}$$

Then, since $S^{(\alpha)}$ and $S^{(\beta)}$ are arbitrary we may move to the necessary and sufficient conditions

$$\begin{aligned} &\sum_k \langle n'|[\exp(\mathcal{L}\tau)|m\rangle\langle k|]|n\rangle \rho^{ss}_{m',k} \\ &= \sum_k \langle m'|[\exp(\tilde{\mathcal{L}}\tau)|k\rangle\langle n'|]|m\rangle \tilde{\rho}^{ss}_{k,n} \end{aligned} \tag{4.4.8}$$

where we have introduced ρ^{ss} in matrix element form

$$\rho^{ss} = \sum_{k,k'} |k\rangle\langle k'|\rho^{ss}_{k,k'} \tag{4.4.9}$$

Using once again the antiunitary property of the time reversal operator, this may further be written as

$$\sum_k \langle n'|[\exp(\mathcal{L}\tau)|m\rangle\langle k|]|n\rangle \rho^{ss}_{m',k}$$
$$= \sum_k \langle \tilde{m}|[\exp(\mathcal{L}\tau)|\tilde{n}'\rangle\langle\tilde{k}|]|\tilde{m}'\rangle \rho^{ss}_{\tilde{n},\tilde{k}} \qquad (4.4.10)$$

Now we require this result to hold for all values of τ and hence a set of conditions equivalent to (4.4.10) is given by

$$\sum_k \langle n'|(\mathcal{L}^r|m\rangle\langle k|)|n\rangle \rho^{ss}_{m',k}$$
$$= \sum_k \langle \tilde{m}|(\mathcal{L}^r|\tilde{n}'\rangle\langle\tilde{k}|)|\tilde{m}'\rangle \rho^{ss}_{\tilde{n},\tilde{k}} \qquad (4.4.11)$$

which must hold for all integral powers $r \geq 0$. Clearly from this necessary conditions for fulfilment of full quantum detailed balance are

$$\rho^{ss}_{n,m} = \rho^{ss}_{\tilde{m},\tilde{n}} \qquad (4.4.12)$$

$$\sum_k \gamma^{n',n}_{m,k}\, \rho^{ss}_{m',k} = \sum_k \gamma^{\tilde{m},\tilde{m}'}_{\tilde{n}',\tilde{k}}\, \rho^{ss}_{\tilde{n},\tilde{k}} \qquad (4.4.13)$$

We may show further that these are also sufficient for the fulfilment of (4.4.11) to all higher orders. Indeed this follows by induction; if we adopt (4.4.11) by hypothesis and consider then the same expression for the order $r + 1$, we have

$$\sum_k \langle n'|(\mathcal{L}^{r+1}|m\rangle\langle k|)|n\rangle \rho^{ss}_{m',k}$$
$$= \sum_k \langle n'|\mathcal{L}(\mathcal{L}^r|m\rangle\langle k|)|n\rangle \rho^{ss}_{m',k} \qquad (4.4.14)$$

which, if we expand the operator $(\mathcal{L}^r|m\rangle\langle k|)$ in matrix element form

$$(\mathcal{L}^r|m\rangle\langle k|) = \sum_{pq} \langle p|(\mathcal{L}^r|m\rangle\langle k|)|q\rangle\ |p\rangle\langle q| \qquad (4.4.15)$$

becomes

$$\sum_k \langle n'|(\mathcal{L}^{r+1}|m\rangle\langle k|)|n\rangle \rho^{ss}_{m',k}$$
$$= \sum_{pq}\sum_k \gamma^{p,q}_{n',n}\langle p|(\mathcal{L}^{r}|m\rangle\langle k|)|q\rangle \rho^{ss}_{m',k} \qquad (4.4.16)$$

Then, with (4.4.11), this may be written

$$\sum_k \langle n'|(\mathcal{L}^{r+1}|m\rangle\langle k|)|n\rangle \rho^{ss}_{m',k}$$
$$= \sum_{pq}\sum_k \gamma^{p,q}_{n',n}\langle \tilde{m}|(\mathcal{L}^{r}|\tilde{p}\rangle\langle \tilde{k}|)|\tilde{m}'\rangle \rho^{ss}_{\tilde{q},\tilde{k}} \qquad (4.4.17)$$

and further, with (4.4.12) and (4.4.13)

$$\sum_k \langle n'|(\mathcal{L}^{r+1}|m\rangle\langle k|)|n\rangle \rho^{ss}_{m',k}$$
$$= \sum_{pq}\sum_k \gamma^{p,q}_{n',n}\,\rho^{ss}_{k,q}\langle \tilde{m}|(\mathcal{L}^{r}|\tilde{p}\rangle\langle \tilde{k}|)|\tilde{m}'\rangle$$
$$= \sum_{pq}\sum_k \gamma^{\tilde{n}',\tilde{q}}_{\tilde{p},\tilde{k}}\,\rho^{ss}_{\tilde{n},\tilde{q}}\langle \tilde{m}|(\mathcal{L}^{r}|\tilde{p}\rangle\langle \tilde{k}|)|\tilde{m}'\rangle$$
$$= \sum_q \langle \tilde{m}|\mathcal{L}^{r}\Big[\sum_{p,k}\langle \tilde{p}|(\mathcal{L}|\tilde{n}'\rangle\langle \tilde{q}|)|\tilde{k}\rangle\cdot|\tilde{p}\rangle\langle \tilde{k}|\Big]|\tilde{m}'\rangle\, \rho^{ss}_{\tilde{n},\tilde{q}} \qquad (4.4.18)$$

Hence, recognising in this the matrix element expansion of $\mathcal{L}\,|\tilde{n}'\rangle\langle\tilde{q}|$, we have

$$\sum_k \langle n'|(\mathcal{L}^{r+1}|m\rangle\langle k|)|n\rangle \rho^{ss}_{m',k}$$
$$= \sum_k \langle \tilde{m}|(\mathcal{L}^{r+1}|\tilde{n}'\rangle\langle \tilde{k}|)|\tilde{m}'\rangle\, \rho^{ss}_{\tilde{n},\tilde{k}} \qquad (4.4.19)$$

This is just the result (4.4.11) for power r + 1, and thus it is established by induction that (4.4.12) and (4.4.13) are both necessary and sufficient conditions for full quantum detailed balance. We therefore take (4.4.12) and (4.4.13) as the statement of full quantum detailed balance in the energy representation.

4.5 Diagonal Master Equations and Detailed Balance

Here we define the "diagonal master equation" as corresponding to the familiar Pauli situation. We then consider the quantum detailed balance in relation to these special master equations.

From the previous section, the master equation now reads

$$\frac{d\rho_{n,n'}}{dt} = \sum_{m,m'} \gamma^{n,n'}_{m,m'} \rho_{m,m'} \tag{4.5.1}$$

where

$$\gamma^{n,n'}_{m,m'} = \langle n| (\mathcal{L}\, |m\rangle\langle m'|)|n'\rangle \tag{4.5.2}$$

Here we focus our attention on the particular class of master equations whose Liouvillians admit the conditions

$$\langle n| (\mathcal{L}\, |m\rangle\langle m'|)|n\rangle \propto \delta_{m,m'} \tag{4.5.3}$$

for all n

$$\langle n| (\mathcal{L}\, |m\rangle\langle m|)|n'\rangle \propto \delta_{n,n'} \tag{4.5.4}$$

for all m.

For these we adopt the term "diagonal master equations" since (4.4.2) may then be replaced by the pair of equations:

$$\frac{d\rho_{n,n}}{dt} = \sum_{m} \gamma^{n,n}_{m,m} \rho_{m,m} \tag{4.5.5}$$

and, for $n \neq n'$

$$\frac{d\rho_{n,n'}}{dt} = \sum_{\substack{m,m' \\ m \neq m'}} \gamma^{n,n'}_{m,m'} \rho_{m,m'} \tag{4.5.6}$$

In the first instance we have diagonal elements coupled only to diagonal elements, and in the second off diagonals coupled only to off diagonals. It is easily shown that the first of these may be written in the Pauli form. From the requirement that the master equation preserve unit trace it follows that

$$\sum_{m} \gamma^{m,m}_{n,n} = 0 \tag{4.5.7}$$

and hence, isolating $\gamma^{n,n}_{n,n}\,\rho_{n,n}$ in (4.5.5) and writing

$$\frac{d\rho_{n,n}}{dt} = \sum_{m\neq n} \gamma^{n,n}_{m,m}\,\rho_{m,m} + \gamma^{n,n}_{n,n}\,\rho_{n,n} \tag{4.5.8}$$

we find

$$\frac{d\rho_{n,n}}{dt} = \sum_{m\neq n} \gamma^{n,n}_{m,m}\,\rho_{m,m} - \gamma^{m,m}_{n,n}\,\rho_{n,n} \tag{4.5.9}$$

Thus, (4.5.5) may be written

$$\frac{d\rho_{n,n}}{dt} = \sum_{m\neq n} \gamma_{m,n}\,\rho_{m,m} - \gamma_{n,m}\,\rho_{n,n} \tag{4.5.10}$$

with

$$\gamma_{n,m} = \lim_{\tau\to 0} \frac{1}{\tau}\langle m|[\exp(\mathcal{L}\tau)|n\rangle\langle n|]|m\rangle = \gamma^{m,m}_{n,n} \tag{4.5.11}$$

being the probability per unit time of transition from state $|n\rangle$ to $|m\rangle$. Taken in conjunction (4.5.5) and (4.5.6) therefore constitute the familiar Pauli situation, as with an initial random phase assumption (4.5.6) ensures a diagonal density operator at all times. Identification with the discrete classical Markoff process, as in section 3.4 is appropriate.

Let us consider also correlations at two times. Using the formal expression of (2.1.6) we may write

$$\langle S^{(\alpha)}(\tau)\, S^{(\beta)}(0)\rangle_{\rho_{ss}} = tr_s[\exp(\mathcal{L}\tau)\, S^{(\beta)}(0)\,\rho_{ss}]\, S^{(\alpha)}(0)$$

$$= \sum_{n,n'} \sum_{m,m'} S^{(\alpha)}_{n,n'}\, S^{(\beta)}_{m,m'}\,\langle n'|[\exp(\mathcal{L}\tau)|m\rangle\langle m'|]|n\rangle\, \rho^{ss}_{m,m'} \tag{4.5.12}$$

While possession of the full master equation furnishes knowledge of all $\gamma^{n,n'}_{m,m'}$ and hence allows evaluation of this average in the general case, if allusion to the discrete classical Markoff process is to be maintained restriction to operators diagonal in the energy representation is required. We therefore write

$$\langle S^{(\alpha)}(\tau)\, S^{(\beta)}(0)\rangle_{\rho_{ss}} = \sum_{n,m} S^{(\alpha)}_{n,n}\, S^{(\beta)}_{m,m}\, W_{ss}(|n\rangle,\tau\,;|m\rangle,0) \qquad (4.5.13)$$

where $W_{ss}(|n\rangle,\tau;\ |m\rangle,0)$ plays the role of a joint probability function

$$\begin{aligned} W_{ss}(|n\rangle,\tau\,;|m\rangle,0) &= \langle n|\left[\exp(\mathcal{L}\tau)|m\rangle\langle m|\right]|n\rangle\, \rho^{ss}_{m,m} \\ &= (\exp \gamma\tau)_{m,n}\, \rho^{ss}_{m,m} \qquad (4.5.14) \end{aligned}$$

(γ) is a matrix with elements $\gamma_{n,m}$. A reduced form of the quantum detailed balance omitting correlation of nondiagonal operators is therefore clearly implied and follows readily from (4.1.15). Using the identity

$$\langle \tilde{n}|\ \tilde{O}^{\dagger}|\tilde{n}\rangle = \langle n|O|n\rangle \qquad (4.5.15)$$

we find

$$W_{ss}(|n\rangle,\tau\,;|m\rangle,0) = W_{ss}(|\tilde{m}\rangle,\tau\,;|\tilde{n}\rangle,0) \qquad (4.5.16)$$

For states invariant under time reversal this leads to the familiar Pauli detailed balance of (3.1.5)

$$\gamma_{n,m}\, \rho^{ss}_{n,n} = \gamma_{m,n}\, \rho^{ss}_{m,m} \qquad (4.5.17)$$

The status of the Pauli detailed balance with respect to the full quantum detailed balance of (4.4.12) and (4.4.13) is now apparent. It is restricted to the case of diagonal master equations and appears then only as a reduced form of full quantum detailed balance.

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$$\langle\tilde{n}|\,\tilde{O}^{\dagger}|\tilde{n}\rangle = \langle n|O|n\rangle \tag{4.5.15}$$

we find

$$W_{ss}(|n\rangle,\tau\,;|m\rangle,0) = W_{ss}(|\tilde{m}\rangle,\tau\,;|\tilde{n}\rangle,0) \tag{4.5.16}$$

For states invariant under time reversal this leads to the familiar Pauli detailed balance of (3.1.5)

$$\gamma_{n,m}\, \rho^{ss}_{n,n} = \gamma_{m,n}\, \rho^{ss}_{m,m} \tag{4.5.17}$$

The status of the Pauli detailed balance with respect to the full quantum detailed balance of (4.4.12) and (4.4.13) is now apparent. It is restricted to the case of diagonal master equations and appears then only as a reduced form of full quantum detailed balance.

4.6 Examples

We conclude this chapter with a brief discussion of two examples. In the first we illustrate explicitly the distinction between full phase-space conditions and their classical limit with reference to the damped harmonic oscillator. The second then serves to demonstrate the possible failure of quantum detailed balance in a nonequilibrium situation while both classical and Pauli conditions hold. Here our discussion is in relation to the single mode laser.

a) Phase-Space Conditions for the Damped Oscillator

The phase-space description for an harmonic oscillator coupled to a thermal reservoir is well known (Agarwal, 1969; Louisell, 1969). The appropriate quantum-mechanical Fokker-Plank equation takes the form of (4.2.3) with, for normal ($\mu = \nu = 0$, $\lambda = \frac{1}{2}$) and antinormal ($\mu = \nu = 0$, $\lambda = -\frac{1}{2}$) mappings respectively,

$$\mathcal{L}^{(N)}(\alpha,\alpha^*) = (\gamma/2 + i\omega_0)\frac{\partial}{\partial\alpha}\alpha + (\gamma/2 - i\omega_0)\frac{\partial}{\partial\alpha^*}\alpha^* + \gamma(\bar{n}+1)\frac{\partial^2}{\partial\alpha\partial\alpha^*} \tag{4.6.1}$$

$$\mathcal{L}^{(A)}(\alpha,\alpha^*) = (\gamma/2 + i\omega_0)\frac{\partial}{\partial\alpha}\alpha + (\gamma/2 - i\omega_0)\frac{\partial}{\partial\alpha^*}\alpha^* + \gamma\bar{n}\frac{\partial^2}{\partial\alpha\partial\alpha^*} \tag{4.6.2}$$

Here ω_0 is the oscillator frequency, $1/\gamma$ the lifetime, and $\bar{n}$ the reservoir occupation number. From this Fokker-Plank equation, in the steady state we find

$$\begin{aligned} P_{ss}^{(N)}(\alpha,\alpha^*) &= (\bar{n}+1)^{-1}\exp(-\alpha\alpha^*/\bar{n}+1) \\ P_{ss}^{(A)}(\alpha,\alpha^*) &= \bar{n}^{-1}\exp(-\alpha\alpha^*/\bar{n}) \end{aligned} \tag{4.6.3}$$

Now for antinormal mapping we have the result (2.3.3) and thus the phase-space detailed balance conditions (4.3.6) and (4.3.7) read

$$P_{ss}^{(A)}(\alpha,\alpha^*) = P_{ss}^{(A)}(\alpha^*,\alpha) \qquad (4.6.4)$$

$$P_{ss}^{(A)}(\alpha,\alpha^*)\exp\left(-\frac{\overleftarrow{\partial}}{\partial\alpha^*}\frac{\overrightarrow{\partial}}{\partial\alpha}\right)\mathcal{L}^{(N)^+}(\alpha,\alpha^*)$$
$$= \mathcal{L}^{(A)}(\alpha^*,\alpha)\,P_{ss}^{(A)}(\alpha,\alpha^*)\exp\left(-\frac{\overleftarrow{\partial}}{\partial\alpha^*}\frac{\overrightarrow{\partial}}{\partial\alpha}\right) \qquad (4.6.5)$$

From (4.6.3) the first of these is trivially satisfied, and in establishing the fulfilment of the second condition it is only necessary to insert $\mathcal{L}^{(A)}(\alpha^*,\alpha)$ from (4.6.2) and $\mathcal{L}^{(N)^+}(\alpha,\alpha^*)$. The usual definition for adjoint has, for arbitrary functions $F_1\ (\alpha,\alpha^*)$ and $F_2\ (\alpha,\alpha^*)$

$$\int d^2\alpha\, F_1(\alpha,\alpha^*)\mathcal{L}^{(N)}(\alpha,\alpha^*)F_2(\alpha,\alpha^*) = \int d^2\alpha\, F_2(\alpha,\alpha^*)\mathcal{L}^{(N)^+}(\alpha,\alpha^*)\,F_1(\alpha,\alpha^*) \qquad (4.6.6)$$

and hence, from (4.6.1) we find

$$\mathcal{L}^{(N)^+}(\alpha,\alpha^*) = -(\gamma/2+i\omega_0)\,\alpha\frac{\partial}{\partial\alpha} - (\gamma/2-i\omega_0)\,\alpha^*\frac{\partial}{\partial\alpha^*} + \gamma(\bar{n}+1)\frac{\partial^2}{\partial\alpha\partial\alpha^*} \qquad (4.6.7)$$

Then, relegating the tedious algebra to appendix D, it is found that both sides of (4.6.5) admit the operator form

$$P_{ss}^{(A)}(\alpha,\alpha^*)\sum_{n=0}^{\infty}(1/n!)(\alpha/\bar{n})^n\Big[-(\gamma/2+i\omega_0)(\bar{n}+1)/\bar{n}\cdot\alpha\frac{\partial}{\partial\alpha} - (\gamma/2-i\omega_0)\alpha^*\frac{\partial}{\partial\alpha^*} + \gamma(\bar{n}+1)\frac{\partial^2}{\partial\alpha\partial\alpha^*}\Big]\frac{\partial^n}{\partial\alpha^n} \qquad (4.6.8)$$

Full quantum detailed balance is satisfied.

We now introduce the classical limit of section (4.3). Since $\bar{n}$ is given by $[\exp(\omega_0/kT) - 1]^{-1}$ this here corresponds to the limit $(1/\bar{n})\to 0$. Hence, introducing this to (4.6.8) we find in the limit a behaviour as for the operator.

$$P_{ss}^{(A)}(\alpha,\alpha^*)\Big[-(\gamma/2+i\omega_0)\,\alpha\frac{\partial}{\partial\alpha} - (\gamma/2-i\omega_0)\,\alpha^*\frac{\partial}{\partial\alpha^*} + \gamma\bar{n}\frac{\partial^2}{\partial\alpha\partial\alpha^*}\Big] \qquad (4.6.9)$$

Here we have just the operator $P_{ss}^{(A)}(\alpha,\alpha^*)\mathcal{L}^{(A)+}(\alpha,\alpha^*)$ arising in the classical conditions (4.3.8), (4.3.9) demonstrating clearly the classical conditions as a limiting form of the full quantum detailed balance conditions.

The onset of the classical limit for this oscillator example may be further illustrated by a resource to the form for two-time averages with antinormal and normal mapping given in (2.3.25). For the steady state this reads

$$\langle \hat{S}_1(\tau)\hat{S}_2(0)\rangle_{\rho_{ss}} = \int d^2\alpha \int d^2\beta \left[\hat{S}_2^{(A)}\left(\alpha,\alpha^* - \frac{\partial}{\partial\alpha}\right) P_{ss}^{(A)}(\alpha,\alpha^*)\right] \mathcal{P}^{(A)}(\beta,\beta^*,\tau\,|\,\alpha,\alpha^*,0)\, S_1^{(N)}(\beta,\beta^*) \tag{4.6.10}$$

Where $P_{ss}^{(A)}(\alpha,\alpha^*)$ is given by (4.6.3) and we have

$$\mathcal{P}^{(A)}(\beta,\beta^*,\tau\,|\,\alpha,\alpha^*,0) = \left[\bar{n}(1-e^{-\gamma\tau})\right]^{-1} \exp\left[-\frac{|\alpha-\beta e^{-(\gamma/2+i\omega_0)\tau}|^2}{\bar{n}(1-e^{-\gamma\tau})}\right] \tag{4.6.11}$$

Then, with (4.6.3), we find

$$\hat{S}_2^{(A)}\left(\alpha,\alpha^* - \frac{\partial}{\partial\alpha}\right) P_{ss}^{(A)}(\alpha,\alpha^*) = \hat{S}_2^{(A)}\left(\alpha, \frac{\bar{n}+1}{\bar{n}}\alpha^*\right) P_{ss}^{(A)}(\alpha,\alpha^*) \tag{4.6.12}$$

Clearly, as $1/\bar{n} \to 0$

$$\lim_{1/\bar{n}\to 0} \hat{S}_2^{(A)}\left(\alpha,\alpha^* - \frac{\partial}{\partial\alpha}\right) P_{ss}^{(A)}(\alpha,\alpha^*) = \hat{S}_2^{(A)}(\alpha,\alpha^*)\, P_{ss}^{(A)}(\alpha,\alpha^*) \tag{4.6.13}$$

and, with the joint distribution of (4.3.5), (4.6.10) reduces to the classical phase-space integral for two-time averages

$$\langle \hat{S}_1(\tau)\hat{S}_2(0)\rangle_{\rho_{ss}} = \int d^2\alpha \int d^2\beta\; S_1^{(N)}(\beta,\beta^*)\, S_2^{(A)}(\alpha,\alpha^*)\, W_{ss}^{(A)}(\beta,\beta^*,\tau\,;\,\alpha,\alpha^*,0) \tag{4.6.14}$$

A like treatment arising in (2.3.28) yields in addition

$$\langle \tilde{\hat{S}}_1^{\dagger}(\tau)\,\tilde{\hat{S}}_2^{\dagger}(0)\rangle_{\rho_{ss}} = \int d^2\alpha \int d^2\beta\; S_1^{(N)}(\beta,\beta^*)\, S_2^{(A)}(\alpha,\alpha^*)\; P_{ss}^{(N)}(\beta^*,\beta)\,\mathcal{P}^{(N)}(\alpha^*,\alpha,\tau\,|\,\beta^*,\beta,0) \tag{4.6.15}$$

and

$$\mathcal{P}^{(N)}(\beta,\beta^*,\tau\,|\,\alpha,\alpha^*,0) = \left[(\bar{n}+1)(1-e^{-\gamma\tau})\right]^{-1} \exp\left[-\frac{|\alpha-\beta e^{-(\gamma/2+i\omega_0)\tau}|^2}{(\bar{n}+1)(1-e^{-\gamma\tau})}\right] \tag{4.6.16}$$

With (4.6.3), (4.6.11) and (4.6.16), in the limit considered (4.6.15) may also be written in terms of the antinormal joint distribution:

$$\langle \tilde{\hat{S}}_1^{\dagger}(\tau)\,\tilde{\hat{S}}_2^{\dagger}(0)\rangle_{\rho_{ss}} = \int d^2\alpha \int d^2\beta\; S_1^{(N)}(\beta,\beta^*)\, S_2^{(A)}(\alpha,\alpha^*)\; W_{ss}^{(A)}(\alpha^*,\alpha,\tau;\beta^*,\beta,0) \tag{4.6.17}$$

In (4.6.14) and (4.6.17) we then find the full quantum detailed balance (4.2.5) reduced to the classical form

$$W_{ss}^{(A)}(\beta,\beta^*,\tau;\alpha,\alpha^*,0) = W_{ss}^{(A)}(\alpha^*,\alpha,\tau;\beta^*,\beta,0) \tag{4.6.18}$$

b) The Single Mode Laser

While we have shown in section 4.1 that the quantum detailed balance is expected to hold in equilibrium as a consequence of microreversibility, this does not follow for the nonequilibrium situation. The fulfilment of detailed balance in both its classical limit and reduced Pauli forms may, however, be ensured for certain nonequilibrium systems by symmetry considerations (Graham, 1973). It is therefore to be expected that nonequilibrium systems exist for which classical considerations and Pauli conditions hold while full quantum detailed balance fails. We will demonstrate that such is the case for the laser.

In the phase-space formalism, with atomic variables removed, the single mode homogeneously broadened laser satisfies to a very good approximation (Risken, 1970) the rotating wave van der Pol oscillator equation (Lax and Louisell, 1969). We then write for antinormal ordering and scaled α and time variables

$$\mathcal{L}^{(A)}(\alpha,\alpha^*) = -\frac{\partial}{\partial\alpha}(g-i\omega_0-|\alpha|^2)\alpha - \frac{\partial}{\partial\alpha^*}(g+i\omega_0-|\alpha|^2)\alpha^* + 4\frac{\partial^2}{\partial\alpha\partial\alpha^*} \tag{4.6.19}$$

where g is a numerical pumping parameter. In the steady state

$$P_{ss}^{(A)}(\alpha,\alpha^*) = (N/2\pi)\exp[-\tfrac{1}{4}(|\alpha|^2-g)^2] \tag{4.6.20}$$

where N is required for normalisation. Then in considering the classical detailed balance conditions (4.3.8) and (4.3.9), the first of these is trivially satisfied while, with (4.6.6) and (4.6.19), in the second we have the operators

$$\mathcal{L}^{(A)+}(\alpha,\alpha^*) = (g-i\omega_0-|\alpha|^2)\alpha\frac{\partial}{\partial\alpha} + (g+i\omega_0-|\alpha|^2)\alpha^*\frac{\partial}{\partial\alpha^*} + 4\frac{\partial^2}{\partial\alpha\partial\alpha^*} \tag{4.6.21}$$

and

$$\mathcal{L}^{(A)}(\alpha^*,\alpha) = -\frac{\partial}{\partial\alpha^*}(g-i\omega_0-|\alpha|^2)\alpha^* - \frac{\partial}{\partial\alpha}(g+i\omega_0-|\alpha|^2)\alpha + 4\frac{\partial^2}{\partial\alpha\partial\alpha^*} \tag{4.6.22}$$

Following the simple algebraic manipulations presented in appendix D, from these and (4.6.20), the second requirement of the classical detailed balance conditions is also satisfied.

In the energy representation the corresponding equation to (4.6.19) is the Scully-Lamb master equation (Scully and Lamb, 1967), which reads

$$\frac{d\rho_{n,n'}}{dt} = -[(n+1)R_{n,n'} + (n'+1)R^*_{n',n}]\rho_{n,n'} + [R_{n-1,n'-1} + R^*_{n'-1,n-1}](n,n')^{1/2}\rho_{n-1,n'-1} - (1/2)C(n+n')\rho_{n,n'} + C[(n+1)(n'+1)]^{1/2}\rho_{n+1,n'+1} \quad (4.6.23)$$

where $R_{n,n'}$ is defined in terms of laser parameters. For diagonal elements we have

$$\frac{d\rho_{n,n}}{dt} = -A(n+1)[1+(n+1)B/A]^{-1}\rho_{n,n} + An[1+nB/A]^{-1}\rho_{n-1,n-1} - Cn\rho_{n,n} + C(n+1)\rho_{n+1,n+1} \quad (4.6.24)$$

which in the stead state yields

$$\rho^{ss}_{n,n} = \mathcal{N}\prod_{k=0}^{n} A/C[1+kB/A]^{-1} \quad (4.6.25)$$

where A, B, and C are constants. In the steady state all off diagonal elements are zero. Then with

$$\gamma_{n,n+1} = -A(n+1)[1+(n+1)B/A]^{-1}$$
$$\gamma_{n,n-1} = -Cn \quad (4.6.26)$$

it follows simply that the Pauli detailed balance condition (4.5.18) holds.

It has now been seen that both the classical phase-space conditions and the Pauli conditions hold; what, however, is the circumstance for the full quantum detailed balance conditions? These are most easily tested in the energy representation, and, with vanishing nondiagonal elements in the steady state and the invariance of the Fock states under time reversal, the full detailed balance condition (4.5.20) reads

$$\gamma^{n',m'}_{m,n}\rho^{ss}_{n,n} = \gamma^{m,n}_{n',m'}\rho^{ss}_{m',m'} \quad (4.6.27)$$

Then, from (4.5.2), (4.5.3), and (4.6.23), here we have

$$\gamma^{n,n'}_{m,m'} = \{-[(n+1)R_{n,n'}+(n'+1)R^{*}_{n',n}]-1/2\cdot C(n+n')\}\delta_{n,m}\,\delta_{n',m'}$$
$$+(R_{n-1,n'-1}+R^{*}_{n'-1,n-1})(nn')^{1/2}\delta_{n,m+1}\,\delta_{n',m'+1}$$
$$+C[(n+1)(m+1)]^{1/2}\delta_{n,m-1}\,\delta_{n',m'-1} \qquad (4.6.28)$$

and hence the condition (4.6.27) requires

$$A[1+(n+1)B/A]^{-1} = R_{n,n'} + R^{*}_{n',n} \qquad (4.6.29)$$

which, via the full definition of the coefficients $R_{n,n'}$ may be written in the form

$$A[1+(n+1)B/A] = A[1+(\tfrac{n+n'}{2}+1)B/A + (n-n')^2 DB/A] \qquad (4.6.30)$$

where D is a constant. This is clearly satisfied only for the diagonal elements $n = n'$, a circumstance which corresponds simply to the Pauli conditions. Thus we see failure of the full detailed balance conditions and illustrate definitively their distinction from the classical limit and Pauli conditions.

CHAPTER V

THE GENERALISED MASTER EQUATION AND DETAILED BALANCE

5.1 Equilibrium Solution

In this final chapter of Part II we will be considering the generalised master equation of section 1.3 in relation to the full quantum detailed balance. This master equation is applicable to the description of the relaxation towards equilibrium of a general quantum system via arbitrary couplings to thermal reservoirs. We will reveal how the nature of these system reservoir couplings may lead to the failure of detailed balance. As a prelude to this work it is necessary that we consider here the equilibrium solution itself, showing that this is given by the canonical form

$$\rho_{ss} = \exp(-H_s/kT)/tr_s \exp(-H_s/kT) \tag{5.1.1}$$

where H_s is the system Hamiltonian, k is Boltzmann's constant, and T the equilibrium temperature.

We take from section (1.3) the generalised master equation which reads

$$\frac{d\rho}{dt} = \mathcal{L}\rho \tag{5.1.2}$$

where

$$\mathcal{L}\rho = (-i/\hbar)[H_s,\rho] + \mathcal{L}_I\rho \tag{5.1.3}$$

$$\mathcal{L}_I\rho = \sum_\lambda \sum_{\mu,\nu} \gamma_\lambda(\omega_\nu)/2\{[\overset{(+)}{S}_\lambda(\omega_\nu)\rho, S^\dagger_\lambda(\omega_\mu)] + [S_\lambda(\omega_\mu), \rho \overset{(+)}{S}{}^\dagger_\lambda(\omega_\nu)]$$
$$+ \bar{n}_\lambda(\omega_\nu)([\overset{(+)}{S}_\lambda(\omega_\nu)\rho, S^\dagger_\lambda(\omega_\mu)] + [S_\lambda(\omega_\mu), \rho\overset{(+)}{S}{}^\dagger_\lambda(\omega_\nu)]$$
$$+ [\overset{(+)}{S}{}^\dagger_\lambda(\omega_\nu)\rho, S_\lambda(\omega_\mu)] + [S^\dagger_\lambda(\omega_\mu), \rho\overset{(+)}{S}_\lambda(\omega_\nu)])\} \qquad (5.1.4)$$

For our present purpose temperatures are uniform throughout all reservoirs and

$$\bar{n}_\lambda(\omega_\nu) = \bar{n}(\omega_\nu) = [\exp(\hbar\omega_\nu/kT)-1]^{-1} \qquad (5.1.5)$$

In the energy representation the operators $S_\lambda^{(\pm)}(\omega_\mu)$ are defined within the scheme for S_λ:

$$S_\lambda = \overset{(+)}{S}_\lambda + \overset{(-)}{S}_\lambda \qquad (5.1.6)$$

where

$$\overset{(+)}{S}_\lambda = \sum_\mu \overset{(+)}{S}_\lambda(\omega_\mu)$$
$$\overset{(+)}{S}_\lambda(\omega_\mu) = \sum_{\substack{n,m \\ E_n < E_m}} |E_n\rangle\langle E_m| S^\lambda_{n,m}\, \delta_{\omega_{n,m},\omega_\mu} \qquad (5.1.7)$$

and

$$\overset{(-)}{S}_\lambda = \sum_\mu \overset{(-)}{S}_\lambda(\omega_\nu)$$
$$\overset{(-)}{S}_\lambda(\omega_\nu) = \sum_{\substack{n,m \\ E_n > E_m}} |E_n\rangle\langle E_m| S^\lambda_{n,m}\, \delta_{\omega_{n,m},\omega_\nu} \qquad (5.1.8)$$

with

$$\omega_{n,m} = |E_n - E_m|/\hbar \qquad (5.1.9)$$

Now our purpose, in demonstrating (5.1.1) as the stationary solution to (5.1.2), is to show

$$\begin{aligned}\mathcal{L}_I\rho_{ss} = \sum_\lambda\sum_{\mu,\nu}\gamma_\lambda(\omega_\nu)/2\cdot\Big\{&[\overset{(+)}{S}_\lambda(\omega_\nu)\rho_{ss},S_\lambda^\dagger(\omega_\mu)] \\ &+[S_\lambda(\omega_\mu),\rho_{ss}\overset{(+)}{S}{}_\lambda^\dagger(\omega_\nu)] \\ +\bar{n}_\lambda(\omega_\nu)\big([\overset{(+)}{S}_\lambda(\omega_\nu)\rho_{ss},S_\lambda^\dagger(\omega_\mu)]&+[S_\lambda(\omega_\mu),\rho_{ss}\overset{(+)}{S}{}_\lambda^\dagger(\omega_\nu)] \\ +[\overset{(+)}{S}{}_\lambda^\dagger(\omega_\nu)\rho_{ss},S_\lambda(\omega_\mu)]&+[S_\lambda^\dagger(\omega_\mu),\rho_{ss}\overset{(+)}{S}_\lambda(\omega_\nu)]\big)\Big\} \\ &= 0\end{aligned} \tag{5.1.10}$$

the first term of (5.1.3) clearly vanishing. A proof follows from four operator identities which facilitate the transfer of ρ_{ss} to the left hand side of this expression. These simple results, which we verify in appendix E, read

$$\overset{(+)}{S}_\lambda(\omega_\nu)\rho_{ss} = \rho_{ss}\overset{(+)}{S}_\lambda(\omega_\nu)\exp(-\hbar\omega_\nu/kT) \tag{5.1.11}$$

$$\overset{(-)}{S}_\lambda(\omega_\nu)\rho_{ss} = \rho_{ss}\overset{(-)}{S}_\lambda(\omega_\nu)\exp(\hbar\omega_\nu/kT) \tag{5.1.12}$$

$$\overset{(+)}{S}{}_\lambda^\dagger(\omega_\nu)\rho_{ss} = \rho_{ss}\overset{(+)}{S}{}_\lambda^\dagger(\omega_\nu)\exp(\hbar\omega_\nu/kT) \tag{5.1.13}$$

$$\overset{(-)}{S}{}_\lambda^\dagger(\omega_\nu)\rho_{ss} = \rho_{ss}\overset{(-)}{S}{}_\lambda^\dagger(\omega_\nu)\exp(-\hbar\omega_\nu/kT) \tag{5.1.14}$$

Invoking then these results, but omitting the tedious algebra, the four commutators in (5.1.10) reduce to the forms

$$[\overset{(+)}{S}_\lambda(\omega_\nu)\rho_{ss},S_\lambda^\dagger(\omega_\mu)] = \rho_{ss}A^\lambda_{\mu,\nu}\exp(-\hbar\omega_\nu/kT) \tag{5.1.15}$$

$$[S_\lambda(\omega_\mu),\rho_{ss}\overset{(+)}{S}{}_\lambda^\dagger(\omega_\nu)] = -\rho_{ss}B^\lambda_{\mu,\nu} \tag{5.1.16}$$

$$[\overset{(+)}{S}{}_\lambda^\dagger(\omega_\nu)\rho_{ss},S_\lambda(\omega_\mu)] = \rho_{ss}B^\lambda_{\mu,\nu}\exp(\hbar\omega_\nu/kT) \tag{5.1.17}$$

$$[S_\lambda^\dagger(\omega_\mu),\rho_{ss}\overset{(+)}{S}_\lambda(\omega_\nu)] = -\rho_{ss}A^\lambda_{\mu,\nu} \tag{5.1.18}$$

where

$$A^{\lambda}_{\mu,\nu} = \overset{(+)}{S}_{\lambda}(\omega_{\nu})\overset{(+)}{S}{}^{\dagger}_{\lambda}(\omega_{\mu}) + \overset{(+)}{S}_{\lambda}(\omega_{\nu})\overset{(-)}{S}{}^{\dagger}_{\lambda}(\omega_{\mu})$$
$$- \overset{(+)}{S}{}^{\dagger}_{\lambda}(\omega_{\mu})\overset{(+)}{S}_{\lambda}(\omega_{\nu})\exp(\hbar\omega_{\mu}/kT)$$
$$- \overset{(-)}{S}{}^{\dagger}_{\lambda}(\omega_{\mu})\overset{(+)}{S}_{\lambda}(\omega_{\nu})\exp(-\hbar\omega_{\mu}/kT) \qquad (5.1.19)$$

$$B^{\lambda}_{\mu,\nu} = \overset{(+)}{S}{}^{\dagger}_{\lambda}(\omega_{\nu})\overset{(+)}{S}_{\lambda}(\omega_{\mu}) + \overset{(+)}{S}{}^{\dagger}_{\lambda}(\omega_{\nu})\overset{(-)}{S}_{\lambda}(\omega_{\mu})$$
$$- \overset{(+)}{S}_{\lambda}(\omega_{\mu})\overset{(+)}{S}{}^{\dagger}_{\lambda}(\omega_{\nu})\exp(-\hbar\omega_{\mu}/kT)$$
$$- \overset{(-)}{S}_{\lambda}(\omega_{\mu})\overset{(+)}{S}{}^{\dagger}_{\lambda}(\omega_{\nu})\exp(\hbar\omega_{\mu}/kT) \qquad (5.1.20)$$

Thus, we may write

$$\mathcal{L}_{I}\rho_{ss} = \sum_{\lambda}\sum_{\mu\nu}\gamma_{\lambda}(\omega_{\nu})/2\,\left[(\bar{n}(\omega_{\nu})+1) - \exp(\hbar\omega_{\nu}/kT)\,\bar{n}(\omega_{\nu})\right]$$
$$\cdot\rho_{ss}\left[\exp(-\hbar\omega_{\nu}/kT)\,A^{\lambda}_{\mu,\nu} - B^{\lambda}_{\mu,\nu}\right] \qquad (5.1.21)$$

It remains only then to recognise that using (5.1.5) we have

$$\bar{n}(\omega_{\nu}) + 1 = \left[\exp(\hbar\omega_{\nu}/kT) - 1\right]^{-1}\exp(\hbar\omega_{\nu}/kT)$$
$$= \bar{n}(\omega_{\nu})\exp(\hbar\omega_{\nu}/kT) \qquad (5.1.22)$$

and on substituting this into (5.1.21) we are led directly to the desired result (5.1.10).

5.2 Detailed Balance through the Operator Conditions

Having obtained the form of the equilibrium density operator for the generalised master equation, we are now in a position to test this equation for the fulfilment of quantum detailed balance. We have derived in the previous chapter detailed balance conditions in both the phase-space calculus and the energy representation. It is also possible, by working directly from (4.1.15), to achieve these conditions in a simple operator form reading

$$\rho_{ss} = \tilde{\rho}_{ss} \tag{5.2.1}$$

$$\rho_{ss}\bar{\mathcal{L}} = \tilde{\mathcal{L}}\rho_{ss} \tag{5.2.2}$$

where in the second result $\bar{\mathcal{L}}$ and $\tilde{\mathcal{L}}$ are defined as in (4.2.11). The derivation of the operator conditions, originally obtained by Agarwal (1973), is outlined in appendix F. It will be our purpose here to apply (5.2.1) and (5.2.2) to the generalised master equation (5.1.2), (5.1.3), (5.1.4) with its equilibrium solution (5.1.1), in this way achieving a broad view as to those features which give rise to the satisfaction or failure of the full quantum detailed balance at equilibrium.

Clearly, due to the time reversal invariance of the Hamiltonian (4.1.4), our equilibrium solution (5.1.1) satisfies (5.2.1). Further, it is a trivial exercise to show the fulfilment of (5.2.2) by the reversible part of the Liouillian . Thus we are concerned solely with an investigation of the condition

$$\rho_{ss}\bar{\mathcal{L}}_I = \tilde{\mathcal{L}}_I\rho_{ss} \tag{5.2.3}$$

Now, as an assurance of the time reversal invariance of (4.1.4) we take S_λ to be itself invariant and $\tilde{\mathcal{L}}_I$ is therefore unchanged from $\mathcal{L}_I$. We write from (5.1.10) the form

$$\begin{aligned}\tilde{\mathcal{L}}_I\hat{O} = \sum_\lambda\sum_{\mu,\nu}\gamma_\lambda(\omega_\nu)/2\cdot\Big[&(\bar{n}(\omega_\nu)+1)\big(S^{(+)}_\lambda(\omega_\nu)\hat{O}S^{\dagger}_\lambda(\omega_\mu)+S_\lambda(\omega_\mu)\hat{O}S^{(+)\dagger}_\lambda(\omega_\nu)\\ &-\hat{O}S^{(+)\dagger}_\lambda(\omega_\nu)S_\lambda(\omega_\mu)-S^{\dagger}_\lambda(\omega_\mu)S^{(+)}_\lambda(\omega_\nu)\hat{O}\big)\\ &+\bar{n}(\omega_\nu)\big(S^{(+)\dagger}_\lambda(\omega_\nu)\hat{O}S_\lambda(\omega_\mu)+S^{\dagger}_\lambda(\omega_\mu)\hat{O}S^{(+)}_\lambda(\omega_\nu)\\ &-\hat{O}S^{(+)}_\lambda(\omega_\nu)S^{\dagger}_\lambda(\omega_\mu)-S_\lambda(\omega_\mu)S^{(+)\dagger}_\lambda(\omega_\nu)\hat{O}\big)\Big]\end{aligned} \tag{5.2.4}$$

where $\hat{O}$ is some arbitrary operator. Further, applying the second of (4.2.11), via the cyclic property of the trace we have

$$\bar{\mathcal{L}}_I \hat{O} = \sum_\lambda \sum_{\mu,\nu} \gamma_\lambda(\omega_\nu)/2 \, [(\bar{n}(\omega_\nu)+1)(S_\lambda^{(+)\dagger}(\omega_\nu)\hat{O}S_\lambda(\omega_\mu) + S_\lambda^\dagger(\omega_\mu)\hat{O}S_\lambda^{(+)}(\omega_\nu)$$
$$-\hat{O}S_\lambda^\dagger(\omega_\mu)S_\lambda^{(+)}(\omega_\nu) - S_\lambda^{(+)\dagger}(\omega_\nu)S_\lambda(\omega_\mu)\hat{O}$$
$$+\bar{n}(\omega_\nu)(S_\lambda^{(+)}(\omega_\nu)\hat{O}S_\lambda^\dagger(\omega_\mu) + S_\lambda(\omega_\mu)\hat{O}S_\lambda^{(+)\dagger}(\omega_\nu)$$
$$-\hat{O}S_\lambda(\omega_\mu)S_\lambda^{(+)\dagger}(\omega_\nu) - S_\lambda^{(+)}(\omega_\nu)S_\lambda^\dagger(\omega_\mu)\hat{O})] \tag{5.2.5}$$

From here we make recourse to the identities (5.1.11) to (5.1.14) and the method of the previous section. This enables us to take ρ_{ss} through to the left in $\tilde{\mathcal{L}}_I \rho_{ss}$ and hence facilitates a direct comparison of this operator with $\rho_{ss} \bar{\mathcal{L}}_I$. Thus we may write

$$\tilde{\mathcal{L}}_I \rho_{ss} = \rho_{ss} \mathcal{L}_I' \tag{5.2.6}$$

where, with the aid of (5.1.22), we find

$$\mathcal{L}_I' \hat{O} = \sum_\lambda \sum_{\mu,\nu} \gamma_\lambda(\omega_\nu)/2 \cdot [(\bar{n}(\omega_\nu)+1)(S_\lambda^{(+)\dagger}(\omega_\nu)\hat{O}S_\lambda(\omega_\mu) + \mathcal{S}_\lambda^{\prime\mu,\nu\prime}\hat{O}S_\lambda^{(+)}(\omega_\nu)$$
$$-\hat{O}S_\lambda^{(+)\dagger}(\omega_\nu)S_\lambda(\omega_\mu) - \mathcal{S}_\lambda^{\prime\mu,\nu\prime}S_\lambda^{(+)}(\omega_\nu)\hat{O})$$
$$+\bar{n}(\omega_\nu)(S_\lambda^{(+)}(\omega_\nu)\hat{O}S_\lambda^\dagger(\omega_\mu) + \mathcal{S}_\lambda^{\prime\mu,\nu}\hat{O}S_\lambda^{(+)\dagger}(\omega_\nu)$$
$$-\hat{O}S_\lambda^{(+)}(\omega_\nu)S_\lambda^\dagger(\omega_\mu) - \mathcal{S}_\lambda^{\prime\mu,\nu}S_\lambda^{(+)\dagger}(\omega_\nu)\hat{O})] \tag{5.2.7}$$

with

$$\mathcal{S}_\lambda^{\prime\mu,\nu} = S_\lambda^{(+)}(\omega_\mu)\exp[\hbar(\omega_\nu-\omega_\mu)/kT]$$
$$+ S_\lambda^{(-)}(\omega_\mu)\exp[\hbar(\omega_\nu+\omega_\mu)/kT] \tag{5.2.8}$$

$$\mathcal{S}'^{\mu,\nu}_{\lambda} = S^{(+)\dagger}_{\lambda}(\omega_\mu)\exp[-\hbar(\omega_\nu-\omega_\mu)/kT] + S^{(-)}_{\lambda}(\omega_\mu)\exp[-\hbar(\omega_\nu+\omega_\mu)/kT] \quad (5.2.9)$$

In this, our immediate observation is that $\mathcal{L}'_I$ and $\bar{\mathcal{L}}_I$ are not equivalent; there is a variance between the pair of operators $S_\lambda(\omega_\mu)$, $S^\dagger_\lambda(\omega_\mu)$ and the pair $\mathcal{S}^{\mu,\nu}_\lambda$, $\mathcal{S}'^{\mu,\nu}_\lambda$. Fulfilment of full quantum detailed balance is therefore not the general circumstance for the operator master equation at equilibrium.

Looking more deeply into the situation arising in (5.2.8) and (5.2.9) we note that, using (5.1.22), we may write these expressions in the forms

$$\mathcal{S}^{\mu,\nu}_{\lambda} = [\bar{n}(\omega_\nu)^{-1}(\bar{n}(\omega_\nu)+1)][(\bar{n}(\omega_\mu)+1)^{-1}\bar{n}(\omega_\mu)]\,S^{(+)}_{\lambda}(\omega_\mu) + [\bar{n}(\omega_\nu)^{-1}(\bar{n}(\omega_\nu)+1)][\bar{n}(\omega_\mu)^{-1}(\bar{n}(\omega_\mu)+1)]\,S^{(-)}_{\lambda}(\omega_\mu) \quad (5.2.10)$$

$$\mathcal{S}'^{\mu,\nu}_{\lambda} = [(\bar{n}(\omega_\nu)+1)^{-1}\bar{n}(\omega_\nu)][\bar{n}(\omega_\mu)^{-1}(\bar{n}(\omega_\mu)+1)]\,S^{(+)\dagger}_{\lambda}(\omega_\mu) + [(\bar{n}(\omega_\nu)+1)^{-1}\bar{n}(\omega_\nu)][(\bar{n}(\omega_\mu)+1)^{-1}\bar{n}(\omega_\mu)]\,S^{(-)\dagger}_{\lambda}(\omega_\mu) \quad (5.2.11)$$

Thus we see failure of detailed balance here clearly as a manifestation of the quantum nature of our system. Indeed, in the formal classical limit for which $\bar{n}(\omega_\nu) \to \infty$, (5.2.10) and (5.2.11) reduce to expressions for $S_\lambda(\omega_\mu)$ and its Hermitian conjugate and (5.2.7) becomes identical to (5.2.5). It is not our intention to investigate in detail the origin of this problem, merely observing that it lies somewhere within the approximations mediating (4.1.14) and (4.1.15). We can, however, identify particular features of the system-reservoir coupling operators S_λ, which, by their presence, contribute to the failure of detailed balance.

We notice that in (5.2.10) and (5.2.11), if we impose the conditions:

a) all terms for which $\mu \neq \nu$ vanish,

b) $S_\lambda^{(-)}(\omega_\mu)$ vanishes,

we obtain

$$\begin{aligned} \mathcal{S}_\lambda^{\mu,\nu} &= S_\lambda^{(+)}(\omega_\nu) \\ \mathcal{S}_\lambda^{\mu,\nu'} &= S_\lambda^{(+)\dagger}(\omega_\nu) \end{aligned} \tag{5.2.12}$$

while, in (5.2.5), $S_\lambda(\omega_\nu)$ becomes $S_\lambda^{(+)}(\omega_\nu)$ and hence (5.2.7) and (5.2.5) are equal, irrespective of the value of $\bar{n}(\omega_\nu)$. For all such master equations full quantum detailed balance is satisfied. In this we have isolated two features. Firstly, only exchanges of energy with the boson reservoir at a single frequency are allowed which requires $S_\lambda^{(+)}(\omega_\nu)$ to comprise only diadics of equally spaced energy eigenstates. The second requirement is the omission of negative frequency components from S_λ which removes all highly oscillating terms from the system reservoir interaction. These separate classifications may manifest themselves quite eplicitly in separate master equations. This we will demonstrate in a forthcoming section where we present three boson master equations, one satisfying the quantum detailed balance while the other two fail, each in one of the respects noted above.

5.3 The Generalised Master Equation and the Diagonal Form

In section (4.5) we have defined the diagonal master equation by laying two restrictions on the Liouvillian $\mathcal{L}$. In the notation of (5.1.7) and (5.1.8) these read

$$\langle E_n|(\mathcal{L}|E_m\rangle\langle E_{m'}|)|E_n\rangle \propto \delta_{E_m,E_{m'}} \tag{5.3.1}$$

and

$$\langle E_n|(\mathcal{L}|E_m\rangle\langle E_m|)|E_{n'}\rangle \propto \delta_{E_n,E_{n'}} \tag{5.3.2}$$

For such equations, when attention is restricted to correlation of diagonal operators only, the quantum detailed balance statement (4.1.15) reduces to the well known Pauli detailed balance of Chapter 3. In this section we will cast the generalised master equation into a form within the energy representation and see just when this itself takes on the features of a diagonal master equation.

Since the reversible part of $\mathcal{L}$ trivially obeys (5.3.1) and (5.3.2) we consider the requirements

$$\langle E_n|(\mathcal{L}_I|E_m\rangle\langle E_{m'}|)|E_n\rangle \propto \delta_{E_m,E_{m'}} \tag{5.3.3}$$

$$\langle E_n|(\mathcal{L}_I|E_m\rangle\langle E_m|)|E_{n'}\rangle \propto \delta_{E_n,E_{n'}} \tag{5.3.4}$$

where, from the form (5.1.4) for $\mathcal{L}_I$, we have

$$\begin{aligned}\langle E_{n'}|(\mathcal{L}_I|E_m\rangle\langle E_{m'}|)|E_{n'}\rangle = \sum_{\lambda}\sum_{\mu,\nu} \gamma_\lambda(\omega_\nu)/2 \Big[&(\bar{n}(\omega_\nu)+1) \\ &\Big(\overset{(+)}{S}_\lambda(\omega_\nu)_{E_n,E_m} S^\dagger_\lambda(\omega_\mu)_{E_{m'},E_{n'}} + S_\lambda(\omega_\mu)_{E_n,E_m} \overset{(+)}{S}{}^\dagger_\lambda(\omega_\nu)_{E_{m'},E_{n'}} \\ &- \delta_{E_n,E_m}[\overset{(+)}{S}{}^\dagger_\lambda(\omega_\nu) S_\lambda(\omega_\mu)]_{E_{m'},E_{n'}} - \delta_{E_{m'},E_{n'}}[S^\dagger_\lambda(\omega_\mu)\overset{(+)}{S}_\lambda(\omega_\nu)]_{E_n,E_m}\Big) \\ &+ \bar{n}(\omega_\nu) \\ &\Big(\overset{(+)}{S}{}^\dagger_\lambda(\omega_\nu)_{E_n,E_m} S_\lambda(\omega_\mu)_{E_{m'},E_{n'}} + S^\dagger_\lambda(\omega_\mu)_{E_n,E_m} \overset{(+)}{S}_\lambda(\omega_\nu)_{E_{m'},E_{n'}} \\ &- \delta_{E_n,E_m}[\overset{(+)}{S}_\lambda(\omega_\nu) S^\dagger_\lambda(\omega_\mu)]_{E_{m'},E_{n'}} - \delta_{E_{m'},E_{n'}}[S_\lambda(\omega_\mu)\overset{(+)}{S}{}^\dagger_\lambda(\omega_\nu)]_{E_n,E_m}\Big)\Big]\end{aligned} \tag{5.3.5}$$

and, as a corollary to (5.1.7) and (5.1.8), it is found that

$$\overset{(\pm)}{S}_\lambda(\omega_\mu)_{E_n,E_m} = \langle E_n|\overset{(\pm)}{S}_\lambda(\omega_\mu)|E_m\rangle = S^\lambda_{n,m}\,\delta_{E_n\pm\hbar\omega_\mu,E_m} \tag{5.3.6}$$

and

$$\begin{aligned}[\overset{(+)}{S}{}^\dagger_\lambda(\omega_\nu)\overset{(\pm)}{S}_\lambda(\omega_\mu)]_{E_{m'},E_{n'}} &= \langle E_{m'}|\overset{(+)}{S}{}^\dagger_\lambda(\omega_\nu)\overset{(\pm)}{S}_\lambda(\omega_\mu)|E_{n'}\rangle \\ &= S^{\lambda *}_{E_{m'}-\hbar\omega_\nu,E_{m'}}\, S^\lambda_{E_{n'}\mp\hbar\omega_\mu,E_{n'}}\,\delta_{E_{m'}-\hbar\omega_\nu,E_{n'}\mp\hbar\omega_\mu}\end{aligned} \tag{5.3.7}$$

$$[\overset{(\pm)}{S}{}_\lambda^\dagger(\omega_\mu)\overset{(+)}{S}_\lambda(\omega_\nu)]_{E_n,E_m} = \langle E_n|\overset{(\pm)}{S}{}_\lambda^\dagger(\omega_\mu)\overset{(+)}{S}_\lambda(\omega_\nu)|E_m\rangle$$

$$= S^{\lambda^*}_{E_n-\hbar\omega_\mu,E_n} S^{\lambda}_{E_m-\hbar\omega_\nu,E_m}\,\delta_{E_n\mp\hbar\omega_\mu,E_m-\hbar\omega_\nu} \quad (5.3.8)$$

$$[\overset{(+)}{S}_\lambda(\omega_\nu)\overset{(\pm)}{S}{}_\lambda^\dagger(\omega_\mu)]_{E_m',E_n'} = \langle E_m'|\overset{(+)}{S}_\lambda(\omega_\nu)\overset{(\pm)}{S}{}_\lambda^\dagger(\omega_\mu)|E_n'\rangle$$

$$= S^{\lambda}_{E_m',E_m'+\hbar\omega_\nu} S^{\lambda^*}_{E_n',E_n'\pm\hbar\omega_\mu}\,\delta_{E_m'+\hbar\omega_\nu,E_n'\pm\hbar\omega_\mu} \quad (5.3.9)$$

$$[\overset{(\pm)}{S}_\lambda(\omega_\mu)\overset{(+)}{S}{}_\lambda^\dagger(\omega_\nu)]_{E_n,E_m} = \langle E_n|\overset{(\pm)}{S}_\lambda(\omega_\mu)\overset{(+)}{S}{}_\lambda^\dagger(\omega_\nu)|E_m\rangle$$

$$= S^{\lambda}_{E_n,E_n\pm\hbar\omega_\mu} S^{\lambda^*}_{E_m,E_m+\hbar\omega_\nu}\,\delta_{E_n\pm\hbar\omega_\mu,E_m+\hbar\omega_\nu} \quad (5.3.10)$$

Now, in particular, we may write for comparison with (5.3.3)

$$\begin{aligned}\langle E_n|(\mathcal{L}_I|E_m\rangle\langle E_m'|)|E_n'\rangle = \sum_\lambda\sum_{\mu,\nu}\gamma_\lambda(\omega_\nu)/2\cdot\Big[(\bar n(\omega_\nu)+1)\\ (\overset{(+)}{S}_\lambda(\omega_\nu)_{E_n,E_m}S_\lambda^\dagger(\omega_\mu)_{E_m',E_n'} + S_\lambda(\omega_\mu)_{E_n,E_m}\overset{(+)}{S}{}_\lambda^\dagger(\omega_\nu)_{E_m',E_n'}\\ -\delta_{E_n,E_m}[\overset{(+)}{S}{}_\lambda^\dagger(\omega_\nu)S_\lambda(\omega_\mu)]_{E_m',E_n'}-\delta_{E_m',E_n'}[S_\lambda^\dagger(\omega_\mu)\overset{(+)}{S}_\lambda(\omega_\nu)]_{E_n,E_m})\\ +\bar n(\omega_\nu)\\ (\overset{(+)}{S}{}_\lambda^\dagger(\omega_\nu)_{E_n,E_m}S_\lambda(\omega_\mu)_{E_m',E_n'} + S_\lambda^\dagger(\omega_\mu)_{E_n,E_m}\overset{(+)}{S}_\lambda(\omega_\nu)_{E_m',E_n'}\\ -\delta_{E_n,E_m}[\overset{(+)}{S}_\lambda(\omega_\nu)S_\lambda^\dagger(\omega_\mu)]_{E_m',E_n'}-\delta_{E_m',E_n'}[S_\lambda(\omega_\mu)\overset{(+)}{S}{}_\lambda^\dagger(\omega_\nu)]_{E_n,E_m})\Big]\end{aligned} \quad (5.3.11)$$

for which, from (5.3.6) to (5.3.10)

$$\overset{(+)}{S}_\lambda(\omega_\nu)_{E_n,E_m}\overset{(\pm)}{S}{}_\lambda^\dagger(\omega_\mu)_{E_m',E_n'} = S^{\lambda}_{E_m-\hbar\omega_\nu,E_m} S^{\lambda^*}_{E_m'\mp\hbar\omega_\mu,E_m'}\,\delta_{E_n,E_m-\hbar\omega_\nu}$$

$$\delta_{E_m-\hbar\omega_\nu,E_m'\mp\hbar\omega_\mu} \quad (5.3.12)$$

$$\overset{(\pm)}{S}_\lambda(\omega_\mu)_{E_n,E_m}\overset{(+)}{S}{}_\lambda^\dagger(\omega_\nu)_{E_m',E_n'} = S^{\lambda}_{E_m\mp\hbar\omega_\mu,E_m} S^{\lambda^*}_{E_m'-\hbar\omega_\nu,E_m'}\,\delta_{E_n,E_m'-\hbar\omega_\nu}$$

$$\delta_{E_m\mp\hbar\omega_\mu,E_m'-\hbar\omega_\nu} \quad (5.3.13)$$

$$\overset{(+)}{S}{}^{\dagger}_{\lambda}(\omega_\nu)_{E_n,E_m}\overset{(\pm)}{S}_{\lambda}(\omega_\mu)_{E_m',E_n} = S^{\lambda^*}_{E_m,E_m+\hbar\omega_\nu}\, S^{\lambda}_{E_m',E_m'\pm\hbar\omega_\mu}\,\delta_{E_n,E_m+\hbar\omega_\nu}\,\delta_{E_m+\hbar\omega_\nu,\,E_m'+\hbar\omega_\mu} \tag{5.3.14}$$

$$\overset{(\pm)}{S}{}^{\dagger}_{\lambda}(\omega_\mu)_{E_n,E_m}\overset{(+)}{S}_{\lambda}(\omega_\nu)_{E_m',E_n} = S^{\lambda^*}_{E_m,E_m\pm\hbar\omega_\mu}\, S^{\lambda}_{E_m',E_m'+\hbar\omega_\nu}\,\delta_{E_n,E_m'+\hbar\omega_\nu}\,\delta_{E_m\pm\hbar\omega_\mu,\,E_m'+\hbar\omega_\nu} \tag{5.3.15}$$

and

$$\delta_{E_n,E_m}[\overset{(+)}{S}{}^{\dagger}_{\lambda}(\omega_\nu)\overset{(\pm)}{S}_{\lambda}(\omega_\mu)]_{E_m',E_n} = S^{\lambda^*}_{E_m'-\hbar\omega_\nu,E_m'}\, S^{\lambda}_{E_m\mp\hbar\omega_\mu,E_m}\,\delta_{E_n,E_m}\,\delta_{E_m'-\hbar\omega_\nu,\,E_m\mp\hbar\omega_\mu} \tag{5.3.16}$$

$$\delta_{E_m',E_n}[\overset{(\pm)}{S}{}^{\dagger}_{\lambda}(\omega_\mu)\overset{(+)}{S}_{\lambda}(\omega_\nu)]_{E_n,E_m} = S^{\lambda^*}_{E_m'\mp\hbar\omega_\mu,E_m'}\, S^{\lambda}_{E_m-\hbar\omega_\nu,E_m}\,\delta_{E_n,E_m'}\,\delta_{E_m'\mp\hbar\omega_\nu,\,E_m-\hbar\omega_\mu} \tag{5.3.17}$$

$$\delta_{E_n,E_m}[\overset{(+)}{S}_{\lambda}(\omega_\nu)\overset{(\pm)}{S}{}^{\dagger}_{\lambda}(\omega_\mu)]_{E_m',E_n} = S^{\lambda}_{E_m',E_m'+\hbar\omega_\nu}\, S^{\lambda^*}_{E_m,E_m\pm\hbar\omega_\mu}\,\delta_{E_n,E_m}\,\delta_{E_m'+\hbar\omega_\nu,\,E_m\pm\hbar\omega_\mu} \tag{5.3.18}$$

$$\delta_{E_m',E_n}[\overset{(\pm)}{S}_{\lambda}(\omega_\mu)\overset{(+)}{S}{}^{\dagger}_{\lambda}(\omega_\nu)]_{E_n,E_m} = S^{\lambda}_{E_m',E_m'\pm\hbar\omega_\mu}\, S^{\lambda^*}_{E_m,E_m+\hbar\omega_\nu}\,\delta_{E_n,E_m'}\,\delta_{E_m'\pm\hbar\omega_\mu,\,E_m+\hbar\omega_\nu} \tag{5.3.19}$$

In addition, for comparison with (5.3.4)

$$\begin{aligned}\langle E_n|(\mathcal{L}_I|E_m\rangle\langle E_m|)|E_{n'}\rangle = \sum_{\lambda,\mu,\nu}\sum \gamma_\lambda(\omega_\nu)/2\cdot\Big[&(\bar{n}(\omega_\nu)+1)\\ &(\overset{(+)}{S}_{\lambda}(\omega_\nu)_{E_n,E_m}S^{\dagger}_{\lambda}(\omega_\mu)_{E_m,E_{n'}} + S_{\lambda}(\omega_\mu)_{E_n,E_m}\overset{(+)}{S}{}^{\dagger}_{\lambda}(\omega_\nu)_{E_m,E_{n'}}\\ &-\delta_{E_n,E_m}[\overset{(+)}{S}_{\lambda}(\omega_\nu)S^{\dagger}_{\lambda}(\omega_\mu)]_{E_m,E_{n'}}-\delta_{E_m,E_{n'}}[S^{\dagger}_{\lambda}(\omega_\mu)\overset{(+)}{S}_{\lambda}(\omega_\nu)]_{E_n,E_m})\\ &+\bar{n}(\omega_\nu)\\ &(\overset{(+)}{S}{}^{\dagger}_{\lambda}(\omega_\nu)_{E_n,E_m}S_{\lambda}(\omega_\mu)_{E_m,E_{n'}} + S^{\dagger}_{\lambda}(\omega_\mu)_{E_n,E_m}\overset{(+)}{S}_{\lambda}(\omega_\nu)_{E_m,E_{n'}}\\ &-\delta_{E_n,E_m}[\overset{(+)}{S}_{\lambda}(\omega_\nu)S^{\dagger}_{\lambda}(\omega_\mu)]_{E_m,E_{n'}}-\delta_{E_m,E_{n'}}[S_{\lambda}(\omega_\mu)\overset{(+)}{S}{}^{\dagger}_{\lambda}(\omega_\nu)]_{E_n,E_m})\Big]\end{aligned} \tag{5.3.20}$$

where

$$\overset{(+)}{S}_\lambda(\omega_\nu)_{E_n,E_m}\,\overset{(\pm)}{S}_\lambda(\omega_\mu)^\dagger_{E_m,E_n'} = S^{\lambda}_{E_n,E_n+\hbar\omega_\nu}\,S^{\lambda^*}_{E_n',E_n'\pm\hbar\omega_\mu}\,\delta_{E_n+\hbar\omega_\nu,E_m}\,\delta_{E_n+\hbar\omega_\nu,E_n'\pm\hbar\omega_\mu} \tag{5.3.21}$$

$$\overset{(\pm)}{S}_\lambda(\omega_\mu)_{E_n,E_m}\,\overset{(+)}{S}_\lambda(\omega_\nu)^\dagger_{E_m,E_n'} = S^{\lambda}_{E_n,E_n\pm\hbar\omega_\mu}\,S^{\lambda^*}_{E_n',E_n'+\hbar\omega_\nu}\,\delta_{E_n'+\hbar\omega_\nu,E_m}\,\delta_{E_n\pm\hbar\omega_\mu,E_n'+\hbar\omega_\nu} \tag{5.3.22}$$

$$\overset{(+)}{S}_\lambda(\omega_\nu)^\dagger_{E_n,E_m}\,\overset{(\pm)}{S}_\lambda(\omega_\mu)_{E_m,E_n'} = S^{\lambda^*}_{E_n-\hbar\omega_\nu,E_n}\,S^{\lambda}_{E_n'\mp\hbar\omega_\mu,E_n'}\,\delta_{E_n-\hbar\omega_\nu,E_m}\,\delta_{E_n-\hbar\omega_\nu,E_n'\mp\hbar\omega_\mu} \tag{5.3.23}$$

$$\overset{(\pm)}{S}_\lambda(\omega_\mu)^\dagger_{E_n,E_m}\,\overset{(+)}{S}_\lambda(\omega_\nu)_{E_m,E_n'} = S^{\lambda^*}_{E_n\mp\hbar\omega_\mu,E_n}\,S^{\lambda}_{E_n'-\hbar\omega_\nu,E_n'}\,\delta_{E_n'-\hbar\omega_\nu,E_m}\,\delta_{E_n\mp\hbar\omega_\mu,E_n'-\hbar\omega_\nu} \tag{5.3.24}$$

and

$$\delta_{E_n,E_m}\left[\overset{(+)}{S}_\lambda(\omega_\nu)^\dagger\,\overset{(\pm)}{S}_\lambda(\omega_\mu)\right]_{E_m,E_n'} = S^{\lambda^*}_{E_n-\hbar\omega_\nu,E_n}\,S^{\lambda}_{E_n'\mp\hbar\omega_\mu,E_n'}\,\delta_{E_n,E_m}\,\delta_{E_n-\hbar\omega_\nu,E_n'\mp\hbar\omega_\mu} \tag{5.3.25}$$

$$\delta_{E_m,E_n'}\left[\overset{(\pm)}{S}_\lambda(\omega_\mu)^\dagger\,\overset{(+)}{S}_\lambda(\omega_\nu)\right]_{E_n,E_m} = S^{\lambda^*}_{E_n\mp\hbar\omega_\mu,E_n}\,S^{\lambda}_{E_n'-\hbar\omega_\nu,E_n'}\,\delta_{E_n',E_m}\,\delta_{E_n\mp\hbar\omega_\mu,E_n'-\hbar\omega_\nu} \tag{5.3.26}$$

$$\delta_{E_n,E_m}\left[\overset{(+)}{S}_\lambda(\omega_\nu)\,\overset{(\pm)}{S}_\lambda(\omega_\mu)^\dagger\right]_{E_m,E_n'} = S^{\lambda}_{E_n,E_n+\hbar\omega_\nu}\,S^{\lambda^*}_{E_n',E_n'\pm\hbar\omega_\mu}\,\delta_{E_n,E_m}\,\delta_{E_n+\hbar\omega_\nu,E_n'\pm\hbar\omega_\mu} \tag{5.3.27}$$

$$\delta_{E_m,E_n'}\left[\overset{(\pm)}{S}_\lambda(\omega_\mu)\,\overset{(+)}{S}_\lambda(\omega_\nu)^\dagger\right]_{E_n,E_m} = S^{\lambda}_{E_n,E_n\pm\hbar\omega_\mu}\,S^{\lambda^*}_{E_n',E_n'+\hbar\omega_\nu}\,\delta_{E_n',E_m}\,\delta_{E_n\pm\hbar\omega_\mu,E_n'+\hbar\omega_\nu} \tag{5.3.28}$$

Now, examining the terms (5.3.12) to (5.3.19) and (5.3.21) to (5.3.28) we find that, just as the operator master equation does not generally satisfy quantum detailed balance, neither does it in the general case take on the diagonal form specified by (5.3.3) and (5.3.4) in the energy representation. However, as previously, we may impose the requirement that

a) all terms for which $\mu \neq \nu$ vanish,

b) $S_\lambda^{(-)}(\omega_\mu)$ vanishes.

In so doing we find that all remaining terms in (5.3.12) to (5.3.19) are then proportional to $\delta_{m,m'}$, while corresponding terms in (5.3.21) to (5.3.28) are proportional to $\delta_{n,n'}$. This is just the requirement of (5.3.3) and (5.3.4). We therefore conclude that those same terms in the generalised master equation which give rise to the failure of full quantum detailed balance also appear as nondiagonal terms in the energy representation. Thus, for our generalised operator master equation at equilibrium, satisfaction of full quantum detailed balance is equivalent to the acquisition of a master equation in diagonal form in the energy representation.

5.4 Examples

Here we present three boson master equations together with the operators S_λ characterising their associated system-reservoir interactions. In this we illustrate the satisfaction or failure of quantum detailed balance due to the nature of the system-reservoir interaction Hamiltonian.

As a first example, the operator master equation for an harmonic oscillator coupled to a single thermal reservoir at temperature T is familiar (Louisell, 1969; Carmichael, 1972). This follows the scheme (5.1.2) to (5.1.4) with

$$H_S = \hbar\omega_0 a^\dagger a \tag{5.4.1}$$

$$\mathcal{L}_I \rho = (\gamma/2)\{[a\rho, a^\dagger] + [a, \rho a^\dagger] + \bar{n}([a\rho, a^\dagger] + [a, \rho a^\dagger] + [a^\dagger\rho, a] + [a^\dagger, \rho a])\} \tag{5.4.2}$$

Here $a^\dagger$ and a are creation and annihilation operators obeying boson commutation relations $[a,a^\dagger] = 1$, ω_0 is the frequency of free oscillation, γ is a damping constant, and $\bar{n}$ the mean occupation number for reservoir oscillators at ω_0. In this case the reservoir interaction is an interaction between bosons in the rotating-wave approximation, and thus takes the form (1.3.4) with

$$S = a \tag{5.4.3}$$

Now the energy eigenstates are provided by the Fock states, for which

$$H_s|n\rangle = n\hbar\omega_0|n\rangle \tag{5.4.4}$$

In terms of these we may write

$$a = \sum_n n^{1/2}|n-1\rangle\langle n| \tag{5.4.5}$$

which carries into the scheme (5.1.6) to (5.1.9) with the identification

$$\begin{aligned} S^{(+)} &= S^{(+)}(\omega_0) = \sum_n n^{1/2}|n-1\rangle\langle n| \\ S^{(-)} &= 0 \end{aligned} \tag{5.4.6}$$

With this (5.4.2) appears directly in the form (5.1.4). In (5.4.6) we have the requirements which, in section (5.2), assured satisfaction of quantum detailed balance, and further produced a master equation in diagonal form in section (5.3). Indeed there appears here only a single energy spacing between the diadics constituting the expansion of $S^{(+)}$, while in addition $S^{(-)}$ vanishes. The fulfilment of detailed balance by (5.4.2) has, in fact, already been established independently in section (4.6) where we applied phase-space detailed balance conditions.

For our second example we move to resonantly coupled bosons, one of which is damped by coupling to a thermal reservoir (Carmichael and

Walls (1973). Here

$$H_s = \hbar\omega_0 a^\dagger a + \hbar\omega_0 b^\dagger b + \hbar\kappa(a^\dagger b + ab^\dagger) \tag{5.4.7}$$

or alternatively, in terms of normal modes

$$H_s = \hbar(\omega_0+\kappa)A^\dagger A + \hbar(\omega_0-\kappa)B^\dagger B \tag{5.4.8}$$

where

$$\begin{aligned} A &= 1/\sqrt{2}\,(a+b) \\ B &= 1/\sqrt{2}\,(a-b) \end{aligned} \tag{5.4.9}$$

$b^\dagger$ and b, $A^\dagger$ and A, and $B^\dagger$ and B are also boson creation and annihilation operator pairs, while κ is a coupling constant. Now the master equation, taken in terms of normal mode operators, has

$$\begin{aligned}
\mathcal{L}_I\rho = {} & \gamma(\omega_0+\kappa)/2\cdot\tfrac{1}{2}\{[A\rho,A^\dagger]+[A,\rho A^\dagger]+\bar{n}(\omega_0+\kappa)([A\rho,A^\dagger] \\
& \qquad +[A,\rho A^\dagger]+[A^\dagger,\rho A]+[A^\dagger\rho,A])\} \\
& +\gamma(\omega_0+\kappa)/2\cdot\tfrac{1}{2}\{[A\rho,B^\dagger]+[B,\rho A^\dagger]+\bar{n}(\omega_0+\kappa)([A\rho,B^\dagger] \\
& \qquad +[B,\rho A^\dagger]+[A^\dagger\rho,B]+[B^\dagger,\rho A])\} \\
& +\gamma(\omega_0-\kappa)/2\cdot\tfrac{1}{2}\{[B\rho,A^\dagger]+[A,\rho B^\dagger]+\bar{n}(\omega_0-\kappa)([B\rho,A^\dagger] \\
& \qquad +[A,\rho B^\dagger]+[B^\dagger\rho,A]+[A^\dagger,\rho B])\} \\
& +\gamma(\omega_0-\kappa)/2\cdot\tfrac{1}{2}\{[B\rho,B^\dagger]+[B,\rho B^\dagger]+\bar{n}(\omega_0-\kappa)([B\rho,B^\dagger] \\
& \qquad +[B,\rho B^\dagger]+[B^\dagger\rho,B]+[B^\dagger,\rho B])\}
\end{aligned} \tag{5.4.10}$$

Again, this appears as an example of the form (5.1.4) when the reservoir coupling operator a is expressed within the scheme (5.1.6) to (5.1.9). The energy eigenstates are here best given as the Fock states $|N,M\rangle$ for the normal mode operators A and B. Then

$$S^{(+)} = S^{(+)}(\omega_o+\kappa) + S^{(+)}(\omega_o-\kappa)$$
$$= 1/\sqrt{2}\left(\sum_N N^{1/2}|N-1,M\rangle\langle N,M| + \sum_M M^{1/2}|N,M-1\rangle\langle N,M|\right)$$
$$= 1/\sqrt{2}\,(A+B) \qquad (5.4.11)$$

$$S^{(-)} = 0 \qquad (5.4.12)$$

A primary difference arises between this and (5.4.6) for the single boson. The energy spacings for diadics appearing in the expansion for $S^{(+)}$ now demand a separation within this expansion into the two operators $S^{(+)}(\omega_0 + \kappa)$ and $S^{(+)}(\omega_0 - \kappa)$ which correspond to the operators A and B. Thus, in (5.4.10) there appears the two sets of cross-product terms. In line with section (5.2), these bring about a failure of quantum detailed balance and it may be taken as a general property of internally coupled systems that such cross products will arise and the failure of full quantum detailed balance will ensue.

As our final example we cite a master equation first derived by Agarwal (1971) for spontaneous emission without the rotating-wave approximation. This is simply the oscillator master equation (5.4.2), derived from a system reservoir interaction with

$$S = (a + a^\dagger) \qquad (5.4.13)$$

In this instance

$$\mathcal{L}_I\rho = (\gamma/2)\{[a\rho,(a+a^\dagger)] + [(a+a^\dagger),\rho a^\dagger]$$
$$+ \bar{n}\,([a\rho,(a+a^\dagger)] + [(a+a^\dagger),\rho a^\dagger]$$
$$+ [a^\dagger\rho,(a+a^\dagger)] + [(a+a^\dagger),\rho a])\} \qquad (5.4.14)$$

while, with (5.4.5), the identification of (5.4.13) with the scheme

(5.1.6) to (5.1.9) has

$$\begin{aligned} S^{(+)} &= S^{(+)}(\omega_o) = \sum_n n^{1/2} |n-1\rangle\langle n| \\ S^{(-)} &= S^{(-)}(\omega_o) = \sum_n (n+1)^{1/2} |n\rangle\langle n+1| \end{aligned} \qquad (5.4.15)$$

Thus, we see the appearance of a non-vanishing $S^{(-)}(\omega_o)$, and in this have an example of failure of quantum detailed balance at equilibrium in the second manner outlined in section (5.2).

PART III

RESONANCE FLUORESCENCE AND THE DYNAMICAL STARK EFFECT

Our visual perception of the environment is greatly enhanced by the colour and texture imparted through the interaction of light with matter. Working with his understanding of this interaction the theoretical physicist provides description of the many contributing phenomenon. Amongst these is featured that of resonance fluorescence.

CHAPTER VI

RESONANCE FLUORESCENCE - AN EXAMPLE OF AN OPEN QUANTUM MARKOFFIAN SYSTEM

6.1 Introduction

An understanding of the optical phenomena which underly the colouring of our visual environment must have been a primary objective in the earliest studies into the interaction of radiation with matter. In our own experience, for the most part, we are involved with objects coloured through the preferential absorption of light within some spectral range. Colour is imparted through the reflection or transmission of the remaining parts of the spectrum. On the other hand, there is a group of phenomena whose effect arises through preferential scattering. Of these perhaps the most familiar is Rayleigh scattering by particles small in comparison with optical wavelengths, where, in the ω^4 frequency dependence of Rayleigh's scattering formula, there is contained a mechanism providing the blue canopy of our sky. It is within this category of scattering phenomena that we encounter the property of fluorescence. Fluorescence has intrigued physicists for over a century, being first investigated as early as 1845 (Herschel, 1845) and 1846 (Brewster, 1846). To the impressionable observer it remains no less fascinating today, as may be evidenced by the beautiful picture of the red fluorescence accompanying illumination of a sapphire crystal by blue light in the Scientific American article, "The Optical Properties of Materials", by Ali Javan (Javan, 1967). In this and the following chapter we will concern ourselves with the theoretical description of resonance fluorescence within the context of the open quantum

Markoffian system. Particular attention is paid to the evaluation of field correlation functions using the results of chapter II.

The earliest description of the atomic scattering mechanism was provided by the Lorentzian or electron oscillator model for atom-field interaction (Lorentz, 1952), and this remains adequate even today for a wide variety of applications. The Lorentzian concept is simple, and draws solely on classical mechanics and electrodynamics. With the atomic scattering centre idealised as an harmonic electron oscillator, incident radiation induces in this system forced vibrations at a frequency ω, generally differing from the frequency ω_0 for free oscillation, Fig. (6.1.1a). Remission of radiation then accompanies the resulting acceleration of the electron while there is a simultaneous damping of vibration through an interaction back on the harmonic oscillator. The dispersion formula is in this instance familiar, with, for the total cross section ϕ for scattered light

$$\phi \propto \frac{\omega^4}{(\omega^2-\omega_0^2)^2 + \omega^2\gamma^2} \tag{6.1.1}$$

Here γ is a damping constant characterising the interaction between oscillator and emitted light. The case of resonance fluorescence is then contained within this result for $\omega \sim \omega_0$, in which case the scattered intensity becomes very large, and

$$\phi \propto \frac{\omega^2}{(\omega-\omega_0)^2 + (\gamma/2)^2} \tag{6.1.2}$$

The need to move to quantum concepts for the understanding of fluorescence, as throughout optical spectroscopy in general, arises initially, not so much in quantitative considerations, but in qualitative ones. Armed solely with a classical conception we may wonder, for example, at the red fluorescence arising in sapphire crystals as a response to illumination by blue light. Of course,

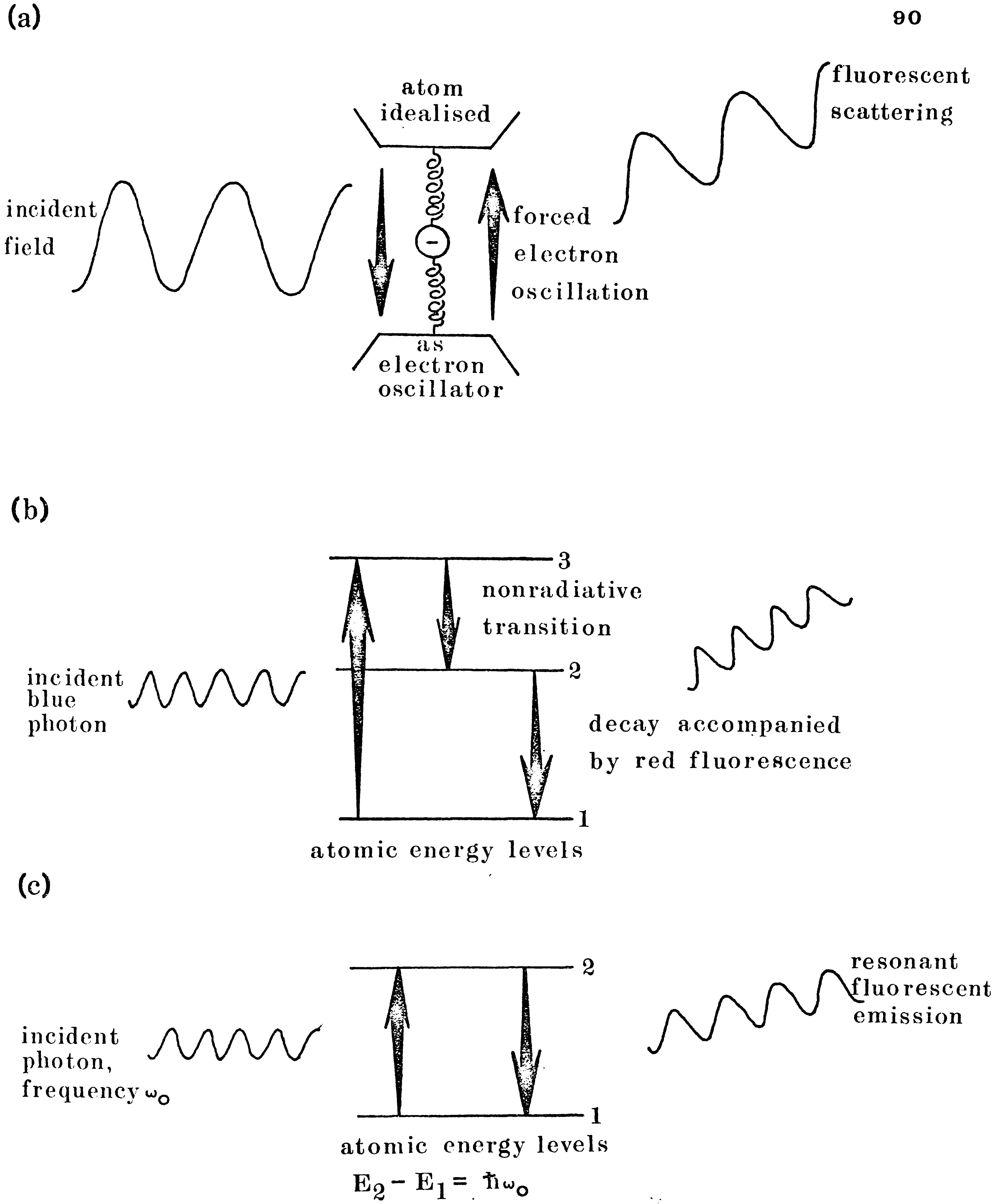


Fig (6·1·1) Fluorescent scattering in; (a) Lorentz's electron oscillator model, (b) the three level chromium atom, (c) a resonant two level atom.

within the quantum theory this is merely indication of the mediation of some intermediate atomic level as illustrated in Fig. (6.1.1b). The early development of the quantum treatment for resonance fluorescence is contained in the work of Weisskopf and Wigner (1930), Weisskopf (1933), and Heitler (1954). In Heitler's book, "The Quantum Theory of Radiation", we find a perturbation calculation substantiating the predictions provided by the oscillator theory, however including conceptions compatible with the quantum view of atomic structure. For irradiation by a continuous spectrum the probability of photon emission with a frequency ω emerges with a lineshape corresponding to spontaneous emission

$$p(\omega) \quad \propto \quad \frac{\gamma/2}{(\omega-\omega_0)^2 + (\gamma/2)^2} \tag{6.1.3}$$

Further, the probability for absorption reveals the shape for the atomic absorption line. Thus, with regard to lineshape, one may take the elementary view of successive, and independent, absorption and emission between two atomic levels Fig. (6.1.1c). However, from the overall perspective, this must be recognised as somewhat naive, since absorbed and emitted photons must remain correlated, a requisite of energy conservation. Thus, if illumination by a sharp line is considered the independence of absorption and emission can no way be upheld. The scattered spectrum then retains the shape of the incident field and incident and scattered radiations are coherent. In this is therefore the view of scattering via a single quantum process, the atom appearing as mediator between incident and scattered fields.

For many years the calculations of Weisskopf, Wigner and Heitler stood as adequate description for resonance fluorescence. However, due to their perturbative basis they cannot lay claim to full quantum electrodynamic description of the phenomena. From Heitler's calculations we are left with a view of coherent scattering. For this it is clear that regular interruption of the atomic scattering process, for example, by inelastic collision, will destroy this coherence. Hence, the familiar feature of collision broadening. Armed with this observation, we must then recognise within resonance fluorescence the potential for destruction of coherence by the incident beam itself. Indeed, the atom may not act as a free scattering centre for successive photons, but is all the time interacting with the incident field. Throughout there exists therefore a finite probability for photon exchange with the irradiating field accompanied by a reorientation of atomic state. It should be expected then that the quantum fluctuations arising in these coupled dynamics be reflected in the statistics and spectrum of the scattered field. In this arises an area of fluorescent phenomena denoted A.C., or dynamical, Stark effect where a three-peaked scattered spectrum is characteristic (Mollow, 1969, 1970; Oliver *et al.*, 1971; Stroud, 1971; Carmichael and Walls, 1976b). Of course, the significance of incoherent effects must be viewed in relation to the intensity of incident radiation, this determining the distance which the atomic scatterer is taken from its ground state. Equivalently, the criterion is the degree of deviation by the atomic oscillator from linear classical behaviour. With the light intensities available in modern laser technology the régime of nonlinear atomic dynamics is readily accessible.

In recent years considerable attention has been paid to the re-evaluation of the quantum electrodynamic theory of resonance fluorescence. Aside from some general interest (Apanasevich, 1964; Bergmann, 1967; Newstein, 1968; Morozov, 1969; Mollow, 1969, 1970), the stimulus of these studies has largely been provided by the proposition, of Stroud and Jaynes (1970), that here there may be a test for quantum electrodynamics, as against the neoclassical theory for atom-field interactions (Jaynes and Cummings, 1963, Crisp and Jaynes, 1969; Stroud and Jaynes, 1970). In the wake of their suggestion a stream of theoretical publications has flowed (Chang and Stehle, 1971; Gush and Gush, 1972; Smithers and Freedhoff, 1974, 1975; Hassan and Bullough, 1975; Mollow, 1975a, b; Carmichael and Walls, 1975, 1976a, b; Swain, 1975; Cohen-Tannoudji, 1975, 1976), both quantum statistical and quantum dynamical in standpoint. Interest throughout this 'dialogue' has been sustained by the diversity of predictions for spectral detail appearing at the outset. Of particular importance amongst spectral characteristics is the long-time spectral behaviour since it is in this that the neoclassical and Q.E.D. calculations are expected to disagree. Specific attention has also been paid to the differing views of Stroud (1971) on the one hand, and Mollow (1970) on the other, as regards linewidths and relative peak heights (Carmichael and Walls, 1975). The consensus that has emerged finds substantiation in its qualitative features in the recent results of various experimental groups (Schuda *et al.*, 1974; Walther, 1975, Wu *et al.*, 1975; Hartig *et al.*, 1976).

Part III of this thesis is devoted to the results of three publications on the dynamical Stark effect presented to "Journal of Physics B" (Carmichael and Walls, 1975, 1976a, b). The perspective taken is that of an open quantum Markoffian system, as formulated in section 6.5, and therefore may be identified as a quantum-statistical or stochastic treatment. The tutorial advantage of this over the quantum dynamical treatments of others (Smithers and Freedhoff, 1974, 1975; Mollow, 1975a, b) is considered full justification in itself for our approach. However, there is, in particular, a further bonus in an easy route to the second order field correlation function. This has only recently been calculated by others (Cohen-Tannoudji, 1976;

Kimble and Mandel, 1976) and provides novel possibilities for the experimental investigation of spectral features, together with further potential for direct comparison of Q.E.D. with neoclassical results.

The remaining three sections of the present chapter are devoted to introduction, and formulation of resonance fluorescence and the dynamical Stark effect within the context of the open quantum Markoffian system. After outlining our model in section 6.2, the field operator for scattered light is, in section 6.3, expressed as a function solely of atomic operators. The master equation which is to form the basis for the study of atomic dynamics is then derived in section 6.4 from the general formulation of section 1.2. In chapter VII we turn to explicit calculation of those features characterising fluorescence within the model of the open quantum Markoffian system. An overall view is presented with general formulations for atomic matrix elements and field correlation functions in section 7.1. Then, in section 7.2. specific solution of atomic dynamics is tackled which yields directly the semiclassical results for the scattered spectrum. For comparison we find the first-order field correlation function and the full quantum-mechanical result for the scattered spectrum, including the region of transient atomic dynamics in section 7.3. A one-photon approximation to these results is investigated in section 7.4 which directly reveals the failings of early quantum-dynamical calculations by Stroud (1971). We then conclude this chapter in section 7.5 with the evaluation of the second-order field correlation function and the revelation of the potential this function holds as a basis for measurements of spectral characteristics via photon-correlation techniques.

6.2 Formulation as an Open Quantum Markoffian system

Our approach is to view the whole fluorescent phenomena within the framework of the open quantum Markoffian system, as developed in Part I of this thesis. Diagrammatically, the physical system to which our calculations pertain is presented in Fig. (6.2.1); substantially an outline of the experimental situation of Schuda *et al.* (1974). Our quantum-mechanical model for approaching this system theoretically is given schematically by Fig. (6.2.2). As a polarised plane wave, a single, highly populated, mode of the radiation field irradiates an atomic dipole transition at resonance. Together these constitute a familiar coupled system, denoted S, such as is found in the single mode laser. With these origins, fluorescent light then appears as scattering to other modes of the vacuum and may be regarded as energy radiated from the coupled system to a reservoir R at zero temperature. The dynamics of the whole is then governed by a Hamiltonian H, which, in the language of chapter I, may be written

$$H = H_S + H_R + H_{SR} \tag{6.2.1}$$

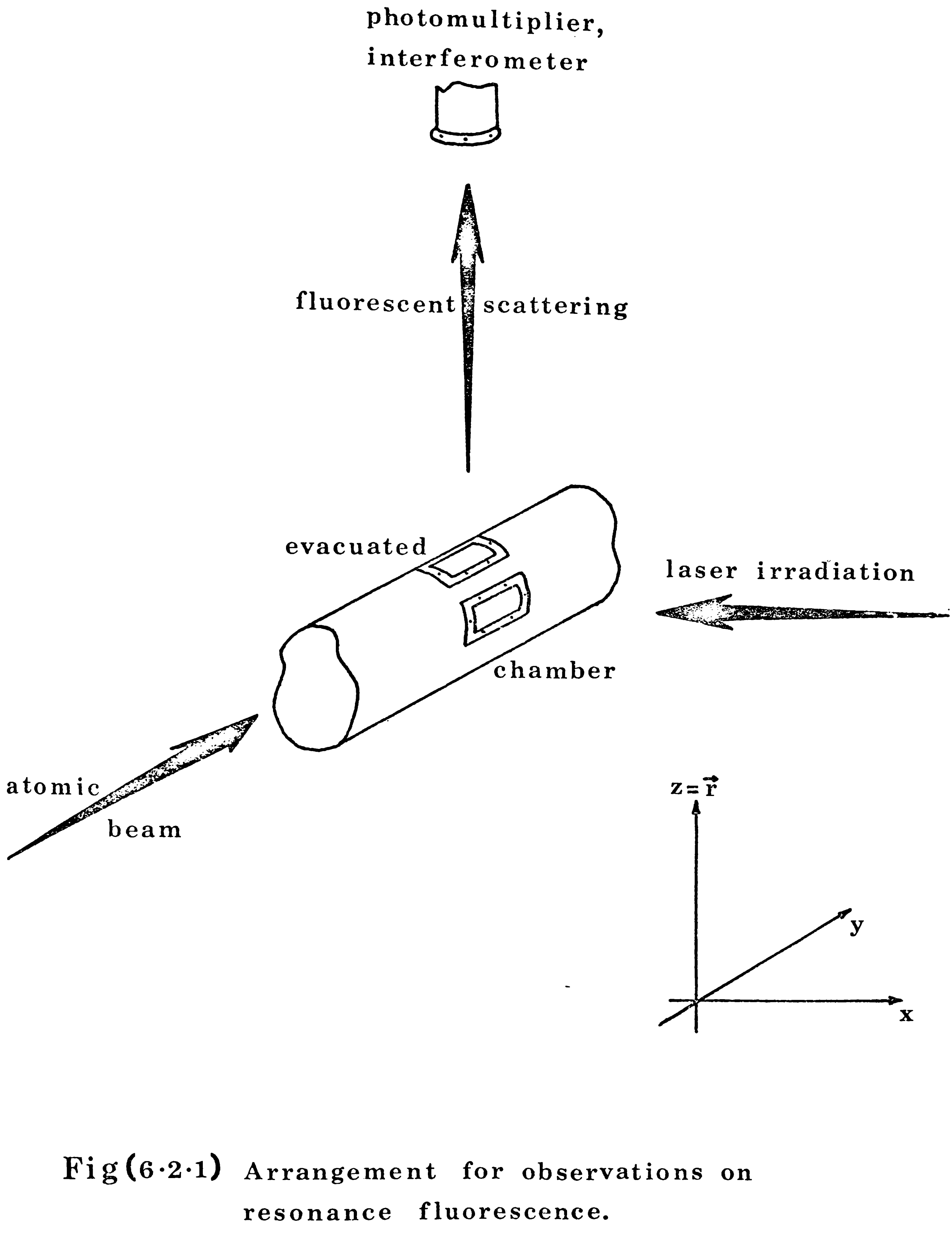
H_S governs dynamics for the resonantly coupled incident field plus atomic dipole, H_R describes the vacuum field, and H_{SR} mediates between incident and scattered fields as an interaction term.

With this designation it is appropriate to view the field operator $\vec{E}(\vec{r},t)$ as a composite of incident field $\vec{E}_I(\vec{r},t)$ and scattered field $\vec{E}_S(\vec{r},t)$:

$$\vec{E}(\vec{r},t) = \vec{E}_I(\vec{r},t) + \vec{E}_S(\vec{r},t) \tag{6.2.2}$$

where

$$\vec{E}_I(\vec{r},t) = i(\hbar\omega_o/2\varepsilon_o V)^{1/2}\hat{e}_o(a(t)e^{i\vec{k}_o\cdot\vec{r}} - a^\dagger(t)e^{-i\vec{k}_o\cdot\vec{r}}) \tag{6.2.3}$$



Fig(6·2·1) Arrangement for observations on resonance fluorescence.

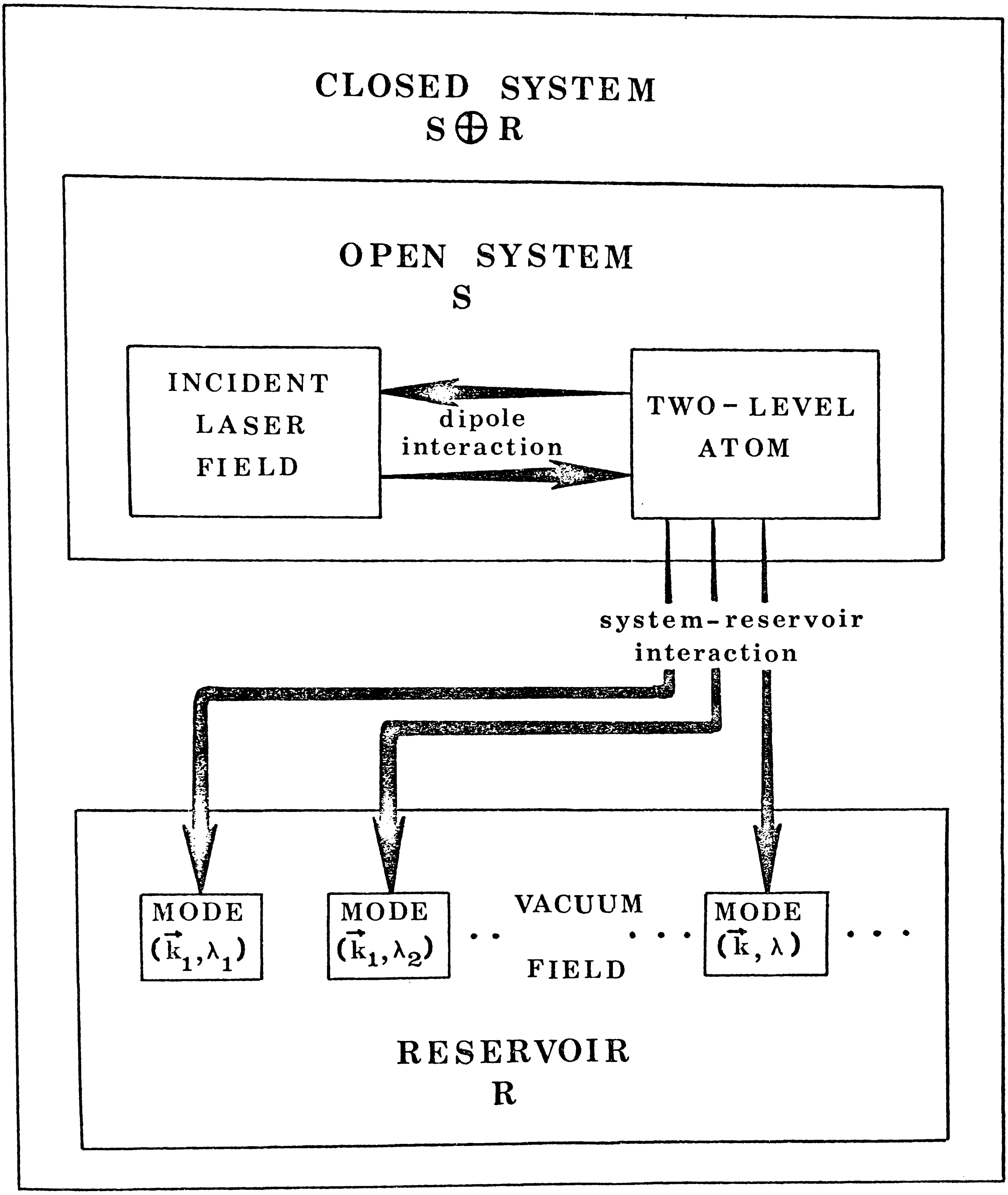


Fig (6·2·2) Schematic presentation of theoretical model for resonance fluorescence.

$$\vec{E}_s(\vec{r},t) = i\sum_{\vec{k},\lambda}(\hbar\omega_k/2\varepsilon_0 V)^{1/2}\hat{e}_{\vec{k},\lambda}(b_{\vec{k},\lambda}(t)e^{i\vec{k}\cdot\vec{r}} - b^{\dagger}_{\vec{k},\lambda}(t)e^{-i\vec{k}\cdot\vec{r}}) \tag{6.2.4}$$

Here $a^{\dagger}$ and a create and annihilate photons in the incident beam with frequency ω_0, wavevector $\vec{k}_0$ and polarisation $\hat{e}_0$; $b^{\dagger}_{\vec{k},\lambda}$ and $b_{\vec{k},\lambda}$ fulfil a similar purpose for the vacuum mode of frequency ω_k, wavevector $\vec{k}$ and polarisation $\hat{e}_{\vec{k},\lambda}$; ε_0 and V have their usual meanings as vacuum permittivity and normalisation volume. Then, writing σ_z, σ_+ and σ_- as the usual Pauli pseudo spin operators, the Hamiltonian (6.2.1), taken in the rotating-wave and dipole approximations, has

$$H_S = \hbar\omega_0 a^{\dagger}a + (1/2)\hbar\omega_0\sigma_z + \hbar(\kappa_0^* a^{\dagger}\sigma_- + \kappa_0 a\sigma_+) \tag{6.2.5}$$

$$H_R = \sum_{\vec{k},\lambda}\hbar\omega_k b^{\dagger}_{\vec{k},\lambda}b_{\vec{k},\lambda} \tag{6.2.6}$$

$$H_{SR} = \sum_{\vec{k},\lambda}\hbar(\kappa^*_{\vec{k},\lambda}b^{\dagger}_{\vec{k},\lambda}\sigma_- + \kappa_{\vec{k},\lambda}b_{\vec{k},\lambda}\sigma_+) \tag{6.2.7}$$

Coupling constants κ_0 and $\kappa_{\vec{k},\lambda}$ for dipole interaction are given, in their usual form, by

$$\kappa_0 = -i\omega_0 e^{-i\vec{k}_0\cdot\vec{r}_a}(1/2\hbar\omega_0\varepsilon_0 V)^{1/2}\hat{e}_0\cdot\vec{\mu} \tag{6.2.8}$$

$$\kappa_{\vec{k},\lambda} = -i\omega_0 e^{-i\vec{k}\cdot\vec{r}_a}(1/2\hbar\omega_k\varepsilon_0 V)^{1/2}\hat{e}_{\vec{k},\lambda}\cdot\vec{\mu} \tag{6.2.9}$$

$\vec{\mu}$ being the atomic dipole matrix element and $\vec{r}_a$ the position vector for location of the atomic nucleus. Throughout the above all operators obey standard commutation relations, the nonvanishing commutators reading

$$\begin{aligned} [a,a^{\dagger}] &= 1 \\ [b_{\vec{k},\lambda}, b^{\dagger}_{\vec{k},\lambda}] &= 1 \\ [\sigma_+,\sigma_-] &= \sigma_z \\ [\sigma_{\pm},\sigma_z] &= \mp 2\sigma_{\pm} \end{aligned} \tag{6.2.10}$$

6.3 Electric Field operator

In contrast to the usual circumstance in which application of master equation techniques is to be found, here, superficially, there appears to be an obstacle. While it is routine to obtain a master equation corresponding to the Hamiltonian (6.2.5) to (6.2.7), our interest is with the scattered field, and thus, in effect, the reservoir R, rather than the coupled atomic system. Nevertheless, from this section it will become clear that atomic dynamics are quite adequate for our purposes. By virtue of the close relationship between the scattered field operator and atomic pseudo-spin operators, field correlations may be obtained via the calculation of atomic correlations at retarded times.

It is usual to divide the electric field into positive and negative components, and for $\vec{E}_S(\vec{r},t)$ we write

$$\vec{E}_s(\vec{r},t) = \vec{E}_s^{(+)}(\vec{r},t) + \vec{E}_s^{(-)}(\vec{r},t) \tag{6.3.1}$$

with

$$\vec{E}_s^{(-)}(\vec{r},t) = \vec{E}_s^{(+)}(\vec{r},t)^{\dagger} \tag{6.3.2}$$

$$\vec{E}_s^{(+)}(\vec{r},t) = i\sum_{\vec{k},\lambda} (\hbar\omega_k/2\varepsilon_0 V)^{1/2}\, \hat{e}_{\vec{k},\lambda}\, b_{\vec{k},\lambda}(t)\, e^{i\vec{k}\cdot\vec{r}} \tag{6.3.3}$$

Now, for a freely propogating field each mode obeys a Heisenberg equation of motion,

$$\frac{d b_{\vec{k},\lambda}}{dt} = -i\omega_k\, b_{\vec{k},\lambda} \tag{6.3.4}$$

which, in the presence of atomic scattering is modified to include a source term attributable to the interaction Hamiltonian (6.2.7):

$$\begin{aligned}\frac{d b_{\vec{k},\lambda}}{dt} &= -i\omega_k b_{\vec{k},\lambda} - (i/\hbar)[b_{\vec{k},\lambda}, H_{SR}] \\ &= -i\omega_k b_{\vec{k},\lambda} - i\,\kappa^{*}_{\vec{k},\lambda}\,\sigma_- \end{aligned} \tag{6.3.5}$$

Then, viewing similarly the pseudo spin operator σ_-, for a free atom

$$\frac{d\sigma_-}{dt} = -i\omega_0 \sigma_- \tag{6.3.6}$$

which also, neglecting coupling to the incident field*, finds modification in the interaction with the vacuum field:

$$\frac{d\sigma_-}{dt} = -i\omega_0 \sigma_- - (i/\hbar)[\sigma_-, H_{SR}] \tag{6.3.7}$$

Now, in anticipation of manipulations further on, it is convenient to isolate the free dynamics, writing

$$\begin{aligned} b_{\vec{k},\lambda}(t) &= e^{-i\omega_k t}\,\hat{b}_{\vec{k},\lambda}(t) \\ \sigma_-(t) &= e^{-i\omega_0 t}\,\hat{\sigma}_-(t) \end{aligned} \tag{6.3.8}$$

whence $\hat{b}_{\vec{k},\lambda}(t)$ and $\hat{\sigma}_-(t)$ are operators slowly developing in time. From (6.3.5) we have

$$\frac{d\hat{b}_{\vec{k},\lambda}}{dt} = -i K^*_{\vec{k},\lambda}\, e^{i(\omega_k-\omega_0)t}\,\hat{\sigma}_-(t) \tag{6.3.9}$$

and the scattering component in mode $(\vec{k},\lambda)$ follows, in terms of atomic source operators, by formal integration:

$$b_{\vec{k},\lambda}(t) = b_{\vec{k},\lambda}(0)e^{-i\omega_k t} - iK^*_{\vec{k},\lambda}e^{-i\omega_0 t}\int_0^t dt'\, e^{i(\omega_k-\omega_0)(t'-t)}\,\hat{\sigma}_-(t') \tag{6.3.10}$$

The full field operator now has

$$\begin{aligned} \vec{E}_s^{(+)}(\vec{r},t) = \vec{\mathcal{E}}_s^{(+)}(\vec{r},t) &+ \sum_{\vec{k},\lambda} (\hbar\omega_k/2\varepsilon_0 V)^{1/2}\,\hat{e}_{\vec{k},\lambda} K^*_{\vec{k},\lambda}\, e^{i\vec{k}\cdot\vec{r}} \\ &\cdot e^{-i\omega_0 t}\int_0^t dt'\, e^{i(\omega_k-\omega_0)(t'-t)}\,\hat{\sigma}_-(t') \end{aligned} \tag{6.3.11}$$

---

*The calculations of the present section may be carried through allowing for incident beam coupling in the free dynamics. However, any differences are negligible other than for extremely intense fields.

where $\vec{\varepsilon}_S^{(+)}(\vec{r},t)$ is just the freely propagating field with

$$\vec{\mathcal{E}}_s^{(+)}(\vec{r},t) = i\sum_{\vec{k},\lambda}(\hbar\omega_k/2\mathcal{E}_o V)^{1/2}\hat{e}_{\vec{k},\lambda}\, b_{\vec{k},\lambda}(0)\, e^{-i(\omega_k t-\vec{k}\cdot\vec{r})} \tag{6.3.12}$$

The problem of mode summation may now be tackled with recourse to the density of modes for a cavity of volume V. If $\rho_\omega\, d\omega$ is the number of modes in the frequency interval ω to $\omega + d\omega$, the familiar result is given by

$$\rho_\omega\, d\omega = (\omega^2 V/8\pi^3 c^3)\, d\omega \tag{6.3.13}$$

With this we take care of the magnitude $k = \frac{\omega}{c}$ of momentum, while in terms of the unit vector $\frac{\vec{k}}{k}$ the mode density is isotropic. Adopting, then, the geometry of Fig. (6.3.1) we may move in (6.3.11) to an integration overall $\vec{k}$, retaining the summation over λ. We write

$$\begin{aligned}\vec{E}_s^{(+)}(\vec{r},t) = \vec{\mathcal{E}}_s^{(+)}(\vec{r},t) + i(\omega_o/16\pi^3c^3\mathcal{E}_o)\, e^{-i\omega_o t}\sum_\lambda\int_0^{2\pi}d\phi\int_0^{\pi}d\theta\,\sin\theta \\ \int_0^\infty d\omega\,\omega^2\,\hat{e}_{\vec{k},\lambda}(\hat{e}_{\vec{k},\lambda}\cdot\vec{\mu})\, e^{i\omega\frac{r}{c}\cos\theta} \\ \int_0^t dt'\, e^{i(\omega-\omega_o)(t'-t)}\,\hat{\sigma}_-(t')\end{aligned} \tag{6.3.14}$$

where, for $\kappa_{\vec{k},\lambda}$ we have substituted from (6.2.9), taking note that the atomic nucleus is located at the origin. To make explicit the summation over λ we must choose two perpendicular polarisation vectors $\hat{e}_{\vec{k},\lambda}$ for each wavevector $\vec{k}$. It is expedient then to take the first perpendicular to $\vec{\mu}$ ensuring the dot product $\hat{e}_{\vec{k},\lambda_1}\cdot\vec{\mu}$ vanishes. In this $\hat{e}_{\vec{k},\lambda_2}$ is also defined and with the geometry of Fig. (6.3.2) it readily follows that

$$\hat{e}_{\vec{k},\lambda_2}(\hat{e}_{\vec{k},\lambda_2}\cdot\vec{\mu}) = -\left(\vec{\mu}\times\frac{\vec{k}}{k}\right)\times\frac{\vec{k}}{k} \tag{6.3.15}$$

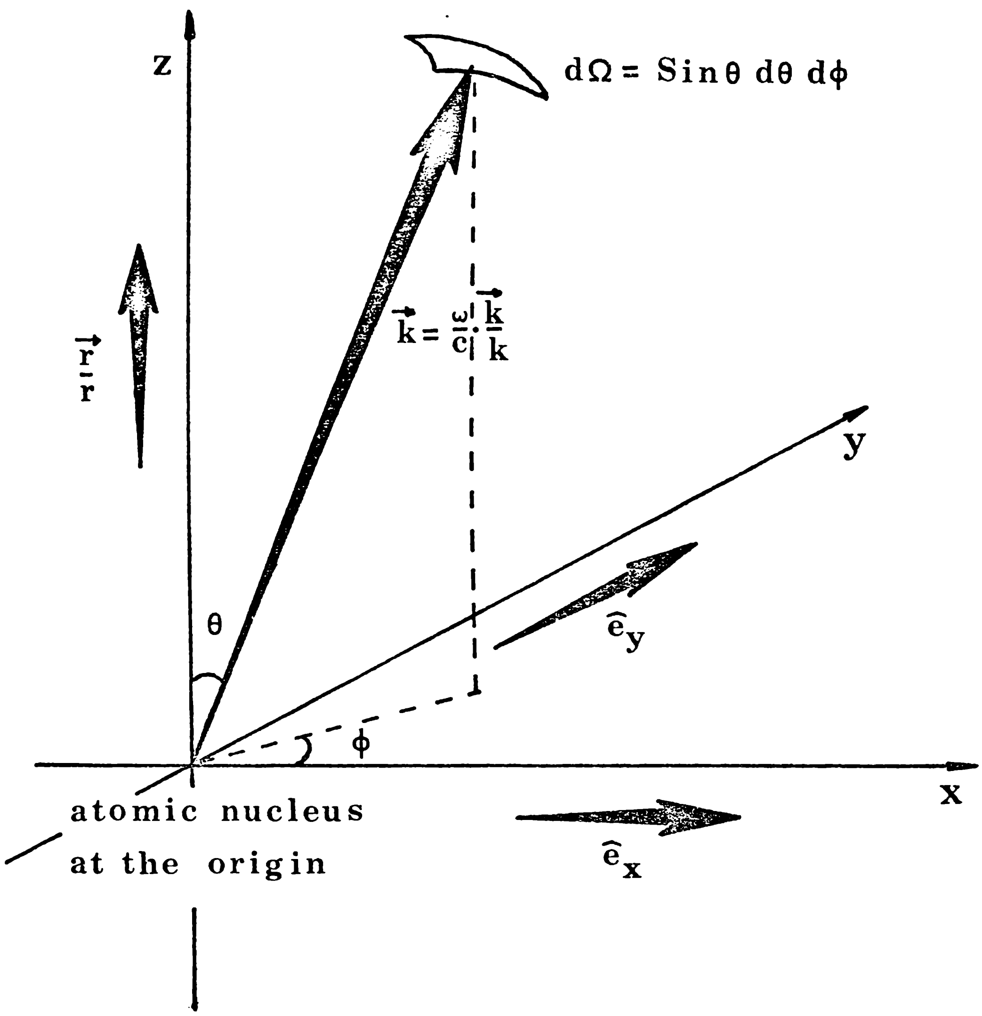


Fig (6·3·1) Scattering geometry for modal summation.

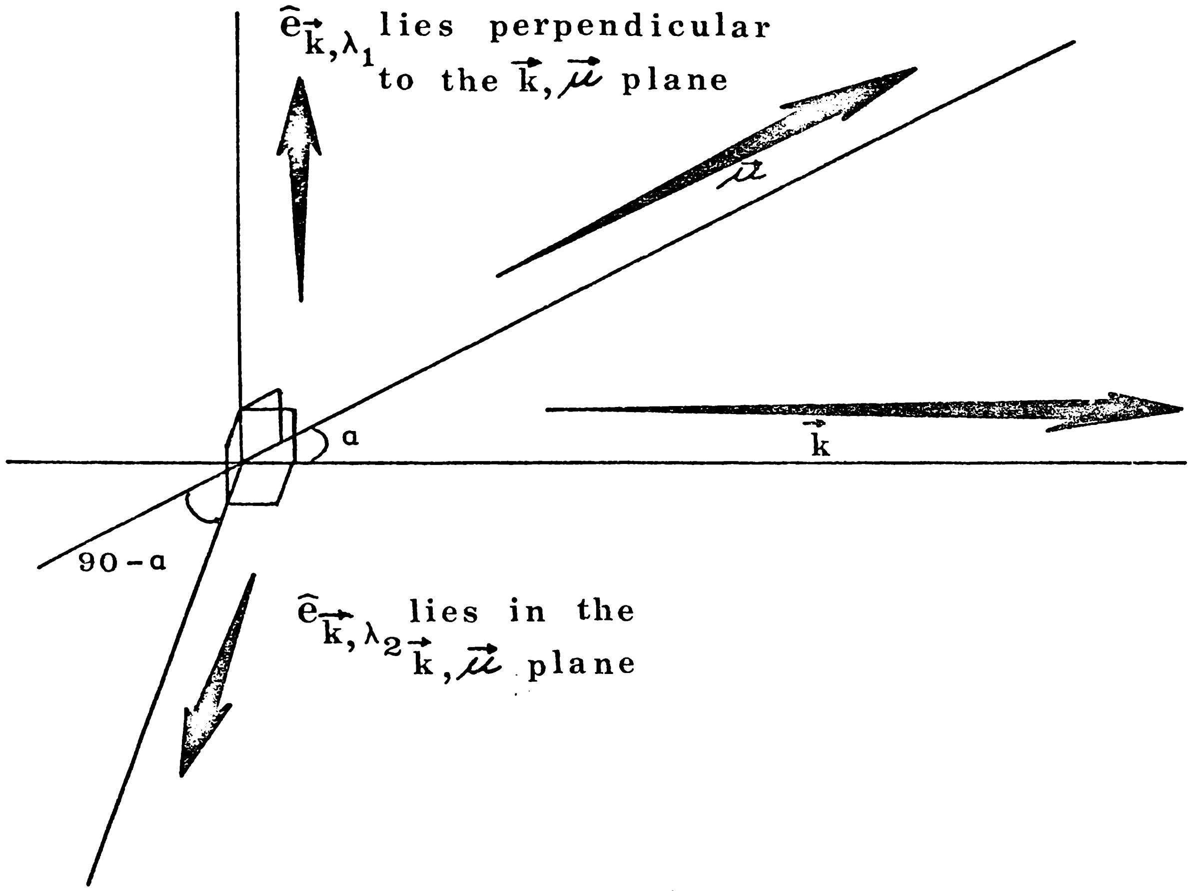


Fig (6·3·2) Geometry of polarisation vectors for modal summation.

Thus mode summation is reduced to the evaluation of the expression

$$\vec{E}_s^{(+)}(\vec{r},t) = \vec{\mathcal{E}}_s^{(+)}(\vec{r},t) - i(\omega_0/16\pi^3 c^3 \varepsilon_0) e^{-i\omega_0 t} \int_0^{2\pi} d\phi \int_0^{\pi} d\theta \operatorname{Sin}\theta$$
$$\int_0^{\infty} d\omega\, \omega^2 \left(\vec{\mu} \times \frac{\vec{k}}{k}\right) \times \frac{\vec{k}}{k}\, e^{i\omega \frac{r}{c} \operatorname{Cos}\theta}$$
$$\int dt' e^{i(\omega_k - \omega_0)(t'-t)} \hat{\sigma}_-(t') \tag{6.3.16}$$

The further reduction of this expression follows readily from a re-expression of $\vec{k}$ as a component vector with unit vectors $\hat{e}_x$, $\hat{e}_y$ and $\frac{\vec{r}}{r}$ along cartesian axes. With

$$\frac{\vec{k}}{k} = \operatorname{Sin}\theta \operatorname{Cos}\phi\, \hat{e}_x + \operatorname{Sin}\theta \operatorname{Sin}\phi\, \hat{e}_y + \operatorname{Cos}\theta \frac{\vec{r}}{r} \tag{6.3.17}$$

we find

$$\left(\vec{\mu} \times \frac{\vec{k}}{k}\right) \times \frac{\vec{k}}{k} = \operatorname{Sin}^2\theta \operatorname{Cos}^2\phi (\vec{\mu} \times \hat{e}_x) \times \hat{e}_x$$
$$+ \operatorname{Sin}^2\theta \operatorname{Sin}^2\phi (\vec{\mu} \times \hat{e}_y) \times \hat{e}_y + \operatorname{Cos}^2\theta \left(\vec{\mu} \times \frac{\vec{r}}{r}\right) \times \frac{\vec{r}}{r}$$
$$+ \operatorname{Sin}^2\theta \operatorname{Cos}\phi \operatorname{Sin}\phi (\vec{\mu} \times \hat{e}_x) \times \hat{e}_y + \operatorname{Cos}\theta \operatorname{Sin}\theta \operatorname{Cos}\phi (\vec{\mu} \times \hat{e}_x) \times \frac{\vec{r}}{r}$$
$$+ \operatorname{Sin}\theta \operatorname{Cos}\theta \operatorname{Sin}\phi (\vec{\mu} \times \hat{e}_y) \times \frac{\vec{r}}{r} + \operatorname{Sin}^2\theta \operatorname{Cos}\phi \operatorname{Sin}\phi (\vec{\mu} \times \hat{e}_y) \times \hat{e}_x$$
$$+ \operatorname{Cos}\theta \operatorname{Sin}\theta \operatorname{Cos}\phi \left(\vec{\mu} \times \frac{\vec{r}}{r}\right) \times \hat{e}_x + \operatorname{Cos}\theta \operatorname{Sin}\theta \operatorname{Sin}\phi \left(\vec{\mu} \times \frac{\vec{r}}{r}\right) \times \hat{e}_y \tag{6.3.18}$$

from which we require angular integrals:

$$\int_0^{2\pi} d\phi \operatorname{Cos}\phi = \int_0^{2\pi} d\phi \operatorname{Sin}\phi = \int_0^{2\pi} d\phi \operatorname{Cos}\phi \operatorname{Sin}\phi = 0$$
$$\int_0^{2\pi} d\phi = 2\pi, \quad \int_0^{2\pi} d\phi \operatorname{Cos}^2\phi = \int_0^{2\pi} d\phi \operatorname{Sin}^2\phi = \pi \tag{6.3.19}$$

and

$$\int_0^{\pi} d\theta \operatorname{Sin}\theta \operatorname{Cos}^2\theta\, e^{i\omega \frac{r}{c} \operatorname{Cos}\theta} = -i(c/\omega r)\left(e^{i\omega \frac{r}{c}} - e^{-i\omega \frac{r}{c}}\right)$$
$$\int_0^{\pi} d\theta \operatorname{Sin}\theta \operatorname{Sin}^2\theta\, e^{i\omega \frac{r}{c} \operatorname{Cos}\theta} = 0 \tag{6.3.20}$$

where, in these we have retained only first order terms in $(c/\omega r)$. Combining then (6.3.16) with (6.3.18), (6.3.19) and (6.3.20), we have

$$\vec{E}_s^{(+)}(\vec{r},t) = \vec{\mathcal{E}}_s^{(+)}(\vec{r},t) - (\omega_0/8\varepsilon_0\pi^2c^2r)(\vec{\mu}\times\frac{\vec{r}}{r})\times\frac{\vec{r}}{r}\int_0^\infty d\omega\,\omega \int_0^t dt'\, e^{i(\omega-\omega_0)[t'-(t-\frac{r}{c})]}\hat{\sigma}_-(t')\,e^{-i\omega_0(t-\frac{r}{c})} - e^{i(\omega-\omega_0)[t'-(t+\frac{r}{c})]}\hat{\sigma}_-(t')\,e^{-i\omega_0(t+\frac{r}{c})} \tag{6.3.21}$$

Now in considering the temporal integrations, while $\hat{\sigma}_-(t')$ varies slowly on some suitable time scale between o and t, $e^{i(\omega-\omega_0)[t'-(t+\frac{r}{c})]}$ is highly oscillatory throughout this range. $e^{i(\omega-\omega_0)[t'-(t-\frac{r}{c})]}$ exhibits similar oscillatory behaviour except for $t' \sim t - \frac{r}{c}$. With a view to this it is a standard procedure to take

$$\int_0^t dt'\, e^{i(\omega-\omega_0)[t'-(t+\frac{r}{c})]}\hat{\sigma}_-(t')\,e^{-i\omega_0(t+\frac{r}{c})} = 0 \tag{6.3.22}$$

and

$$\begin{aligned}\int_0^t dt'\, e^{i(\omega-\omega_0)[t'-(t-\frac{r}{c})]}\hat{\sigma}_-(t')\,e^{-i\omega_0(t-\frac{r}{c})} &= \sigma_-(t-\tfrac{r}{c})\int_0^t dt'\, e^{i(\omega-\omega_0)[t'-(t-\frac{r}{c})]} \\ &= \sigma_-(t-\tfrac{r}{c})2\pi\,\delta(\omega-\omega_0)\end{aligned} \tag{6.3.23}$$

where in this second result we recognise

$$\sigma_-(t-\tfrac{r}{c}) = \hat{\sigma}_-(t-\tfrac{r}{c})\,e^{-i\omega_0(t-\frac{r}{c})} \tag{6.3.24}$$

Using (6.3.22) and (6.3.23) in (6.3.21) we then find as expression for $\vec{E}_S^{(+)}(\vec{r},t)$ in terms of retarded atomic operators the form

$$\vec{E}_s^{(+)}(\vec{r},t) = \vec{\mathcal{E}}_s^{(+)}(\vec{r},t) - (\omega_0^2/4\pi\varepsilon_0c^2r)(\vec{\mu}\times\frac{\vec{r}}{r})\times\frac{\vec{r}}{r}\,\sigma_-(t-\tfrac{r}{c}) \tag{6.3.25}$$

As we should expect, this is just the retarded field generated by a point dipole with the classical dipole moment replaced by the atomic lowering operator σ_- (Landau and Lifshitz, 1961). This general result is familiar in the literature (Agarwal, 1974; Ackerhalt and Eberly, 1974; Kimble and Mandel, 1975a, b; Saunders *et al.*, 1975).

6.4 Master equation for Atomic Dynamics

We have already outlined the formulation of the fluorescence problem within the scope of the open quantum Markoffian system. Then in the previous section we have seen how field operators may be related directly to atomic dynamics. It remains now to obtain explicit expression for the operator master equation governing these dynamics.

For the arbitrary open system S coupled to a thermal reservoir R, derivation of the master equation for the reduced density operator ρ from the microscopic Hamiltonian for $S \oplus R$ has been tackled in its formal aspects in section 1.2. We have the specification

$$\frac{d\rho(t)}{dt} = \mathcal{L}\,\rho(t) \tag{6.4.1}$$

where for the generalised Liouville operator $\mathcal{L}$,

$$\mathcal{L} = L_S + \mathcal{L}_I \tag{6.4.2}$$

the irreversible component $\mathcal{L}_I$ adopting the form

$$\mathcal{L}_I = e^{-iL_St}\int_0^t tr_R(\hat{L}_{SR}(t)\hat{L}_{SR}(t-\tau)R_{TH})\,e^{iL_St} \tag{6.4.3}$$

Here the Liouvillian L takes its usual definition,

$$L = L_S + L_R + L_{SR} = (1/\hbar)[H_S+H_R+H_{SR},\ ] = (1/\hbar)[H,\ ] \tag{6.4.4}$$

and for system-reservoir coupling in the interaction picture

$$\hat{L}_{SR} = e^{-i(L_S+L_R)t}\,L_{SR}\,e^{i(L_S+L_R)t} \tag{6.4.5}$$

Now, in the Hamiltonian of (6.2.5) to (6.2.7) we have a case exhibiting internal coupling, the consequences of which, in relation to the irreversible term (6.4.3), have been studied for both the damped harmonic and atomic oscillators (Carmichael and Walls, 1973, 1974). These are significant only at extremely high intensities*, and consequently, here we will neglect this coupling in relation to the irreversible dynamics. For (6.4.3) and (6.4.5) we take

$$\mathcal{L}_I \rho = e^{-iL_A t} \int_0^t d\tau \, \mathrm{tr}_R(\hat{L}_{SR}(t)\hat{L}_{SR}(t-\tau)R_{TH})\hat{\rho}(t) \tag{6.4.6}$$

and

$$\hat{L}_{SR}(t) = e^{-i(L_A+L_R)t} L_{SR}\, e^{i(L_A+L_R)t} \tag{6.4.7}$$

where L_A governs free atomic dynamics alone;

$$L_A = (1/\hbar)[H_A,\ ] = (1/\hbar)[(1/2)\hbar\omega_0\sigma_z,\ ] \tag{6.4.8}$$

and $\hat{\rho}(t)$ is the reduced density operator in the interaction picture:

$$\hat{\rho}(t) = e^{-iL_A t}\rho(t) \tag{6.4.9}$$

Explicit evaluation of $\mathcal{L}_I$ may now be made in view of the microscopic Hamiltonian (6.2.5) to (6.2.7). Using the identity

$$e^{-iLt} O_1 O_2 = (e^{-iLt} O_1)(e^{-iLt} O_2) \tag{6.4.10}$$

where O_1 and O_2 are arbitrary operators, it follows from (6.4.7) and the interaction Hamiltonian (6.2.7) that

$$\hat{L}_{SR}(t) = \sum_{\vec{k},\lambda} [\kappa^*_{\vec{k},\lambda} b^\dagger_{\vec{k},\lambda}(t)\,\sigma_-(t) + \kappa_{\vec{k},\lambda} b_{\vec{k},\lambda}(t)\sigma_+(t),\ \ ] \tag{6.4.11}$$

---

*The criterion for high intensity here is $\bar{n}^{1/2}\kappa_0 \sim \omega_0$ far greater than that required for any of the effects considered in the following chapter.

where $b_{\vec{k},\lambda}(t)$ and $\sigma_-(t)$ are merely solutions to the Heisenberg equations (6.3.4) and (6.3.6) for free dynamics

$$b_{\vec{k},\lambda}(t) = e^{-iL_s t} b_{\vec{k},\lambda} = e^{-i\omega_k t} b_{\vec{k},\lambda} \tag{6.4.12}$$

$$\sigma_-(t) = e^{-iL_A t} \sigma_- = e^{-i\omega_0 t} \sigma_- \tag{6.4.13}$$

The integrand of (6.4.6) is then expressible in terms of reservoir correlations as

$$\begin{aligned}
&\hbar^2 \mathrm{tr}_R(\hat{L}_{SR}(t)\hat{L}_{SR}(t-\tau)R_{TH})\hat{\rho}(t) = \\
&\langle \vec{\mathcal{E}}_s^{(-)}(\vec{r}_a,t)\cdot\vec{\mu}\, \vec{\mathcal{E}}_s^{(-)}(\vec{r}_a,t-\tau)\cdot\vec{\mu}\rangle(\sigma_-(t)\sigma_-(t-\tau)\hat{\rho}(t) - \sigma_-(t-\tau)\hat{\rho}(t)\sigma_-(t)) \\
&+\langle \vec{\mathcal{E}}_s^{(-)}(\vec{r}_a,t)\cdot\vec{\mu}\, \vec{\mathcal{E}}_s^{(+)}(\vec{r}_a,t-\tau)\cdot\vec{\mu}\rangle(\sigma_-(t)\sigma_+(t-\tau)\hat{\rho}(t) - \sigma_+(t-\tau)\hat{\rho}(t)\sigma_-(t)) \\
&+\langle \vec{\mathcal{E}}_s^{(+)}(\vec{r}_a,t)\cdot\vec{\mu}\, \vec{\mathcal{E}}_s^{(-)}(\vec{r}_a,t-\tau)\cdot\vec{\mu}\rangle(\sigma_+(t)\sigma_-(t-\tau)\hat{\rho}(t) - \sigma_-(t-\tau)\hat{\rho}(t)\sigma_+(t)) \\
&+\langle \vec{\mathcal{E}}_s^{(+)}(\vec{r}_a,t)\cdot\vec{\mu}\, \vec{\mathcal{E}}_s^{(+)}(\vec{r}_a,t-\tau)\cdot\vec{\mu}\rangle(\sigma_+(t)\sigma_+(t-\tau)\hat{\rho}(t) - \sigma_+(t-\tau)\hat{\rho}(t)\sigma_+(t)) \\
&-\langle \vec{\mathcal{E}}_s^{(-)}(\vec{r}_a,t-\tau)\cdot\vec{\mu}\, \vec{\mathcal{E}}_s^{(-)}(\vec{r}_a,t)\cdot\vec{\mu}\rangle(\sigma_-(t)\hat{\rho}(t)\sigma_-(t-\tau) - \hat{\rho}(t)\sigma_-(t-\tau)\sigma_-(t)) \\
&-\langle \vec{\mathcal{E}}_s^{(+)}(\vec{r}_a,t-\tau)\cdot\vec{\mu}\, \vec{\mathcal{E}}_s^{(-)}(\vec{r}_a,t)\cdot\vec{\mu}\rangle(\sigma_-(t)\hat{\rho}(t)\sigma_+(t-\tau) - \hat{\rho}(t)\sigma_+(t-\tau)\sigma_-(t)) \\
&-\langle \vec{\mathcal{E}}_s^{(-)}(\vec{r}_a,t-\tau)\cdot\vec{\mu}\, \vec{\mathcal{E}}_s^{(+)}(\vec{r}_a,t)\cdot\vec{\mu}\rangle(\sigma_+(t)\hat{\rho}(t)\sigma_-(t-\tau) - \hat{\rho}(t)\sigma_-(t-\tau)\sigma_+(t)) \\
&-\langle \vec{\mathcal{E}}_s^{(+)}(\vec{r}_a,t-\tau)\cdot\vec{\mu}\, \vec{\mathcal{E}}_s^{(+)}(\vec{r}_a,t)\cdot\vec{\mu}\rangle(\sigma_+(t)\hat{\rho}(t)\sigma_-(t-\tau) - \hat{\rho}(t)\sigma_+(t-\tau)\sigma_+(t))
\end{aligned} \tag{6.4.14}$$

where $\vec{\varepsilon}_s(\vec{r},t)$ is the freely propagating field of (6.3.12). These reservoir correlations are readily evaluated with recourse to the canonical form for the reservoir density operator. In view of our choice of a reservoir at zero temperature the complications of thermal statistics are avoided and only two nonvanishing correlations remain:

$$\begin{aligned}
(1/\hbar)^2\langle \vec{\mathcal{E}}_s^{(+)}(\vec{r}_a,t)\cdot\vec{\mu}\, \vec{\mathcal{E}}_s^{(-)}(\vec{r}_a,t-\tau)\cdot\vec{\mu}\rangle &= \sum_{\vec{k},\lambda} |K_{\vec{k},\lambda}|^2\, e^{-i\omega_k\tau} \\
(1/\hbar)^2\langle \vec{\mathcal{E}}_s^{(+)}(\vec{r}_a,t-\tau)\cdot\vec{\mu}\, \vec{\mathcal{E}}_s^{(-)}(\vec{r}_a,t)\cdot\vec{\mu}\rangle &= \sum_{\vec{k},\lambda} |K_{\vec{k},\lambda}|^2\, e^{i\omega_k\tau}
\end{aligned} \tag{6.4.15}$$

where $b_{\vec{k},\lambda}(t)$ and $\sigma_{-}(t)$ are merely solutions to the Heisenberg equations (6.3.4) and (6.3.6) for free dynamics

$$b_{\vec{k},\lambda}(t) = e^{-iL_s t} b_{\vec{k},\lambda} = e^{-i\omega_k t} b_{\vec{k},\lambda} \tag{6.4.12}$$

$$\sigma_{-}(t) = e^{-iL_A t} \sigma_{-} = e^{-i\omega_0 t} \sigma_{-} \tag{6.4.13}$$

The integrand of (6.4.6) is then expressible in terms of reservoir correlations as

$$\begin{aligned}
&\hbar^2 \mathrm{tr}_R(\hat{L}_{SR}(t)\hat{L}_{SR}(t-\tau)R_{TH})\hat{\rho}(t) = \\
&\langle \vec{\mathcal{E}}_s^{(-)}(\vec{r}_a,t)\cdot\vec{\mu}\,\vec{\mathcal{E}}_s^{(-)}(\vec{r}_a,t-\tau)\cdot\vec{\mu}\rangle(\sigma_-(t)\sigma_-(t-\tau)\hat{\rho}(t) - \sigma_-(t-\tau)\hat{\rho}(t)\sigma_-(t)) \\
&+\langle \vec{\mathcal{E}}_s^{(-)}(\vec{r}_a,t)\cdot\vec{\mu}\,\vec{\mathcal{E}}_s^{(+)}(\vec{r}_a,t-\tau)\cdot\vec{\mu}\rangle(\sigma_-(t)\sigma_+(t-\tau)\hat{\rho}(t) - \sigma_+(t-\tau)\hat{\rho}(t)\sigma_-(t)) \\
&+\langle \vec{\mathcal{E}}_s^{(+)}(\vec{r}_a,t)\cdot\vec{\mu}\,\vec{\mathcal{E}}_s^{(-)}(\vec{r}_a,t-\tau)\cdot\vec{\mu}\rangle(\sigma_+(t)\sigma_-(t-\tau)\hat{\rho}(t) - \sigma_-(t-\tau)\hat{\rho}(t)\sigma_+(t)) \\
&+\langle \vec{\mathcal{E}}_s^{(+)}(\vec{r}_a,t)\cdot\vec{\mu}\,\vec{\mathcal{E}}_s^{(+)}(\vec{r}_a,t-\tau)\cdot\vec{\mu}\rangle(\sigma_+(t)\sigma_+(t-\tau)\hat{\rho}(t) - \sigma_+(t-\tau)\hat{\rho}(t)\sigma_+(t)) \\
&-\langle \vec{\mathcal{E}}_s^{(-)}(\vec{r}_a,t-\tau)\cdot\vec{\mu}\,\vec{\mathcal{E}}_s^{(-)}(\vec{r}_a,t)\cdot\vec{\mu}\rangle(\sigma_-(t)\hat{\rho}(t)\sigma_-(t-\tau) - \hat{\rho}(t)\sigma_-(t-\tau)\sigma_-(t)) \\
&-\langle \vec{\mathcal{E}}_s^{(+)}(\vec{r}_a,t-\tau)\cdot\vec{\mu}\,\vec{\mathcal{E}}_s^{(-)}(\vec{r}_a,t)\cdot\vec{\mu}\rangle(\sigma_-(t)\hat{\rho}(t)\sigma_+(t-\tau) - \hat{\rho}(t)\sigma_+(t-\tau)\sigma_-(t)) \\
&-\langle \vec{\mathcal{E}}_s^{(-)}(\vec{r}_a,t-\tau)\cdot\vec{\mu}\,\vec{\mathcal{E}}_s^{(+)}(\vec{r}_a,t)\cdot\vec{\mu}\rangle(\sigma_+(t)\hat{\rho}(t)\sigma_-(t-\tau) - \hat{\rho}(t)\sigma_-(t-\tau)\sigma_+(t)) \\
&-\langle \vec{\mathcal{E}}_s^{(+)}(\vec{r}_a,t-\tau)\cdot\vec{\mu}\,\vec{\mathcal{E}}_s^{(+)}(\vec{r}_a,t)\cdot\vec{\mu}\rangle(\sigma_+(t)\hat{\rho}(t)\sigma_-(t-\tau) - \hat{\rho}(t)\sigma_+(t-\tau)\sigma_+(t))
\end{aligned} \tag{6.4.14}$$

where $\vec{\mathcal{E}}_s(\vec{r},t)$ is the freely propagating field of (6.3.12). These reservoir correlations are readily evaluated with recourse to the canonical form for the reservoir density operator. In view of our choice of a reservoir at zero temperature the complications of thermal statistics are avoided and only two nonvanishing correlations remain:

$$\begin{aligned}
(1/\hbar)^2\langle \vec{\mathcal{E}}_s^{(+)}(\vec{r}_a,t)\cdot\vec{\mu}\,\vec{\mathcal{E}}_s^{(-)}(\vec{r}_a,t-\tau)\cdot\vec{\mu}\rangle &= \sum_{\vec{k},\lambda} |K_{\vec{k},\lambda}|^2\, e^{-i\omega_k\tau} \\
(1/\hbar)^2\langle \vec{\mathcal{E}}_s^{(+)}(\vec{r}_a,t-\tau)\cdot\vec{\mu}\,\vec{\mathcal{E}}_s^{(-)}(\vec{r}_a,t)\cdot\vec{\mu}\rangle &= \sum_{\vec{k},\lambda} |K_{\vec{k},\lambda}|^2\, e^{i\omega_k\tau}
\end{aligned} \tag{6.4.15}$$

Here the modal summation may be tackled in a fashion similar to that of the previous section with introduction of the density of states (6.3.13) and integration over frequency. Further, with the appropriate choice of polarisation vectors, as indicated in Fig. (6.3.2), we may write

$$(1/\hbar)^2\langle \vec{\mathcal{E}}_s^{(+)}(\vec{r}_a,t)\cdot\vec{\mu}\,\vec{\mathcal{E}}_s^{(-)}(\vec{r}_a,t-\tau)\cdot\vec{\mu}\rangle = (1/4\pi\varepsilon_0)(\omega_0^2\mu^2/4\pi^2\hbar c^3)\int_0^{2\pi}d\phi\int_0^{\pi}d\theta\,\mathrm{Sin}\,\theta\left(\hat{e}_{\vec{k},\lambda_2}\cdot\frac{\vec{\mu}}{\mu}\right)^2\int_0^{\infty}d\omega\,\omega\,e^{i\omega\tau} \tag{6.4.16}$$

Clearly, from Fig. (6.3.2), for the angular integration

$$\begin{aligned}\left(\hat{e}_{\vec{k},\lambda_2}\cdot\frac{\vec{\mu}}{\mu}\right)^2 &= \mathrm{Sin}^2\alpha = 1-\mathrm{Cos}^2\alpha\\ &= 1-\left(\frac{\vec{\mu}}{\mu}\cdot\frac{\vec{k}}{k}\right)^2\end{aligned} \tag{6.4.17}$$

whence, with

$$\begin{aligned}\frac{\vec{\mu}}{\mu} &= \frac{\mu_x}{\mu}\hat{e}_x+\frac{\mu_y}{\mu}\hat{e}_y+\frac{\mu_z}{\mu}\hat{e}_z\\ \frac{\vec{k}}{k} &= \mathrm{Sin}\,\theta\,\mathrm{Cos}\,\phi\,\hat{e}_x+\mathrm{Sin}\,\theta\,\mathrm{Sin}\,\phi\,\hat{e}_y+\mathrm{Cos}\,\theta\,\hat{e}_z\end{aligned} \tag{6.4.18}$$

it follows that

$$\begin{aligned}\left(\hat{e}_{\vec{k},\lambda_2}\cdot\frac{\vec{\mu}}{\mu}\right)^2 = 1-(1/\mu^2)\big[&\mu_x^2\,\mathrm{Sin}^2\theta\,\mathrm{Cos}^2\phi+\mu_y^2\,\mathrm{Sin}^2\theta\,\mathrm{Sin}^2\phi\\ &+\mu_z^2\,\mathrm{Cos}^2\theta\\ &+\mu_x\mu_y\,\mathrm{Sin}^2\theta\,\mathrm{Cos}\,\phi\,\mathrm{Sin}\,\phi+\mu_y\mu_z\,\mathrm{Sin}\,\theta\,\mathrm{Cos}\,\theta\,\mathrm{Cos}\,\phi\\ &+\mu_y\mu_z\,\mathrm{Sin}\,\theta\,\mathrm{Cos}\,\theta\,\mathrm{Sin}\,\phi+\mu_y\mu_x\,\mathrm{Sin}^2\theta\,\mathrm{Sin}\,\phi\,\mathrm{Cos}\,\phi\\ &+\mu_z\mu_x\,\mathrm{Sin}\,\theta\,\mathrm{Cos}\,\theta\,\mathrm{Cos}\,\phi+\mu_z\mu_y\,\mathrm{Sin}\,\theta\,\mathrm{Cos}\,\theta\,\mathrm{Sin}\,\phi\,\big]\end{aligned} \tag{6.4.19}$$

Thus, using the angular integrals (6.3.19) together with

$$\int_0^{\pi}d\theta\,\mathrm{Sin}\,\theta\,\mathrm{Cos}^2\theta = 2/3,\quad \int_0^{\pi}d\theta\,\mathrm{Sin}\,\theta = 2,\quad \int_0^{\pi}d\theta\,\mathrm{Sin}\,\theta\,\mathrm{Sin}^2\theta = 4/3, \tag{6.4.20}$$

we find

$$\int_0^{2\pi} d\phi \int_0^{\pi} d\theta \, \sin\theta \left(\hat{e}_{\vec{k},\lambda_2} \cdot \frac{\vec{\mu}}{\mu}\right)^2 = 8\pi/3 \tag{6.4.21}$$

Substitution in (6.4.16) yields

$$(1/\hbar)^2 \langle \vec{\mathcal{E}}_s^{(+)}(\vec{r}_a,t)\cdot\vec{\mu}\,\vec{\mathcal{E}}_s^{(-)}(\vec{r}_a,t-\tau)\cdot\vec{\mu}\rangle = (1/4\pi\mathcal{E}_0)(4\omega_0^2\mu^2/3\hbar c^3)$$
$$(1/2\pi)\int d\omega\,\omega\, e^{i\omega\tau} \tag{6.4.22}$$

In view of the highly oscillatory nature of the integrand we then make the standard approximation yielding infinitely short correlations and write

$$(1/\hbar)^2 \langle \vec{\mathcal{E}}_s^{(+)}(\vec{r}_a,t)\cdot\vec{\mu}\,\vec{\mathcal{E}}_s^{(-)}(\vec{r}_a,t-\tau)\cdot\vec{\mu}\rangle = \gamma\,\delta(\tau) \tag{6.4.23}$$

where γ is the Einstein A coefficient

$$\gamma = (4\omega_0^3\mu^2/3\hbar c^3)(1/4\pi\mathcal{E}_0) \tag{6.4.24}$$

We may now introduce (6.4.23) to (6.4.14) to obtain our master equation. With (6.4.6), (6.4.14) and (6.4.23) it follows immediately that

$$\mathcal{L}_I\rho = (\gamma/2)(2\sigma_-\rho\sigma_+ - \sigma_+\sigma_-\rho - \rho\sigma_+\sigma_-) \tag{6.4.25}$$

In reaching this form the time dependence of atomic operators is made explicit in (6.4.13), and for the integration of (6.4.6)

$$\int_0^t d\tau\,\delta(\tau) = 1/2 \tag{6.4.26}$$

With (6.4.1), (6.4.2) and (6.4.25) the full master equation therefore reads

$$\frac{d\rho}{dt} = (-i/\hbar)[H_S,\rho] + (\gamma/2)(2\sigma_-\rho\sigma_+ - \sigma_+\sigma_-\rho - \rho\sigma_+\sigma_-) \tag{6.4.27}$$

CHAPTER VII

FLUORESCENT SPECTRA AND DYNAMICS

7.1 Formal apparatus

Having as a foundation the result (6.3.25), we must now formulate the atomic dynamics arising in the master equation of the previous section with a mind to the extraction of field correlations and spectral characteristics. It is convenient that we work in the energy representation corresponding to the Hamiltonian for S. Here the familiar eigenstates of H_S derived in appendix G have

$$\begin{aligned} H_s|E_n^2\rangle &= \hbar\left[\omega_0(n+1/2)+|\kappa_0|(n+1)^{1/2}\right]|E_n^2\rangle \\ H_s|E_n^2\rangle &= \hbar\left[\omega_0(n+1/2)-|\kappa_0|(n+1)^{1/2}\right]|E_n^1\rangle \end{aligned} \tag{7.1.1}$$

where

$$\begin{aligned} |E_n^2\rangle &= (1/\sqrt{2})(|n,+\rangle + |n+1,-\rangle) \\ |E_n^1\rangle &= (1/\sqrt{2})(|n,+\rangle - |n+1,-\rangle) \end{aligned} \tag{7.1.2}$$

$|n\rangle$ are the Fock states and $|+\rangle$ and $|-\rangle$ the upper and lower atomic states respectively. Within this representation the master equation (6.4.26) takes the form

$$\begin{aligned} \frac{d\rho_{n,\eta;m,\xi}}{dt} &= -(i/\hbar)(E_n^\eta - E_m^\xi)\rho_{n,\eta;m,\xi} \\ &\quad +(-1)^{\eta+\xi}(\gamma/4)\sum_{\mu,\nu}\rho_{n+1,\mu;m+1,\nu} \\ &\quad -(\gamma/4)\sum_{\nu}(\rho_{n,\eta;m,\nu}+\rho_{n,\nu;m,\xi}) \end{aligned} \tag{7.1.3}$$

where

$$\rho_{n,\eta;m,\xi} = \langle E_n^\eta | \rho | E_m^\xi \rangle \qquad \eta = 1,2 \qquad \xi = 1,2 \tag{7.1.4}$$

We have here an infinite set of coupled equations indicating the successive scattering of photons from the incident beam and the consequent eventual decay of all matrix elements to those of lower photon number. It will nevertheless be unnecessary, so far as the fluorescent field itself is concerned, to solve such a complex system. We may put the detailed dynamical structure aside and deal only with atomic matrix elements summed over the incident field. We define $\rho_{\eta,\xi}$ by

$$\rho_{\eta,\xi} = \sum_n \rho_{n,\eta;n,\xi} \tag{7.1.5}$$

The determination of the matrix elements (7.1.5) is readily made from the solution of a simple set of four coupled equations. We merely assume an incident field of high intensity, possessing a sharply peaked photon-number distribution. We may, for example, take a coherent state corresponding to laser radiation:

$$|\alpha\rangle = \exp(-1/2 \cdot |\alpha|^2) \sum_n [\alpha^n/(n!)^{1/2}] |n\rangle \tag{7.1.6}$$

Here $\bar{n} = |\alpha|^2$, which for the Poisson distribution also gives the variance. Then for $\bar{n} \sim 10^8$ a fractional change of 10^{-4} only arises in photon number over one standard deviation. Combined with a parametric approximation the result of this circumstance is that in summing matrix elements over the field, the factor $-i(E_n^\eta - E_n^\xi)/\hbar$ in (7.1.3) may be taken through the summation as $-i(E_{\bar{n}}^\xi - E_{\bar{n}}^\xi)/\hbar$. We find for $\rho_{\eta,\xi}$ the coupled equations

$$\frac{d}{dt}\begin{pmatrix}\rho_{22}\\ \rho_{11}\\ \rho_{21}\\ \rho_{12}\end{pmatrix} = \begin{pmatrix} -\gamma/4 & \gamma/4 & 0 & 0 \\ \gamma/4 & -\gamma/4 & 0 & 0 \\ -\gamma/2 & -\gamma/2 & -(3\gamma/4+2i\bar{n}^{1/2}|X_0|) & -\gamma/4 \\ -\gamma/2 & -\gamma/2 & -\gamma/4 & -(3\gamma/4-2i\bar{n}^{1/2}|X_0|) \end{pmatrix}\begin{pmatrix}\rho_{22}\\ \rho_{11}\\ \rho_{21}\\ \rho_{12}\end{pmatrix} \tag{7.1.7}$$

Now our interest in the scattered field $\vec{E}_S(\vec{r},t)$ begins with the mean field of the semiclassical approximation. For this, with (6.3.25) we may write

$$\langle \vec{E}_S^{(+)}(\vec{r},t)\rangle = -I_0(\vec{r})^{1/2}\langle \sigma_-(\hat{t})\rangle \tag{7.1.8}$$

where t = t - r/c, and

$$I_0(\vec{r}) = \left[\omega_0^2/4\pi\varepsilon_0 c^2 r\cdot(\vec{\mu}\times\frac{\vec{r}}{r})\times\frac{\vec{r}}{r}\right]^2 \tag{7.1.9}$$

is the intensity detected at position $\vec{r}$ and retarded time $\hat{t} = 0$ for spontaneous emission. For evaluation of $\langle\sigma_-(\hat{t})\rangle$ we may define, in addition to (7.1.5), the matrix elements $\psi_{\eta,\xi}$, with

$$\psi_{\eta,\xi} = \sum_n \rho_{n+1,\eta;n,\xi} \tag{7.1.10}$$

whence it readily follows that

$$\langle\sigma_-(\hat{t})\rangle = (1/2)(\psi_{22}(\hat{t})-\psi_{11}(\hat{t}))-(1/2)(\psi_{21}(\hat{t})-\psi_{12}(\hat{t})) \tag{7.1.11}$$

Solutions for these are also available from (7.1.3) by virtue of the nature of the chosen incident field. Indeed, resorting to the transformation $\psi_{\eta,\xi}(t) = e^{-i\omega_0 t}\hat{\psi}_{\eta,\xi}(t)$, the matrix equation (7.1.7) may be taken over identically for the $\hat{\psi}_{\eta,\xi}$.

Our further interest in the scattered field lies with the first-and second-order correlation functions:

$$G^{(1)}(\vec{r},t;\vec{r},t+\tau) = \langle \vec{E}_s^{(-)}(\vec{r},t)\vec{E}_s^{(+)}(\vec{r},t+\tau)\rangle \tag{7.1.12}$$

and

$$G^{(2)}(\vec{r},t;\vec{r},t+\tau) = \langle \vec{E}_s^{(-)}(\vec{r},t)\vec{E}_s^{(-)}(\vec{r},t+\tau)\vec{E}_s^{(+)}(\vec{r},t+\tau)\vec{E}_s^{(+)}(\vec{r},t)\rangle \tag{7.1.13}$$

With recourse to (6.3.25), for these we may write respectively

$$G^{(1)}(\vec{r},t;\vec{r},t+\tau) = I_o(\vec{r})\langle \sigma_+(\hat{t})\sigma_-(\hat{t}+\tau)\rangle \tag{7.1.14}$$

and

$$G^{(2)}(\vec{r},t;\vec{r},t+\tau) = I_o(\vec{r})^2\langle \sigma_+(\hat{t})\sigma_+(\hat{t}+\tau)\sigma_-(\hat{t}+\tau)\sigma_-(\hat{t})\rangle \tag{7.1.15}$$

Our concern must then be with the evaluation of the two-time averages $<\sigma_+(\hat{t})\sigma_-(\hat{t} + \tau)>$ and $<\sigma_+(\hat{t})\sigma_+(\hat{t} + \tau)\sigma_-(\hat{t} + \tau)\sigma_-(\hat{t})>$. For this purpose we have firstly, in relation to $<\sigma_+(\hat{t})\sigma_-(\hat{t} + \tau)>$, the result (2.1.18) of chapter II. For the time-ordered average $<\sigma_+(\hat{t})\sigma_+(\hat{t} + \tau)\sigma_-(\hat{t} + \tau)\sigma_-(\hat{t})>$ an argument analogous to that of section 2.1 leads to a similar result. Thus, for $\tau \geq 0$

$$\langle \sigma_+(\hat{t})\sigma_-(\hat{t}+\tau)\rangle = tr_s[e^{\mathcal{L}\tau}\rho(\hat{t})\sigma_+]\sigma_- \tag{7.1.16}$$

and

$$\langle \sigma_+(\hat{t})\sigma_+(\hat{t}+\tau)\sigma_-(\hat{t}+\tau)\sigma_-(\hat{t})\rangle = tr_s[e^{\mathcal{L}\tau}\sigma_-\rho(\hat{t})\sigma_+]\sigma_+\sigma_- \tag{7.1.17}$$

Now, with a mind to solving for these two averages we may define respectively the two operators $\Sigma(\tau)$ and $\Pi(\tau)$ by

$$\begin{aligned} \Sigma(\tau) &= e^{\mathcal{L}\tau}\Sigma(o) \\ \Sigma(o) &= \rho(\hat{t})\sigma_+ \end{aligned} \tag{7.1.18}$$

and

$$\Pi(\tau) = e^{\mathcal{L}\tau}\,\Pi(0)$$
$$\Pi(0) = \sigma_-\,\rho(\hat{t})\,\sigma_+ \tag{7.1.19}$$

It then clearly follows that we may write

$$\frac{d\,\Sigma(\tau)}{d\tau} = \mathcal{L}\,\Sigma(\tau)$$
$$\frac{d\,\Pi(\tau)}{d\tau} = \mathcal{L}\,\Pi(\tau) \tag{7.1.20}$$

and that matrix elements of these operators are consequently determined by equations formally equivalent to (7.1.3). Further, it is readily shown that for the traces of (7.1.16) and (7.1.17) we may write

$$\langle\sigma_+(\hat{t})\,\sigma_-(\hat{t}+\tau)\rangle = (1/2)(\Sigma_{22}(\tau) - \Sigma_{11}(\tau)) - (1/2)(\Sigma_{21}(\tau) - \Sigma_{12}(\tau)) \tag{7.1.21}$$

and

$$\langle\sigma_+(\hat{t})\,\sigma_+(\hat{t}+\tau)\,\sigma_-(\hat{t}+\tau)\,\sigma_-(\hat{t})\rangle = (1/2)(\Pi_{22}(\tau) + \Pi_{11}(\tau))$$
$$+ (1/2)(\Pi_{21}(\tau) + \Pi_{12}(\tau)) \tag{7.1.22}$$

where the matrix elements $\Sigma_{\eta,\xi}(\tau)$ and $\Pi_{\eta,\xi}(\tau)$ are respectively defined by

$$\Sigma_{\eta,\xi}(\tau) = \sum_n \langle E^{\eta}_{n+1}|\,\Sigma(\tau)\,|E^{\xi}_n\rangle \tag{7.1.23}$$

and

$$\Pi_{\eta,\xi}(\tau) = \sum_n \langle E^{\eta}_n|\,\Pi(\tau)\,|E^{\xi}_n\rangle \tag{7.1.24}$$

Here the matrix elements $\Pi_{\eta,\xi}$ and $\Sigma_{\eta,\xi}$ appear formally equivalent to the $\rho_{\eta,\xi}$ of (7.1.5) and the $\psi_{\eta,\xi}$ of (7.1.10) respectively. Thus, after making the transformation $\Sigma_{\eta,\xi}(\tau) = e^{-i\omega_0\tau}\,\hat{\Sigma}_{\eta,\xi}(\tau)$, these also find solution via equations identical to (7.1.7). Overall then, we are simply required to solve a single set of four equations in order to evaluate atomic matrix elements, the mean scattered field, and both first- and second-order field correlation functions.

7.2 Atomic Dynamics and the Semiclassical Field

The coupled equations (7.1.14) are easily solved for atomic matrix elements. With algebraic detail relegated to appendix H, defining the vector $\vec{\rho}$ by

$$\vec{\rho} = (1/2)\begin{pmatrix} \rho_{22} + \rho_{11} \\ \rho_{22} - \rho_{11} \\ \rho_{21} + \rho_{12} \\ \rho_{21} - \rho_{12} \end{pmatrix} \tag{7.2.1}$$

we find the formal solution

$$\vec{\rho}(t) = S\exp(\Lambda t)S^{-1}\vec{\rho}(0) \tag{7.2.2}$$

where for the matrix S

$$S = \begin{pmatrix} S_1 & 0 & 0 & 0 \\ 0 & S_2 & 0 & 0 \\ -(\gamma^2/\gamma^2+8\bar{n}|X_0|^2)S_1 & 0 & S_3 & -(2i\bar{n}^{1/2}|X_0|/\tfrac{1}{4}\gamma-\Omega)S_4 \\ (2i\bar{n}^{1/2}|X_0|/\tfrac{1}{2}\gamma)(\gamma^2/\gamma^2+8\bar{n}|X_0|^2)S_1 & 0 & (2i\bar{n}^{1/2}|X_0|/\tfrac{1}{4}\gamma-\Omega)S_3 & S_4 \end{pmatrix} \tag{7.2.3}$$

and S_1, S_2, S_3 and S_4 are aribtrary constants. Λ is the 4 x 4 diagonal matrix

$$\Lambda = D(0, -\tfrac{1}{2}\gamma, -\tfrac{3}{4}\gamma+\Omega, -\tfrac{3}{4}\gamma-\Omega) \tag{7.2.4}$$

with

$$\Omega = [(\tfrac{1}{4}\gamma)^2 - 4\bar{n}|K_0|^2]^{1/2} \tag{7.2.5}$$

If we begin with an atom in its lower state, the solutions, written in terms of the energy representation of the free atom are

$$\rho_{+,+}(t) = (4\bar{n}|K_0|^2/\gamma^2 + 8\bar{n}|K_0|^2)[1 - e^{-\frac{3}{4}\gamma t}(\cosh\Omega t + (\tfrac{3}{4}\gamma/\Omega)\sinh\Omega t)] \tag{7.2.6}$$

and

$$\rho_{+,-}(t) = -2i\bar{n}^{1/2}|K_0|\{(\gamma/\gamma^2 + 8\bar{n}|K_0|^2)[1 - e^{-\frac{3}{4}\gamma t}(\cosh\Omega t + (\tfrac{3}{4}\gamma/\Omega)\sinh\Omega t)] + (1/2)\, e^{-\frac{3}{4}\gamma t}\sinh\Omega t/\Omega \tag{7.2.7}$$

where we have defined $\rho_{+,+}(t)$ and $\rho_{+,-}(t)$ by

$$\begin{aligned} \rho_{+,+}(t) &= \sum_n \langle n,+|\,\rho(t)\,|n,+\rangle \\ \rho_{+,-}(t) &= \sum_n \langle n,+|\,\rho(t)\,|n+1,-\rangle \end{aligned} \tag{7.2.8}$$

Naturally

$$\begin{aligned} \rho_{-,-}(t) &= 1 - \rho_{+,+}(t) \\ \rho_{-,+}(t) &= \rho_{+,-}(t)^* \end{aligned} \tag{7.2.9}$$

Similar solutions follow for an initially excited atom and include an additional contribution accounting, in the weak-coupling limit, for normal spontaneous emission. These solutions are

$$\rho_{+,+}(t) = (4\bar{n}|K_0|^2/\gamma^2 + 8\bar{n}|K_0|^2)[1 - e^{-\frac{3}{4}\gamma t}(\cosh\Omega t + (\tfrac{3}{4}\gamma/\Omega)\sinh\Omega t)]$$

$$+ e^{-\frac{3}{4}\gamma t}(\mathrm{Cosh}\,\Omega t - (\tfrac{1}{4}\gamma/\Omega)\,\mathrm{Sinh}\,\Omega t) \qquad (7.2.10)$$

and

$$\rho_{+,-}(t) = -2i\bar{n}^{1/2}|\mathcal{K}_0|\{(\gamma/\gamma^2 + 8\bar{n}|\mathcal{K}_0|^2)[1 - e^{-\frac{3}{4}\gamma t}(\mathrm{Cosh}\,\Omega t + (\tfrac{3}{4}\gamma/\Omega)\,\mathrm{Sinh}\,\Omega t)] - (1/2)\,e^{-\frac{3}{4}\gamma t}\,\mathrm{Sinh}\,\Omega t/\Omega \qquad (7.2.11)$$

Similar solutions to the semiclassical Bloch equations for a spin $\frac{1}{2}$ system in a combination of static and RF magnetic fields were first given by Torrey (1949).

In these solutions we see the dynamics separating into an initial transient régime, followed by a saturation steady state. For weak coupling, the saturated atom settles close to its lower level and we expect the behaviour of a classical electron oscillator. With increased incident intensity, however, we find the saturation steady state moves into the nonlinear region;

$$\rho_{+,+}^{ss} = 4\bar{n}|\mathcal{K}_0|^2/\gamma^2 + 8\bar{n}|\mathcal{K}_0|^2 \qquad (7.2.12)$$

For very intense illumination a limit is reached midway between the upper and lower levels;

$$\lim_{\bar{n}\to\infty} \rho_{+,+}^{ss} = 1/2 \qquad (7.2.13)$$

Quantum fluctuations may therefore be expected to become important with intense illumination while remaining of no consequence for weak scattering.

To appreciate now the predictions of the semiclassical perspective on the scattered field we calculate the mean field, which, from (6.3.25) reads

$$\langle \vec{E}_s^{(+)}(\vec{r},t)\rangle = -[\omega_0^2/4\pi\varepsilon_0 c^2 r.(\vec{\mu}\times\frac{\vec{r}}{r})\times\frac{\vec{r}}{r}]\langle\sigma_-(\hat{t})\rangle \tag{7.2.14}$$

where $\langle\sigma_-(\hat{t})\rangle$ is given by (7.1.11), or equivalently

$$\langle\sigma_-(\hat{t})\rangle = \sum_n \langle n,+|\,\rho(\hat{t})|n,-\rangle = \psi_{+,-}(\hat{t}) \tag{7.2.15}$$

In the previous section we saw that $\psi_{\eta,\xi}(t)$ may be calculated from equations identical to (7.1.7), and therefore, allowing for the differing initial values of $\rho_{\eta,\xi}$ and $\psi_{\eta,\xi}$, may make direct recourse to the solutions (7.2.7) and (7.2.11). We write

$$\psi_{+,-}(\hat{t}) = e^{-i(\omega_0\hat{t}+\phi)}\rho_{+,-}(\hat{t}) \tag{7.2.16}$$

where $e^{-i\phi}$ is a phase factor arising in the initial value $\psi_{\eta,\xi}(0)$. Thus, we find for the mean field

$$\langle \vec{E}_s^{(+)}(\vec{r},t)\rangle = -[\omega_0^2/4\pi\varepsilon_0 c^2 r.(\vec{\mu}\times\frac{\vec{r}}{r})\times\frac{\vec{r}}{r}]\rho_{+,-}(\hat{t})\,e^{-i(\omega_0\hat{t}+\phi)} \tag{7.2.17}$$

where $\rho_{+,-}(\hat{t})$ is given by (7.2.7) and (7.2.11) for an atom prepared in lower and upper states respectively.

Here again is the initial transient region followed by saturation. For very intense incident radiation and the régime of the dynamical Stark effect, with

$$4\bar{n}|K_0|^2 >> (\gamma/4)^2 \tag{7.2.18}$$

we may write

$$\begin{aligned}\langle \vec{E}_s^{(+)}(\vec{r},t)\rangle = {} & [\omega_0^2/4\pi\varepsilon_0 c^2 r.(\vec{\mu}\times\frac{\vec{r}}{r})\times\frac{\vec{r}}{r}]\,2i\bar{n}^{1/2}|K_0| \\ & (\gamma/\gamma^2+8\bar{n}|K_0|^2)[e^{-i(\omega_0\hat{t}+\phi)} - (1/2)e^{-\frac{3}{4}\gamma\hat{t}} \\ & (e^{-i[(\omega_0+2\bar{n}^{1/2}|K_0|)\hat{t}+\phi]} + e^{-i[(\omega_0-2\bar{n}^{1/2}|K_0|)\hat{t}+\phi]})]\end{aligned} \tag{7.2.19}$$

which would predict semiclassically a three-component spectrum through the transient region reducing to a sharp line for long times. This is an erroneous prediction, however, as has been confirmed by recent experiments (Schuda *et al.*, 1974; Wu *et al.*, 1975; Walther, 1975; Hartig *et al.*, 1976). We will see in the following section how this inadequacy arises in neglecting quantum fluctuations. For weak illumination the sharp spectrum is as expected from classical electron-oscillator theory.

7.3 The First-Order Correlation Function and Scattered Spectrum

In this section we present solutions for the first-order correlation function (7.1.14) from which we derive the scattered spectrum. This spectrum is defined in terms of the probability $P(\omega,\vec{r},T)$ for photon detection by a monochromatic detector during interval T. We have the result (Glauber, 1964, 1970)

$$P(\omega,\vec{r},T) \propto \int_{r/c}^{T} dt_1 \int_{r/c}^{T} dt_2\, e^{i\omega(t_2-t_1)}\, G^{(1)}(\vec{r},t_1;\vec{r},t_2) \qquad (7.3.1)$$

and with normalisation so the integrated spectrum gives the intensity $I(\vec{r},T) = G^{(1)}(\vec{r},T;\vec{r},T)$ we may define the spectrum $I(\omega,\vec{r},T)$ by

$$I(\omega,\vec{r},T) = (1/2\pi)\Big(I(\vec{r},T)\Big/\int_{r/c}^{T} dt\, I(\vec{r},T)\Big)\, P(\omega,\vec{r},T) \qquad (7.3.2)$$

Since

$$G^{(1)}(\vec{r},t_1;\vec{r},t_2) = G^{(1)}(\vec{r},t_2;\vec{r},t_1)^* \qquad (7.3.3)$$

it readily follows that

$$I(\omega;\vec{r},T) = (1/2\pi)\Big(I(\vec{r},T)\Big/\int_{r/c}^{T} dt\, I(\vec{r},t)\Big) \cdot 2\,\mathrm{Re}\int_{r/c}^{T} dt \int_{0}^{t-r/c} d\tau\, e^{i\omega\tau}\, G^{(1)}(\vec{r},t;\vec{r},t+\tau) \qquad (7.3.4)$$

where we have introduced t and τ with $t_1 \to t$ and $\tau = t_2 - t$.

Now the first-order correlation function, as given by (7.1.14) and (7.1.21), follows from the solution of equations equivalent to (7.1.7):

$$\frac{d}{d\tau}\begin{pmatrix}\Sigma_{22}\\ \Sigma_{11}\\ \Sigma_{21}\\ \Sigma_{12}\end{pmatrix} = \begin{pmatrix} -(\gamma/4+i\omega_0) & \gamma/4 & 0 & 0 \\ \gamma/4 & -(\gamma/4+i\omega_0) & 0 & 0 \\ -\gamma/2 & -\gamma/2 & -[3\gamma/4+i(\omega_0+2\bar{n}^{1/2}|X_0|)] & -\gamma/4 \\ -\gamma/2 & -\gamma/2 & -\gamma/4 & -[3\gamma/4+i(\omega_0-2\bar{n}^{1/2}|X_0|)] \end{pmatrix}\begin{pmatrix}\Sigma_{22}\\ \Sigma_{11}\\ \Sigma_{21}\\ \Sigma_{12}\end{pmatrix} \qquad (7.3.5)$$

The solution is then as in (7.2.2) and thus, defining

$$\vec{\Sigma} = (1/2)\begin{pmatrix}\Sigma_{22}+\Sigma_{11}\\ \Sigma_{22}-\Sigma_{11}\\ \Sigma_{21}+\Sigma_{12}\\ \Sigma_{21}-\Sigma_{12}\end{pmatrix} \qquad (7.3.6)$$

we write

$$\vec{\Sigma}(\tau) = e^{-i\omega_0\tau}\, S\exp(\Lambda\tau)S^{-1}\,\vec{\Sigma}(0) \qquad (7.3.7)$$

where, from (7.1.18) and (7.1.23), the initial vector $\vec{\Sigma}(0)$ is given by

$$\vec{\Sigma}(0) = (1/2)\begin{pmatrix} \rho_{-,+}(\hat{t}) \\ \rho_{+,+}(\hat{t}) \\ -\rho_{-,+}(\hat{t}) \\ -\rho_{+,+}(\hat{t}) \end{pmatrix} \tag{7.3.8}$$

Due to the complexity of the solutions for atomic matrix elements, and hence $\vec{\Sigma}(0)$, a general solution would serve no purpose here. If we concern ourselves now solely with the steady state however, from (7.2.6), (7.2.7) and (7.2.9), we may define $\vec{\Sigma}(0)$ through

$$\begin{aligned}
\rho^{ss}_{+,+} &= 4\bar{n}|K_0|^2/\gamma^2+8\bar{n}|K_0|^2 \\
\rho^{ss}_{-,-} &= 1 - 4\bar{n}|K_0|^2/\gamma^2+8\bar{n}|K_0|^2 \\
\rho^{ss}_{+,-} &= -2i\,\bar{n}^{1/2}|K_0|(\gamma/\gamma^2+8\bar{n}|K_0|^2) \\
\rho^{ss}_{-,+} &= 2i\,\bar{n}^{1/2}|K_0|(\gamma/\gamma^2+8\bar{n}|K_0|^2)
\end{aligned} \tag{7.3.9}$$

and hence find

$$\begin{aligned}
G^{(1)}_{ss}(\tau) = {} & I_0(\vec{r})(4\bar{n}|K_0|^2/\gamma^2+8\bar{n}|K_0|^2)[(\gamma^2/\gamma^2+8\bar{n}|K_0|^2)\,e^{-i\omega_0\tau} \\
& +(1/2)\,e^{-(\frac{1}{2}\gamma+i\omega_0)\tau} \\
& -(1/2)\left(\frac{\gamma^2}{\gamma^2+8\bar{n}|K_0|^2}\,\frac{\frac{3}{4}\gamma+\Omega}{\Omega} - \frac{\frac{1}{2}\gamma}{\Omega} - (1/2)\,\frac{\frac{1}{4}\gamma+\Omega}{\Omega}\right) \\
& \cdot e^{-[(\frac{3}{4}\gamma-\Omega)+i\omega_0]\tau} \\
& +(1/2)\left(\frac{\gamma^2}{\gamma^2+8\bar{n}|K_0|^2}\,\frac{\frac{3}{4}\gamma-\Omega}{\Omega} - \frac{\frac{1}{2}\gamma}{\Omega} - (1/2)\,\frac{\frac{1}{4}\gamma-\Omega}{\Omega}\right) \\
& \cdot e^{-[(\frac{3}{4}\gamma+\Omega)+i\omega_0]\tau}
\end{aligned} \tag{7.3.10}$$

Taking the limit for $T \to \infty$ the spectrum (7.3.4) is given by the Fourier transform

$$I(\omega,\vec{r},\infty) = (1/2\pi)\; 2\,\mathrm{Re}\int_0^{\infty} e^{i\omega\tau}\, G^{(1)}_{ss}(\tau) \tag{7.3.11}$$

Thus, in the definition (7.2.5) for Ω we find a threshold at

$$2\bar{n}^{1/2}\,|K_0| = \gamma/4 \tag{7.3.12}$$

Here the spectrum splits, changing from a single peak to a central peak plus equally spaced sidebands. For weak illumination;

$$4\bar{n}|K_0|^2 \ll (\gamma/4)^2 \tag{7.3.13}$$

the correlation function (7.3.10) becomes

$$G^{(1)}_{ss}(\tau) = I_0(\vec{r})(4\bar{n}|K_0|^2/\gamma^2 + 8\bar{n}|K_0|^2)\, e^{-i\omega_0\tau} \tag{7.3.14}$$

and in the strong coupling limit of (7.2.18) we have

$$G^{(1)}_{ss}(\tau) = (1/2)I_0(\vec{r})\Big[(\gamma^2/\gamma^2+8\bar{n}|K_0|^2)e^{-i\omega_0\tau} + (1/2)e^{-(\frac{1}{2}\gamma+i\omega_0)\tau} + (1/4)\,e^{-[\frac{1}{4}\gamma+i(\omega_0+2\bar{n}^{1/2}|K_0|)]\tau} + (1/4)\,e^{-[\frac{1}{4}\gamma+i(\omega_0-2\bar{n}^{1/2}|K_0|)]\tau}\Big] \tag{7.3.15}$$

Thus, from (7.3.11), for weak scattering we find

$$I(\omega,\vec{r},\infty) = I_0(\vec{r})(4\bar{n}|K_0|^2/\gamma^2+8\bar{n}|K_0|^2)\,\delta(\omega-\omega_0) \tag{7.3.16}$$

and we regain the sharp spectrum predicted by the semiclassical theory in (7.2.19). This is just the coherent scattering obtained by Heitler (1954). For very intense fields however (7.3.11) and (7.3.15) give

$$I(\omega,\vec{r},\infty) = (1/2\pi)I_0(\vec{r})\Big[2\pi(\gamma/\gamma^2+8\bar{n}|K_0|^2)\delta(\omega-\omega_0) + (1/2)\frac{\frac{1}{2}\gamma}{(\frac{1}{2}\gamma)^2+(\omega-\omega_0)^2}$$

$$+(1/4)\frac{\frac{3}{4}\gamma}{(\frac{3}{4}\gamma)^2+[\omega-(\omega_0+2\bar{n}^{1/2}|K_0|)]^2}+(1/4)\frac{\frac{3}{4}\gamma}{(\frac{3}{4}\gamma)^2+[\omega-(\omega_0-2\bar{n}^{1/2}|K_0|)]^2}\Bigg]\Bigg\} \tag{7.3.17}$$

Here the régime of the dynamical Stark effect is apparent, and added to the coherent scattering we see three peaks arising from the quantum fluctuations. This sharply contrasts the picture presented for the steady state by (7.2.19). Illustrations of the steady-state spectrum throughout the range of incident intensities is presented in Figs. (7.3.1) to (7.3.4).

As an example of the behaviour of this first-order correlation function and spectrum in the transient region, let us now consider the time-dependent matrix elements in (7.3.8) but make a restriction to the intense field limit (7.2.18). For an initially excited atom (7.2.10), (7.2.11) and (7.2.9) give

$$\begin{aligned}
\rho_{+,+}(\hat{t}) &= (1/2)(1 + e^{-\frac{3}{4}\gamma\hat{t}}\operatorname{Cos}2\bar{n}^{1/2}|K_0|\hat{t})\\
\rho_{-,-}(\hat{t}) &= (1/2)(1 - e^{-\frac{3}{4}\gamma\hat{t}}\operatorname{Cos}2\bar{n}^{1/2}|K_0|\hat{t})\\
\rho_{+,-}(\hat{t}) &= -(1/2)\,i\,e^{-\frac{3}{4}\gamma\hat{t}}\operatorname{Sin}2\bar{n}^{1/2}|K_0|\hat{t}\\
\rho_{-,+}(\hat{t}) &= (1/2)\,i\,e^{-\frac{3}{4}\gamma\hat{t}}\operatorname{Sin}2\bar{n}^{1/2}|K_0|\hat{t}
\end{aligned} \tag{7.3.18}$$

Introducing these to (7.3.8), we then find from (7.3.7), with (7.1.14) and (7.1.21)

$$\begin{aligned}
G^{(1)}(\vec{r},t;\vec{r},t+\tau) = (1/2)I_0(\vec{r})\Big[&(1/2)(1+e^{-\frac{3}{4}\gamma\hat{t}}\operatorname{Cos}2\bar{n}^{1/2}|K_0|\hat{t})\,e^{-(\frac{1}{2}\gamma+i\omega_0)\tau}\\
&+(1/4)(1+e^{-(\frac{3}{4}\gamma-2i\bar{n}^{1/2}|K_0|)\tau})\,e^{-(\frac{3}{4}\gamma+i\omega_{21})\tau}\\
&+(1/4)(1+e^{-(\frac{3}{4}\gamma+2i\bar{n}^{1/2}|K_0|)\tau})\,e^{-(\frac{3}{4}\gamma+i\omega_{12})\tau}\Big]
\end{aligned} \tag{7.3.19}$$

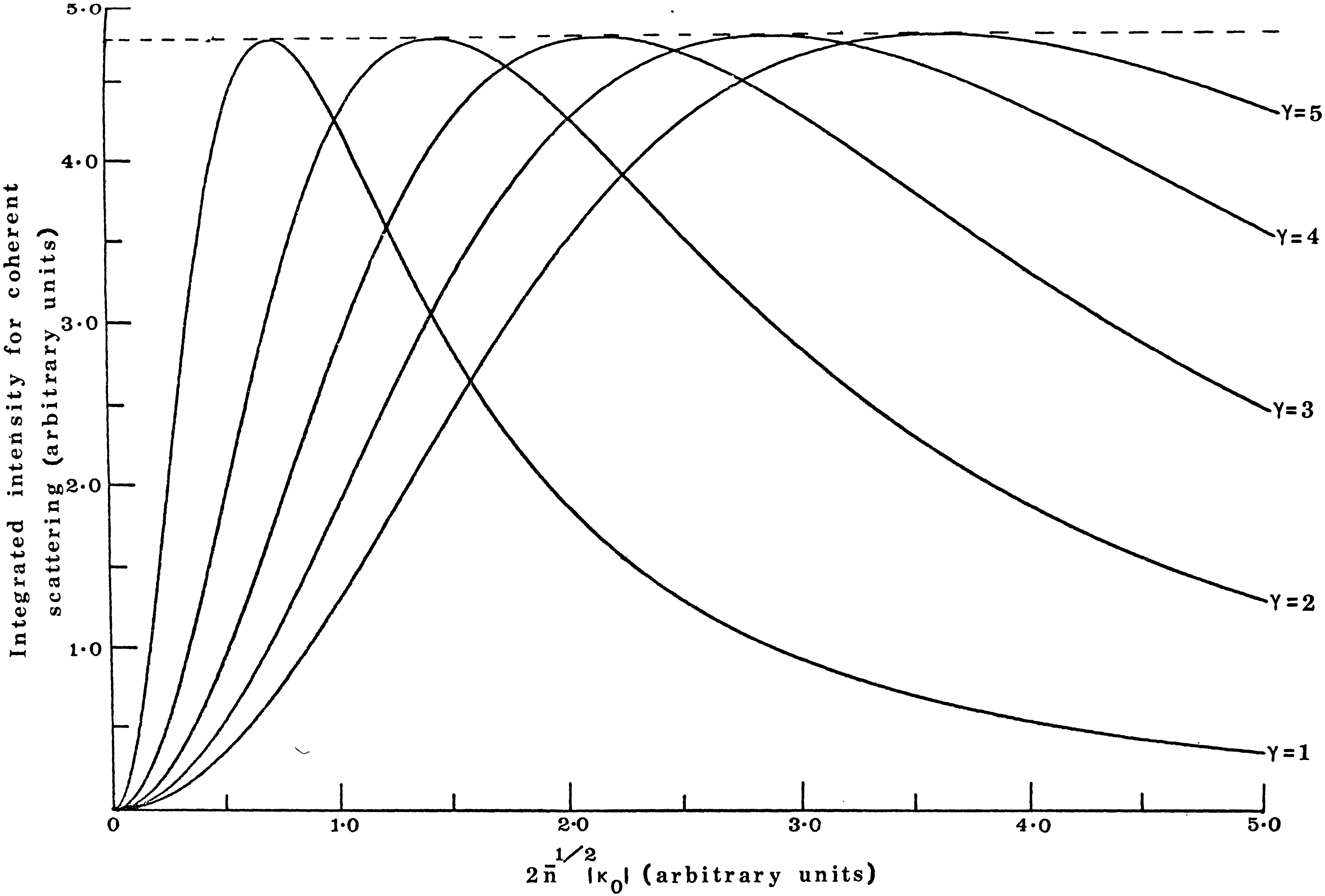
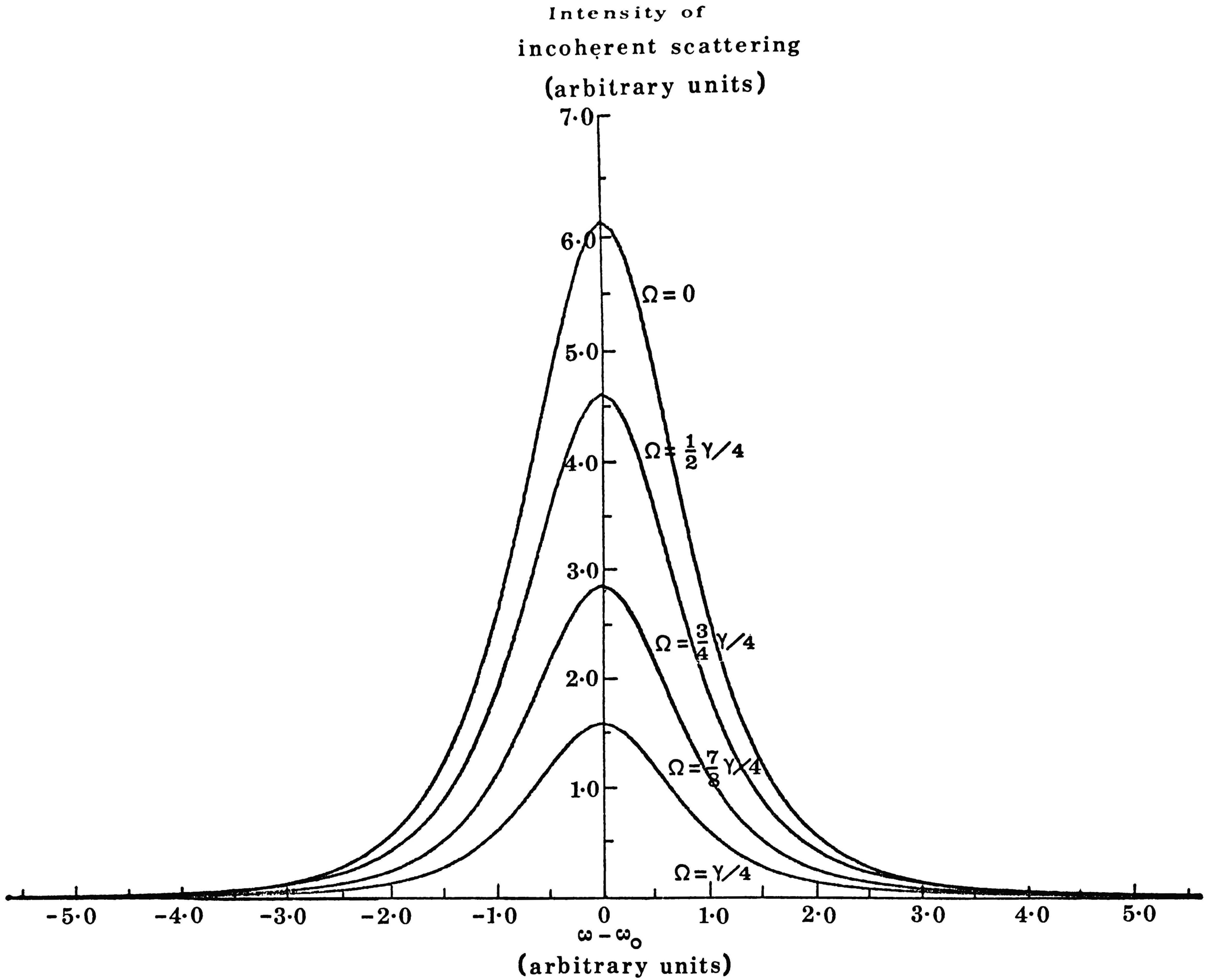


Fig (7·3·1) Variation in integrated coherent scattering with incident intensity.



Fig(7·3·2) Incoherent fluorescent spectrum for the steady state in the range $0 < 2\bar{n}^{-1/2}|\kappa_0| \leqslant \gamma/4$.

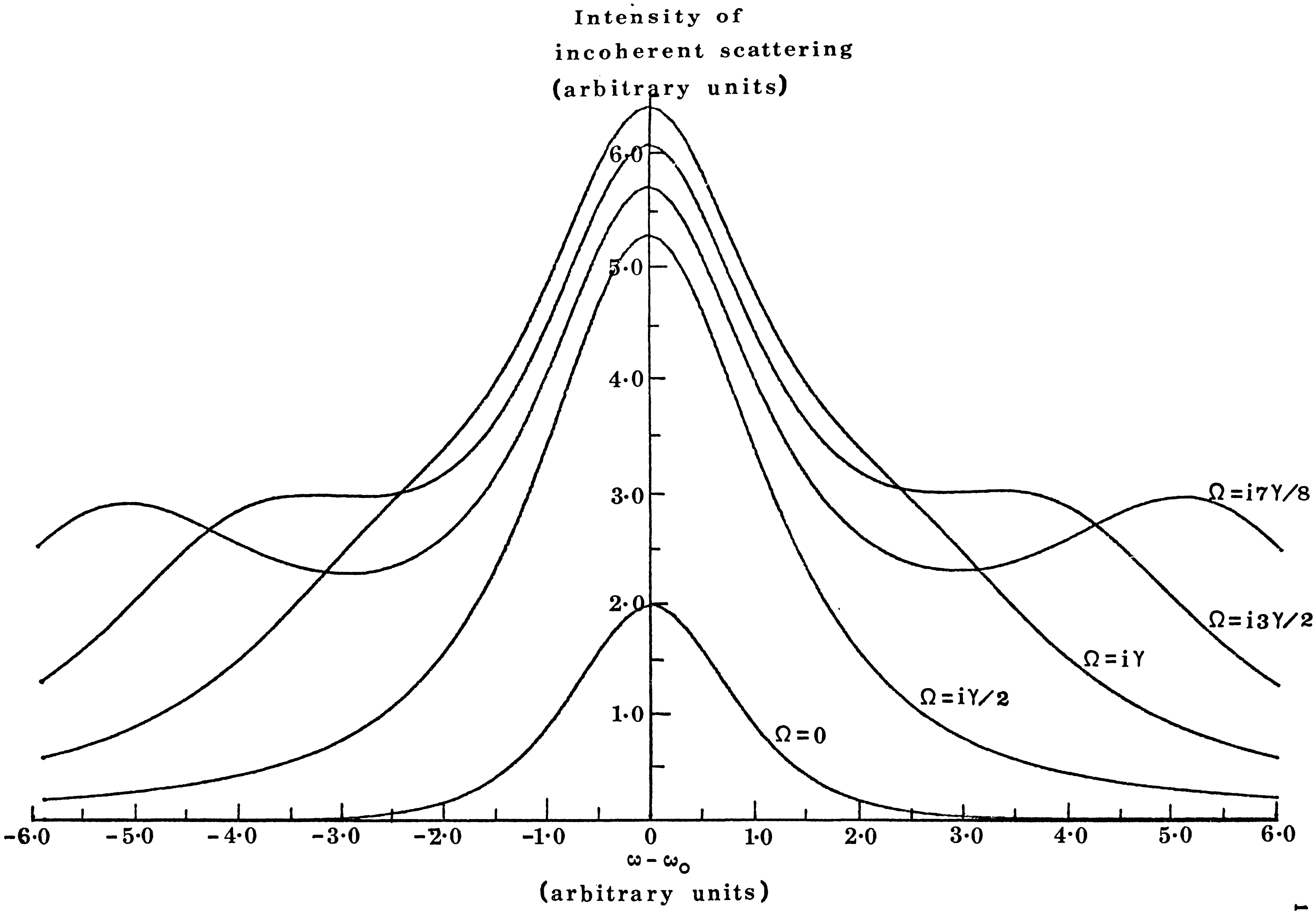


Fig (7·3·3) Incoherent fluorescent spectrum for the steady state in the range $2\bar{n}^{-1/2}|\kappa_0| \geq \gamma/4$.

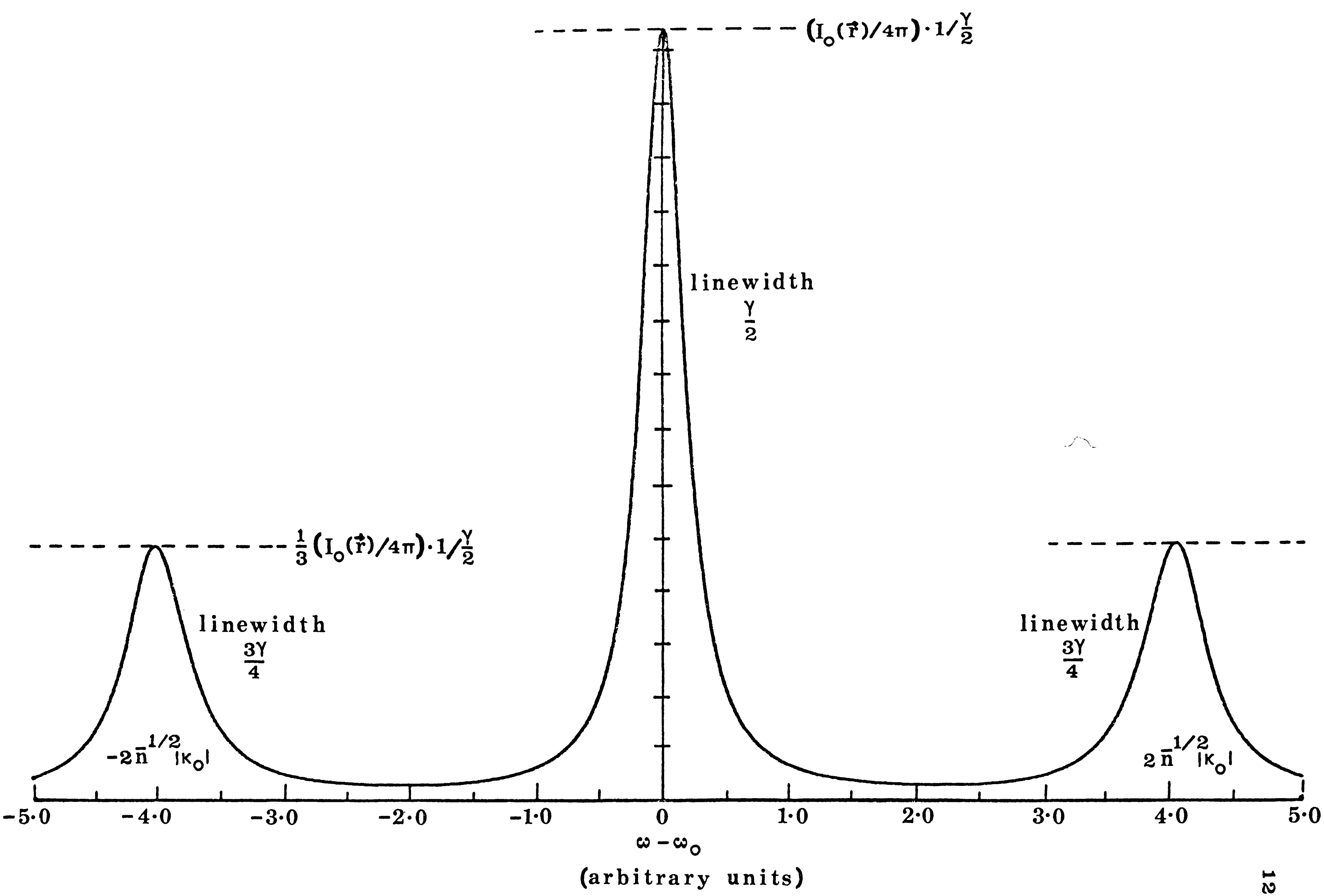


Fig (7·3·4) Steady state fluorescent spectrum for intense illumination.

where we define

$$\begin{aligned} \omega_{21} &= \omega_0 + 2\bar{n}^{1/2}|K_0| \\ \omega_{12} &= \omega_0 - 2\bar{n}^{1/2}|K_0| \end{aligned} \tag{7.3.20}$$

To obtain the time-dependent spectrum the task is in principle simple. We merely evaluate the integrals arising in (7.3.4) with substitution of (7.3.19), and at the outset, for normalisation we find an intensity

$$I(\vec{r},T) = (1/2)I_0(\vec{r})(1 + e^{-\frac{3}{4}\gamma\hat{T}}\cos 2\bar{n}^{1/2}|K_0|\hat{T}) \tag{7.3.21}$$

and

$$\int_{r/c}^{T} dt\, I(\vec{r},t) = (1/2)I_0(\vec{r})[\hat{T} + (\tfrac{3}{4}\gamma/(\tfrac{3}{4}\gamma)^2 + 4\bar{n}|K_0|^2)(1 - e^{-\frac{3}{4}\gamma\hat{T}}\cos 2\bar{n}^{1/2}|K_0|\hat{T})$$
$$+ (2\bar{n}^{1/2}|K_0|/(\tfrac{3}{4}\gamma)^2 + 4\bar{n}|K_0|^2)e^{-\frac{3}{4}\gamma\hat{T}}\sin 2\bar{n}^{1/2}|K_0|\hat{T}] \tag{7.3.22}$$

with $\hat{T} = T - r/c$. A full discussion of this scattered intensity is given by Kimble and Mandel (1975a). Now the time dependence of the probability $P(\omega,\vec{r},T)$ has a double origin. There is a nonstationary emission process and a superimposed T dependence associated with the finite time for detection. For the steady-state probability $P_{ss}(\omega,\vec{r},T)$ arising from (7.3.19) in the limit $\hat{t}\to\infty$ only the second consideration remains. The integrals are elementary and we find

$$P_{ss}(\omega,\vec{r},T) = P_{ss}^{\omega_0}(\omega,\vec{r},T) + P_{ss}^{\omega_{21}}(\omega,\vec{r},T) + P_{ss}^{\omega_{12}}(\omega,\vec{r},T) \tag{7.3.23}$$

with

$$P_{ss}^{\omega_0}(\omega,\vec{r},T) = (1/2)I_0(\vec{r})\left[\frac{\tfrac{1}{2}\gamma}{(\tfrac{1}{2}\gamma)^2 + (\omega-\omega_0)^2}\hat{T} - \frac{(\tfrac{1}{2}\gamma)^2 - (\omega-\omega_0)^2}{[(\tfrac{1}{2}\gamma)^2 + (\omega-\omega_0)^2]^2}(1 - e^{-\frac{1}{2}\gamma\hat{T}}\cos(\omega-\omega_0)\hat{T})\right.$$
$$\left. - \frac{\gamma(\omega-\omega_0)}{[(\tfrac{1}{2}\gamma)^2 + (\omega-\omega_0)^2]^2}e^{-\frac{1}{2}\gamma\hat{T}}\sin(\omega-\omega_0)\hat{T}\right] \tag{7.3.24}$$

$$P_{ss}^{\omega_{21}}(\omega,\vec{r},T) = (1/4)I_0(\vec{r})\Big[\frac{\frac{3}{4}\gamma}{(\frac{3}{4}\gamma)^2+(\omega-\omega_{21})^2}\hat{T} - \frac{(\frac{3}{4}\gamma)^2-(\omega-\omega_{21})^2}{[(\frac{3}{4}\gamma)^2+(\omega-\omega_{21})^2]^2}(1-e^{-\frac{3}{4}\gamma\hat{T}}\cos(\omega-\omega_{21})\hat{T})$$
$$-\frac{\frac{3}{2}\gamma(\omega-\omega_{21})}{[(\frac{3}{4}\gamma)^2+(\omega-\omega_{21})^2]^2}e^{-\frac{3}{4}\gamma\hat{T}}\sin(\omega-\omega_{21})\hat{T}\Big] \tag{7.3.25}$$

$$P_{ss}^{\omega_{12}}(\omega,\vec{r},T) = (1/4)I_0(\vec{r})\Big[\frac{\frac{3}{4}\gamma}{(\frac{3}{4}\gamma)^2+(\omega-\omega_{12})^2}\hat{T} - \frac{(\frac{3}{4}\gamma)^2-(\omega-\omega_{12})^2}{[(\frac{3}{4}\gamma)^2+(\omega-\omega_{12})^2]^2}(1-e^{-\frac{3}{4}\gamma\hat{T}}\cos(\omega-\omega_{12})\hat{T})$$
$$-\frac{\frac{3}{2}\gamma(\omega-\omega_{12})}{[(\frac{3}{4}\gamma)^2+(\omega-\omega_{12})^2]^2}e^{-\frac{3}{4}\gamma\hat{T}}\sin(\omega-\omega_{12})\hat{T}\Big] \tag{7.3.26}$$

These results may be combined with (7.3.21) and (7.3.22) in the form

$$I(\vec{r},T)\Big/\int_{r/c}^{T} dt\, I(\vec{r},\infty) = 1/\hat{T} \tag{7.3.27}$$

to obtain via (7.3.2) the spectrum arising for a saturated atom after a finite observation time T. Clearly for $T\to\infty$ we regain (7.3.17). For the complete nonstationary correlation function (7.3.19) the spectrum is extremely complicated. We find it possible, however, to make a general prescription in the form

$$I(\omega,\vec{r},T) = I(\omega,\vec{r},\infty)\frac{I(\vec{r},T)}{I(\vec{r},\infty)} + I'(\omega,\vec{r},T) \tag{7.3.28}$$

The first term is simply the three-peaked spectrum of the steady state modulated by the scattered intensity. The term $I'(\omega,\vec{r},T)$, which through the transient region modifies the shape of these three peaks contains zero integrated intensity. A full expression of $I'(\omega,\vec{r},T)$ is given in appendix I.

7.4 One-Photon Approximation

In the calculations of the previous section no restriction was made on the number of photon emissions and (7.3.17) agrees with the results of others whose work includes full photon cascades (Mollow, 1969, 1975a,b;

Swain, 1975; Smithers and Freedhoff, 1975). We will proceed now to show how a one-photon approximation may be introduced into our formalism and thus demonstrate the inadequacy of an approach based on single photon emission (Stroud, 1971) for very intense fields.

Taking matrix elements diagonal in the field only, the master equation (7.1.3) reads

$$\frac{d}{dt}\rho_{n,\eta;n,\xi} = (-i/\hbar)(E_n^{\eta}-E_n^{\xi})\rho_{n,\eta;n,\xi} + (-1)^{\eta+\xi}(\gamma/4)\sum_{\mu,\nu}\rho_{n+1,\mu;n+1,\nu} - (\gamma/4)\sum_{\nu}(\rho_{n,\eta;n,\nu}+\rho_{n,\nu;n,\xi}) \tag{7.4.1}$$

Let us now take the incident field initially in a Fock state with N photons and the atom excited. All other modes are initially in the vacuum and therefore the possible subsequent states of the coupled field plus atom are $|N-n,+\rangle$, $|N-n+1,-\rangle$, n varying from o to N. Correspondingly, in the H_S representation we have states $|E^2_{N-n}\rangle$ and $|E^1_{N-n}\rangle$ for which, from (7.4.1), we may write a set of N + 1 equations. As $\rho_{N+1,\eta;\ N+1,\xi}$ must remain zero at all times, there are N coupled equations formally the same as (7.4.1), while for $\rho_{N,\eta;N,\xi}$

$$\frac{d}{dt}\rho_{N,\eta;N,\xi} = (-i/\hbar)(E_N^{\eta}-E_N^{\xi})\rho_{N,\eta;N,\xi} - (\gamma/4)\sum_{\nu}(\rho_{N,\eta;N,\nu}+\rho_{N,\nu;N,\xi}) \tag{7.4.2}$$

which yields

$$\frac{d}{dt}\begin{pmatrix}\rho_{N,2;N,2}\\ \rho_{N,1;N,1}\\ \rho_{N,2;N,1}\\ \rho_{N,1;N,2}\end{pmatrix} = \begin{pmatrix}-\gamma/2 & 0 & -\gamma/4 \\ 0 & -\gamma/2 & -\gamma/4 \\ -\gamma/4 & -\gamma/4 & -(\gamma/2+2iN^{1/2}|K_0|) \\ -\gamma/4 & 0 & 0\end{pmatrix}$$

$$\begin{matrix} -\gamma/4 \\ -\gamma/4 \\ 0 \\ -(\gamma/2 - 2iN^{1/2}|X_0|) \end{matrix} \Bigg) \begin{pmatrix} \rho_{N,2;N,2} \\ \rho_{N,1;N,1} \\ \rho_{N,2;N,1} \\ \rho_{N,1;N,2} \end{pmatrix} \qquad (7.4.3)$$

Solving this system by the method of section 7.2 and appendix H we find

$$\rho_{N,2;N,2}(t) = \rho_{N,1;N,1}(t) = -(1/2)e^{-\frac{1}{2}\gamma t}\left[(2N^{1/2}|X_0|/\Omega')^2 - (\tfrac{1}{2}\gamma/\Omega')^2 \operatorname{Cosh}\Omega' t + (\tfrac{1}{2}\gamma/\Omega') \operatorname{Sinh}\Omega' t\right] \qquad (7.4.4)$$

and

$$\rho_{N,2;N,1}(t) = \rho_{N,1;N,2}(t)^* = -(1/2)e^{-\frac{1}{2}\gamma t}\Big[(2iN^{1/2}|X_0|/\Omega')(\tfrac{1}{2}\gamma/\Omega') - [1 + (2iN^{1/2}|X_0|/\Omega')(\tfrac{1}{2}\gamma/\Omega')]\operatorname{Cosh}\Omega' t + [(\tfrac{1}{2}\gamma/\Omega') + (2iN^{1/2}|X_0|/\Omega')]\operatorname{Sinh}\Omega' t\Big] \qquad (7.4.5)$$

where

$$\Omega' = [(\tfrac{1}{2}\gamma)^2 - 4N^2|X_0|^2]^{1/2} \qquad (7.4.6)$$

We will concern ourselves from here only with the limit of intense illumination for which

$$4N|X_0|^2 >> (\gamma/2)^2 \qquad (7.4.7)$$

In this instance

$$\begin{aligned} \rho_{N,2;N,2}(t) &= \rho_{N,1;N,1}(t) = (1/2)\, e^{-\frac{1}{2}\gamma t} \\ \rho_{N,2;N,1}(t) &= \rho_{N,1;N,2}(t)^* = (1/2)\, e^{-(\frac{1}{2}\gamma + 2iN^{1/2}|X_0|)t} \end{aligned} \qquad (7.4.8)$$

which, using (7.1.2), gives for the probability that the coupled atom plus field remains in its initial state

$$\rho_{N,+;N,+}(t) = e^{-\frac{1}{2}\gamma t} \cos^2 N^{1/2} |\chi_0| t \tag{7.4.9}$$

This is just the result obtained by Stroud (1971) in a one-photon approximation. This is not, however, the probability that the first photon emission has not taken place, as stated by Stroud. We must recognise the possibility with an initially excited atom of realising the state $|N+1,-\rangle$ without emission to the vacuum. We find from (7.4.8)

$$\rho_{N,+;N,+}(t) + \rho_{N+1,-;N+1,-}(t) = e^{-\frac{1}{2}\gamma t} \tag{7.4.10}$$

The probability for no emission therefore decays exponentially, without modulation. This is missed in Stroud's analysis since his basis does not include the state $|N+1,-\rangle$.

We now calculate the first-order correlation function and spectrum in a one-photon approximation by restricting ourselves to the reduced set of basis states $|E_N^2\rangle$, $|E_N^1\rangle$ and $|E_{N-1}^2\rangle$, $|E_{N-1}^1\rangle$ corresponding to the possible states involved in the scattering of the first photon. The calculation proceeds essentially as before with (7.1.23) replaced by

$$\Sigma_{\eta,\xi}(\tau) = \langle E_N^{\eta} | \Sigma(\tau) | E_{N-1}^{\xi} \rangle \tag{7.4.11}$$

This obeys an equation formally equivalent to (7.4.2), and after a transformation $\Sigma_{\eta,\xi}(\tau) = \hat{\Sigma}_{\eta,\xi}(\tau)e^{-i\omega_0\tau}$ the equations for these matrix elements reduce to a form equivalent to (7.4.3).

Now in (7.4.8) there is no steady state as the matrix elements decay eventually to those of lower photon number. In the knowledge that a steady-state spectrum does arise however, in the one-photon approximation we see it maintained by a series of independent emissions from the atomic excited state, each followed by pumping back to the excited state by the incident field. The spectrum is then built up from a large number of identical contributions arising in separate transitions. In solving the equation for $\Sigma_{\eta,\xi}(\tau)$ we then take for the initial conditions required by (7.1.19) and (7.4.11) those defined by (7.4.8) with $\hat{t} = o$. We find from (7.1.14) and (7.1.21)

$$G^{(1)}_{ss}(\tau) = (1/2)\, I_o(\vec{r})\Big(e^{-(\frac{1}{2}\gamma + i\omega_o)\tau} + (1/2)\, e^{-[\frac{1}{2}\gamma + i(\omega_o + 2N^{1/2}|\chi_o|)]\tau} + (1/2)\, e^{-[\frac{1}{2}\gamma + i(\omega_o - 2N^{1/2}|\chi_o|)]\tau}\Big) \tag{7.4.12}$$

Substituting into (7.3.11) then gives

$$I(\omega,\vec{r},\infty) = (1/2\pi)\, I_o(\vec{r})\left[\frac{\frac{1}{2}\gamma}{(\frac{1}{2}\gamma)^2 + (\omega-\omega_o)^2} + (1/2)\frac{\frac{1}{2}\gamma}{(\frac{1}{2}\gamma)^2 + [\omega-(\omega_o+2N^{1/2}|\chi_o|)]^2} + (1/2)\frac{\frac{1}{2}\gamma}{(\frac{1}{2}\gamma)^2 + [\omega-(\omega_o-2N^{1/2}|\chi_o|)]^2}\right] \tag{7.4.13}$$

This spectrum is the same as that obtained by Stroud (1971) with the absence of his linewidth narrowing (for the origin of this feature see Mollow, 1975a, b). In comparison with (7.3.17) (neglecting the coherent scattering) we see then the inadequacy of the one-photon approximation in predicting linewidths and peak heights. In (7.3.17) sidebands are broadened from the central peak by a factor of 3:2 and peak heights are in the ratio of 3:1. The one-photon approximation omits the sideband broadening and gives the ratio of peak heights as 2:1 (Carmichael and Walls, 1975a). With the scattering intensity given by $G^{(1)}_{ss}(o)$ we might note also the reduction of intensity by a factor of 1/2 in (7.3.17) as compared with (7.4.13). This arises in the depletion of the initial scattered intensity to half its value at saturation, as evidenced by (7.3.21).

We note in conclusion that while we have restricted ourselves here for the most part to the limit (7.4.7), for arbitrary intensities we will obtain the general features of the previous section with splitting of the spectrum at a threshold. Clearly, from (7.4.6), however, the one-photon approximation sets this threshold at

$$\gamma/4 = 2N^{1/2}|\kappa_0| \tag{7.4.14}$$

indicating twice the intensity required by (7.3.12).

7.5 The Second-Order Correlation Function

We turn in this final section to the second-order correlation function and intensity correlations. Calculation of this function is readily available in our formalism. With (7.1.15) and (7.1.22) we require only the solution to the equation

$$\frac{d}{d\tau}\begin{pmatrix}\Pi_{22}\\ \Pi_{11}\\ \Pi_{21}\\ \Pi_{12}\end{pmatrix} = \begin{pmatrix} -\gamma/4 & \gamma/4 & 0 & 0\\ \gamma/4 & -\gamma/4 & 0 & 0\\ -\gamma/2 & -\gamma/2 & -(3\gamma/4+2i\bar{n}^{1/2}|\kappa_0|) & -\gamma/4\\ -\gamma/2 & -\gamma/2 & -\gamma/4 & -(3\gamma/4-2i\bar{n}^{1/2}|\kappa_0|)\end{pmatrix}\begin{pmatrix}\Pi_{22}\\ \Pi_{11}\\ \Pi_{21}\\ \Pi_{12}\end{pmatrix} \tag{7.5.1}$$

This follows again from the scheme outlined for atomic dynamics in appendix H, and defining

$$\vec{\Pi} = (1/2)\begin{pmatrix}\Pi_{22}+\Pi_{11}\\ \Pi_{22}-\Pi_{11}\\ \Pi_{21}+\Pi_{12}\\ \Pi_{21}-\Pi_{12}\end{pmatrix} \tag{7.5.2}$$

we may write

$$\vec{\Pi}(\tau) = S\exp(\Lambda\tau)S^{-1}\vec{\Pi}(0) \tag{7.5.3}$$

where S and Λ are as defined in (7.2.3) and (7.2.4), and from (7.1.19) we find

$$\vec{\Pi}(0) = (1/2) \begin{pmatrix} \rho_{+,+}(t) \\ 0 \\ -\rho_{+,+}(t) \\ 0 \end{pmatrix} \tag{7.5.4}$$

We will concern ourselves only with the steady state, and therefore, from (7.2.12), the initial condition is specified by

$$\rho^{ss}_{+,+} = 4\bar{n}|X_0|^2/\gamma^2 + 8\bar{n}|X_0|^2 \tag{7.5.5}$$

Substitution in (7.5.3) then yields

$$G^{(2)}_{ss}(\tau) = G^{(1)}_{ss}(0)^2\left[1 - e^{-\frac{3}{4}\gamma\tau}(\text{Cosh}\,\Omega\tau + \tfrac{3}{4}\gamma/\Omega \cdot \text{Sinh}\,\Omega\tau)\right] \tag{7.5.6}$$

where $G^{(1)}_{ss}(0)$ is simply the steady-state intensity

$$G^{(1)}_{ss}(0) = I_0(\vec{r})\, 4\bar{n}|X_0|^2/\gamma^2 + 8\bar{n}|X_0|^2 \tag{7.5.7}$$

Now the familiar demonstration of second-order correlation effects is, of course, the photon-bunching phenomenon in the Hanbury Brown and Twiss experiment (Hanbury-Brown and Twiss, 1956). In contrast to this there are also fields for which photons tend to be separated on the average, producing second-order correlations which fall below $G^{(1)}_{ss}(0)^2$ as τ approaches zero. Such an effect has been termed photon antibunching and arises, for example, in parametric subharmonic generation (Stoler, 1974). Turning then to (7.5.6) we find just this behaviour, where, for $\tau = 0$, the second-order correlation function vanishes. The interpretation of this is simple and, of course, depends solely on the quantum nature of the scattering.

Consider a photon detected at a position $\vec{r}$ and time $\hat{t}$. This then serves to identify the atom at its lower state at time t so that we may view this emission as preparing the atom in its ground state. Knowing that any subsequent emission must begin with an excited atom, a delay corresponding to the time taken to regain this excited condition is naturally expected. We ask therefore: what is the probability for finding an initially unexcited atom in its upper state? The answer is given by (7.2.6) and is just the expression (7.5.6) for $G^{(2)}_{ss}(\tau)$. It has been pointed out by Cohen-Tannoudji (1976) that this provides an example of the principle of reduction of the wavepacket in quantum mechanics.

In the weak field limit (7.5.6) takes the form displayed in Fig. (7.5.1):

$$G^{(2)}_{ss}(\tau) = G^{(1)}_{ss}(0)^2 \left(1 - e^{-\frac{1}{2}\gamma\tau}\right)^2 \tag{7.5.8}$$

Here this antibunching phenomenon is particularly significant since it provides a purely Q.E.D. prediction in a region which is otherwise adequately described in a semiclassical treatment. Measurement of $G^{(2)}_{ss}(\tau)$ therefore presents the possibilities for a further test of Q.E.D. For strong illumination the correlation function is presented in Fig. (7.5.2) and corresponds to the form

$$G^{(2)}_{ss}(\tau) = G^{(1)}_{ss}(0)^2 \left(1 - e^{-\frac{3}{4}\gamma\tau} \cos 2\bar{n}^{1/2}|X_0|\tau\right) \tag{7.5.9}$$

Here we have both photon bunching and antibunching displayed in the one situation. This, of course, corresponds to the oscillation of the probability for excitation through the transient régime between values above and below that attained at saturation.

Let us now consider $G^{(2)}_{ss}(\tau)$ from another perspective; that which sees in it a possible source for the experimental measurement of

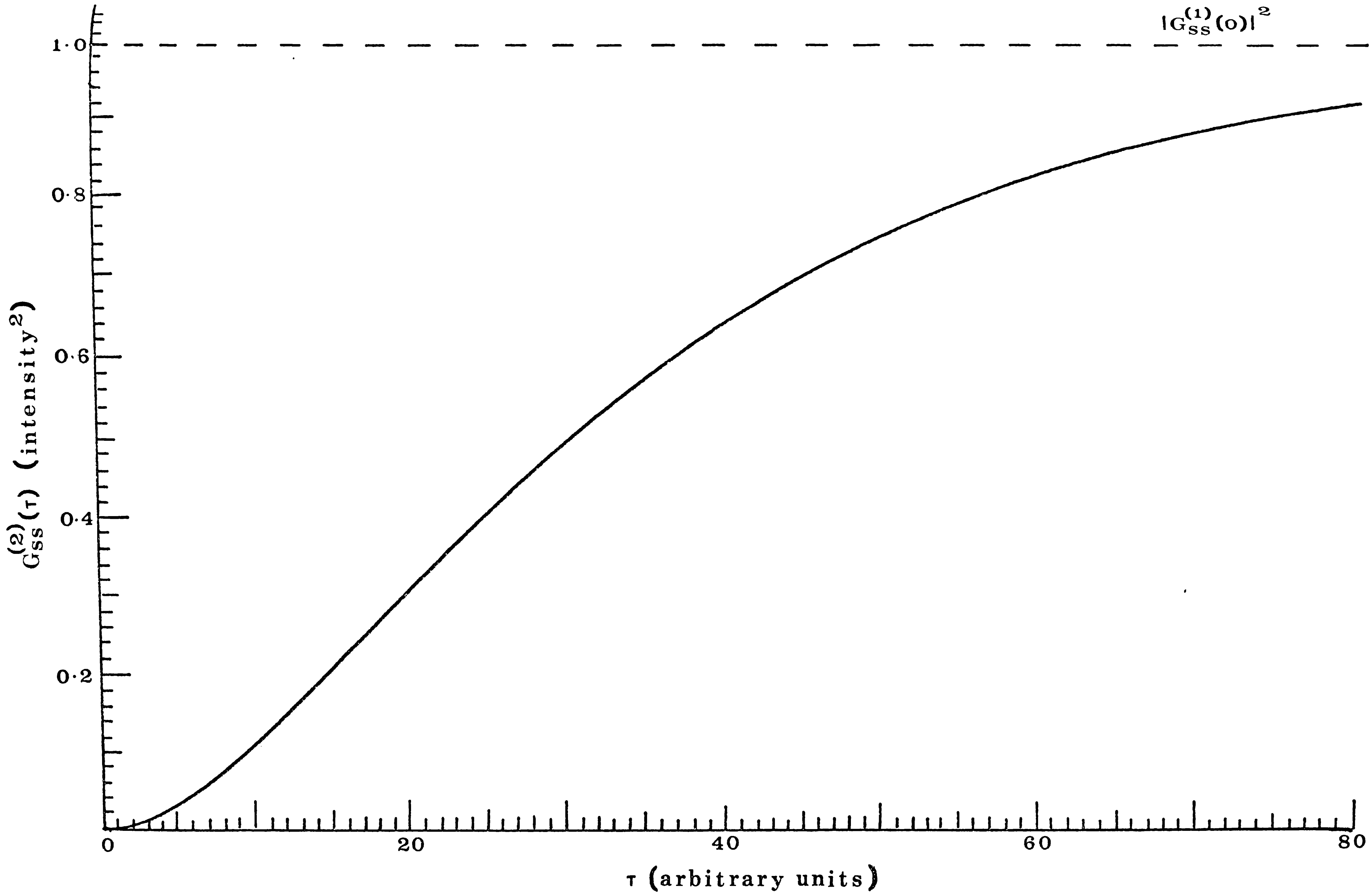


Fig (7·5·1) Second-order correlation function for single atom fluorescence, $2\bar{n}^{1/2}|\kappa_0| \ll \gamma/4$.

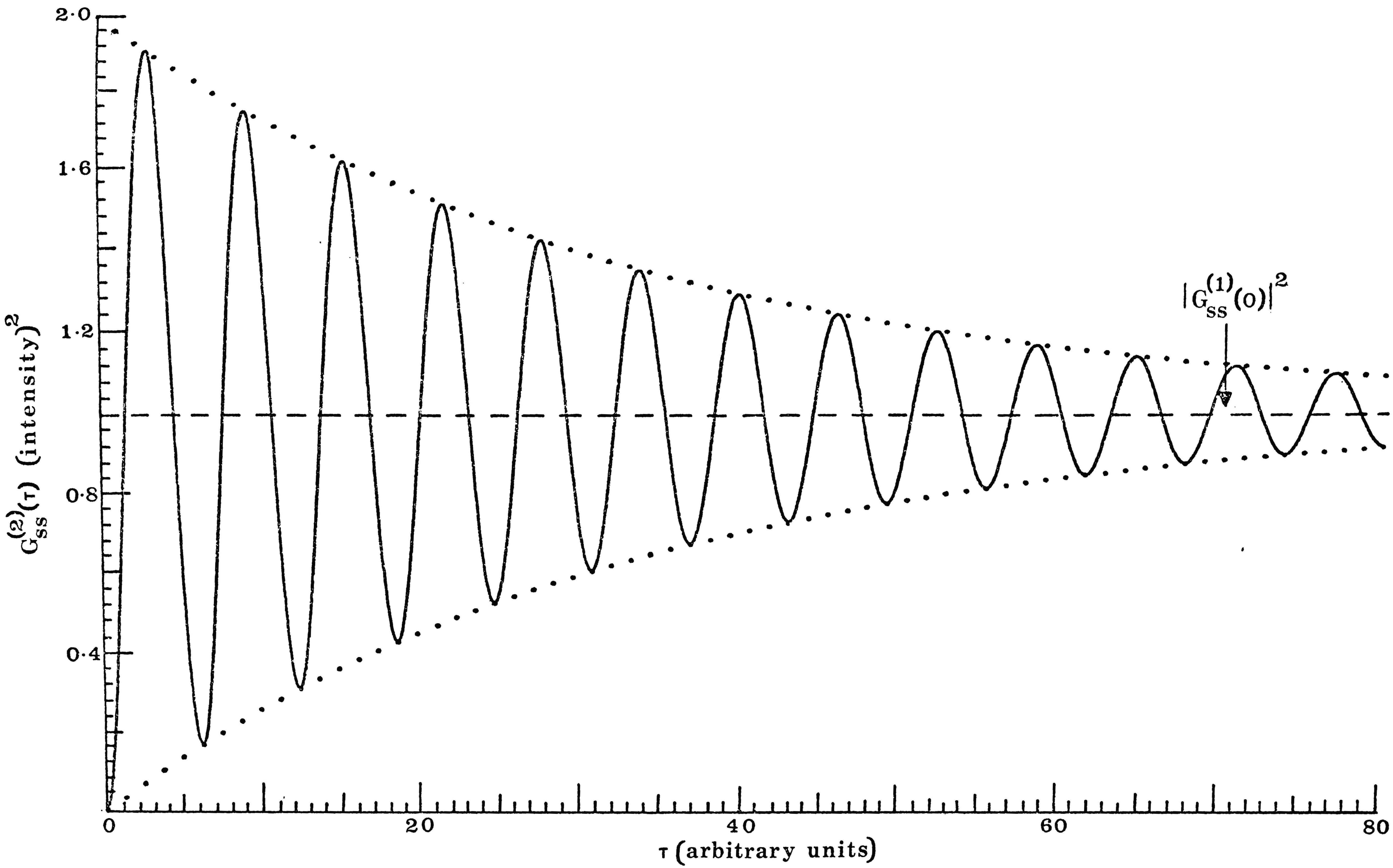


Fig (7·5·2) Second-order correlation function for single-atom fluorescence, $2\bar{n}^{1/2}|\kappa_0| \gg \gamma/4$.

spectral parameters. We are concerned then particularly with the form (7.5.9) which is associated with the three-peaked spectrum given in (7.3.17). We see directly that contained in this function is information on both the peak widths and the splitting frequency. With the measurement of $G_{ss}^{(2)}(\tau)$ we can then extract spectral detail which has to this stage been unavailable (Carmichael and Walls, 1976a).

Now (7.5.6) applies to the scattering from a single atom repeatedly absorbing and re-emitting photons from the incident field. There is clearly then going to be a major problem obtaining sufficient scattered intensity in any attempt to measure this correlation function. In view of this fact we might usefully consider the simultaneous illumination of many atoms. This corresponds to the experiments of Schuda *et al.* (1974), Walther (1975), Wu *et al.* (1975), and Hartig *et al.* (1976), where an atomic beam is arranged to cross a laser light so that many atoms experience irradiation at one time. The scattered field $\vec{E}_s(\vec{r},t)$ becomes the sum of fields $\vec{E}_{s_k}(\vec{r},t)$ arising from individual atoms:

$$\vec{E}_s(\vec{r},t) = \sum_k \vec{E}_{s_k}(\vec{r},t) \tag{7.5.10}$$

Now these scattering centres enter the laser field at random times and may be taken to act independently. This means that the component of $G_{ss}^{(2)}(\tau)$ corresponding to the second-order correlations for individual atoms is swamped in the limit of many atoms by that corresponding to the product of first-order correlations. This reflects the introduction of Gaussian statistics which follow from the central limit theorem. We find a result which holds generally for Gaussian signals (Glauber, 1963):

$$G^{(2)}(\vec{r},t;\vec{r},t+\tau) = G^{(1)}(\vec{r},t;\vec{r},t)^2 + |G^{(1)}(\vec{r},t;\vec{r},t+\tau)|^2 \tag{7.5.11}$$

We may take the illuminated atoms in their saturated state and then the summed field will be stationary and the first-order correlation function simply proportional to that for a single atom. For weak and strong illumination respectively we may therefore write from (7.3.14) and (7.3.15)

$$|G_{ss}^{(1)}(\tau)| \propto I_0(\vec{r})\, 4\bar{n}|\kappa_0|^2/\gamma^2 + 8\bar{n}|\kappa_0|^2 \tag{7.5.12}$$

and

$$|G_{ss}^{(1)}(\tau)| \propto (1/4)I_0(\vec{r})\left(e^{-\frac{1}{2}\gamma\tau} + e^{-\frac{3}{4}\gamma\tau}\cos 2\bar{n}^{1/2}|\kappa_0|\tau\right) \tag{7.5.13}$$

The property of photon antibunching is lost here as we expect, nonetheless (7.5.13) still contains the spectral information hoped for. Indeed it bears possibilities over and above those found in (7.5.9). In Figs. (7.5.3) and (7.5.4) clearly the addition and subtraction of the upper and lower envelopes makes available both the curves $e^{-\frac{1}{2}\gamma\tau}$ and $e^{-\frac{3}{4}\gamma\tau}$. Here, then, is the potential for a direct test of the predicted ratio 3:2 for sideband broadening.

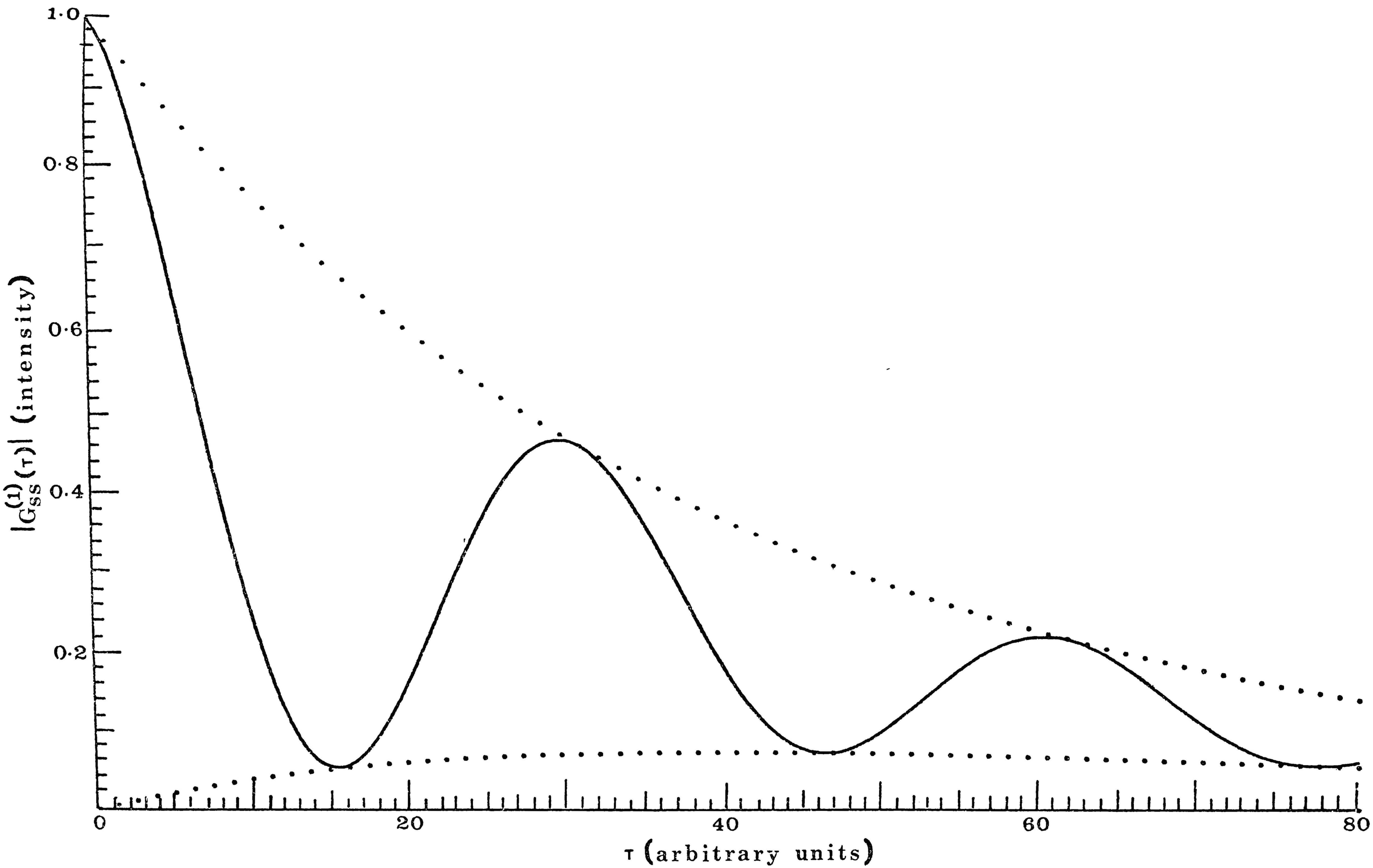


Fig (7·5·3) Second-order correlation function for light scattered by many atoms, $2\bar{n}^{1/2}|\kappa_0|:\gamma/4 = 20:1$. Sideband displacement 10 times natural linewidth.

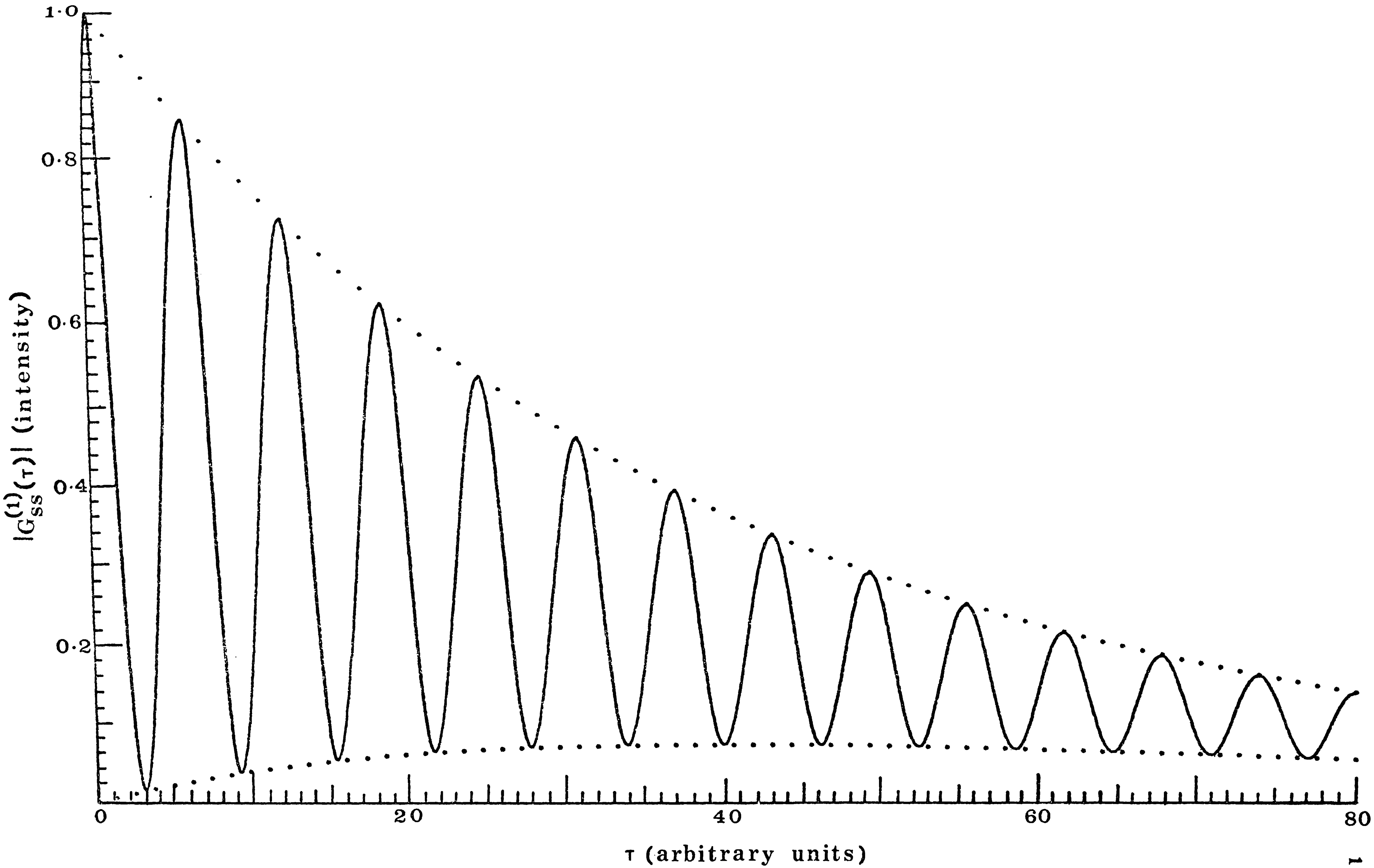


Fig (7·5·4) Second-order correlation function for light scattered by many atoms, $2\bar{n}^{1/2}|\kappa_0| : \gamma/4 = 100:1$. Sideband displacement 50 times natural linewidth.

APPENDIX A

TWO-TIME AVERAGES AND THE QUANTUM REGRESSION THEOREM OF LAX

We have in section (2.1) obtained the result

$$\langle S^{(\alpha)}(t+\tau)\, S^{(\beta)}(t)\rangle_{\rho(t)} = tr_s\left[e^{\mathcal{L}\tau} S^{(\beta)}(0)\,\rho(t)\right] S^{(\alpha)}(0) \tag{A.1}$$

for evaluating a two-time average after tracing over reservoir variables. We will demonstrate here how a direct relationship may be drawn between this result and the quantum regression theorem of Lax (1963, 1967, 1968). This theorem states that assuming $S^{(\alpha)}(t + \tau)$ and $S^{(\beta)}(t)$ belong to a linear set of Markoffian operators, in the sense that

$$\langle S^{(\alpha)}(t+\tau)\rangle = \sum_{\gamma} G^{\alpha,\gamma}(t+\tau, t)\,\langle S^{(\gamma)}(t)\rangle \tag{A.2}$$

where the $G^{\alpha,\gamma}(t + \tau, t)$ are some constants, then

$$\langle S^{(\alpha)}(t+\tau)\, S^{(\beta)}(t)\rangle = \sum_{\gamma} G^{\alpha,\gamma}(t+\tau, t)\,\langle S^{(\gamma)}(t)\, S^{(\beta)}(t)\rangle \tag{A.3}$$

Thus, once the $G^{\alpha,\gamma}(t + \tau, t)$ are known two-time averages are available from single time averages.

If we proceed from (2.1.1) we may write

$$\langle S^{(\alpha)}(t+\tau)\rangle = tr_s\, \rho(t+\tau)\, S^{(\alpha)}(0) \tag{A.4}$$

and in view of the formal solution to (1.2.35)

$$\langle S^{(\alpha)}(t+\tau)\rangle = tr_s\left[e^{\mathcal{L}\tau}\rho(t)\right] S^{(\alpha)}(0) \tag{A.5}$$

We now define an operator $\bar{\mathcal{L}}$ by the general requirement

$$\mathrm{tr}_s\, S^{(\alpha)}\, \mathcal{L}\, S^{(\beta)} = \mathrm{tr}_s\, S^{(\beta)}\, \bar{\mathcal{L}}\, S^{(\alpha)} \tag{A.6}$$

for any two operators $S^{(\alpha)}$ and $S^{(\beta)}$. We also introduce the energy representation $|\varepsilon,\{\lambda\}>$ with

$$H_s|\mathcal{E},\{\lambda\}\rangle = \mathcal{E}|\mathcal{E},\{\lambda\}\rangle \tag{A.7}$$

$\{\lambda\}$ being a set of quantum numbers accounting for an energy degeneracy. For simplicity we may distinguish the $|\varepsilon,\{\lambda\}>$ by a single index, writing the energy eigenstate $|n>$. Then using (A.6) and adopting the energy representation we find from (A.5)

$$\begin{aligned}\langle S^{(\alpha)}(t+\tau)\rangle &= \mathrm{tr}_s\big[e^{\bar{\mathcal{L}}\tau}\, S^{(\alpha)}(0)\big]\,\rho(t)\\ &= \sum_{\substack{n,m,\\ n',m'}} L^{n',m'}_{n,m}(\tau)\, S^{(\alpha)}(0)_{n,m}\, \rho(t)_{n',m'}\end{aligned} \tag{A.8}$$

where $L^{n',m'}_{n,m}(\tau)$ is defined by

$$L^{n',m'}_{n,m}(\tau) = \langle m'|\big(e^{\bar{\mathcal{L}}\tau}|n\rangle\langle m|\big)|n'\rangle \tag{A.9}$$

Thus, since from (2.1.1)

$$\begin{aligned}\langle S^{(\gamma)}(t)\rangle &= \mathrm{tr}_s\, S^{(\gamma)}(0)\,\rho(t)\\ &= \sum_{n',m'} S^{(\gamma)}(0)_{m',n'}\,\rho(t)_{n',m'}\end{aligned} \tag{A.10}$$

the assumption (A.2), when combined with (A.8) and (A.10), leads to the requirement

$$\begin{aligned}&\sum_{n',m'}\sum_{n,m} L^{n',m'}_{n,m}(\tau)\, S^{(\alpha)}(0)_{n,m}\,\rho(t)_{n',m'}\\ &\qquad = \sum_{n',m'}\sum_{\gamma} G^{\alpha,\gamma}(t+\tau,t)\, S^{(\gamma)}(0)_{m',n'}\,\rho(t)_{n',m'}\end{aligned} \tag{A.11}$$

For this a sufficient condition is

$$\sum_{n,m} L_{n,m}^{n',m'}(\tau)\, \overset{(\alpha)}{S}(0)_{n,m} = \sum_{\gamma} G^{\alpha,\gamma}(t+\tau,t)\, \overset{(\gamma)}{S}(0)_{m',n'} \tag{A.12}$$

Let us now turn to the average (A.1). Using (A.6) and introducing the energy representation we may write

$$\begin{aligned}\langle \overset{(\alpha)}{S}(t+\tau)\overset{(\beta)}{S}(t)\rangle &= tr_S[e^{\bar{\mathcal{L}}\tau}\, \overset{(\alpha)}{S}(0)]\, \overset{(\beta)}{S}(0)\, \rho(t) \\ &= \sum_{\substack{n,m \\ n',m'}} L_{n,m}^{n',m'}(\tau)\, \overset{(\alpha)}{S}(0)_{n,m}\, [\overset{(\beta)}{S}(0)\rho(t)]_{n',m'}\end{aligned} \tag{A.13}$$

Inserting now the requirement (A.12) we find

$$\langle \overset{(\alpha)}{S}(t+\tau)\overset{(\beta)}{S}(t)\rangle = \sum_{n',m'}\sum_{\gamma} G^{\alpha,\gamma}(t+\tau,t)\, \overset{(\gamma)}{S}(0)_{m',n'}\, [\overset{(\beta)}{S}(0)\rho(t)]_{n',m'} \tag{A.14}$$

The summation over n' and m' may then be simply carried out yielding the result

$$\langle \overset{(\alpha)}{S}(t+\tau)\overset{(\beta)}{S}(t)\rangle = \sum_{\gamma} G^{\alpha,\gamma}(t+\tau,t)\, \langle \overset{(\gamma)}{S}(t)\overset{(\beta)}{S}(t)\rangle \tag{A.15}$$

This is just the form given by the regression theorem of Lax.

APPENDIX B

THE PHASE-SPACE CALCULUS OF AGARWAL AND WOLF

We present here those results from the phase-space calculus of Agarwal and Wolf (1970) which are fundamental to the work of sections 2.2 and 4.1. At the outset, we restate the equations from section 2.2 relating the phase-space Ω-equivalent $S^{(\Omega)}(\alpha,\alpha^*)$ for an arbitrary operator $\hat{S}(a,a^\dagger)$ of a single boson system to the operator form. For the mapping operator $\hat{\Omega}$, we have

$$\hat{\Omega}\, S^{(\Omega)}(\alpha,\alpha^*) = \hat{S}(a,a^\dagger) \tag{B.1}$$

and with

$$S^{(\Omega)}(\alpha,\alpha^*) = \int d^2\beta\, F^{(\Omega)}(\beta,\beta^*)\exp(\beta\alpha^* - \beta^*\alpha) \tag{B.2}$$

$$F^{(\Omega)}(\beta,\beta^*) = (1/\pi^2)\int d^2\alpha\, S^{(\Omega)}(\alpha,\alpha^*)\exp(\beta^*\alpha - \beta\alpha^*) \tag{B.3}$$

we require

$$\hat{S}(a,a^\dagger) = \int d^2\beta\, G(\beta,\beta^*)\exp(\beta a^\dagger - \beta^* a) \tag{B.4}$$

$$G(\beta,\beta^*) = (1/\pi)\,\mathrm{tr}_s\, \hat{S}(a,a^\dagger)\exp(\beta^* a - \beta a^\dagger) \tag{B.5}$$

where

$$G(\beta,\beta^*) = \Omega(\beta,\beta^*)\, F^{(\Omega)}(\beta,\beta^*) \tag{B.6}$$

Here $\Omega(\beta,\beta^*)$ is referred to as the filter function for the Ω-mapping and is restricted to the form

$$\Omega(\beta,\beta^*) = \exp(\mu\beta^2 + \nu\beta^{*2} + \lambda\beta\beta^*) \tag{B.7}$$

It then possesses in particular the property

$$\Omega(\beta,\beta^*) = \Omega(-\beta,-\beta^*) \tag{B.8}$$

We also define the inverse to the Ω-mapping as that mapping having filter function $\bar{\Omega}(\beta,\beta^*)$, with

$$\Omega(\beta,\beta^*)\,\bar{\Omega}(\beta,\beta^*) = 1 \tag{B.9}$$

Let us consider now two uses of the Dirac δ-function in relation to the above scheme. We take first $\delta^2(\beta-\beta_0)$ to be the function $F^{(\Omega)}(\beta,\beta^*)$ of (B.2) and (B.3). We have

$$S_\delta^{(\Omega)}(\alpha,\alpha^*) = \int d^2\beta\ \delta^2(\beta-\beta_0)\exp(\beta\alpha^*-\beta^*\alpha) \tag{B.10}$$

and

$$\delta^2(\beta-\beta_0) = (1/\pi^2)\int d^2\alpha\ S_\delta^{(\Omega)}(\alpha,\alpha^*)\exp(\beta^*\alpha-\beta\alpha^*) \tag{B.11}$$

In view of the Fourier expansion for $\delta^2(\beta-\beta_0)$; namely

$$\delta^2(\beta-\beta_0) = (1/\pi^2)\int d^2\alpha\ \exp[\alpha(\beta^*-\beta_0^*)-\alpha^*(\beta-\beta_0)] \tag{B.12}$$

we may write

$$S_\delta^{(\Omega)}(\alpha,\alpha^*) = \exp(\alpha^*\beta_0-\alpha\beta_0^*) \tag{B.13}$$

Thus, from (B.1), (B.4), (B.6) and (B.13)

$$\begin{aligned}\hat{\Omega}[\exp(\alpha^*\beta_0-\alpha\beta_0^*)] &= \int d^2\beta\ \Omega(\beta,\beta^*)\,\delta^2(\beta-\beta_0) \\ &\qquad\cdot\exp(\beta a^\dagger-\beta^* a) \\ &= \Omega(\beta_0,\beta_0^*)\exp(\beta_0 a^\dagger-\beta_0^* a)\end{aligned} \tag{B.14}$$

Recognising $\exp(\beta a^\dagger - \beta^* a)$ as the familiar displacement operator $\hat{D}(\beta)$ for the coherent states (Glauber, 1963), we therefore have

$$\hat{\Omega}[\exp(\alpha^*\beta-\alpha\beta^*)] = \Omega(\beta,\beta^*)\,\hat{D}(\beta) \tag{B.15}$$

Let us consider now the Ω-mapping of the Dirac δ-function itself. Thus, in contrast to the above, here we take $s^{(\Omega)}(\alpha,\alpha^*)$ to be $\delta^2(\alpha - \alpha_0)$ with

$$\delta^2(\alpha-\alpha_0) = (1/\pi^2)\int d^2\beta \exp[\beta(\alpha^*-\alpha_0^*)-\beta^*(\alpha-\alpha_0)] \quad \text{(B.16)}$$

In the context of (B.2) and (B.3) we may write

$$\delta^2(\alpha-\alpha_0) = \int d^2\beta\, F_\delta^{(\Omega)}(\beta,\beta^*) \exp[\beta(\alpha^*-\alpha_0^*)-\beta^*(\alpha-\alpha_0)] \quad \text{(B.17)}$$

where $F_\delta^{(\Omega)}(\beta,\beta^*)$ is a complex function appropriate to the Fourier expansion of $\delta^2(\alpha - \alpha_0)$. Clearly (B.12) indicates

$$F_\delta^{(\Omega)}(\alpha,\alpha^*) = (1/\pi^2) \quad \text{(B.18)}$$

and hence, from (B.6)

$$G_\delta^{(\Omega)}(\beta,\beta^*) = (1/\pi^2)\,\Omega(\beta,\beta^*) \quad \text{(B.19)}$$

We then specify $\Delta^{(\Omega)}(a - \alpha_0, a^\dagger - \alpha_0^*)$ such that

$$\hat{\Omega}\,\delta^2(\alpha-\alpha_0) = \Delta^{(\Omega)}(a-\alpha_0, a^\dagger-\alpha_0^*) \quad \text{(B.20)}$$

Aligning this with (B.4) we have

$$\begin{aligned}\Delta^{(\Omega)}(a-\alpha_0, a^\dagger-\alpha_0^*) &= (1/\pi^2)\int d^2\beta\, \Omega(\beta,\beta^*)\exp[\beta(a^\dagger-\alpha_0^*) \\ &\qquad -\beta^*(a-\alpha_0)] \\ &= (1/\pi^2)\int d^2\beta\, \Omega(\beta,\beta^*)\exp(\alpha_0\beta^*-\alpha_0^*\beta)\,\hat{D}(\beta) \quad \text{(B.21)}\end{aligned}$$

Further on we will note the significance of this Ω-equivalent of the Dirac δ-function.

Now the displacement operator $\hat{D}(\beta)$ possesses the properties

$$\hat{D}(\beta)\hat{D}(\beta_0) = \exp[\tfrac{1}{2}(\beta\beta_0^*-\beta^*\beta_0)]\,\hat{D}(\beta+\beta_0) \quad \text{(B.22)}$$

and

$$\mathrm{tr}_S\, \hat{D}(\beta) = \pi\, \delta^2(\beta) \tag{B.23}$$

This imparts an important property to the operators $\Delta^{(\Omega)}[(a - \alpha),(a^\dagger - \alpha^*)]$. Specifically, we take

$$\begin{aligned}\mathrm{tr}_S\, &\Delta^{(\Omega)}[(a-\alpha),(a^\dagger-\alpha^*)]\,\Delta^{(\bar{\Omega})}[(a+\alpha_0),(a^\dagger+\alpha_0^*)] \\ &= [(1/\pi^2)\int d^2\beta\, \Omega(\beta,\beta^*)\, \exp(\beta^*\alpha-\beta\alpha^*)] \\ &\quad\cdot[(1/\pi^2)\int d^2\beta_0\, \bar{\Omega}(-\beta_0,-\beta_0^*)\exp(\beta_0\alpha^*-\beta_0^*\alpha)]\,\mathrm{tr}_S\, \hat{D}(\beta)\hat{D}(-\beta_0)\end{aligned} \tag{B.24}$$

Invoking then (B.22) and (B.23) we find

$$\begin{aligned}\mathrm{tr}_S\, &\Delta^{(\Omega)}[(a-\alpha),(a^\dagger-\alpha^*)]\,\Delta^{(\bar{\Omega})}[(a+\alpha_0),(a^\dagger+\alpha_0^*)] \\ &= (1/\pi)(1/\pi^2)\int d^2\beta\, \Omega(\beta,\beta^*)\bar{\Omega}(-\beta,-\beta^*)\exp[\beta^*(\alpha-\alpha_0)-\beta(\alpha^*-\alpha_0^*)]\end{aligned} \tag{B.25}$$

With (B.8) and (B.9) we may therefore write

$$\mathrm{tr}_S\, \Delta^{(\Omega)}[(a-\alpha),(a^\dagger-\alpha^*)]\Delta^{(\bar{\Omega})}[(a+\alpha_0),(a^\dagger+\alpha_0^*)] = (1/\pi)\,\delta^2(\alpha-\alpha_0) \tag{B.26}$$

We now turn to consider a closed expression for $\hat{S}(a,a^\dagger)$ in terms of its Ω-equivalent $S^{(\Omega)}(\alpha,\alpha^*)$.

We write from (B.4) and (B.6)

$$\hat{S}(a,a^\dagger) = \int d^2\beta\; \Omega(\beta,\beta^*)F^{(\Omega)}(\beta,\beta^*)\exp(\beta a^\dagger-\beta^* a) \tag{B.27}$$

and, introducing (B.3), this yields

$$\begin{aligned}\hat{S}(a,a^\dagger) &= \int d^2\beta\, \Omega(\beta,\beta^*)[(1/\pi)\int d^2\alpha\, S^{(\Omega)}(\alpha,\alpha^*)\exp(\beta^*\alpha-\beta\alpha^*)] \\ &\qquad\cdot\exp(\beta a^\dagger-\beta^* a) \\ &= \int d^2\alpha\, S^{(\Omega)}(\alpha,\alpha^*)(1/\pi^2)\int d^2\beta\, \Omega(\beta,\beta^*) \\ &\qquad\cdot\exp(\beta^*\alpha-\beta\alpha^*)\,\hat{D}(\beta)\end{aligned} \tag{B.28}$$

Taken with the statement of (B.21) we then have

$$\hat{S}(a,a^\dagger) = \int d^2\alpha\, S^{(\Omega)}(\alpha,\alpha^*)\Delta^{(\Omega)}[(a-\alpha),(a^\dagger-\alpha^*)] \qquad (B.29)$$

and the operators $\Delta^{(\Omega)}[(a-\alpha),\ (a^\dagger-\alpha^*)]$ appear in a capacity relating directly $\hat{S}(a,a^\dagger)$ and its Ω-equivalent $S^{(\Omega)}(\alpha,\alpha^*)$.

It is now our purpose to establish the fundamental result (2.2.8) for the trace of the product of two operators $\hat{S}_1(a,a^\dagger)$ and $\hat{S}_2(a,a^\dagger)$. We begin with the expression (B.29) and write

$$\hat{S}_1(a,a^\dagger) = \int d^2\beta\, S_1^{(\Omega)}(\beta,\beta^*)\Delta^{(\Omega)}[(a-\beta),(a^\dagger-\beta^*)] \qquad (B.30)$$

$$\hat{S}_2(a,a^\dagger) = \int d^2\beta_0\, S_2^{(\bar{\Omega})}(-\beta_0,-\beta_0^*)\Delta^{(\bar{\Omega})}[(a+\beta_0),(a^\dagger+\beta_0^*)] \qquad (B.31)$$

It follows that

$$\begin{aligned} tr_S\, \hat{S}_1(a,a^\dagger)\,\hat{S}_2(a,a^\dagger) &= \int d^2\beta\, S_1^{(\Omega)}(\beta,\beta^*)\int d^2\beta_0\, S_2^{(\bar{\Omega})}(-\beta_0,-\beta_0^*) \\ &\quad tr_S\,\Delta^{(\Omega)}[(a-\beta),(a^\dagger-\beta^*)]\Delta^{(\bar{\Omega})}[(a+\beta_0),(a^\dagger+\beta_0^*)] \end{aligned} \qquad (B.32)$$

which, taken with (B.26), and in view of (B.8), yields

$$\begin{aligned} tr_S\, \hat{S}_1(a,a^\dagger)\hat{S}_2(a,a^\dagger) &= \int d^2\beta\, S_1^{(\Omega)}(\beta,\beta^*)\int d^2\beta_0\, S_2^{(\bar{\Omega})}(\beta_0,\beta_0^*) \\ &\qquad\qquad \cdot(1/\pi)\,\delta^2(\beta-\beta_0) \\ &= (1/\pi)\int d^2\beta\, S_1^{(\Omega)}(\beta,\beta^*)\, S_2^{(\bar{\Omega})}(\beta,\beta^*) \end{aligned} \qquad (B.33)$$

Our objective is thus fulfilled in the result

$$tr_S\, \hat{S}_1(a,a^\dagger)\,\hat{S}_2(a,a^\dagger) = (1/\pi)\int d^2\beta\, S_1^{(\Omega)}(\beta,\beta^*)\, S_2^{(\bar{\Omega})}(\beta,\beta^*) \qquad (B.34)$$

We are interested in one further aspect of the phase-space calculus. For the product of two operators $\hat{S}_1(a,a^\dagger)$ and $\hat{S}_2(a,a^\dagger)$ we may define an Ω-equivalent $S_{12}^{(\Omega)}(\alpha,\alpha^*)$, such that

$$\hat{\Omega}\, S_{12}^{(\Omega)}(\alpha,\alpha^*) = \hat{S}_1(a,a^\dagger)\,\hat{S}_2(a,a^\dagger) \qquad (B.35)$$

It is to be shown then that $S_{12}^{(\Omega)}(\alpha,\alpha^*)$ may be expressed in terms of $S_1^{(\Omega)}(\alpha,\alpha^*)$ and $S_2^{(\Omega)}(\alpha,\alpha^*)$ as stated in equations (2.2.23) and (2.2.24) of section 2. We begin from (B.4) and write

$$\hat{S}_1(a,a^\dagger) = \int d^2\beta\, G_1(\beta,\beta^*)\,\hat{D}(\beta) \tag{B.36}$$

and

$$\hat{S}_2(a,a^\dagger) = \int d^2\beta_0\, G_2(\beta_0,\beta_0^*)\,\hat{D}(\beta_0) \tag{B.37}$$

Thus

$$\hat{S}_1(a,a^\dagger)\,\hat{S}_2(a,a^\dagger) = \iint d^2\beta\, d^2\beta_0\, G_1(\beta,\beta^*)G_2(\beta_0,\beta_0^*) \cdot \hat{D}(\beta)\,\hat{D}(\beta_0) \tag{B.38}$$

and invoking (B.22)

$$\hat{S}_1(a,a^\dagger)\,\hat{S}_2(a,a^\dagger) = \iint d^2\beta\, d^2\beta_0\, G_1(\beta,\beta^*)\,G_2(\beta_0,\beta_0^*) \exp[\tfrac{1}{2}(\beta\beta_0^* - \beta^*\beta_0)]\,\hat{D}(\beta+\beta_0) \tag{B.39}$$

Now we may introduce $\Omega(\beta + \beta_0,\ \beta^* + \beta_0^*)$ and $\bar{\Omega}(\beta + \beta_0,\ \beta^* + \beta_0^*)$ using the result

$$\Omega(\beta+\beta_0,\ \beta^*+\beta_0^*)\,\bar{\Omega}(\beta+\beta_0,\ \beta^*+\beta_0^*) = 1 \tag{B.40}$$

We then have

$$\hat{S}_1(a,a^\dagger)\,\hat{S}_2(a,a^\dagger) = \iint d^2\beta\, d^2\beta_0\, G_1(\beta,\beta^*)G_2(\beta_0,\beta_0^*)\bar{\Omega}(\beta+\beta_0,\beta^*+\beta_0^*) \exp[\tfrac{1}{2}(\beta\beta_0 - \beta_0\beta)]\,\Omega(\beta+\beta_0,\beta^*+\beta_0^*)\,\hat{D}(\beta+\beta_0) \tag{B.41}$$

and, noting (B.15), this yields, on the assertion of the linearity of $\hat{\Omega}$

$$\hat{S}_1(a,a^\dagger)\,\hat{S}_2(a,a^\dagger) = \hat{\Omega}\Big\{\iint d^2\beta\, d^2\beta_0\, G_1(\beta,\beta^*)G_2(\beta_0,\beta_0^*)\bar{\Omega}(\beta+\beta_0,\beta^*+\beta_0^*) \cdot \exp[\tfrac{1}{2}(\beta\beta_0^* - \beta^*\beta_0)]\exp[\alpha(\beta^*+\beta_0^*) - \alpha^*(\beta+\beta_0)]\Big\} \tag{B.42}$$

Hence, from (B.35)

$$S_{12}^{(\Omega)}(\alpha,\alpha^*) = \iint d^2\beta\, d^2\beta_0\, G_1(\beta,\beta^*)\, G_2(\beta_0,\beta_0^*)\, \bar{\Omega}(\beta+\beta_0,\beta^*+\beta_0^*)\exp[\tfrac{1}{2}(\beta\beta_0^*-\beta^*\beta_0)]\, \exp[\alpha(\beta^*+\beta_0^*)-\alpha^*(\beta+\beta_0)] \tag{B.43}$$

A simplification of this expression is required, and therefore in line with (B.40), we introduce $\Omega(\beta,\beta^*)\bar{\Omega}(\beta,\beta^*) = 1$ and $\Omega(\beta_0,\beta_0^*)\bar{\Omega}(\beta_0,\beta_0^*) = 1$ and write

$$S_{12}^{(\Omega)}(\alpha,\alpha^*) = \iint d^2\beta\, d^2\beta_0 [G_1(\beta,\beta^*)\bar{\Omega}(\beta,\beta^*)\exp(\alpha\beta^*-\alpha^*\beta)]\, \Omega(\beta,\beta^*)\bar{\Omega}(\beta+\beta_0,\beta^*+\beta_0^*)\Omega(\beta_0,\beta_0^*)\exp[\tfrac{1}{2}(\beta\beta_0^*-\beta^*\beta_0)]\, [G_2(\beta_0,\beta_0^*)\bar{\Omega}(\beta_0,\beta_0^*)\exp(\alpha\beta_0^*-\alpha^*\beta_0)] \tag{B.44}$$

We then introduce differential operators $\overleftarrow{\frac{\partial}{\partial\alpha}}$, $\overrightarrow{\frac{\partial}{\partial\alpha}}$ and $\overleftarrow{\frac{\partial}{\partial\alpha^*}}$, $\overrightarrow{\frac{\partial}{\partial\alpha^*}}$ where the arrows indicate respectively differentiation of the function to the right and left of the operator. Clearly

$$\Omega(\beta,\beta^*) = \exp(\alpha\beta^*-\alpha^*\beta)\,\Omega\Big(-\overleftarrow{\frac{\partial}{\partial\alpha^*}},\overleftarrow{\frac{\partial}{\partial\alpha}}\Big) \tag{B.45}$$

$$\Omega(\beta_0,\beta_0^*) = \Omega\Big(-\overrightarrow{\frac{\partial}{\partial\alpha^*}},\overrightarrow{\frac{\partial}{\partial\alpha}}\Big)\exp(\alpha\beta_0^*-\alpha^*\beta_0) \tag{B.46}$$

$$\bar{\Omega}(\beta+\beta_0,\beta^*+\beta_0^*) = \exp(\alpha\beta^*-\alpha^*\beta)\,\bar{\Omega}\Big(-\overleftarrow{\frac{\partial}{\partial\alpha^*}}-\overrightarrow{\frac{\partial}{\partial\alpha^*}},\ \overleftarrow{\frac{\partial}{\partial\alpha}}+\overrightarrow{\frac{\partial}{\partial\alpha}}\Big)\exp(\alpha\beta_0^*-\alpha^*\beta_0) \tag{B.47}$$

$$\exp[\tfrac{1}{2}(\beta\beta_0^*-\beta^*\beta_0)] = \exp(\alpha\beta^*-\alpha^*\beta)\exp\Big[\tfrac{1}{2}\Big(-\overleftarrow{\frac{\partial}{\partial\alpha^*}}\overrightarrow{\frac{\partial}{\partial\alpha}}+\overleftarrow{\frac{\partial}{\partial\alpha}}\overrightarrow{\frac{\partial}{\partial\alpha^*}}\Big)\Big]\exp(\alpha\beta_0^*-\alpha^*\beta_0) \tag{B.48}$$

Omitting the tedious algebra, it follows, for filter functions given by (2.2.7), that

$$\Omega(\beta,\beta^*)\bar{\Omega}(\beta+\beta_0,\beta^*+\beta_0^*)\Omega(\beta_0,\beta_0^*)\exp[\tfrac{1}{2}(\beta\beta_0^*-\beta^*\beta_0)] = \exp(\alpha\beta^*-\alpha^*\beta)\exp[\overleftrightarrow{\Lambda}^{(\Omega)}(\alpha,\alpha^*)]\exp(\alpha\beta_0^*-\alpha^*\beta_0) \tag{B.49}$$

where

$$\overleftrightarrow{\Lambda}^{(\Omega)}(\alpha,\alpha^*) = -2\nu \frac{\overleftarrow{\partial}}{\partial\alpha}\frac{\overrightarrow{\partial}}{\partial\alpha} - 2\mu \frac{\overleftarrow{\partial}}{\partial\alpha^*}\frac{\overrightarrow{\partial}}{\partial\alpha^*} + (\lambda+\tfrac{1}{2})\frac{\overleftarrow{\partial}}{\partial\alpha}\frac{\overrightarrow{\partial}}{\partial\alpha^*} + (\lambda-\tfrac{1}{2})\frac{\overleftarrow{\partial}}{\partial\alpha^*}\frac{\overrightarrow{\partial}}{\partial\alpha} \qquad \text{(B.50)}$$

Thus, from (B.44)

$$S_{12}^{(\Omega)}(\alpha,\alpha^*) = \int d^2\beta\, G_1(\beta,\beta^*)\bar{\Omega}(\beta,\beta^*)\exp(\alpha\beta^*-\alpha^*\beta) \exp[\overleftrightarrow{\Lambda}^{(\Omega)}(\alpha,\alpha^*)] \int d^2\beta_0\, G_2(\beta_0,\beta_0^*)\bar{\Omega}(\beta_0,\beta_0^*)\exp(\alpha\beta_0^*-\alpha^*\beta_0) \qquad \text{(B.51)}$$

In view of equations (B.2) and (B.6) we have therefore completed the proof, and

$$S_{12}^{(\Omega)}(\alpha,\alpha^*) = S_1^{(\Omega)}(\alpha,\alpha^*)\exp[\overleftrightarrow{\Lambda}^{(\Omega)}(\alpha,\alpha^*)]\,S_2^{(\Omega)}(\alpha,\alpha^*) \qquad \text{(B.52)}$$

APPENDIX C

TWO DIFFERENTIAL OPERATOR IDENTITIES

We present in this appendix the proofs for two identities, applied in section 2.3 in reference to the phase-space integrals for two-time averages. These two results appear in the text as equations (2.3.8) and (2.3.9) and read as follows:

$$\sum_{n=0}^{\infty} (-1)^n/n! \cdot \frac{\partial^n}{\partial\alpha^{*n}} \left(\alpha^{*q} \frac{\partial^n}{\partial\alpha^n}\right) = \left(\alpha^* - \frac{\partial}{\partial\alpha}\right)^q \exp\left(-\frac{\partial^2}{\partial\alpha\,\partial\alpha^*}\right) \tag{C.1}$$

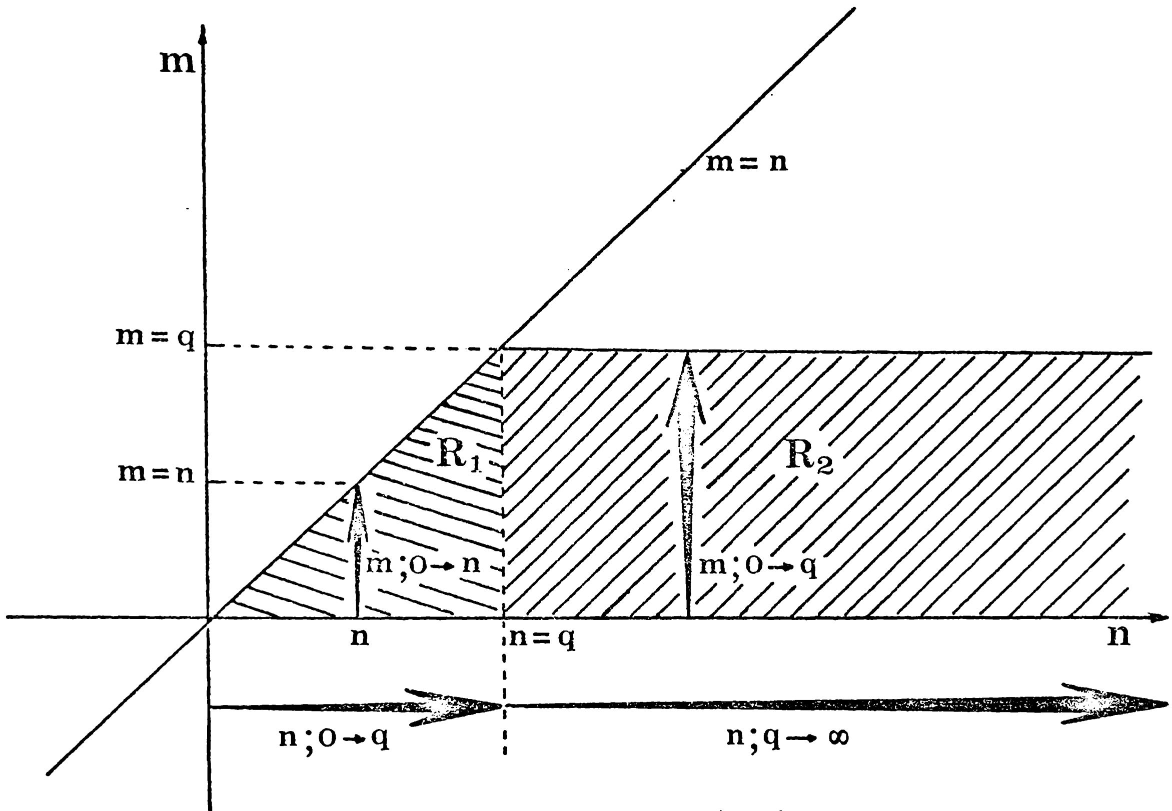
and

$$\left(\alpha^* - \frac{\partial}{\partial\alpha}\right)^p \phi_1(\alpha,\alpha^*)\,\phi_2(\alpha,\alpha^*) = \sum_{r=0}^{p} [(-1)^r p!/r!(p-r)!] \left[\frac{\partial^r}{\partial\alpha^r}\phi_1(\alpha,\alpha^*)\right]\left[\left(\alpha^* - \frac{\partial}{\partial\alpha}\right)^{p-r}\phi_2(\alpha,\alpha^*)\right] \tag{C.2}$$

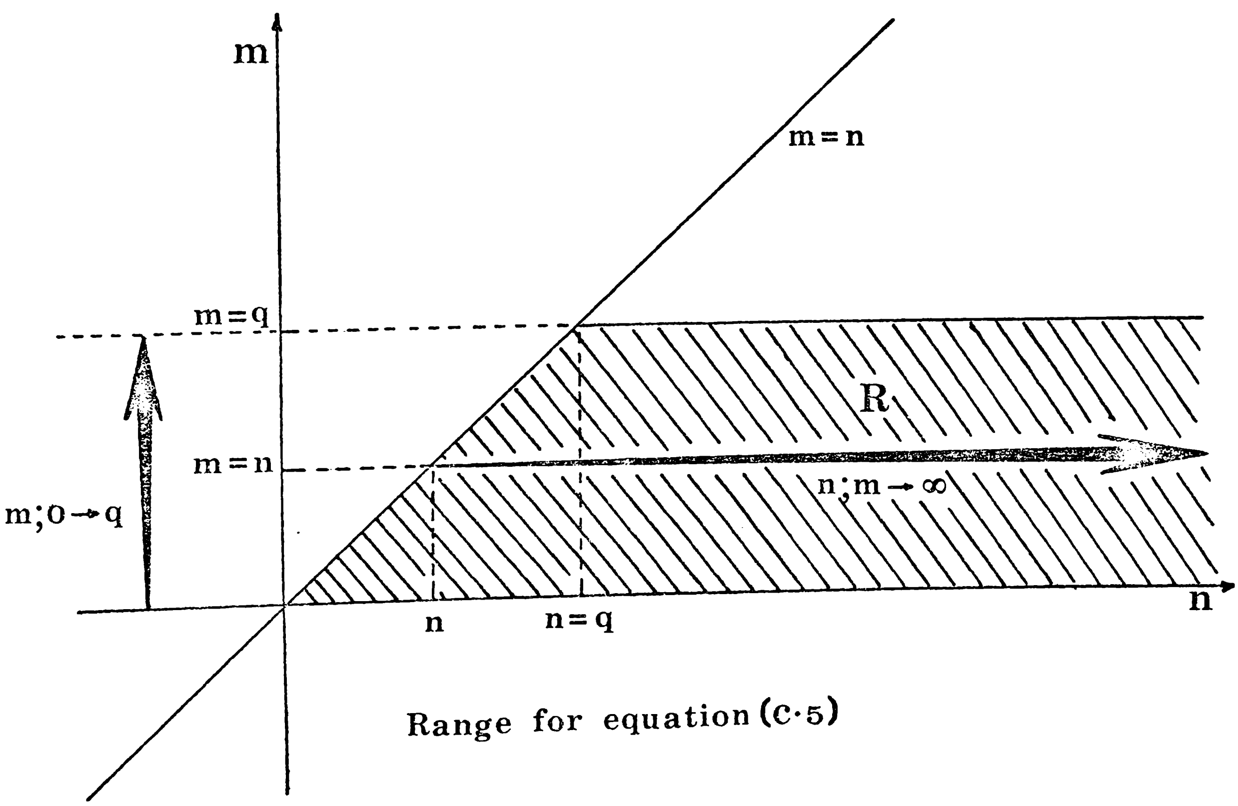
We begin with (C.1) for which we may write

$$\sum_{n=0}^{\infty} (-1)^n/n! \cdot \frac{\partial^n}{\partial\alpha^{*n}} \left(\alpha^{*q} \frac{\partial^n}{\partial\alpha^n}\right) = \sum_{n=0}^{\infty} \sum_{m=0}^{n} [(-1)^n/m!(n-m)!] \cdot \left(\frac{\partial^m}{\partial\alpha^{*n}}\alpha^{*q}\right) \frac{\partial^{n-m}}{\partial\alpha^{*n-m}} \frac{\partial^n}{\partial\alpha^n} \tag{C.3}$$

where we have simply applied that general result of differential calculus relating to the repeated differentiation of a product. The form revealed by (C.3) clearly dictates that we divide the range of summation into two. With $n; 0 \to q$, $m; 0 \to n$ we set R_1, and with $n; q+1 \to \infty$, $m; 0 \to q$ R_2 is formed, as shown schematically in Fig.(C.1). We then write (C.3) in the



Range for equation (c·4)



Range for equation (c·5)

Fig (c·1) Conversion of summation ranges between equations (c·4) and (c·5).

form

$$\sum_{n=0}^{\infty}(-1)^n/n!\cdot\frac{\partial^n}{\partial\alpha^{*n}}\left(\alpha^{*q}\frac{\partial^n}{\partial\alpha^n}\right)=\left(\sum_{n=0}^{q}\sum_{m=0}^{\infty}+\sum_{n=q+1}^{\infty}\sum_{m=0}^{q}\right)$$

$$[(-1)^n q!/m!(n-m)!(q-m)!]\alpha^{*q-m}\frac{\partial^m}{\partial\alpha^m}\left(\frac{\partial^2}{\partial\alpha\partial\alpha^*}\right)^{n-m} \quad (C.4)$$

It is now a simplification to view the combined range $R_1 + R_2$ as the single range R, for which $m;0\to q$, $n;m\to\infty$. We may then write

$$\sum_{n=0}^{\infty}(-1)^n/n!\cdot\frac{\partial^n}{\partial\alpha^{*n}}\left(\alpha^{*q}\frac{\partial^n}{\partial\alpha^n}\right)=\sum_{m=0}^{q}\sum_{n=m}^{\infty}[(-1)^n q!/m!(n-m)!(q-m)!]$$

$$\cdot\alpha^{*q-m}\frac{\partial^n}{\partial\alpha^m}\left(\frac{\partial^2}{\partial\alpha\partial\alpha^*}\right)^{n-m} \quad (C.5)$$

Further, defining $r = n - m$, we have

$$\sum_{n=0}^{\infty}(-1)^n/n!\cdot\frac{\partial^n}{\partial\alpha^{*n}}\left(\alpha^{*q}\frac{\partial^n}{\partial\alpha^n}\right)=\sum_{m=0}^{q}\sum_{r=0}^{\infty}[(-1)^{m+r}q!/m!r!(q-m)!]$$

$$\alpha^{*q-m}\frac{\partial^m}{\partial\alpha^m}\left(\frac{\partial^2}{\partial\alpha\partial\alpha^*}\right)^r$$

$$=\left[\sum_{m=0}^{q}(-1)^m q!/m!(q-m)!\cdot\alpha^{*q-m}\frac{\partial^m}{\partial\alpha^m}\right]\left[\sum_{r=0}^{\infty}(-1)^r/r!\left(\frac{\partial^2}{\partial\alpha\partial\alpha^*}\right)^r\right] \quad (C.6)$$

$$=\left(\alpha^*-\frac{\partial}{\partial\alpha}\right)^q\exp\left(-\frac{\partial^2}{\partial\alpha\partial\alpha^*}\right)$$

and our proof is completed.

We turn now to (C.2) and its verification by induction. To begin, since

$$\left(\alpha^*-\frac{\partial}{\partial\alpha}\right)\phi_1(\alpha,\alpha^*)\phi_2(\alpha,\alpha^*) = \phi_1(\alpha,\alpha^*)\left(\alpha^*-\frac{\partial}{\partial\alpha}\right)\phi_2(\alpha,\alpha^*)$$

$$-\left[\frac{\partial}{\partial\alpha}\phi_1(\alpha,\alpha^*)\right]\phi_2(\alpha,\alpha^*)$$

$$=\sum_{r=0}^{1}[(-1)^r/r!(1-r)!]\left[\frac{\partial^r}{\partial\alpha^r}\phi_1(\alpha,\alpha^*)\right]$$

$$\cdot\left[\left(\alpha^*-\frac{\partial}{\partial\alpha}\right)^{1-r}\phi_2(\alpha,\alpha^*)\right] \quad (C.7)$$

this statement is clearly true for $p = 1$. We now adopt, by hypothesis, its validity for an arbitrary p, and consider then the statement for p + 1. In this instance we therefore take

$$\left(\alpha^* - \frac{\partial}{\partial\alpha}\right)^{p+1} \phi_1(\alpha,\alpha^*)\phi_2(\alpha,\alpha^*) = \left(\alpha^* - \frac{\partial}{\partial\alpha}\right)\left[\left(\alpha^* - \frac{\partial}{\partial\alpha}\right)^p \phi_1(\alpha,\alpha^*)\phi_2(\alpha,\alpha^*)\right] \quad \text{(C.8)}$$

which, by hypothesis, yields

$$\begin{aligned}
&\left(\alpha^* - \frac{\partial}{\partial\alpha}\right)^{p+1} \phi_1(\alpha,\alpha^*)\phi_2(\alpha,\alpha^*) \\
&= \left(\alpha^* - \frac{\partial}{\partial\alpha}\right)\sum_{r=0}^{p}\left[(-1)^r p!/r!(p-r)!\right]\left[\frac{\partial^r}{\partial\alpha^r}\phi_1(\alpha,\alpha^*)\right]\left[\left(\alpha^* - \frac{\partial}{\partial\alpha}\right)\phi_2(\alpha,\alpha^*)\right] \\
&= \sum_{r=0}^{p}\left[(-1)^r p!/r!(p-r)!\right]\left\{\left[\frac{\partial^r}{\partial\alpha^r}\phi_1(\alpha,\alpha^*)\right]\left[\left(\alpha^* - \frac{\partial}{\partial\alpha}\right)^{p+1-r}\phi_2(\alpha,\alpha^*)\right]\right. \\
&\qquad\qquad \left. - \left[\frac{\partial^{r+1}}{\partial\alpha^{r+1}}\phi_1(\alpha,\alpha^*)\right]\left[\left(\alpha^* - \frac{\partial}{\partial\alpha}\right)^{p-r}\phi_2(\alpha,\alpha^*)\right]\right\} \\
&= \sum_{r=0}^{p}\left[(-1)^r p!/r!(p-r)!\right]\left[\frac{\partial^r}{\partial\alpha^r}\phi_1(\alpha,\alpha^*)\right]\left[\left(\alpha^* - \frac{\partial}{\partial\alpha}\right)^{p+1-r}\phi_2(\alpha,\alpha^*)\right] \\
&\quad + \sum_{s=1}^{p+1}\left[(-1)^s p!/(s-1)!(p+1-s)!\right]\left[\frac{\partial^s}{\partial\alpha^s}\phi_1(\alpha,\alpha^*)\right] \\
&\qquad\qquad \left[\left(\alpha^* - \frac{\partial}{\partial\alpha}\right)^{p+1-s}\phi_2(\alpha,\alpha^*)\right] \quad \text{(C.9)}
\end{aligned}$$

where we have defined s = r + 1 for the expression of the second term. Now the first and last terms of the respective summations may be extracted so we view the form

$$\begin{aligned}
&\left(\alpha^* - \frac{\partial}{\partial\alpha}\right)^{p+1} \phi_1(\alpha,\alpha^*)\phi_2(\alpha,\alpha^*) = \phi_1(\alpha,\alpha^*)\left(\alpha^* - \frac{\partial}{\partial\alpha}\right)^{p+1}\phi_2(\alpha,\alpha^*) \\
&\quad + \sum_{r=1}^{p}(-1)^r p!\left[1/r!(p-r)! + 1/(r-1)!(p+1-r)!\right]\left[\frac{\partial^r}{\partial\alpha^r}\phi_1(\alpha,\alpha^*)\right] \\
&\quad \left[\left(\alpha^* - \frac{\partial}{\partial\alpha}\right)^{p+1-r}\phi_2(\alpha,\alpha^*)\right] + (-1)^{p+1}\left[\frac{\partial^{p+1}}{\partial\alpha^{p+1}}\phi_1(\alpha,\alpha^*)\right]\phi_2(\alpha,\alpha^*) \quad \text{(C.10)}
\end{aligned}$$

Then, since

$$1/r!(p-r)! + 1/(r-1)!(p+1-r)! = (p+1)/r!(p+1-r)! \quad \text{(C.11)}$$

it readily follows that

$$\left(\alpha^* - \frac{\partial}{\partial \alpha}\right)^{p+1} \phi_1(\alpha,\alpha^*)\,\phi_2(\alpha,\alpha^*) = \sum_{r=0}^{p+1} \left[(-1)^r (p+1)!/r!\,(p+1-r)!\right] \left[\frac{\partial^r}{\partial \alpha^r}\phi_1(\alpha,\alpha^*)\right]\left[\left(\alpha^* - \frac{\partial}{\partial \alpha}\right)^{p+1-r}\phi_2(\alpha,\alpha^*)\right] \tag{C.12}$$

Having thus shown that the truth of (C.2) for p implies also its truth for (p + 1), by induction, our proof is completed.

APPENDIX D

TWO APPLICATIONS OF DETAILED BALANCE CONDITIONS

We present in this appendix the manipulative algebra belonging to the application of full phase-space detailed balance conditions to the harmonic oscillator Fokker-Plank equation in section IV 6a, and classical detailed balance conditions to the laser Fokker-Plank equation in section IV 6b.

a) *Full Phase-Space Detailed Balance Conditions for the Harmonic Oscillator*

We are concerned here with the detailed balance condition (4.6.5) for the harmonic oscillator. This reads

$$\begin{aligned} P_{ss}^{(A)}(\alpha,\alpha^*)\exp\left(-\overleftarrow{\frac{\partial}{\partial\alpha^*}}\overrightarrow{\frac{\partial}{\partial\alpha}}\right)\overset{+}{\mathcal{L}}^{(N)}(\alpha,\alpha^*) \\ = \mathcal{L}^{(A)}(\alpha^*,\alpha)\,P_{ss}^{(A)}(\alpha,\alpha^*)\exp\left(-\overleftarrow{\frac{\partial}{\partial\alpha^*}}\overrightarrow{\frac{\partial}{\partial\alpha}}\right) \end{aligned} \tag{D.1}$$

where

$$P_{ss}^{(A)}(\alpha,\alpha^*) = \bar{n}^{-1}\exp(-\alpha\alpha^*/\bar{n}) \tag{D.2}$$

and

$$\mathcal{L}^{(A)}(\alpha^*,\alpha) = (\gamma/2+i\omega_0)\frac{\partial}{\partial\alpha^*}\alpha^* + (\gamma/2-i\omega_0)\frac{\partial}{\partial\alpha}\alpha + \gamma\bar{n}\frac{\partial^2}{\partial\alpha\partial\alpha^*} \tag{D.3}$$

$$\overset{+}{\mathcal{L}}^{(N)}(\alpha,\alpha^*) = -(\gamma/2+i\omega_0)\alpha\frac{\partial}{\partial\alpha} - (\gamma/2-i\omega_0)\alpha^*\frac{\partial}{\partial\alpha^*} + \gamma(\bar{n}+1)\frac{\partial^2}{\partial\alpha\partial\alpha^*} \tag{D.4}$$

The fulfilment of this condition will be established in the demonstration that both sides of (D.1) may adopt the form

$$P_{ss}^{(A)}(\alpha,\alpha^*)\sum_{n=0}^{\infty}(1/n!)(\alpha/\bar{n})^n\Big[-(\gamma/2+i\omega_0)(\bar{n}+1)/\bar{n}\cdot\alpha\frac{\partial}{\partial\alpha} \\ -(\gamma/2-i\omega_0)\alpha^*\frac{\partial}{\partial\alpha^*}+\gamma(\bar{n}+1)\frac{\partial^2}{\partial\alpha\partial\alpha^*}\Big]\frac{\partial^n}{\partial\alpha^n} \tag{D.5}$$

We take firstly the left hand side of (D.1), for which, on expanding the exponential, we may write

$$P_{ss}^{(A)}(\alpha,\alpha^*)\exp\left(-\frac{\overleftarrow{\partial}}{\partial\alpha^*}\frac{\overrightarrow{\partial}}{\partial\alpha}\right)\mathcal{L}^{(N)^+}(\alpha,\alpha^*) \\ =\sum_{n=0}^{\infty}(-1)^n/n!\cdot\Big[\frac{\partial^n}{\partial\alpha^{*n}}P_{ss}^{(A)}(\alpha,\alpha^*)\Big]\frac{\partial^n}{\partial\alpha^n}\mathcal{L}^{(N)^+}(\alpha,\alpha^*) \tag{D.6}$$

Then, with (D.2), clearly

$$\frac{\partial^n}{\partial\alpha^{*n}}P_{ss}^{(A)}(\alpha,\alpha^*)=(-\alpha/\bar{n})^n P_{ss}^{(A)}(\alpha,\alpha^*) \tag{D.7}$$

Further, from (D.4), and using the identity

$$\frac{\partial^n}{\partial\alpha^n}\alpha = n\frac{\partial^{n-1}}{\partial\alpha^{n-1}}+\alpha\frac{\partial^n}{\partial\alpha^n} \tag{D.8}$$

we have

$$\frac{\partial^n}{\partial\alpha^n}\mathcal{L}^{(N)^+}(\alpha,\alpha^*)=\Big[-(\gamma/2+i\omega_0)(n+\alpha\frac{\partial}{\partial\alpha})-(\gamma/2-i\omega_0)\alpha^*\frac{\partial}{\partial\alpha^*} \\ +\gamma(\bar{n}+1)\frac{\partial^2}{\partial\alpha\partial\alpha^*}\Big]\frac{\partial^n}{\partial\alpha^n} \tag{D.9}$$

Thus, (D.6) becomes

$$P_{ss}^{(A)}(\alpha,\alpha^*)\exp\left(-\frac{\overleftarrow{\partial}}{\partial\alpha^*}\frac{\overrightarrow{\partial}}{\partial\alpha}\right)\mathcal{L}^{(N)^+}(\alpha,\alpha^*) \\ =P_{ss}^{(A)}(\alpha,\alpha^*)\sum_{n=0}^{\infty}(1/n!)(\alpha/\bar{n})^n \\ \cdot\Big[-(\gamma/2+i\omega_0)(n+\alpha\frac{\partial}{\partial\alpha})-(\gamma/2-i\omega_0)\alpha^*\frac{\partial}{\partial\alpha^*}+\gamma(\bar{n}+1)\frac{\partial^2}{\partial\alpha\partial\alpha^*}\Big]\frac{\partial^n}{\partial\alpha^n} \tag{D.10}$$

Now one further step is required to achieve the result (D.5). We find

$$\sum_{n=0}^{\infty}(1/n!)(\alpha/\bar{n})^n n\frac{\partial^n}{\partial\alpha^n}=\sum_{n=1}^{\infty}[1/(n-1)!](\alpha/\bar{n})^{n-1}(1/\bar{n})\alpha\frac{\partial}{\partial\alpha}\frac{\partial^{n-1}}{\partial\alpha^{n-1}} \\ =\sum_{n=0}^{\infty}(1/n!)(\alpha/\bar{n})^n(1/\bar{n})\alpha\frac{\partial}{\partial\alpha}\frac{\partial^n}{\partial\alpha^n} \tag{D.11}$$

and with this

$$\overset{(A)}{P}_{ss}(\alpha,\alpha^*)\exp\left(-\frac{\overleftarrow{\partial}}{\partial\alpha^*}\frac{\overrightarrow{\partial}}{\partial\alpha}\right)\overset{(N)^+}{\mathcal{L}}(\alpha,\alpha^*)$$

$$=\overset{(A)}{P}_{ss}(\alpha,\alpha^*)\sum_{n=0}^{\infty}(1/n!)(\alpha/\bar{n})^n$$

$$[-(\gamma/2+i\omega_0)(\bar{n}+1)/\bar{n}\cdot\alpha\frac{\partial}{\partial\alpha}-(\gamma/2-i\omega_0)\alpha^*\frac{\partial}{\partial\alpha^*}+\gamma(\bar{n}+1)\frac{\partial^2}{\partial\alpha\partial\alpha^*}]\frac{\partial^n}{\partial\alpha^n} \tag{D.12}$$

Turning now to the right hand side of (D.1), with an expansion of the exponential, we write

$$\overset{(A)}{\mathcal{L}}(\alpha^*,\alpha)\overset{(A)}{P}_{ss}(\alpha,\alpha^*)\exp\left(-\frac{\overleftarrow{\partial}}{\partial\alpha^*}\frac{\overrightarrow{\partial}}{\partial\alpha}\right)$$

$$=\sum_{n=0}^{\infty}(-1)^n/n!\,\overset{(A)}{\mathcal{L}}(\alpha^*,\alpha)\left[\frac{\partial^n}{\partial\alpha^{*n}}\overset{(A)}{P}_{ss}(\alpha,\alpha^*)\right]\frac{\partial^n}{\partial\alpha^n} \tag{D.13}$$

which, with (D.7), becomes

$$\overset{(A)}{\mathcal{L}}(\alpha^*,\alpha)\overset{(A)}{P}_{ss}(\alpha,\alpha^*)\exp\left(-\frac{\overleftarrow{\partial}}{\partial\alpha^*}\frac{\overrightarrow{\partial}}{\partial\alpha}\right)$$

$$=\sum_{n=0}^{\infty}(1/n!)\,\overset{(A)}{\mathcal{L}}(\alpha^*,\alpha)(\alpha/\bar{n})^n\,\overset{(A)}{P}_{ss}(\alpha,\alpha^*)\frac{\partial^n}{\partial\alpha^n} \tag{D.14}$$

Then, from (D.3) it easily follows that

$$\overset{(A)}{\mathcal{L}}(\alpha^*,\alpha)(\alpha/\bar{n})^n=(\alpha/\bar{n})^n[(\gamma/2-i\omega_0)(n+1+\alpha\frac{\partial}{\partial\alpha})+(\gamma/2+i\omega_0)$$

$$(1+\alpha^*\frac{\partial}{\partial\alpha^*})+\gamma\bar{n}(n/\alpha\cdot\frac{\partial}{\partial\alpha^*}+\frac{\partial^2}{\partial\alpha\partial\alpha^*})] \tag{D.15}$$

and from this, with (D.2)

$$\overset{(A)}{\mathcal{L}}(\alpha^*,\alpha)(\alpha/\bar{n})^n\,\overset{(A)}{P}_{ss}(\alpha,\alpha^*)=\overset{(A)}{P}_{ss}(\alpha,\alpha^*)(\alpha/\bar{n})^n[(\gamma/2-i\omega_0)$$

$$\cdot(n+1-\alpha\alpha^*/\bar{n}+\alpha\frac{\partial}{\partial\alpha})+(\gamma/2+i\omega_0)(1-\alpha\alpha^*/\bar{n}+\alpha^*\frac{\partial}{\partial\alpha^*})$$

$$+\gamma\bar{n}(-n+1/\bar{n}+\alpha\alpha^*/\bar{n}^2+n/\alpha\cdot\frac{\partial}{\partial\alpha^*}-\alpha^*/\bar{n}\frac{\partial}{\partial\alpha^*}-\alpha/\bar{n}\frac{\partial}{\partial\alpha}+\frac{\partial^2}{\partial\alpha\partial\alpha^*})$$

$$= \overset{(A)}{P}_{ss}(\alpha,\alpha^*)(\alpha/\bar{n})^n \left[-(\gamma/2+i\omega_0)\left(n+\alpha\frac{\partial}{\partial\alpha}\right)-(\gamma/2-i\omega_0)\,\alpha^*\frac{\partial}{\partial\alpha}\right.$$

$$\left.+\gamma\bar{n}\left(n/\alpha\cdot\frac{\partial}{\partial\alpha^*}+\frac{\partial^2}{\partial\alpha\partial\alpha^*}\right)\right] \tag{D.16}$$

Now, in a like manner to the result (D.11)

$$\sum_{n=0}^{\infty}(1/n!)(\alpha/\bar{n})^n\,\gamma\bar{n}(n/\alpha)\frac{\partial}{\partial\alpha^*}\frac{\partial^n}{\partial\alpha^n}$$

$$=\sum_{n=1}^{\infty}[1/(n-1)!](\alpha/\bar{n})^{n-1}\gamma\frac{\partial^2}{\partial\alpha\partial\alpha^*}\frac{\partial^{n-1}}{\partial\alpha^{n-1}}$$

$$=\sum_{n=0}^{\infty}(1/n!)(\alpha/\bar{n})^n\,\gamma\frac{\partial^2}{\partial\alpha\partial\alpha^*}\frac{\partial^n}{\partial\alpha^n} \tag{D.17}$$

Hence, substituting (D.16) into (D.14) and drawing on the results (D.11) and (D.17), we find

$$\overset{(A)}{\mathcal{L}}(\alpha^*,\alpha)\overset{(A)}{P}_{ss}(\alpha,\alpha^*)\exp\left(-\frac{\overleftarrow{\partial}}{\partial\alpha^*}\frac{\overrightarrow{\partial}}{\partial\alpha}\right)=\overset{(A)}{P}_{ss}(\alpha,\alpha^*)\sum_{n=0}^{\infty}(1/n!)(\alpha/\bar{n})^n$$

$$\cdot\left[-(\gamma/2+i\omega_0)\frac{\bar{n}+1}{\bar{n}}\alpha\frac{\partial}{\partial\alpha}-(\gamma/2-i\omega_0)\alpha^*\frac{\partial}{\partial\alpha^*}+\gamma(\bar{n}+1)\frac{\partial^2}{\partial\alpha\partial\alpha^*}\right]\frac{\partial^n}{\partial\alpha^n} \tag{D.18}$$

In this our objective is achieved since the results (D.12) and (D.18) have both the right and left hand sides of (D.1) admitting the form (D.5).

b) *Classical Detailed Balance Conditions for the Laser Fokker-Plank Equation*

For the laser Fokker-Plank equation, we have from (4.6.21), (4.6.22) and (4.6.20)

$$\overset{(A)}{\mathcal{L}}{}^{+}(\alpha,\alpha^*)=(g-i\omega_0-|\alpha|^2)\alpha\frac{\partial}{\partial\alpha}+(g+i\omega_0-|\alpha|^2)\alpha^*\frac{\partial}{\partial\alpha^*}$$

$$+4\frac{\partial^2}{\partial\alpha\partial\alpha^*} \tag{D.19}$$

$$\overset{(A)}{\mathcal{L}}(\alpha^*,\alpha)=-\frac{\partial}{\partial\alpha^*}(g-i\omega_0-|\alpha|^2)\alpha^*-\frac{\partial}{\partial\alpha}(g+i\omega_0-|\alpha|^2)\alpha$$

$$+4\frac{\partial^2}{\partial\alpha\partial\alpha^*} \tag{D.20}$$

and

$$P_{ss}^{(A)}(\alpha,\alpha^*) = (N/2\pi)\exp[-\tfrac{1}{4}(|\alpha|^2-g)^2] \tag{D.21}$$

It is our purpose here to show that the classical detailed balance condition

$$P_{ss}^{(A)}(\alpha,\alpha^*)\mathcal{L}^{(A)^+}(\alpha,\alpha^*) = \mathcal{L}^{(A)}(\alpha^*,\alpha)P_{ss}^{(A)}(\alpha,\alpha^*) \tag{D.22}$$

is fulfilled.

We begin with the right hand side of (D.22), and, on substituting from (D.20), may write

$$\begin{aligned}\mathcal{L}^{(A)}(\alpha^*,\alpha)P_{ss}^{(A)}(\alpha,\alpha^*) &= -\frac{\partial}{\partial\alpha^*}[(g-i\omega_0-|\alpha|^2)\alpha^* P_{ss}^{(A)}(\alpha,\alpha^*)]\\ &\quad -P_{ss}^{(A)}(\alpha,\alpha^*)(g-i\omega_0-|\alpha|^2)\alpha^*\frac{\partial}{\partial\alpha^*}\\ &\quad -\frac{\partial}{\partial\alpha}[(g+i\omega_0-|\alpha|^2)\alpha P_{ss}^{(A)}(\alpha,\alpha^*)]\\ &\quad -P_{ss}^{(A)}(\alpha,\alpha^*)(g+i\omega_0-|\alpha|^2)\alpha\frac{\partial}{\partial\alpha}\\ &\quad +4\frac{\partial^2}{\partial\alpha\partial\alpha^*}P_{ss}^{(A)}(\alpha,\alpha^*)\end{aligned} \tag{D.23}$$

Now, we may expand the last term in this expression to obtain

$$\begin{aligned}\frac{\partial^2}{\partial\alpha\partial\alpha^*}P_{ss}^{(A)}(\alpha,\alpha^*) &= [\frac{\partial^2}{\partial\alpha\partial\alpha^*}P_{ss}^{(A)}(\alpha,\alpha^*)] + [\frac{\partial}{\partial\alpha^*}P_{ss}^{(A)}(\alpha,\alpha^*)]\frac{\partial}{\partial\alpha}\\ &\quad + [\frac{\partial}{\partial\alpha}P_{ss}^{(A)}(\alpha,\alpha^*)]\frac{\partial}{\partial\alpha^*} + P_{ss}^{(A)}(\alpha,\alpha^*)\frac{\partial^2}{\partial\alpha\partial\alpha^*}\end{aligned} \tag{D.24}$$

Then, invoking (D.21), we have

$$4[\frac{\partial}{\partial\alpha^*}P_{ss}^{(A)}(\alpha,\alpha^*)]\frac{\partial}{\partial\alpha} = 2P_{ss}^{(A)}(\alpha,\alpha^*)(g-|\alpha|^2)\alpha\frac{\partial}{\partial\alpha} \tag{D.25}$$

$$4[\frac{\partial}{\partial\alpha}P_{ss}^{(A)}(\alpha,\alpha^*)]\frac{\partial}{\partial\alpha^*} = 2P_{ss}^{(A)}(\alpha,\alpha^*)(g-|\alpha|^2)\alpha^*\frac{\partial}{\partial\alpha^*} \tag{D.26}$$

while

$$4\left[\frac{\partial^2}{\partial\alpha\partial\alpha^*}\overset{(A)}{P}_{ss}(\alpha,\alpha^*)\right] = \frac{\partial}{\partial\alpha}(g-|\alpha|^2)\alpha\overset{(A)}{P}_{ss}(\alpha,\alpha^*) + \frac{\partial}{\partial\alpha^*}(g-|\alpha|^2)\alpha^*\overset{(A)}{P}_{ss}(\alpha,\alpha^*) \qquad (D.27)$$

On substitution, these results give for (D.23)

$$\overset{(A)}{\mathcal{L}}(\alpha^*,\alpha)\overset{(A)}{P}_{ss}(\alpha,\alpha^*) = \overset{(A)}{P}_{ss}(\alpha,\alpha^*)\Big[(g-i\omega_0-|\alpha|^2)\alpha\frac{\partial}{\partial\alpha} + (g+i\omega_0-|\alpha|^2)\alpha^*\frac{\partial}{\partial\alpha^*} + 4\frac{\partial^2}{\partial\alpha\partial\alpha^*}\Big] + i\omega_0\left[\frac{\partial}{\partial\alpha^*}\alpha^*\overset{(A)}{P}_{ss}(\alpha,\alpha^*) - \frac{\partial}{\partial\alpha}\alpha\overset{(A)}{P}_{ss}(\alpha,\alpha^*)\right] \qquad (D.28)$$

and it remains only to note that

$$\frac{\partial}{\partial\alpha^*}\alpha^*\overset{(A)}{P}_{ss}(\alpha,\alpha^*) = \overset{(A)}{P}_{ss}(\alpha,\alpha^*)\left[1+\tfrac{1}{2}(g-|\alpha|^2)\alpha^*\alpha\right] \qquad (D.29)$$

$$\frac{\partial}{\partial\alpha}\alpha\overset{(A)}{P}_{ss}(\alpha,\alpha^*) = \overset{(A)}{P}_{ss}(\alpha,\alpha^*)\left[1+\tfrac{1}{2}(g-|\alpha|^2)\alpha\alpha^*\right] \qquad (D.30)$$

whence the final term of (D.28) vanishes leaving

$$\overset{(A)}{\mathcal{L}}(\alpha^*,\alpha)\overset{(A)}{P}_{ss}(\alpha,\alpha^*) = \overset{(A)}{P}_{ss}(\alpha,\alpha^*)\Big[(g-i\omega_0-|\alpha|^2)\alpha\frac{\partial}{\partial\alpha} + (g+i\omega_0-|\alpha|^2)\alpha^*\frac{\partial}{\partial\alpha^*} + 4\frac{\partial^2}{\partial\alpha\partial\alpha^*}\Big] \qquad (D.31)$$

This, in the light of (D.19), completes our proof of the fulfilment of (D.22) and the satisfaction of the classical detailed balance conditions by the laser Fokker-Plank equation.

APPENDIX E

THE OPERATOR IDENTITIES OF CHAPTER V

In this appendix we provide the proof of the four results (5.1.11) to (5.1.14), these reading

$$\overset{(+)}{S}_{\lambda}(\omega_\nu)\rho_{ss} = \rho_{ss}\overset{(+)}{S}_{\lambda}(\omega_\nu)\exp(-\hbar\omega_\nu/kT) \tag{E.1}$$

$$\overset{(-)}{S}_{\lambda}(\omega_\nu)\rho_{ss} = \rho_{ss}\overset{(-)}{S}_{\lambda}(\omega_\nu)\exp(\hbar\omega_\nu/kT) \tag{E.2}$$

$$\overset{(+)\dagger}{S}_{\lambda}(\omega_\nu)\rho_{ss} = \rho_{ss}\overset{(+)\dagger}{S}_{\lambda}(\omega_\nu)\exp(\hbar\omega_\nu/kT) \tag{E.3}$$

$$\overset{(-)\dagger}{S}_{\lambda}(\omega_\nu)\rho_{ss} = \rho_{ss}\overset{(-)\dagger}{S}_{\lambda}(\omega_\nu)\exp(-\hbar\omega_\nu/kT) \tag{E.4}$$

For the first, from (5.1.1) and (5.1.7), we may write

$$\overset{(+)}{S}_{\lambda}(\omega_\nu)\rho_{ss} = \Big(\sum_{\substack{n,m\\E_n<E_m}} |E_n\rangle\langle E_m|\, S^{\lambda}_{n,m}\,\delta_{\omega_{n,m},\omega_\nu}\Big) \cdot\exp(-H_s/kT)/\mathrm{tr}_s\exp(-H_s/kT) \tag{E.5}$$

which, on the introduction of the product $\exp(-H_s/kT)\exp(H_s/kT)$ as unity, may be restated

$$\overset{(+)}{S}_{\lambda}(\omega_\nu)\rho_{ss} = \rho_{ss}\exp(H_s/kT)\Big(\sum_{\substack{n,m\\E_n<E_m}} |E_n\rangle\langle E_m|\, S^{\lambda}_{n,m}\,\delta_{\omega_{n,m},\omega_\nu}\Big)\exp(-H_s/kT) \tag{E.6}$$

Now $|E_n\rangle$ and $|E_m\rangle$ are eigenstates of the Hamiltonian H_s and thus

$$\overset{(+)}{S}_{\lambda}(\omega_\nu)\rho_{ss} = \rho_{ss}\Big(\sum_{\substack{n,m\\E_n<E_m}} |E_n\rangle\langle E_m|\, S^{\lambda}_{n,m}\,\delta_{\omega_{n,m},\omega_\nu} \cdot\exp[-(E_m-E_n)/kT]\Big) \tag{E.7}$$

Further, with the definition (5.1.9)

$$\overset{(+)}{S}_\lambda(\omega_\nu)\,\rho_{ss} = \rho_{ss}\Big(\sum_{\substack{n,m\\E_n<E_m}} |E_n\rangle\langle E_m|\,\overset{\lambda}{S}_{n,m}\,\delta_{\omega_{n,m},\omega_\mu}\Big)\cdot\exp(-\hbar\omega_\nu/kT)$$

$$= \rho_{ss}\,\overset{(+)}{S}_\lambda(\omega_\nu)\exp(-\hbar\omega_\nu/kT) \qquad \text{(E.8)}$$

In this our objective is fulfilled. The relationships (E.2) to (E.4) follow via analogous arguments.

APPENDIX F

DETAILED BALANCE CONDITIONS IN OPERATOR FORM

From section (4.1) the statement of quantum detailed balance is found in (4.1.15) and reads

$$\left\langle \overset{(\alpha)}{S}(\tau)\, \overset{(\beta)}{S}(0) \right\rangle_{\rho_{ss}} = \left\langle \overset{\widetilde{(\beta)}}{S}{}^{\dagger}(\tau)\, \overset{\widetilde{(\alpha)}}{S}{}^{\dagger}(0) \right\rangle_{\rho_{ss}} \tag{F.1}$$

where $S^{(\alpha)}$ and $S^{(\beta)}$ are arbitrary system operators. In this appendix we will show that equivalent to this is the requirement of the operator conditions

$$\rho_{ss} = \tilde{\rho}_{ss} \tag{F.2}$$

$$\rho_{ss} \bar{\mathcal{L}}\, S^{(\alpha)} = \tilde{\mathcal{L}} \rho_{ss}\, S^{(\alpha)} \tag{F.3}$$

Here the operators $\tilde{\mathcal{L}}$ and $\bar{\mathcal{L}}$ find definition in (4.2.11):

$$\widetilde{\mathcal{L} S^{(\alpha)}} = \tilde{\mathcal{L}}\, \widetilde{S^{(\alpha)}} \tag{F.4}$$

$$tr_s\, S^{(\alpha)} \mathcal{L} S^{(\beta)} = tr_s\, S^{(\beta)} \bar{\mathcal{L}}\, S^{(\alpha)} \tag{F.5}$$

We begin with the expression for two-time averages given in (2.1.16), with this we may write (F.1) in the form

$$tr_s[\exp(\mathcal{L}\tau)\, \overset{(\beta)}{S}(0)\rho_{ss}]\, \overset{(\alpha)}{S}(0) = tr_s[\exp(\mathcal{L}\tau)\, \overset{\widetilde{(\alpha)}}{S}{}^{\dagger}(0)\rho_{ss}]\, \overset{\widetilde{(\beta)}}{S}{}^{\dagger}(0) \tag{F.6}$$

and, from (4.1.11) and (F.4)

$$tr_s[\exp(\mathcal{L}\tau)\, \overset{(\beta)}{S}(0)\, \rho_{ss}]\, \overset{(\alpha)}{S}(0) = tr_s[\exp(\tilde{\mathcal{L}}\tau)\, \tilde{\rho}_{ss}\, \overset{(\alpha)}{S}(0)]\, \overset{(\beta)}{S}(0) \tag{F.7}$$

Then, with (F.5) and the cyclic property of the trace, we have

$$\mathrm{tr}_s\,\rho_{ss}[\exp(\bar{\mathcal{L}}\tau)\overset{(\alpha)}{S}(0)]\overset{(\beta)}{S}(0) = \mathrm{tr}_s[\exp(\tilde{\mathcal{L}}\tau)\tilde{\rho}_{ss}\overset{(\alpha)}{S}(0)]\overset{(\beta)}{S}(0) \qquad \text{(F.8)}$$

Now, since $S^{(\beta)}(0)$ is arbitrary we may choose for it any dyadic and replace (F.8) by the operator requirement

$$\rho_{ss}\exp(\bar{\mathcal{L}}\tau)\overset{(\alpha)}{S}(0) = \exp(\tilde{\mathcal{L}}\tau)\tilde{\rho}_{ss}\overset{(\alpha)}{S}(0) \qquad \text{(F.9)}$$

Clearly, then, necessary and sufficient conditions are

$$\rho_{ss} = \tilde{\rho}_{ss} \qquad \text{(F.10)}$$

$$\rho_{ss}\bar{\mathcal{L}}^{n}\overset{(\alpha)}{S}(0) = \tilde{\mathcal{L}}^{n}\tilde{\rho}_{ss}\overset{(\alpha)}{S}(0) \qquad \text{(F.11)}$$

where n is any positive integer. It remains only to show by induction that these may be equivalently replaced by the simple first order conditions (F.2), (F.3). For this, we adopt (F.11) by hypothesis and take

$$\rho_{ss}\bar{\mathcal{L}}^{n+1}\overset{(\alpha)}{S}(0) = \rho_{ss}\bar{\mathcal{L}}[\bar{\mathcal{L}}^{n}\overset{(\alpha)}{S}(0)] \qquad \text{(F.12)}$$

Using (F.3) this yields

$$\begin{aligned}\rho_{ss}\bar{\mathcal{L}}^{n+1}\overset{(\alpha)}{S}(0) &= \tilde{\mathcal{L}}\rho_{ss}[\bar{\mathcal{L}}^{n}\overset{(\alpha)}{S}(0)]\\ &= \tilde{\mathcal{L}}[\rho_{ss}\bar{\mathcal{L}}^{n}\overset{(\alpha)}{S}(0)]\end{aligned}$$

and then, invoking (F.10)

$$\begin{aligned}\rho_{ss}\bar{\mathcal{L}}^{n+1}\overset{(\alpha)}{S}(0) &= \tilde{\mathcal{L}}[\tilde{\mathcal{L}}^{n}\rho_{ss}\overset{(\alpha)}{S}(0)]\\ &= \tilde{\mathcal{L}}^{n+1}\rho_{ss}\overset{(\alpha)}{S}(0)\end{aligned} \qquad \text{(F.13)}$$

It follows that (F.3) ensures all higher orders, and hence (F.2) and (F.3) become necessary and sufficient conditions for the fulfilment of (F.1).

In this way they qualify as the detailed balance conditions in operator form.

APPENDIX G

ENERGY REPRESENTATION FOR THE COUPLED ATOM-FIELD MODE SYSTEM

Here we evaluate the energy eigenvalues and eigenstates for the system comprising a two-level atom coupled at resonance to a single mode of the electromagnetic field. The Hamiltonian H_S, as given by (6.2.5), has

$$H_s = \hbar\omega_0 a^\dagger a + (1/2)\hbar\omega_0\sigma_z + \hbar(\chi_0^* a^\dagger \sigma_- + \chi_0 a \sigma_+) \tag{G.1}$$

As a complete set of states we may start with $|n,+\rangle$ and $|n+1,-\rangle$, where $|n\rangle$ are the usual Fock states;

$$\hbar\omega_0 a^\dagger a|n\rangle = n\hbar\omega_0|n\rangle \tag{G.2}$$

and $|+\rangle$ and $|-\rangle$ represent upper and lower atomic levels respectively:

$$(1/2)\hbar\omega_0\sigma_z|\pm\rangle = \pm(1/2)\hbar\omega_0|\pm\rangle \tag{G.3}$$

Together these constitute the energy eigenstates for free dynamics. On introduction of atom-field interaction, from (G.2) and (G.3) we move to the coupled equations

$$H_s\begin{pmatrix}|n,+\rangle \\ |n+1,-\rangle\end{pmatrix} = \hbar\begin{pmatrix}\omega_0(n+1/2) & (n+1)^{1/2}\chi_0^* \\ (n+1)^{1/2}\chi_0 & \omega_0(n+1/2)\end{pmatrix}\begin{pmatrix}|n,+\rangle \\ |n+1,-\rangle\end{pmatrix} \tag{G.4}$$

or

$$H_s\begin{pmatrix}|n,+\rangle \\ |n+1,-\rangle\end{pmatrix} = M\begin{pmatrix}|n,+\rangle \\ |n+1,-\rangle\end{pmatrix} \tag{G.5}$$

where the matrix M is given by

$$M = \hbar \begin{pmatrix} \omega_0(n+1/2) & (n+1)^{1/2} K_0^* \\ (n+1)^{1/2} K_0 & \omega_0(n+1/2) \end{pmatrix} \tag{G.6}$$

Now M May be diagonalised by a collineatory transformation (Eisenschitz, 1966)

$$S^{-1} M S = \Lambda \tag{G.7}$$

where Λ is a diagonal matrix whose diagonal elements are the eigenvalues λ_1 and λ_2 of M and S is a 2 x 2 matrix whose columns form the corresponding eigenvectors $\vec{S}_1$ and $\vec{S}_2$ of M. Achieving such a diagonalisation (G.5) may be written

$$H_s \begin{pmatrix} |E_n^2\rangle \\ |E_n^1\rangle \end{pmatrix} = \Lambda \begin{pmatrix} |E_n^2\rangle \\ |E_n^1\rangle \end{pmatrix} \tag{G.8}$$

where

$$\begin{pmatrix} |E_n^2\rangle \\ |E_n^1\rangle \end{pmatrix} = S^{-1} \begin{pmatrix} |n,+\rangle \\ |n+1,-\rangle \end{pmatrix} \tag{G.9}$$

will be energy eigenstates for H_S with respective eigenvalues

$$E_n^{2,1} = \lambda_{1,2} \tag{G.10}$$

The eigenvalues λ_1 and λ_2 are to be found by solution of the characteristic equation

$$\det(M - \lambda I_2) = 0 \tag{G.11}$$

I_2 being the 2 x 2 identity matrix. Thus, from (G.6) we have

$$[\hbar\omega_0(n+1/2) - \lambda]^2 - \hbar^2(n+1)|K_0|^2 = 0 \tag{G.12}$$

whence

$$E_n^{2,1} = \lambda_{1,2} = \hbar\left[\omega_0(n+1/2) \pm (n+1)^{1/2}|K_0|\right] \tag{G.13}$$

Writing then

$$S = \begin{pmatrix} S_{11} & S_{12} \\ S_{21} & S_{22} \end{pmatrix} \tag{G.14}$$

we have

$$M\begin{pmatrix} S_{11} \\ S_{21} \end{pmatrix} = \lambda_1 \begin{pmatrix} S_{11} \\ S_{21} \end{pmatrix} \tag{G.15}$$

and

$$M\begin{pmatrix} S_{12} \\ S_{22} \end{pmatrix} = \lambda_2 \begin{pmatrix} S_{12} \\ S_{22} \end{pmatrix} \tag{G.16}$$

which, from (G.6) and G.13) yields

$$\begin{aligned} S_{21} &= S_{11} \\ S_{12} &= -S_{22} \end{aligned} \tag{G.17}$$

and we write

$$S = \begin{pmatrix} S_{11} & -S_{22} \\ S_{11} & S_{22} \end{pmatrix} \tag{G.18}$$

It then remains only to invert S;

$$S^{-1} = (1/\sqrt{2})\begin{pmatrix} 1/S_{11} & 1/S_{11} \\ -1/S_{22} & 1/S_{22} \end{pmatrix} \tag{G.19}$$

and with (G.9)

$$\begin{aligned} |E_n^2\rangle &= (1/\sqrt{2})(|n,+\rangle + |n+1,-\rangle) \\ |E_n^1\rangle &= (1/\sqrt{2})(|n,+\rangle - |n+1,-\rangle) \end{aligned} \tag{G.20}$$

where S_{11} and S_{22} are chosen for normalisation to unity. In (G.13) and (G.20) we find fulfilment of our objective.

APPENDIX H

SOLUTION OF MATRIX EQUATIONS FOR RESONANCE FLUORESCENCE

In the text we have developed a formalism permitting solution for atomic matrix elements, the semiclassical scattered field, and first- and second-order correlation functions by way of a single set of four coupled equations. Here we will approach the solution of these equations, working specifically with the atomic matrix elements $\rho_{\eta,\xi}$ of (7.1.5).

From section 7.1, in resonance fluorescence the master equation for atomic dynamics may be written as a set of four coupled equations, these reading

$$\frac{d}{dt}\begin{pmatrix}\rho_{22}\\ \rho_{11}\\ \rho_{21}\\ \rho_{12}\end{pmatrix} = \begin{pmatrix} -\gamma/4 & \gamma/4 & 0 & 0\\ \gamma/4 & -\gamma/4 & 0 & 0\\ -\gamma/2 & -\gamma/2 & -(3\gamma/4+2i\bar{n}^{1/2}|\mathcal{K}_0|) & -\gamma/4\\ -\gamma/2 & -\gamma/2 & -\gamma/2 & -(3\gamma/4-2i\bar{n}^{1/2}|\mathcal{K}_0|)\end{pmatrix}\begin{pmatrix}\rho_{22}\\ \rho_{11}\\ \rho_{21}\\ \rho_{12}\end{pmatrix} \tag{H.1}$$

For their solution it is convenient that we define the vector $\vec{\rho}$ by

$$\vec{\rho} = (1/2)\begin{pmatrix}\rho_{22}+\rho_{11}\\ \rho_{22}-\rho_{11}\\ \rho_{21}+\rho_{12}\\ \rho_{21}-\rho_{12}\end{pmatrix} \tag{H.2}$$

and then write

$$\frac{d\vec{\rho}}{dt} = M\vec{\rho} \tag{H.3}$$

where the matrix M reads

$$M = \begin{pmatrix} 0 & 0 & 0 & 0 \\ 0 & -\gamma/2 & 0 & 0 \\ -\gamma & 0 & -\gamma & -2i\bar{n}^{1/2}|K_0| \\ 0 & 0 & -2i\bar{n}^{1/2}|K_0| & -\gamma/2 \end{pmatrix} \tag{H.4}$$

Now the solution of (H.3) may be achieved via the diagonalisation of M using the usual collineatory transformation (Eisenschitz, 1966). With this, for some matrix S, to be determined, we have

$$S^{-1} M S = \Lambda \tag{H.5}$$

where Λ is a diagonal with diagonal elements λ_i, i = 1 to 4, given by the characteristic equation

$$\det(M - \lambda I_4) = 0 \tag{H.6}$$

I_4 is the 4 x 4 identity matrix. The columns $\vec{s}_i$, i = 1 to 4, are then the eigenvectors of M corresponding to respective eigenvalues λ_i and

$$M\vec{s}_i = \lambda_i \vec{s}_i \tag{H.7}$$

Having this scheme $\vec{\sigma}$ is defined by

$$\vec{\sigma} = S^{-1}\vec{\rho} \tag{H.8}$$

and the matrix equation (H.3) written

$$\frac{d\vec{\sigma}}{dt} = \Lambda\vec{\sigma} \tag{H.9}$$

Thus we have recourse to the simple solution

$$\vec{\sigma}(t) = \exp(\Lambda t)\vec{\sigma}(0) \tag{H.10}$$

which, with inversion of (H.8), yields

$$\vec{\rho}(t) = S \exp(\Lambda t) S^{-1} \vec{\rho}(0) \tag{H.11}$$

It remains to solve (H.6) and (H.7) for the eigenvalues λ_i and eigenvectors $\vec{s}_i$ of M, and taking first the characteristic equation (H.6), it readily follows from (H.4) that this admits the form

$$\lambda(\lambda + \gamma/2)(\lambda^2 + \lambda 3\gamma/2 + \gamma^2/2 + 4\bar{n}|K_0|^2) = 0 \tag{H.12}$$

Thus, solving the quadratic,

$$\Lambda = \begin{pmatrix} 0 & 0 & 0 & 0 \\ 0 & -\gamma/2 & 0 & 0 \\ 0 & 0 & -3\gamma/4 + \Omega & 0 \\ 0 & 0 & 0 & -3\gamma/4 - \Omega \end{pmatrix} \tag{H.13}$$

where for Ω we have

$$\Omega = [(\gamma/4)^2 - 4\bar{n}|K_0|^2]^{1/2} \tag{H.14}$$

Turning then to (H.7) we may write

$$M \begin{pmatrix} S_{11} \\ S_{21} \\ S_{31} \\ S_{41} \end{pmatrix} = 0 \tag{H.15}$$

$$M \begin{pmatrix} S_{12} \\ S_{22} \\ S_{32} \\ S_{42} \end{pmatrix} = -\gamma/2 \begin{pmatrix} S_{12} \\ S_{22} \\ S_{32} \\ S_{42} \end{pmatrix} \tag{H.16}$$

$$M\begin{pmatrix} S_{13} \\ S_{23} \\ S_{33} \\ S_{43} \end{pmatrix} = -(3\gamma/4 - \Omega)\begin{pmatrix} S_{13} \\ S_{23} \\ S_{33} \\ S_{43} \end{pmatrix} \tag{H.17}$$

and

$$M\begin{pmatrix} S_{14} \\ S_{24} \\ S_{34} \\ S_{44} \end{pmatrix} = -(3\gamma/4 + \Omega)\begin{pmatrix} S_{14} \\ S_{24} \\ S_{34} \\ S_{44} \end{pmatrix} \tag{H.18}$$

and from these we find

$$\vec{S}_1 = \begin{pmatrix} S_1 \\ 0 \\ -(\gamma^2/\gamma^2 + 8\bar{n}|K_0|^2)S_1 \\ (2i\bar{n}^{1/2}|K_0|/\tfrac{1}{2}\gamma)(\gamma^2/\gamma^2 + 8\bar{n}|K_0|^2)S_1 \end{pmatrix} \tag{H.19}$$

$$\vec{S}_2 = \begin{pmatrix} 0 \\ S_2 \\ 0 \\ 0 \end{pmatrix} \tag{H.20}$$

$$\vec{S}_3 = \begin{pmatrix} 0 \\ 0 \\ S_3 \\ (2i\bar{n}^{1/2}|K_0|/\tfrac{1}{4}\gamma - \Omega)S_3 \end{pmatrix} \tag{H.21}$$

$$\vec{S}_4 = \begin{pmatrix} 0 \\ 0 \\ -(2i\bar{n}^{1/2}|K_0|/\frac{1}{4}\gamma - \Omega)\, S_4 \\ S_4 \end{pmatrix} \tag{H.22}$$

where S_i, i = 1 to 4, are arbitrary constants. We may therefore write

$$S = \begin{pmatrix} S_1 & 0 & 0 & 0 \\ 0 & S_2 & 0 & 0 \\ -(\gamma^2/\gamma^2 + 8\bar{n}|K_0|^2)S_1 & 0 & S_3 & -(2i\bar{n}^{1/2}|K_0|/\frac{1}{4}\gamma - \Omega)S_4 \\ (2i\bar{n}^{1/2}|K_0|/\frac{1}{2}\gamma)(\gamma^2/\gamma^2 + 8\bar{n}|K_0|^2)S_1 & 0 & (2i\bar{n}^{1/2}|K_0|/\frac{1}{4}\gamma - \Omega)S_3 & S_4 \end{pmatrix} \tag{H.23}$$

and after rather tedious algebra

$$S^{-1} = \begin{pmatrix} 1/S_1 & 0 \\ 0 & 1/S_2 \\ -\frac{1}{2}\gamma/\Omega \cdot 1/S_3 & 0 \\ (\frac{1}{2}\gamma/2i\bar{n}^{1/2}|K_0|)(\frac{1}{4}\gamma - \Omega)/\Omega \cdot 1/S_4 & 0 \end{pmatrix}$$

$$
\left.\begin{matrix}
0 & 0 \\
0 & 0 \\
-1/2\cdot\left(\frac{1}{4}\gamma-\Omega\right)/\Omega\cdot 1/S_3 & 1/2\cdot 2i\bar{n}^{1/2}|\kappa_0|/\Omega\cdot 1/S_3 \\
-1/2\cdot 2i\bar{n}^{1/2}|\kappa_0|/\Omega\cdot 1/S_4 & -1/2\cdot\left(\frac{1}{4}\gamma-\Omega\right)/\Omega\cdot 1/S_4
\end{matrix}\right) \qquad \text{(H.24)}
$$

Solutions for atomic matrix elements with arbitrary initial conditions are then available from (H.11) with (H.13), (H.23) and (H.24).

APPENDIX I

TRANSIENT SPECTRAL FEATURES

We present here the full form for the term $I^{'}(\omega,\vec{r},T)$ in (7.3.28). From (7.3.2) we write

$$I^{'}(\omega,\vec{r},T) = (1/2\pi)\left(I(\vec{r},T)\Big/\int_{r/c}^{T} dt\, I(\vec{r},t)\right) P^{'}(\omega,\vec{r},T) \qquad \text{(I.1)}$$

with the intensity and integrated intensity given by (7.3.21) and (7.3.22) respectively. $P^{'}(\omega,\vec{r},T)$ is conveniently written as the sum of four terms

$$P^{'}(\omega,\vec{r},T) = -(1/2)\, I_0(\vec{r}) \sum_{k=1}^{4} \mathcal{P}_k(\omega,T) \qquad \text{(I.2)}$$

We have

$$\begin{aligned}
\mathcal{P}_1(\omega,T) = (1/2)&\left[\frac{\omega-\omega_{21}}{(\frac{3}{4}\gamma)^2+(\omega-\omega_{21})^2} - \frac{\omega-\omega_{12}}{(\frac{3}{4}\gamma)^2+(\omega-\omega_{12})^2}\right]\frac{2n^{1/2}|X_0|}{(\frac{3}{4}\gamma)^2+4n|X_0|^2} \\
&\cdot\left(1-e^{-\frac{3}{4}\gamma\hat{T}}\cos 2n^{1/2}|X_0|\hat{T}\right) \\
-(1/2)&\left[\frac{\omega-\omega_{21}}{(\frac{3}{4}\gamma)^2+(\omega-\omega_{21})^2} - \frac{\omega-\omega_{12}}{(\frac{3}{4}\gamma)^2+(\omega-\omega_{12})^2}\right]\frac{\frac{3}{4}\gamma}{(\frac{3}{4}\gamma)^2+4n|X_0|^2} \\
&\cdot e^{-\frac{3}{4}\gamma\hat{T}}\sin 2n^{1/2}|X_0|\hat{T}
\end{aligned} \qquad \text{(I.3)}$$

$$\begin{aligned}
\mathcal{P}_2(\omega,T) = &\left[\frac{\frac{1}{2}\gamma}{(\frac{1}{2}\gamma)^2+(\omega-\omega_0)^2}\frac{\frac{1}{2}\gamma}{(\frac{1}{2}\gamma)^2+(\omega-\omega_0)^2} - \frac{(\omega-\omega_0)}{(\frac{1}{2}\gamma)^2+(\omega-\omega_0)^2}\frac{(\omega-\omega_0)}{(\frac{1}{2}\gamma)^2+(\omega-\omega_0)^2}\right] \\
&\cdot\left(1-e^{-\frac{1}{2}\gamma\hat{T}}\cos(\omega-\omega_0)\hat{T}\right) \\
+&\left[\frac{\frac{1}{2}\gamma}{(\frac{1}{2}\gamma)^2+(\omega-\omega_0)^2}\frac{(\omega-\omega_0)}{(\frac{1}{2}\gamma)^2+(\omega-\omega_0)^2} + \frac{\frac{1}{2}\gamma}{(\frac{1}{2}\gamma)^2+(\omega-\omega_0)^2}\frac{(\omega-\omega_0)}{(\frac{1}{2}\gamma)^2+(\omega-\omega_0)^2}\right] \\
&\cdot e^{-\frac{1}{2}\gamma\hat{T}}\sin(\omega-\omega_0)\hat{T}
\end{aligned}$$

$$
\begin{aligned}
&+(1/2)\left[\frac{\frac{3}{2}\gamma}{(\frac{3}{2}\gamma)^2+(\omega-\omega_0)^2}\,\frac{\frac{3}{4}\gamma}{(\frac{1}{2}\gamma)^2+(\omega-\omega_0)^2}-\frac{(\omega-\omega_{21})}{(\frac{3}{4}\gamma)^2+(\omega-\omega_{21})^2}\,\frac{(\omega-\omega_0)}{(\frac{3}{2}\gamma)^2+(\omega-\omega_0)^2}\right]\\
&\qquad\cdot\left(1-e^{-\frac{3}{2}\gamma\hat{T}}\operatorname{Cos}(\omega-\omega_0)\hat{T}\right)\\
&+(1/2)\left[\frac{\frac{3}{2}\gamma}{(\frac{3}{2}\gamma)^2+(\omega-\omega_0)^2}\,\frac{(\omega-\omega_{21})}{(\frac{3}{4}\gamma)^2+(\omega-\omega_{21})^2}+\frac{\frac{3}{4}\gamma}{(\frac{3}{4}\gamma)^2+(\omega-\omega_{21})^2}\,\frac{(\omega-\omega_0)}{(\frac{3}{2}\gamma)^2+(\omega-\omega_0)^2}\right]\\
&\qquad\cdot e^{-\frac{3}{2}\gamma\hat{T}}\operatorname{Sin}(\omega-\omega_0)\hat{T}\\
&+(1/2)\left[\frac{\frac{3}{2}\gamma}{(\frac{3}{2}\gamma)^2+(\omega-\omega_0)^2}\,\frac{\frac{3}{4}\gamma}{(\frac{3}{4}\gamma)^2+(\omega-\omega_{12})^2}-\frac{(\omega-\omega_{12})}{(\frac{3}{4}\gamma)^2+(\omega-\omega_{12})^2}\,\frac{(\omega-\omega_0)}{(\frac{3}{2}\gamma)^2+(\omega-\omega_0)^2}\right]\\
&\qquad\cdot\left(1-e^{-\frac{3}{2}\gamma\hat{T}}\operatorname{Cos}(\omega-\omega_0)\hat{T}\right)\\
&+(1/2)\left[\frac{\frac{3}{2}\gamma}{(\frac{3}{2}\gamma)^2+(\omega-\omega_0)^2}\,\frac{(\omega-\omega_{12})}{(\frac{3}{4}\gamma)^2+(\omega-\omega_{12})^2}+\frac{\frac{3}{4}\gamma}{(\frac{3}{4}\gamma)^2+(\omega-\omega_{12})^2}\,\frac{(\omega-\omega_0)}{(\frac{3}{2}\gamma)^2+(\omega-\omega_0)^2}\right]\\
&\qquad\cdot e^{-\frac{3}{2}\gamma\hat{T}}\operatorname{Sin}(\omega-\omega_0)\hat{T}
\end{aligned}
\tag{I.4}
$$

$$
\begin{aligned}
\mathcal{P}_3(\omega,T)=&\,(1/2)\left[\frac{\frac{3}{4}\gamma}{(\frac{3}{4}\gamma)^2+(\omega-\omega_{21})^2}\,\frac{\frac{3}{4}\gamma}{(\frac{3}{4}\gamma)^2+(\omega-\omega_{21})^2}-\frac{(\omega-\omega_{21})}{(\frac{3}{4}\gamma)^2+(\omega-\omega_{21})^2}\,\frac{(\omega-\omega_{21})}{(\frac{3}{4}\gamma)^2+(\omega-\omega_{21})^2}\right]\\
&\qquad\cdot\left(1-e^{-\frac{3}{4}\gamma\hat{T}}\operatorname{Cos}(\omega-\omega_{21})\hat{T}\right)\\
&+(1/2)\left[\frac{\frac{3}{4}\gamma}{(\frac{3}{4}\gamma)^2+(\omega-\omega_{21})^2}\,\frac{(\omega-\omega_{21})}{(\frac{3}{4}\gamma)^2+(\omega-\omega_{21})^2}+\frac{\frac{3}{4}\gamma}{(\frac{3}{4}\gamma)^2+(\omega-\omega_{21})^2}\,\frac{(\omega-\omega_{21})}{(\frac{3}{4}\gamma)^2+(\omega-\omega_{21})^2}\right]\\
&\qquad\cdot e^{-\frac{3}{4}\gamma\hat{T}}\operatorname{Sin}(\omega-\omega_{21})\hat{T}\\
&+(1/2)\left[\frac{\frac{1}{2}\gamma}{(\frac{1}{2}\gamma)^2+(\omega-\omega_0)^2}\,\frac{\frac{5}{4}\gamma}{(\frac{5}{4}\gamma)^2+(\omega-\omega_{21})^2}-\frac{(\omega-\omega_{21})}{(\frac{5}{4}\gamma)^2+(\omega-\omega_{21})^2}\,\frac{(\omega-\omega_0)}{(\frac{1}{2}\gamma)^2+(\omega-\omega_0)^2}\right]\\
&\qquad\cdot\left(1-e^{-\frac{5}{4}\gamma\hat{T}}\operatorname{Cos}(\omega-\omega_{21})\hat{T}\right)\\
&+(1/2)\left[\frac{\frac{1}{2}\gamma}{(\frac{1}{2}\gamma)^2+(\omega-\omega_0)^2}\,\frac{(\omega-\omega_{21})}{(\frac{5}{4}\gamma)^2+(\omega-\omega_{21})^2}+\frac{\frac{5}{4}\gamma}{(\frac{5}{4}\gamma)^2+(\omega-\omega_{21})^2}\,\frac{(\omega-\omega_0)}{(\frac{1}{2}\gamma)^2+(\omega-\omega_0)^2}\right]\\
&\qquad\cdot e^{-\frac{5}{4}\gamma\hat{T}}\operatorname{Sin}(\omega-\omega_{21})\hat{T}
\end{aligned}
\tag{I.5}
$$

$$
\begin{aligned}
\mathcal{P}_4(\omega,T) = (1/2)\Bigg[& \frac{\frac{3}{4}\gamma}{(\frac{3}{4}\gamma)^2+(\omega-\omega_{12})^2}\cdot\frac{\frac{3}{4}\gamma}{(\frac{3}{4}\gamma)^2+(\omega-\omega_{12})^2} - \frac{(\omega-\omega_{12})}{(\frac{3}{4}\gamma)^2+(\omega-\omega_{12})^2}\,\frac{(\omega-\omega_{12})}{(\frac{3}{4}\gamma)^2+(\omega-\omega_{12})^2}\Bigg] \\
& \cdot\left(1-e^{-\frac{3}{4}\gamma\hat{T}}\,\mathrm{Cos}(\omega-\omega_{12})\hat{T}\right) \\
+(1/2)\Bigg[& \frac{\frac{3}{4}\gamma}{(\frac{3}{4}\gamma)^2+(\omega-\omega_{12})^2}\cdot\frac{(\omega-\omega_{12})}{(\frac{3}{4}\gamma)^2+(\omega-\omega_{12})^2} + \frac{\frac{3}{4}\gamma}{(\frac{3}{4}\gamma)^2+(\omega-\omega_{12})^2}\cdot\frac{(\omega-\omega_{12})}{(\frac{3}{4}\gamma)^2+(\omega-\omega_{12})^2}\Bigg] \\
& \cdot e^{-\frac{3}{4}\gamma\hat{T}}\,\mathrm{Sin}(\omega-\omega_{12})\hat{T} \\
+(1/2)\Bigg[& \frac{\frac{1}{2}\gamma}{(\frac{1}{2}\gamma)^2+(\omega-\omega_0)^2}\cdot\frac{\frac{5}{4}\gamma}{(\frac{5}{4}\gamma)^2+(\omega-\omega_{12})^2} - \frac{(\omega-\omega_{12})}{(\frac{5}{4}\gamma)^2+(\omega-\omega_{12})^2}\,\frac{(\omega-\omega_0)}{(\frac{1}{2}\gamma)^2+(\omega-\omega_0)^2}\Bigg] \\
& \cdot\left(1-e^{-\frac{5}{4}\gamma\hat{T}}\,\mathrm{Cos}(\omega-\omega_{12})\hat{T}\right) \\
+(1/2)\Bigg[& \frac{\frac{1}{2}\gamma}{(\frac{1}{2}\gamma)^2+(\omega-\omega_0)^2}\cdot\frac{(\omega-\omega_{12})}{(\frac{5}{4}\gamma)^2+(\omega-\omega_{12})^2} + \frac{\frac{5}{4}\gamma}{(\frac{5}{4}\gamma)^2+(\omega-\omega_{21})^2}\cdot\frac{(\omega-\omega_0)}{(\frac{1}{2}\gamma)^2+(\omega-\omega_0)^2}\Bigg] \\
& \cdot e^{-\frac{5}{4}\gamma\hat{T}}\,\mathrm{Sin}(\omega-\omega_{12})\hat{T} \qquad \text{(I.6)}
\end{aligned}
$$

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